

Interactive comment on “Simulating age of air and distribution of SF₆ in the stratosphere with SILAM model” by Rostislav Kouznetsov et al.

Anonymous Referee #1

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Recommendation: Major Revision

This study presents simulations of SF₆, and determinations of age of air from this tracer, carried out with a chemical transport model (SILAM). It contains useful material that should be suitable for publication. However, it also includes lengthy discussions of molecular diffusion effects that do not appear to be relevant, while omitting essential information on other aspects of transport that are essential for understanding the distribution of SF₆.

The model's upper boundary is at 65 km (0.1 hPa). At this altitude, the effects of molecular diffusion are essentially negligible compared to the strong vertical mixing generated by breaking gravity waves, and to advection by the mean meridional circulation forced by wave breaking. Thus, the discussion of molecular diffusion, and of simula-

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tions that prescribe unrealistically low values of diffusivity in the upper stratosphere and mesosphere are not very useful and should be omitted in a revised version.

On the other hand, there is little if any discussion of mean meridional advection and how the vertical flux due to the mean meridional circulation is handled in SILAM. The model uses dynamical fields from ERA-Interim, which presumably include the effects of whatever gravity wave parameterization is used in that reanalysis. Thus, vertical fluxes due to mean meridional advection should not be negligible near the upper boundary of SILAM, but the paper does not mention advective transport at all, or how mean advective fluxes are handled at the upper boundary.

Finally, the study does not emphasize enough the role of SF₆ loss by the electron attachment mechanism, which becomes fast in the mesosphere (Fig. 1) and is essential for simulating the distribution of SF₆, as the WACCM results shown in Fig. 6 (which do not include this loss mechanism) make clear.

In view of all of this, I do not believe the paper is suitable for publication as it stands, but could be made so if revised to (1) eliminate irrelevant material on molecular diffusion; (2) use realistic profiles of eddy diffusion that could be obtained from any high-top model that parameterizes gravity wave breaking; (3) explain explicitly how mean meridional advective fluxes are handled at the upper boundary of SILAM; (4) document how these fluxes affect the distribution of SF₆; and (5) emphasize the role of SF₆ loss via electron attachment, which is evidently much more important than photolysis. Specific comments on these and other issues can be found below.

Specific comments (line number)

(72) “Silam”: This is an undefined acronym. If you are going to use it here you need to define it here, not in the next section. Note also that you write “Silam” here and “SILAM” elsewhere. Please pick one form and stick with it throughout the text.

(129) “10 hPa”: The conventional units of pressure in the atmospheric sciences are

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hPa (which are equivalent to the now deprecated mb). You might wish to consider changing references to pressure levels to units of hPa to avoid confusion (thus, 0.1 hPa in this instance).

(131) “effect of diffusion of SF₆ to the upper layers”: Transport through the 0.1 hPa (10 Pa) surface is not solely (or at some latitudes even mainly) due to diffusion; mean meridional advection is important, especially in the polar regions.

(167) “higher than . . . accepted in models”: It is not clear what this means. What models are you referring to? Global models run at practical horizontal resolution do not produce large vertical diffusivity due to the explicitly resolved motions. However, all recent such models include parameterizations of (unresolved) mesoscale gravity wave breaking. Vertical diffusion coefficients, K_{zz} , can be estimated from these parameterizations, and they produce values of K_{zz} that vary strongly with altitude, latitude and season. Thus, a single, global K_{zz} profile is unlikely to capture accurately the role of vertical diffusion. See also comment at line 387.

(168) “in order to cover the whole range of K_z ”: I think what you mean to say here is “to cover a range of vertical profiles of K_z ”. Is that so?

(169) “whose upper part was scaled”: what do you mean by “upper part”?

(170) “The three prescribed. . . profiles”: This is confusing. Fig. 2 shows profiles labeled K_z , 0.1 K_z and 0.01 K_z , but here in the text you refer to 0.03 K_z and 0.001 K_z . Which is right? The figure legend or the text?

(200) “the difference of equilibrium mixing ratio of SF₆”: How is this relevant in a range of pressure (0.1-0.2 hPa) where molecular diffusion is essentially negligible? The equilibrium profile defined by Eq. (5) is relevant for the upper mesosphere and above, which is beyond the upper boundary of the model used here. In fact, Eq. (5) and related discussion do not add anything useful to the problem of modeling SF₆ below the lower mesosphere.

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(203) “in the upper stratosphere heavy gases can no longer be considered as tracers and the molecular diffusion should be treated explicitly”: I do not believe this is right. Molecular diffusion effects should be small compared to eddy mixing and mean meridional advection below the upper mesosphere (75-80 km), and certainly within the range of altitude of the present simulations (top boundary at 0.1 hPa, about 65 km).

(225) “flux decreased by several orders of magnitude . . . at the level of a few Pa”: But in Fig. 3 all flux profiles increase with altitude. What is the definition of the flux shown in that figure? Does it not include a density factor?

(226) “shown in Fig. 3 with solid lines”: Flux profiles in Fig. 3 are labeled K_z , $0.1K_z$, $0.01K_z$ and $0.001K_z$. Do these correspond to the K_z profiles of Fig. 2, except that $0.001K_z$ is not shown in that figure?

(234) “For higher eddy diffusivity . . . molecular diffusion . . . becomes negligible”: This should be the situation in the middle atmosphere up to about 75-80 km. Gravity wave parametrizations yield values of K_{zz} of order $10 \text{ m}^2 \text{ s}^{-1}$ in the lower mesosphere (around 65 km); and mean vertical advection is also large at these altitudes. Therefore, for all practical purposes the effects of molecular diffusion and gravitational separation should be negligible over the range of altitude considered in this study. Note also that, according to Fig. 3, molecular diffusion effects are essentially irrelevant even for unrealistically weak values of eddy diffusion near the upper boundary ($0.1 K_z$ and $0.01 K_z$).

(246) “uppermost layer”: What is the upper boundary condition on the circulation? Does it force the vertical velocity to be zero at the top boundary? If so, note that the effect of mean meridional transport on SF6 distribution and lifetime will not be modeled realistically.

(265) “‘ones’ tracer”: “unity tracer” might be better.

(304) “the southern polar region”: What range of latitude does this cover?

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(320) “inter-annual variability”: This strongest variability seen in Fig. 4 is annual, presumably associated with the cycle of downwelling in winter and upwelling in summer. This again brings up the question what is the upper boundary condition on the dynamics (cf. comment at line 246), and how realistically mean vertical advection is modeled near the upper boundary. Note also that the mean meridional circulation in the mesosphere depends strongly on the contribution of gravity wave drag to the zonal-mean momentum budget, which would depend on how this is parameterized in the ERA-I reanalysis. Details on all of these points are needed.

(324) “simulations with 0.01 Kz”: Do you mean 0.001 Kz? That is what the legends in the left column panels of Fig. 4 indicate.

(330) “molecular diffusion . . . maintains the upward flux . . . even if eddy diffusivity ceases”: But in the real world, the flux at 65 km (0.1 hPa) is controlled principally by the combined effects of eddy diffusion and mean vertical advection.

(341) “vertical exchange is a key controlling factor”: This is correct, but note again that flux due to mean vertical advection is also important and may or may not be modeled properly in the present study, depending on how the upper boundary condition is handled.

(357) “way and rate of SF6 destruction”: What does “way of SF6 destruction” mean? You have varied the effective loss rate by changing the flux at the upper boundary, but as far as I can tell the loss mechanism was not changed.

(365) “the most diffusive case . . . overstated SF6”: This is likely due to the fact that the “1 Kz” profile has too large values in the lower stratosphere (although it has more reasonable values in the upper stratosphere and lower mesosphere).

(373) “largest deviation below 20 km”: See previous comment.

(375) “WACCM . . . under-representing the depletion of SF6 inside the polar vortex”: The problem with the WACCM result is that the standard version of the model does not

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include loss due to electron attachment, only photolysis. I would expect WACCM to simulate SF₆ quite accurately if all loss terms were included. What this result demonstrates is that it is essential to include loss via electron attachment.

(386) “In all the above cases, the ‘1 Kz’ profile is . . . too diffusive”: I don’t see this in all cases. The 1 Kz profile produces good agreement in the upper stratosphere in Fig. 6 b and d.

(387) “The ‘0.03 Kz’ profiles appear to be most realistic”: Actually, none of the profiles is realistic. In particular, the range of K_{zz} as a function of altitude obtained from gravity wave parameterizations is much larger than shown for the 1 Kz profiles of Fig. 2, where K_z varies from a little under 1 m² s⁻¹ at 20 km to less than 10 m² s⁻¹ at 65 km. In models that include a gravity wave parameterization, K_{zz} is estimated to vary between less than 10⁻³ m² s⁻¹ and more than 10 m² s⁻¹ over the same range of altitude. For a recent example, see Zhu et al. (JAS, 67, 2520, 2010).

(432) “lack of a pole-to-pole circulation”: Is this a result of the way the upper boundary condition is handled in these simulations? You need to show the Transformed Eulerian mean circulation as a function of altitude and latitude, at least for the solstice seasons, so the reader can understand the role of mean meridional transport near the top boundary of the model. Explicit description of how the upper boundary flux is handled in SILAM is also necessary.

(433) “understate it above 40 km”: In this instance, one could also question the observations, especially the ones that show an increase in mixing ratio with altitude. It is unclear how such profiles could be generated for a tracer that has a source in the troposphere and a sink in the mesosphere.

(435) Figure 8: Is the “de-biased RMSE” in the figure caption the same thing as “STD” in the ordinate label of the top panel? It would be desirable to keep the terminology consistent to avoid confusion.

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(436) “the difference in statistical scores of the three selected simulations is quite minor”: What “statistical scores” are you referring to?

(440) “standard deviation of model-measurement difference”: How does this eliminate the influence of model bias, assuming that is what you mean to say here? Doesn’t the model-measurement difference contain the bias? A formal definition of this quantity, similar to what is done in Eq. (11) for the NMB, would be useful.

(444) “RMS error of the observations due to retrieval noise in the original MIPAS data”: Is this what you mean by the legend “MIPAS noise” in the top panel of Fig. 8?

(450) Figure 9: This needs labels for the various curves, as in Fig. 8.

(452) “for the upper troposphere”: What does this refer to? This paragraph discusses results for 30-60 km. What does this have to do with the upper troposphere?

(460) “Three other profiles of Kz result in practically identical distribution of AoA”: This would imply that vertical mixing is irrelevant for the small Kz cases, and raises the question what controls AoA in these simulations.

(484) “The resulting model-based apparent AoA [is] much older than the “ideal-age” AoA and pretty close to the values derived from MIPAS”: This is an important result that highlights the role of fast mesospheric destruction of SF6 due to the electron attachment mechanism.

(503) “The reason is...”: You should reference Stiller et al. (2012) here, who already pointed this out.

(512) “‘ideal age’ and “passive” tracers: Are the results for the “ideal age” tracer the set of points labeled “time lag” in Figure 12? Again, consistency in terminology would be desirable.

(537) “eddy-diffusivity profile of Hunten (1975) scaled down”: The Hunten profile almost certainly overestimates diffusivity in the lower stratosphere, but reducing it by a factor

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of 0.03 will not reflect the behavior of vertical mixing in the upper stratosphere and the mesosphere. Ideally, one would estimate vertical mixing (as a function of altitude, latitude and season) from a gravity wave parameterization. Since such a parameterization was not available in the context of the present study, the conclusions regarding the role of K_z in determining age of air cannot be taken at face value.

Typos, etc.:

(28) “that presents an analogy of Lagrangian clock” -> “that is analogous to a Lagrangian clock”

(33) “are not possible, therefore. . .” -> “are not possible; therefore, . . .”

(285) “Eulerian analogy” -> “Eulerian analog”

(318) “is by more than an order of magnitude stronger than one of gravity separation” -> “is stronger than diffusive separation by more than one order of magnitude”

(319) “Regardless the used K_z profiles” -> “Regardless of the K_z profiles used”

(344) “depleting SF6” -> “SF6 that undergoes chemical destruction”

(344) “start to fall down” -> “begin to decrease”

(452) “on pair” -> “on par”

(482) “nor its mixing ration” -> “nor does its mixing ratio”

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