Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2019-592-RC2, 2019

Atmospheric Chemistry and Physics Discussions

Interactive comment on "Simulating age of air and distribution of SF_6 in the stratosphere with SILAM model" *by* Rostislav Kouznetsov et al.

Anonymous Referee #2

Received and published: 8 August 2019

The study by Kouznetsov et al investigates the impact of the vertical diffusion and of the mesospheric sink of SF6 and the SF6 climatology and its trends using a chemistry-transport model. While the mesospheric transport is not explicitly included (due to lack of ERA-Interim data above 0.1 hPa), a parametrization of eddy diffusivity as well as molecular diffusivity is included to mimic transport to the mesosphere. The subject of the study is of high relevance, as SF6 is used frequently to estimate Age-of-Air, and the role of its sinks needs to be better understood. The study is overall well presented and the methods are overall appropriate, but some clarifications are needed (see comments below). Overall, I recommend the authors revise the paper minorly before it can be considered for publication.

General comments:

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1. In lines 45 ff, you correctly mention that a correction has to be applied when deriving AoA from a non-linear increasing tracer, as SF6, as has been done by observational studies. However, it is not entirely clear to me how you calculated AoA from SF6 - simply as time lag, as for the linear increasing tracer? It certainly is known that just calculating the time lag leads to deviations from the true AoA values. If you choose not to include a correction method in the calculation of AoA, you certainly should stress this fact, and I suggest you to refer to the SF6-derived "AoA" as "time lag" rather than AoA. The comparison of the SF6-derived time-lag with / without chemical sink is still valid, but I caution you on the conclusions you draw from the difference of the passive sf6 tracer and the ideal age /linearly increasing tracer: as long as no correction method for the non-linearity is implied, you cannot conclude on whether the non-linear increasing tracer can be used to deduce AoA values in general.

2. While the parametrizations of eddy diffusivity, gravitational separation by molecular diffusion and of SF6 loss are described well in detail, the way they are actually implemented in the model is not entirely clear to me. According to Section 3.4, the overall budget equation of the abundance of a tracer (SF6) is solved for steady state, and this steady state solution scaled by the actual tracer concentration is used above the model top - is this correct? And how exactly is this implemented - as loss due to the lifetime given in line 238 ? Furthermore, it was not clear to me whether the diffusive parametrizations are also applied in the actual model domain, or only for the parametrization above the top level? It could be helpful if you describe the overall approach at the beginning of section 3 (i.e. parametrization of upward transport above $\sim$10Pa by vertical diffusion, where SF6 is depleted, and thus there is no downward transport of SF6).

3. Related to the above comment, I wonder how sensitive your results are to the fact that you represent transport above the model top only as vertical diffusive process, i.e. the actual transport circulation is missing (which circulates air, and thus SF6 from pole-to-pole, as opposed to your assumption of all SF6 that is transported diffusively

upward being lost). Probably the lack of advective transport also affects the results of the evaluation of different values for Kz? Or is this more based on the layers within the model domain (if diffusion is applied there too, see comment above)? Please add discussion of those issues to your study.

Specific comments:

- line 25: you describe here the estimation of AoA with Lagrangian trajectories, but without inter-parcel mixing. The inter-parcel mixing does affect AoA, and there are studies that account for this mixing in Lagrangian frameworks (e.g. Brinkop et al., 2019, Plöger et al., 2015). Thus estimates of AoA with Eularian methods might differ from Lagrangian methods due to the way inter-parcel mixing is calulated. This methodological point should be mentioned somewhere.

(References: Brinkop and Jöckel, ATTILA 4.0: Lagrangian advective and convective transport of passive tracers within the ECHAM5/MESSy (2.53.0) chemistry–climate model, Geosci. Model Dev., 12, 1991–2008, https://doi.org/10.5194/gmd-12-1991-2019, 2019

Ploeger, F.; Riese, M.; Haenel, F.; Konopka, P.; Müller, R. & Stiller, G. Variability of stratospheric mean age of air and of the local effects of residual circulation and eddy mixing Journal of Geophysical Research: Atmospheres, 2015, 120, 716-733)

- line 27: "above-mentioned observational method": I dont see you mention the observational method above this statement?

- line 44: Garcia et al did show that the corrections improve the trend estimate, and they do not use the exact same correction method than what was applied to the observations. So I would not argue that the tracers are "ambiguous proxies" for AoA, but rather that the correction methods accounting for the non-linearity need to be investigated more deeply.

- line 95 ff: Maybe you can mention here which variables from ERA-Interim you use - I

was wondering at this point how vertical transport is calculated, and this became clear only in section 3.5.

- line 122 ff, general: How certain are the SF6 destruction rates, i.e. how do the results by Totterdill et al compare to other studies? Please add a short statement.

- line 156: its not clear to me what the "limiting value" is, and why Kz is "practically always" set to it? Please be more specific here.

- line 159: Kz does not fall below the molecular diffusivity is the lower stratosphere, below ∼40 km, according to Fig. 2, so please refine the statement.

- line 196: do you mean mixing ratio differences between the two layers? Why two layers, and not at one layer? Or do you mean the mean mixing ration in the layer bounded by an upper and lower pressure? it might be easier to put down the equation rather than describing it.

- line 212: I assume you use the US standard atmosphere because at the levels where it matters, ERA-Interim is not available any more? Again, it is not entirely clear if / how you apply this parametrization only at the "top layer", or also throughout the model domain. If the latter is true, the actual ERA-Interim temperatures could be used in the model domain (even tough you could argue that it does not make much of a difference there, as molecular diffusion does not play a role).

- line 224: please be more specific and describe how you obtain the flux F(p) from the steady-state solution of the mixing ratios.

- line 236: see general comment: please be more specific on how exactly the different parametrizations are used in the different areas, and how the upper boundary parametrization is implemented (via the lifetimes?)

- line 250: which "other parameters" do you use?

- line 267: were the other tracers corrected using the "ones" tracer, or just the error

"evaluated"?

- line 343 ff: is this the best way to estimate lifetimes, or couldn't you just average the inverse destruction rate mass-weighted over the entire atmosphere? Also, at line 348, you write that the delay of SF6 between troposphere and upper layers is about 5-6 years, and then use the value 5 years previous to the emission stop to evaluate the lifetime - is this quantitative, or just a rough estimate?

- line 361: "we have found in literature"-> be more specific, e.g. observations that were published by ...

- line 365: "strong exchange through the troposphere"? do you mean too strong upward transport by the diffusion?

- line 384: what is the dynamical reason for the minimum in SF6, and why do you think it is weaker in the model?

- line 464: "practically"? please be more specific

- line 471 ff: as mentioned in the general comment, you should clarify how AoA was calculated from the SF6 tracers, and possibly change the naming to "time-lag".

- line 486: you mentioned earlier that you use a new version of the MIPAS SF6 data, but do not show its AoA, but instead refer to the older published AoA figures. Why don't you add the new MIPAS AoA to Figs. 11 and 12?

- line 515: "non-uniformity" of ERA-Interim, what do you mean? Couldn't this just be the trend in AoA over the period, or why do you think it is an artefact? Further, in line 519, you state that ERA-Interim was not recommended for climatological studies. I'm surprised by this statement, given that ERA-Interim is the basis for a lot of studies of climatologies and trends in various variables. Can you specify which source you quote here, and what exactly should not be done?

- line 521: The trends over the MIPAs period could be compared to other CTM results,

e.g. by Plöger et al, 2015b, who showed that their CTM was capable to reproduce the MIPAS AoA trend rather well.

(Ploeger, F.; Abalos, M.; Birner, T.; Konopka, P.; Legras, B.; Müller, R. & Riese, M. Quantifying the effects of mixing and residual circulation on trends of stratospheric mean age of air Geophysical Research Letters, 2015, 42, 2047-2054)

- line 525: why comparable with Lagrangian simulations? As pointed out before, one difference is the accounting for inter-parcel mixing. I'd rather argue that your results are comparable to other state-of-the-art CTM simulations of AoA.

- line 542: Are those "best" estimates in the upper stratosphere based on the "upper layer", where advective transport is not accounted for? Or dou you refer to the results in the model domain?

-line 549: I dont understand the sentence on the standard deviation controlled by noise. Do you mean to say that the standard deviation between model and MIPAS is about as large as the error on the satellite data?

- line 551: you might want to add the range of lifetimes you obtain.

- line 560: as stated in the general comments, as long as you do not apply corrections for the non-linear growth, you can not conclude on the suitability of the non-linear tracers in general. You can conclude here that without correcting for the non-linear growth, the apparent AoA and its trends deviate strongly, and that this motivates the investigation of correction methods.

Typos/ Language / Technical:

- Abstract, line 11: ".. does not exceed 6-6.5 years": it is not clear to me what this statement refers to - is this the "true" (ideal age) maximum value for AoA?

- line 18: what do you mean by "polar circulation" ?

- line 37, and general: check the parenthesis around references, they are incorrect at

several places, for example here it should read (Waugh, 2009 ; Stiller et al., 2012)

- line 86: "transformation procedure" - do you mean the chemical "transformation"? -> change to "chemical sinks" (?)

- line 122: over 60 km -> above 60 km; "that fall..." -> "i.e. within and above the top most..."

- line 159: please avoid using the word "practically", as it is not very specific

- line 168: "than ones accepted.."- Do you mean "than the ones usually used in models"?

- line 176: "the mesosphere" (add the)

- line 196: "in the vertical, one obtains that the ..."

- line 202: the overwhelming" (add "the")

- line 247: remove "been"

- line 267: "rations" -> "ratios";

- line 300: "downdraught" -> "downwelling"

- line 344: fall down -> decrease

- line 383: "the one in Fig.." (add the)

- line 386: "furthermost" -> furthest

- line 419: to the polar (replace "a" by "the")

- line 426: overstating -> overestimating

- line 452: do you mean upper stratosphere?

- line 482: "nor its ..."-> "nor does its mixing ratio" (remove "n")

- line 484: replace second "well" with "with"

- line 490: pointed out (add "out")

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