Comments on acp-2020-95

General comments

This paper extends the investigation in earlier papers of the gravitational separation (hereafter GS) and its relationship with age of air in the stratosphere. What is novel about this paper is its focus on availability of Ar/N_2 ratio as an independent age tracer. This is a new idea because Ar/N_2 ratio is totally different from "clock" tracers that are often used for age observations based on monotonic increase trends in troposphere. Moreover, Ar/N_2 has an advantage as an age tracer because it will be free from unfavorable influences of the propagation of seasonal variations and chemical processes on age estimation. In the previous studies, the relationships between age and GS have been examined, but observation data was quite limited and showed somewhat large scatter and there was no idea to use it for age tracer. This study shows tight correlations between AoA and GS by using aircraft data although it is limited to the lower stratosphere, which is supported by suitable 3-D model simulations. Another good point is sufficient theoretical development for the molecular diffusion process in atmosphere. In a sense, the atmospheric GS is somewhat new science, although the theory of molecular diffusion itself is well established. The authors presented a careful consideration of the molecular diffusion process in a ternary mixture by reviewing the theory from the basics and derived an unambiguous and generalized definition of the molecular diffusion coefficient as well as a simple method of GS simulation. These results will be very useful for future GS research, especially for model study.

This paper is worthy of publication in principle, subject to attention to a few issues as follows.

Specific comments

(1) As described in section 2.1, inlet fractionation is problematic for Ar/N_2 data obtained by airborne observations. The authors eliminated some data considering apparent biases or bad quality. I recommend that a little more details of data quality check or a criterion will be explained. The reason I care about this is that we can see clear difference of AoA-GS relationship between SH and NH (Fig. 4 and Lines

236-240). Is there a possibility that the data is still affected by the inlet fractionation and/or sample deterioration?

(2) Needless to say, it is useful to show the differences of vertical distributions between the model results and the observation for the model studies. Actually, AoA calculated by TOMCAT has been compared with observations in the previous studies (e.g. Chabrillat et al., 2018). However, the comparisons with the observation results are only shown as the AoA-GS relationships in this paper and we cannot know how TOMCAT reproduces vertical distributions of GS. It would be better to show a direct comparison with the balloon GS data. Particularly, the authors emphasize that TOMCAT includes GS enhancement in the upper stratosphere and mesosphere, which is superior to the previous model in this respect. This advantage will bring a significant improvement especially in the vertical structure of GS. That is one of the reasons why it is recommended to compare with the observations for not only the AoA-GS relationships in the lower stratosphere but also the vertical distributions observed by balloons.

(3) The authors hypothesize that an adequate representation of the mesosphere in models is critical for the AoA-GS relationship (Line 257). In fact, TOMCAT has a great advantage, because the top of model atmosphere is much higher than that in previous model study. However, I cannot fully understand that neglecting some components of molecular diffusion processes is really adequate even in mesosphere. In this model study, the molecular diffusion processes arisen from the concentration gradient and the temperature gradient are neglected. As summarized in Table A2, those components will be quite smaller than the pressure gradient term and negligible below around 35 km. However, the molecular diffusion coefficient becomes large in the upper stratosphere and mesosphere, which means that Peclet number will be small and thus component of concentration gradient (corresponds to the 3rd term RHS of eq. B2) is not negligible. In such a case, the assumption of the steady-state solution (eq. B4) is also not true. In this connection, magnitude of GS shown in Fig. 6 is much smaller than that of Fig.4 in Ishidoya et al. (2013) in the mid-stratosphere over high latitudes where the mesospheric GS will strongly influence due to the downward transport. It would be better to show that the influences of ignoring some terms and the assumption of a steady state are small even in the upper stratosphere and mesosphere.

(4) Another question arises also about the steady-state assumption concerning the seasonal variations. We can find that large seasonal variations of Ar/N_2 ratio occur in stratosphere and mesosphere as shown in supplement movie (Lines 249-251). This simply means that the time derivative of δ is not zero in eq. B3 and that there should be some restrictions for the approximation (eq. B4) to be applied. I just feel that an effect of the diffusive separation in the non-steady state will be small for Ar/N_2, because difference of the molecular diffusion coefficients between Ar and N_2 is not so large compared with those of heavy noble gases. However I don't know how far this steady-state assumption can be generalized.

Technical comments

Minor comments are listed below.

(L-109): "...are available from the START-08 campaign on the NSF/NCAR GV, but we have not used these here because the $\delta(Ar/N_2)$ data quality is considerably worse. "
What is the reason why the quality of START-08 data is worse?

(Fig. 1): Some symbols (rotated triangles?) do not seem to match those in the legend.

(Eq. 3): "λ" in the 2nd term of RHS should be λ_2. $G(t'|(\Gamma, \lambda))$ should be $G(t'|(\Gamma_1, \Gamma_2,\lambda_1,\lambda_2))$, to be exact.

(L-152): "Γ_1, Γ_1, λ_1 and λ_2 are... " should be "Γ_1, Γ_2, λ_1 and λ_2 are ...."

(L-179): "(ii)" should be "(iii)".

(Section 3.1): Parameters Γ_1, Γ_2, and A are shown in Fig. 3 and discussed in detail. But there is no description how the shape parameters λ_1 and λ_2 of the inverse-Gaussian distribution were as a result. It would be better to give us information about widths (Δ) or values of $\gamma(=\Delta^2/\Gamma)$ of age spectra obtained by this method.

(L-183 to Eq. 5): "....atmosphere δ_{GST} with a molecular mass 1 amu greater than that of air ..."
GST seems to be a kind of virtual tracer. It may be better to mention how the molecular diffusion volume was defined for D_{GST}.

(L-236 to 240): Again, with this AoA-GS plots alone, it is somewhat difficult to understand the difference between NH and SH. Vertical distributions of AoA and GS may be helpful.

(L-276 and Fig. S2): "...the small seasonal cycle amplitude of δ(Ar/N2) <6 per meg..."
Certainly the average seasonal amplitude seems to be small, but we can see large scatters. Fluctuations of Ar/N_2 ratio in short time will be partly smoothed by the mixing process during the upward transport from the tropical upper troposphere to the lower stratosphere via TTL. Thus, it may not be a big obstacle to AoA estimation from Ar/N_2. Problem is, rather, that this scatter is real atmospheric signal or not.

(Eq. 7 in Appendix A): This number should be something like "(A1)". Also please check eq. numbers in Appendix B and C.

(L-411):"ΔM_i the molecular mass difference to air "
ΔM_i appears in eq. (A21) for the first time.

(Eqs. B1, B2, and B3): Please check the signs (plus/minus) of the molecular diffusion terms in these equations. In the conservation equation, term of the flux divergence (3rd term RHS in eq. B1) should be a form like $-\nabla \cdot [f]$ if we put it on RHS, just the same as the eddy diffusion term. The 3rd and 4th terms of RHS in eq. B2 might have the same signs after using the chain rule. The 3rd term of RHS in eq. B3 might have the same sign with 4th term of RHS in eq. B2 after eliminating a small term.

(L-418): "D" in the expression of Peclet number should be "D_e".

(L-432): "Eq. (6)" should be Eq. (5).