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Interactive comment

Interactive comment on "Transport model diagnosis of the mean age of air derived from stratospheric samples in the tropics" *by* Hanh T. Nguyen et al.

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Reply to Referee 2

We sincerely appreciate Referees 1 and 2 for their review of the manuscript and valuable comments and criticism on it. We understand the problem and have made substantial changes to the manuscript in response to the comments from both Referees. These revisions have significantly improved the manuscript, and we hope we have answered all of the concerns. Our reply to Referee 2 is shown below in blue following the comments cited in italics.

The paper by Nguyen et al. investigates different methods with which observations of mean age tracers and mean age of air (AoA) can be reconstructed. While the study is certainly in the scope of ACP and the subject is of significant scientific interest, there are a number of problems I see in the investigations presented here. In my view, the authors misinterpret some aspects of AoA, as detailed below. Further, in my view some aspects are presented (e.g. related to Figures 1 and 2) which are not taken up in the discussion or conclusions of the manuscript and are not necessary for the understanding. Other aspects like experimental details (e.g. where, when and how were the samples taken) and how was AoA calculated from the observations is omitted. There are also a number of important recent papers, which are not included in the discussion (see details given below). Due to these issues, I believe that the paper is not ready for publication, but needs major revisions before it can be considered for publication.

We really appreciate detailed review and valuable comments to the manuscript. We believe that the application of two independent methods, the boundary impulse response (BIR) method and back trajectories, to the ACTM wind field successfully achieved our research goal of interpreting the vertical profiles of CO_2 and SF_6 ages obtained by cryogenic air sampling in CUBE/Biak campaign. We think our revisions detailed below are enough to satisfy the reviewer and hopefully make the manuscript suitable for publication in ACP.

Major comments

Clock tracers and derivation of AoA

I believe that there is a misinterpretation on what is commonly understood by "clock tracers". Clock tracers are (artificial) tracers which increase not only monotonically, but

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also linearly. Neither SF_6 nor CO_2 fulfill this criterion. SF_6 increases monotonically, but not linearly, CO_2 has in addition a seasonal cycle, thus does not even increase monotonically (except if annually averaged). Therefore, the shape of the assumed age spectrum plays a significant role in deriving AoA from such tracers. This is not sufficiently discussed in the manuscript. The authors may want to consult e.g. a recent paper by Fritsch et al. (2020) in ACP on the issue of how AoA can be derived from such tracers and how that agrees with ideal clock tracers. Using such a clock tracer, AoA can be derived as a lag time without needing any knowledge on the age spectrum. While I have not checked all the papers reference on p.2., l. 22, at least Haenel et al. did not use lag time but did take into account the age spectrum. As in the end the main focus is on the comparison of AoA derived in different ways, the use of clear language and correct referencing is necessary. More details are needed on the calculation of AoA, including which tropospheric reference time series have been used, have these been fitted and AoA derived as in Volk et al., (1997)? Or has AoA been derived by convolution of age spectrum and time series? How many years where taken into account in fitting or in convolution etc. Has CO_2-production by oxidation of CH_4 been taken into account? These are extremely important details which are needed to understand possible discrepancies. Also, I would strongly suggest to include a real clock tracer in the model, from which AoA can then be derived without any assumptions and which can serve as a reference.

Appreciating that CO_2 and SF_6 are conveniently used to visualize stratospheric general circulation under the scope of pseudo-“clock tracers,” we understand it is necessary to distinguish them from the idealized concept of “clock tracers.” The phrases such as “observations of clock tracer” are replaced by “observations of tracer”, for example, wherever necessary. We also understand the importance of age spectra to estimate mean AoA from tracer observations. In the present study, the BIR method and the back trajectory calculations are applied to estimate age spectra and they are used to calculate the mean age by convolution. We understand the descriptions were not sufficient, and revised the manuscript by adding more explanation. As for additional model experiments that use a real clock tracer, we should say it is not possible in the foreseeable future because of the lack of computing resources; the model experiments presented in this study were conducted when one of the coauthors (KI) was affiliated in JAMSTEC before his move to MRI. Following the above comments, description on our field experiments and the method of mean age estimation from the observed CO_2 and SF_6 mole fractions intensionally omitted referring to our publications (Hasebe et al. 2018, Sugawara et al. 2018), are made in some details. All cited references are rechecked and revised based on the reviewer's comments.

Structure of the paper

The paper presents many aspects, many of which are a repetition of previous work, before finally coming to what is really new, the comparison of CO_2 and SF_6 reconstructed with the two different methods (BIR and Lagrangian). In my view, many parts of section 2 are not necessary, while other parts are missing. Note that none of the aspects discussed with respect to Figure 1 and 2 are in any way mentioned in the discussion, conclusion or summary. Missing parts are details about the observations and how AoA has been derived from them, but also explanation of methods, e.g. the BIR methods should be explained in brief. Section 2 also is called "Model and Experiment", so I was expecting the usual explanations of which model has been used in which set-up and details about the observations. As it stands now, it is a mixture of model description and interpretation, but does not have any experimental part at all.

The manuscript has been reorganized as follows:

1. Introduction
   Our research questions are stated clearly to meet the comments from Referee 1. Recent publications in related topics are also added. Some more descriptions

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on our campaign CUBE/Biak have been given as well. An introduction of the atmospheric general circulation model-based chemistry transport model is made with its abbreviation ACTM. The use of “clock tracers” is eliminated in response to the comments by Referee 2.

2. Model experiments

   2.1 Description of the model and simulation design
   We try to interpret the vertical profiles of observationally estimated CO_2- and SF_6-ages referring to transport model calculations. The use of ACTM is thus a key to our analysis. Explanation on the use of ACTM is given here.

   2.2 Evaluation of the model performance
   Our results deeply rely on the performance of the transport model. The model performance is briefly investigated by looking at the distribution of tracers that are released as a “pulse” at the tropical surface. This method of tracer release constitutes the basis of the BIR method.

   2.3 Estimation of age spectra and mean age of air
   We employ BIR method and back trajectories to estimate age spectra and mean age of air in the stratosphere. A brief review of the theoretical foundation of both methods are given here before their application to the tropical stratosphere.

3. Application to CUBE/Biak observations

   3.1 BIR method
   The mean age estimation relies on unobservable age spectrum. The age spectra estimated from BIR method are described.

   3.2 Lagrangian method
   Back trajectory calculations are often conducted to describe the tracer transport from a Lagrangian point of view. The method is one of the important

tools to study stratospheric tracers including water vapor. The use of one-hour averaged one-hour interval wind field, together with additional pressure levels assigned near the tropical tropopause, proved useful to better reproduce the observed profiles of CO_2, SF_6, and water vapor "tape recorder."

3.3 Assessment of the mean age profiles

The mean age profiles derived by applying above two methods are compared against those estimated by using observed CO_2 and SF_6 mole fractions.

4. Discussion

The results obtained above are discussed focusing on the interpretation of the differences between the ACTM-derived and observationally estimated mean ages, Δ^2/Γ-ratio and the shape of age spectra, and the advantage of using one-hour averaged one-hour interval data available from ACTM in trajectory calculation.

5. Summary

The overall results are summarized.

Appendix A: Supplementary notes on the age spectra

The effect of tail correction and fine structure reflecting the pathway difference are discussed emphasizing the importance of using accurate age spectrum for mean age estimation.

Appendix B: The effect of quasi-biennial oscillation (QBO)

The modulation of BIR map over the equator due to QBO is briefly described.

Figures are rearranged and reorganized as follows:

Section 2

Fig. 1: Latitude-height section of the mixing ratio of January-released pulse tracers in (a) February of the first year, (b) February of the second year, and evolution of pulse tracer concentrations (c) over the equator and (d) at some representative latitudes on 50 hPa pressure surface. Panels (a) and (b) come from original Fig. 1, and panel (c) comes from the upper panels of original Fig. 3. Panel (d) consists of lower panels of original Fig. 3. The original Fig. 2 is deleted.

Fig. 2: Zonal mean distribution of three-year averaged mean age in NH winter (DJF) and summer (JJA). This comes from original Fig. 5. Original Fig. 4 goes to Fig. A1 in Appendix A.

Section 3

Fig. 3: (a) BIR map at 50 hPa over the equator, and (b) latitude-height section of the mean age in March 2015. Panel (a) comes from original Fig. 6 (a), while panel (b) is original Fig. 7. Original Fig. 6 (b) goes to Fig. B1 in Appendix B.

Fig. 4: Age spectra derived from BIR method corresponding to the altitudes of eight cryogenic air samples acquired during CUBE/Biak 2015. This is the same as original Fig. 8.

Fig. 5: Examples of (a) age spectrum and (b) water mixing ratio spectrum estimated from back trajectory method. These panels come from original Fig. 9 (a), (b). Those of original Fig. 9 (c), (d) are deleted.

Fig. 6: Vertical profiles of mole fractions of (a) CO_2 (ppm), (b) SF_6 (ppt), and (c) water vapor mixing ratio (ppmv) estimated by back trajectories. This figure comes from original Fig. 10. Original Fig. 11 appears in snapshots in a movie provided by Supplementary Material.

Section 4

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Fig. 7: Comparison of the vertical profiles of (a) mean age and (b) ratio of moments (Δ^2/Γ) estimated by the BIR method, back trajectories, and cryogenic samples. Panel (a) comes from original Fig. 13 after removing horizontal bars for $\Gamma_{\rm bir}$ and $\Gamma_{\rm trj}$. Panel (b) is newly plotted from Table 2.

Fig. 8: Time series of the zonal (u), meridional (v), and vertical (ω) wind components at grid points $0°$ longitude near the equator. This figure comes from original Fig. 12.

Appendix A

Fig. A1: Multi-year averaged age spectra with tail correction estimated by BIR method. This comes from the original Fig. 4.

Fig. A2: (Left) age spectra and (right) meridional projection of back trajectories. This comes from the original Fig. A1.

Appendix B

Fig. B1: A time-height section of mean zonal wind over the equator. This comes from the original Fig. 6 (b).

A supplementary material has been attached with the revised manuscript. It contains an animated GIF showing a meridional projection of air parcels associated with the backward trajectory calculations for one year since the initialization on 27 February 2015.

Specific comments

P2, L6: *I think that this is a very unlucky formulation and explanation of AoA, as it suggests that an air parcel keeps its integrity during transport.*

We are afraid we may not understand you correctly, but Kida (1983, JMSJ, 61, 517) is referred by many studies such as Hall and Plumb (1994) and Waugh and Hall (2002), and we understand his schematic illustration (Fig. 4) is well-accepted as a basic concept in our community. The sentence is retained as it is.

P2, L9: *see discussion above: SF_6 and CO_2 are not clock-tracers.*

As is mentioned in the reply on Major commnets, we no longer call SF_6 and CO_2 as clock tracers in the whole manuscript.

P2, L13: *Note that the results of Engel et al. (2009) have been updated in Engel et al., 2017 and Fritsch et al. 2020. (both in ACP)*

Thank you for the comment. Both papers are cited in Sect. 4.

P2, L22: *see discussion above: AoA cannot be derived from SF_6 or CO_2 using the lag-time approach. While this may have been done in the early years of AoA it is certainly not applied in more recent studies.*

We agree with the referee and the lag-time method is not used in our study. This sentence has been removed along with the revisions.

P2, L25: *a clock tracer must increase linearly, not only monotonically.*

We agree with the referee. This sentence is deleted along with the revisions.

P3, L15: *this is only about models, not about experiments. It should include some basic information about the measurements.*

Section 2 is now entitled “Model experiments” and revised to include description of the model and simulation design (Sect. 2.1), evaluation of the model performance (Sect. 2.2), and estimation of age spectra and mean age of air (Sect. 2.3). The method of estimating mean age from air samples is discussed in the second paragraph of Sect. 4 (Discussion), although we do not describe anything on the measurements such as how to collect air samples and how to measure mole fraction of CO_2 from them.

P3, L26: *can this evaluation be summarized?*

The evaluation of our model performance is made in Sect. 2.2. The results by Krol et al. (2018) are briefly introduced in Sect. 2.3.

P4, L13: *I do not understand this sentence.*

The sentence is rephrased to: "The Northern winter transport field is investigated as an example in the top panels of Fig. 1 by examining the zonal-mean distribution of January-released tracers in (a) February of the first year (i.e., the next month) and (b) February of the second year (i.e., 13th month since release)." in Sect. 2.2.

P4, L30 – P5, L13: *Is this necessary to understand the rest of the paper?*

Thank you for the comment. This part is totally revised into new Sect. 2.2, along with the elimination of Fig. 2 and associated descriptions on the transport features.

P5, L14: *this section should have an introduction to what BIR is.*

Section 2.4 is totally rewritten following your comment. The revised Sect. 2.3 includes a brief introduction on the theoretical foundation of the BIR method.

P6, L7: *I suggest to use larger AoA, not longer.*

We have changed "longer" to "older" referring to preceding studies such as Li et al. (2012a, b) and Ploeger and Birner (2016).

P6, L10: *I find it hard to understand this conclusion from the statements above.*

We understand your concern. This sentence is deleted, and the whole paragraph including Fig. 4 is moved to Appendix A.

P7, L11: *I find this contradictory: doing a reasonable job enables to do a quantitative assessment?*

We think a quantitative assessment can be made only by a good job. Anyway, the sentence is deleted.

P7, L21: *please explain the choice of Tr_{top} of 355 K. This is quite a bit below the tropical tropopause and transport from 355 K to the tropical tropopause should still take at least several weeks to months.*

The 355 K isentrope is surely much lower than the tropical tropopause. On the other hand, it is close to the altitude of tropospheric reference adopted by Sugawara et al. (2018). The meterological reason of the choice is written in Sect. 2.3 as follows: "$\mathrm{Tr}_{\mathrm{top}}$ is taken to be the 355 K isentropic surface, reflecting the fact that the influence of tropical convective motion almost ceases at this level and diabatic forcing gradually changes to radiative heating in and above the TTL (Hasebe and Noguchi, 2016)."

P8, L1: *this is a very large uncertainty range, which is even larger than the central value. Can you explain this large variability and the shape of the distribution (which must be quite unsymmetrical).*

We are really grateful to this comment. There was a mistake in our calculation, and the correct number is 0.24 years rather than 0.69 years.

P8, L6: *Delta may not in any way be mistaken as an uncertainty in AoA. It is the width of the spectrum. If you have a perfect tracer, this is completely unrelated to any uncertainty in AoA.*

We understood and totally agree with the referee about this. Figure 8 is now Fig. 4 in the revised manuscript, and the phrase "as the estimates of uncertainties in Γ_{corr}" is deleted.

P8, L11: *I strongly suggest not to call these experimental conditions: these are model parameters used in the investigation.*

The sentence is changed to: "The model parameters for calculating the kinematic backward trajectories are summarized in Table 1".

P9, L12: *I find it hard to derive this conclusion from the results shown.*

The revised sentence reads, "A remarkable improvement in the use of ACTM-NUDG on H18 and the insufficient performance of ACTM-FREE are further discussed in Sect. 4 investigating the vertical mass transport." (Sect. 3.2). Figure 11 is replaced by a movie to be provided as Supplementary Material.

P10, L20: *This means that only about 3*

We are afraid we do not understand this comment correctly, but we expect 3 years will be an overestimation for the mean age around 25–30 km considering the perceived overestimation of Γ_{Sobs} arising from the mesospheric loss of SF_6. In any case, we would like to focus our evaluation of the observed age profile relative to mean ages obtained by BIR and Lagrangian methods. The sentence is revised to: "The omission of this pathway must result in the underestimation of Γ_{trj} relative to Γ_{bir}. It is also responsible for making Γ_{trj} younger than Γ_{Cobs} and Γ_{Sobs} above 25 km. That is, the absence of mesospheric air parcels in the Lagrangian calculations leads to the higher mole fractions (Fig. 6) and the younger mean age (Fig. 7) than the observational values."
Please note that Fig. 13 (now the left panel of Fig. 7) is revised by eliminating horizontal bars for Γ_{trj} and Γ_{bir} because Δ cannot be used as a measure of uncertainties. The bars for Γ_{Cobs} and Γ_{Sobs} reflect the uncertainties associated with the laboratory analysis to derive CO_2 and SF_6 mole fractions.

P10, L22: *This statement can only be made if the cut-off time is included (I suppose 5 years) and is highly dependent on the region in the stratosphere.*

The sentence is rephrased to: "while more than 50 % of the mean age comes from the tail when the transit time is cut-off at 4 years in the BIR method applied to the extratropical stratosphere (Li et al., 2012b)."

P11, L5: *It has recently been shown that other models have a larger ratio of delta^2 to AoA (Hauck et al., 2019, ACP)*

Thank you for the introduction of recent result. Hauck et al. (2019) is cited together with Fritsch et al. (2020) in Sect. 4 discussing the value of the ratio of moments. Concerning this value, we found an error in the estimation of the width of age spectrum in the BIR method. The corrected results are updated in the revised manuscript (Table 2). The discussion related to this parameter is also revised.

P11, L14: *This statement is not true for clock-tracers, but then as stated above, CO_2 and SF_6 are no clock tracers.*

As is mentioned on our reply to Major comments, we no longer call CO_2 and SF_6 as clock tracers.

P11, L23: *there are more up to date references for mesospheric loss of SF_6, especially Ray et al. (2017, JGR) and Reddmann et al. (2001, JGR).*

We appreciate this input. The suggested papers are cited in the revised manuscript.

P12, L28: *I believe that the authors are wrong: the tail correction is extremely important here, as it has a strong influence on the width of the age spectrum.*

Generally speaking, the tail correction is extremely important. However, here in the tropical lower stratosphere at around 17 to 18 km, the mean ages estimated by trajectory calculations are 0.12 to 0.13 years and the tail corrections are on the order of days. The sentence, "The tail correction is negligible and is ignored here." is retained as it is.

P13, L6: *I am confused: according to Figure A1, AoA is 47 days for sample 03 and delta is 17 days. From this I would derive a ratio of delta^2 to AoA of about 6 (and not 0.08).*

We are sorry to find incorrect citation of the numbers shown in Fig. A1 (now Fig. A2). Correct values are AoA = 49 days and delta = 32 days for Sample 3. From this the ratio of delta^2 to AoA is 20.9 days = 0.06 years. Similarly the ratio is 0.32 years for Sample 1. Table 2 is corrected.

Fig 9: the x-axis of panel a should not be age. This is transit time.

Corrected.

Fig 10: as before: delta is not in any way a measure of the uncertainty of AoA.

The horizontal bars are deleted together with the associated captions.

Fig A1: the x-axis of the left hand panels should not be age. This is transit time.

Corrected (now renumbered to Fig. A2).