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Atmospheric Chemistry and Physics
Discussions

Interactive comment on "Age and gravitational separation of the stratospheric air over Indonesia" *by* Satoshi Sugawara et al.

Anonymous Referee #1

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This manuscript reports a small number of observations of two trace gases, CO2 and SF6, as well as the isotopic composition of N2 and O2, and the ratio of Argon over the former. These observations were carried out with an innovative technique in an interesting and understudied region, i.e. the tropical stratosphere and the authors report some interesting effects. I recommend the manuscript for publication in ACP in general, but a large number of issues will need addressing first; the main ones being

1) a better placing of the results in context of existing literature and comparisons with published results, in particular for the mean age of the air.

2) clarification of many questions related to sample collection, measurements, error bar calculation, and quality assurance.

3) an improved discussion of results relative to common vertical coordinates in the stratosphere, such as the altitude relative to the tropopause, pressures or potential temperatures.

4) a wider consideration of potential age uncertainties and bias and reassessment of the difference between CO2 and SF6-derived mean aged and whether or not it is indeed significant.

5) the modelling part of the manuscript is a nice addition, but currently rather inconclusive as none of the tested scenarios seems to be able to match the observations.

More specific points can be found below.

P1, l15 It is common to open with a statement on the motivation for and wider context of the work presented. I encourage the authors to adopt this practice as it also tends to improve readability.

P1, l17 The concept of the mean age of air needs some introduction here.

P1, l20 Should this be 29 km instead of 25?

P1, l30 Again, the concept of the age of air is not introduced at all.

P2, l1 There is also the more recent work of Engel et al., ACP, 2017 extending the balloon-based trend as well as Stiller et al., ACP, 2017 who show that the observed latitudinal and vertical patterns of the decadal changes of age of air in the lower to middle stratosphere during 2002–2012 are predominantly caused by a southward shift of the circulation pattern of about 5 degrees

P2, l2 Only in the region of constant mean age above 25 km. In fact, both Diallo et al., ACP, 2012 and Boenisch et al., ACP, 2011 found indications for changes in the region below 25 km.

P2, l8-10 Ploeger et al., 2015 demonstrated that both mixing and residual circulation impact on ageing throughout the stratosphere.

P2, l20-22 This is not correct. There is substantially more literature on air sampling in those regions: Volk et al., 1996, Schauffler et al., 1998, Tuck et al., 2004, Kaiser et al., 2006, Laube et al., 2010, and Brinckmann et al., 2012 to name but a few. It would be good to see how the results, in particular the mean ages, compare.

P3, l1-5 Were these samples collected during ascent or descent? In either case, can the authors provide evidence for all species measured that no contamination originated from the balloon (in particular if collection occurred during ascent – Helium leaking into samplers is a known problem) or from the payload?

P3, l20 What was the reason for the improved precision?

P4, l6-7 What range of mole fractions was covered by those primary standards? ECDs are known to be strongly nonlinear detectors, so what methods were employed to ensure that a) detector responses were corrected and b) there is agreement between calibration scales over the entire range of observed mole fractions?

P4, l25 If CO2 mole fractions do indeed change over time, the exact change will be quite influential on the determined age of air. 0.7 μmol mol-1 is equivalent to several months of mean age and it would be good to know whether the authors have tested any potential variability of such a correction from repeats of storage experiments as well as different water vapour contents, both of which are known to be influential factors. A similar question arises for methane – has this been storage-tested at all?

P5, l4-5 This is interesting and unusual as the upper part of the profile implies a vertical transport barrier in the tropical stratosphere. Has this been observed in other tropical data? Could this be a regional phenomenon or related to the onset of the recent QBO anomaly? Or is there any evidence for impact from convection? Moreover, can the authors explain why CO2 and SF6 should exhibit a similar behaviour in the lower part of the profile when one has a strong seasonal cycle and the other has none? CO2 would surely be expected to show some variant of a tape-recorder signal, unless the authors have evidence to the contrary?

P5, l13-16 It is not clear what is being compared here. Similar altitudes cannot be compared directly between tropical and mid latitudes as tropopause heights differ substantially. The authors could use alternative coordinates such as height above tropopause, potential temperature or potential vorticity to address this problem. This would also help answering the question of where the TTL actually was during sampling. In addition, any comparison requires the inclusion of the mid-latitude data set, which could for instance be added to Figure 2.

P6, l9-12 This is not a valid comparison. In the mid-latitudes, 32 km could mean well over 20 km into the stratosphere, whereas in the tropics the tropopause could be around 19 km. Gravitational separation should be compared at least on a per km in the stratosphere basis and the effects of comparing different pressures should also be considered.

P6, l27-28 Why are only the last 5 years shown of 10 or more are available? Are the fits less good for the earlier years? What is the uncertainty range of those fits and how high are the resulting uncertainties in mean ages? And why does the data in Figures 4 and 5 have no error bars?

P7, l11-12 According to the major ground-based networks (AGAGE and NOAA) global SF6 mole fractions did not increase linearly in the last decade. Emissions have been increasing (WMO, 2014) resulting in some curvature of that trend. Have the authors considered the potential impact on mean ages, or, if they cannot resolve these curvatures, the additional uncertainty in their mean ages?

P8, l8 How were these uncertainties calculated?

P8, l9-11 Do the authors actually have any data (e.g. temperature or pressure) to confirm where the tropopause exactly was? And what is meant by “secondary tropopause”?

P8, l24-25 Can the authors present any evidence for this claim?

P8, l29 So other observations do not show a step change in age around 24 km. This reemphasizes my earlier question on an explanation for that phenomenon.

P9, 16 Again, not necessarily relative to the tropopause.

P9, l17-24 There are several more important and also more recent studies, e.g. Waugh and Hall, 2002 and in particular Ray et al., 2017.

P10, l19-20 I suggest showing the actual correlation in Figure 6. I would certainly say the two quantities do not correlate well around the upper part of the profile.

P10, l24-25 This is quite a claim. Are the authors suggesting that the tropical pipe does not pose a significant transport barrier between tropics and extra-tropics?

P11, l6-10 It certainly suggests that the molecular mass difference is dominant, but the molecular diffusion flux could still dampen the signal considerably. A mere proportionality does not rule that out.

P11, l16-17 As pointed out earlier the differences in pressures and temperatures at similar altitudes are quite important here and should be discussed.

P12, l8-9 Did the model-simulated CO2 mole fraction include a realistic seasonal cycle?

P12, l23-25 Figure 7 does not include a comparison with data from Ishidoya et al., 2013. I suggest either including it in the figure or as quantitative statements in the text.

P21&26, figures 2 and 6 Surely the air samples were collected over a range of altitudes? Please add the y-axis uncertainties for all relevant figures.

P23&24, figures 4&5 Some of the scatter observed here is very likely due to changing input from the northern and southern hemisphere as the ITCZ moves through that latitudinal band. Can the authors assess the potential age uncertainty resulting for the exact time of their balloon campaign? This effect might even introduce a bias.

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2017.