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Comment on acp-2021-232

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Referee comment on "The impact of sulfur hexafluoride (SF₆) sinks on age of air climatologies and trends" by Sheena Loeffel et al., Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2021-232-RC2>, 2021

The paper presents a modelling study of the Age-Of-Air and its trends derived from the mixing ratios of SF₆ in the atmosphere.

The authors performed multi-decade simulations of the stratospheric content of several SF₆-type tracers to reveal the influence of the mesospheric sink of SF₆ on the Age-Of-Air (AoA) derived from the SF₆ observations. It is shown that the mesospheric sink introduces a strong positive trend in the apparent SF₆ AoA, that overrides other possible reasons for trends.

Unfortunately, the paper lacks several important details, does not properly describe the current state-of-the-art, and has several methodological issues. Therefore I request a major revision to address the points below.

General comments:

1. The introduction suggests that main objective of the study is to reveal the cause of discrepancy between modelled and observed AoA and their trends. If it is the case, it should be stated explicitly. The lack of clearly formulated objectives and research questions makes it difficult to understand e.g. the model experiment design and justification for the choice of specific setups for the model experiments.
2. Further, the introduction states that "a comprehensive explanation for the trend differences between models and observations is still missing". The effect of the SF₆ destruction on the apparent AoA has been already pointed by Waugh and Hall (2002, Sec 3.2) and addressed by Kouznetsov et al. (2020), who has simulated the effects of the mesospheric sink of SF₆, and concluded that "The apparent over-ageing introduced by the sink is large and variable in space and time. Moreover, the over-ageing due to the sink

increases as the atmospheric burden of SF6 grows". (For more details, please refer to Sec.6.3 of the latter paper.) Therefore, it should be clearly stated what makes the existing explanations non-comprehensive.

3. The conclusions are formulated quite vaguely. It should be clearly stated what are the findings of the present paper, and how they agree/disagree with earlier results, and what of the findings are new. It might make sense to have separate "Discussion/summary" (all references to earlier results etc.) and "Conclusions" (the concise statements that the authors are ready to defend).

4. Modelling studies of long-term evolution of SF6 distribution in the atmosphere have been reported by e.g. Reddmann et al (2001), Kovacs et al. (2017), Kouznetsov et al. (2020). The need for the present study and its similarities and differences from earlier ones should be clearly indicated.

Specific comments:

Sec 2.1: A brief characteristic of the model is missing., e.g. "online spectral chemistry-climate model with hybrid sigma-pressure vertical layers".

Sec 2.2: The description of the SF6 sub-model is very unclear. Probably, most of the reactive species from Table 1 were not implemented as actual tracers in the model. One has to indicate which species were taken as climatology, which were forced from other models, and which were actual tracers. Was the submodel implemented as prescribed destruction rate as a function of altitude, latitude and season, or was it something more sophisticated? The description should be sufficiently detailed to allow for an independent reproduction of the experiment with another model.

Contrary to stated in II. 94-95, Fig.S1 does not show the relative importance of various reactions for SF6 destruction, but rather shows same reactions as in Table 1, but in a graphical form.

Sec 2.3. This is by far not the first simulations of this kind. What are the similarities and differences from the setups used in earlier modelling studies?
What is justification for specific model experiments, i.e. research questions to be addressed with each of the setups?

Sec 3.1: The section has one comparison against 3-year-mean MIPAS profile for a latitude belt of 30N-50N, and one in-situ profile. Those are nice for illustrations, but are insufficient to judge on the model performance in reproducing SF6 distribution in the

stratosphere.

Fig.1a: It is not clear why this specific latitude belt, and these specific years were selected. The MIPAS error bars show standard deviation of individual MIPAS profiles. How those are related to the uncertainty of the average (of millions?) profiles that are shown? The MIPAS averaging kernel and spatio-temporal collocation notably affect the comparison (Kouznetsov et al. 2020). This issue has to be at least discussed.

Fig 2a. Interestingly, Kouznetsov et al. (2020, Fig 5 there) shows very similar offset of the model profiles with respect to the in-situ one. I wonder if it is a coincidence, or an indication of a similar issue with both model setups.

Sec 3.2: The methodology for the lifetime estimate is quite unclear. Instead of explaining the method used, the authors put a reference to a 600-page report (Braesicke et al, 2019). The method, probably, refers to the equation in p. 1.20 of the report. The equation assumes well-mixed atmosphere and implies that the destruction of SF₆ is proportional to its burden, which is not the case: the destruction does not depend on the tropospheric content of SF₆, but rather on its content in mesosphere. In a situation when the change of SF₆ burden is substantial at a time scale of ~10 years (AoA in the stratosphere) the difference leads to "surprising" results like reduction of SF₆ lifetimes by 25% over 100 years.

Given the slow destruction of SF₆ one could still define the lifetime in terms of well-mixed assumption, but that would require a long-term simulation without emissions, to let the mixing ratio relax to its equilibrium distribution and get to the exponential decline of the total burden. Alternatively, as it was done by Kouznetsov et al. (2020), one could use a total burden that corresponds to the mixing ratio next to depletion layers.

Sec 3.4 -- 3.5: Same behaviour of trends in the apparent SF₆ AoA has been pointed out by Waugh and Hall (2002), Waugh et al.(2003) and demonstrated with extensive model simulations (Kouznetsov et al., 2020) for various latitudes and altitudes. Please specify what is a new finding here with respect to those studies.

I.329-365: The fact that SF₆ destruction causes the positive trend in the apparent AoA follows from the simple fact that the SF₆-AoA is proportional to a difference between stratospheric and tropospheric SF₆ mixing ratios. Since the destruction is proportional to the SF₆ mixing ratio, the difference increases together with the increase of the atmospheric SF₆ burden. There is no need to involve any equations or advanced concepts (like Green functions etc.) to explain that.