

## ***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.***

**Hanh T. Nguyen et al.**

hanhnt@ees.hokudai.ac.jp

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### **Reply to Referee 1**

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 1 is shown below in bold.

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following the comments cited in italics.

*In their paper, Nguyen et al. apply two methods (BIR and backward trajectory calculation) to model age of air (spectra) as well as mole fractions of CO<sub>2</sub>, SF<sub>6</sub> and water vapour. In the paper, first the model results are evaluated and subsequently some results are compared with data from a measurement campaign. The comparison works reasonably well, with some discrepancies that mostly can be explained. The overall idea of the paper is good and the method is elaborate. Some of the results are quite interesting and can contribute to foster science in this field, although it is for example not expected that chemical SF<sub>6</sub> depletion will not allow direct comparison in the upper stratosphere if it is not included in the model. It is good to carve out which of the modelling methods is suitable to tackle which science question.*

*However, the paper is chaotic and does not provide the necessary information to evaluate the results. Almost nothing is reported about the measurements of that campaign and where the points that were supposed to be investigated with them. The model description is unclear, I do not understand why sometimes nudging is described, while the authors apply a CTM, which usually is driven (not nudged) by reanalysis data. This is very confusing. The evaluation of the results is pretty lengthy and should be reduced to about two figures. If need be, the rest can be banished to a supplement (maybe together with the appendix). I like the idea of explaining measurements with model results, but at the end, that is only a minor part in the paper and is only partially successful (partly due to the sinks). However, my main point really is that the study does not follow a clear research question. The reader can be lost due to that. What is exactly that is puzzling you about the measurements? Why do you think the applied method can help to answer that question and how do you plan on pursuing that? How can additional information about transport processes be gained through that? What is your contribution to improve the understanding of the underlying processes at the end and how does that fit into existing literature? Some of this information is missing.*

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*lacking, some is spread somewhere across the manuscript, and the reader has to put the pieces together. Further, so many different points are applied, AoA, its spec the CO<sub>2</sub> and SF<sub>6</sub> mole fractions as well as water vapour and the cold point are thrown together, but it does not clearly shine through that all these measures are needed to make the conclusions that are drawn at the end. Also, different models, free running nudged simulations and/or the CTM and the two diagnostic methods, all that lead to confusion and does not help to get to the point. Additionally, intense use of non-intuitive abbreviations complicate reading and also in the results section, many points are thrown together and a clear focus is missing.*

*Hence, I would suggest the authors to completely revise the manuscript and then submit it again. I think the study can help to advance our understanding of stratospheric transport and the methods that can be used to investigate it if it is presented and structured properly. Please start with one or more clear research questions that can be answered with this method and build everything around that. Use only the methods needed, describe them clearly, and then take the reader point by point towards the conclusion. Please also consider my additional comments that I am making below.*

In response to the above comments, the manuscript has been completely revised by setting clear research questions. Before explaining them briefly, let us resolve the confusion on the use of "ACTM" and the application of nudging in the present study. ACTM is an abbreviation of an Atmospheric General Circulation Model (AGCM)-based Chemistry Transport Model (CTM), which was used in the past literature such as Iijima et al. (2010, JGR, 115, D20308, doi:10.1029/2009JD013322). We would like to maintain the use as the continuation of previous studies. The application of ACTM with data assimilation is motivated by our hope that realistic transport field is better represented than the direct use of (re)analysis field, such as ERA-Interim, in higher temporal resolution. We choose nudging as the simplest way for data assimilation. Nudging is frequently applied for the diagnosis of model performance in AoA studies as can

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seen from Krol et al. (2018, Geosci. Model Dev., 11, 3109–3130). The improvement attained by the use of ACTM with nudging is discussed in Section 4.

We agree to the comment that the study must follow a clear research question. We have revised the manuscript to state it clearly in Introduction. That is, how can we interpret the vertical profiles of CO<sub>2</sub> and SF<sub>6</sub> ages obtained by cryogenic air samples in CUBE/Biak campaign, especially from the aspect of the shape of age spectra which is often parameterized by  $\Delta^2/\Gamma$  ratio since Hall and Plumb (1994). We employ three methods, boundary impulse response (BIR) method and Lagrangian backward trajectories, both relying on ACTM wind field. The whole manuscript has been revised along the line to answer the question under a unified story. The description on the evaluation of the results and the number of figures are reduced. Due to the additional description such as the measurements of that campaign, however, the total length of the manuscript remains almost the same. Some of the contents are moved to appendices and supplementary material following the suggestion. The use of abbreviations has also been revised. The details are given below.

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 on our campaign CUBE/Biak have been given as well. An introduction of 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

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We try to interpret the vertical profiles of observationally estimated CO<sub>2</sub>- and SF<sub>6</sub>-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<sub>2</sub>, SF<sub>6</sub>, 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

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against those estimated by using observed CO<sub>2</sub> and SF<sub>6</sub> 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 age spectrum,  $\Delta^2/\Gamma$ -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 differences are discussed emphasizing the importance of using accurate age spectrum 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 panel of original Fig. 3. Panel (d) consists of lower panels of original Fig. 3. The original Fig. 2 is deleted.

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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. 5 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<sub>2</sub> (ppm), (b) SF<sub>6</sub> (ppt), and 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

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

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

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### 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 backward trajectory calculations for one year since the initialization on 27 February 2015.

We believe that the application of two independent methods, BIR and back trajectory to the ACTM wind field successfully achieved our research goal of interpreting vertical profiles of CO<sub>2</sub> and SF<sub>6</sub> ages obtained by cryogenic air sampling in CUBE/Biak campaign. We hope we have made necessary revisions so that the manuscript has reached the required quality for publication in ACP. Detailed revisions associated with this manuscript are provided in the supplementary material. Additional comments follow.

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### **Additional comments:**

P1, L2: *A CTM is not nudged! A CTM uses some meteorological fields for describing transport. This is totally confusing and it is not clear to me what is actually done in this study, because later also you talk about GCM and CTM. Please clarify what is and what you do throughout the paper.*

As was mentioned at the beginning of our reply, we use an Atmospheric general circulation model (AGCM)-based Chemistry Transport Model (CTM), which is named "ACTM" in previous publications. ACTM employs nudging to reproduce realistic transport for atmospheric chemical/non-chemical components in model

P1, L4: *Change "a single" to "the chemistry transport model"*

We have changed "a single model" to "the ACTM."

P1, L3–5: *The sentence is unclear. Are there discrepancies between the two models or between models and observations? And the following sentence starts with a "This", but it is unclear what the "this" refers to, to the usefulness, or to discrepancies.*

This sentence was rewritten together with the following sentence without using the word "discrepancies" and "This.": "Since the BIR method is capable of taking unresolved diffusive processes into account, while the Lagrangian method can distinguish the pathways the air parcels took before reaching the sample site, application of the two methods to the common transport field simulated by ACTM is useful in assessing the CO<sub>2</sub> and SF<sub>6</sub> derived mean ages."

P1, L7: *But where is the connection between the water vapour tape recorder and mean age here?*

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This sentence is revised saying that the advantage is "The capability to examine the reproducibility of the observed values of CO<sub>2</sub>, SF<sub>6</sub>, and water vapour".

P1, L8: *Change "the reality" to "good quality" or something alike.*

The phrase "confirming the reality of the trajectory calculations" is deleted.

P2, L12: *Please consider also the newer publication by Engel et al. from 2017 (10.5194/acp-17-6825-2017)*

Thank you for the suggestion. We have updated and cited the suggested paper in the revised manuscript.

P2, L18: *observations and models*

The sentence is revised as "to resolve this discrepancy by reducing uncertainty in both observational and model estimates".

P2, L18: *With "sampling of clock tracers" do you mean SF<sub>6</sub> and CO<sub>2</sub>? Or more tracers? Can you elaborate a little more on the campaign, please, like what, how long...?*

Yes, they are CO<sub>2</sub> and SF<sub>6</sub>. However, we revised the manuscript to distinguish CO<sub>2</sub> and SF<sub>6</sub> from ideal "clock tracers" following the comments from Referee. The sentence is deleted and a brief description about CUBE/Biak campaign added.

P2, L21: *What exactly is it that is puzzling you about these measurements? This should be central in all parts of the paper.*

We are sorry that we did not explain well about our research question. As mentioned in our reply to your major comments above, our research question is clearly written in Introduction.

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P2, L25: *But that is why the Green's function is used to flatten out these nonlinearities. Please see and possibly mention Frisch et al. 2020 (10.5194/acp-2019-974) and citations therein.*

Thank you for the suggestion. We have added the suggested paper for a discussion in the revised manuscript.

P3, L17–18: *So is it a CTM or a GCM now?*

We are sorry for the confusion, but we do hope this question has been already resolved from our reply at the beginning.

P3, L23: *It would be easier to use the name of the model from here on, instead "ACTM", if the model has a name.*

Again we hope this question has been resolved already; "ACTM" is the name of the model used in this study.

P3, L27: *How did the model perform in that inter-comparison? Was it somewhat around the multi-model mean or was it an outlier?*

As the ACTM was nudged to JRA-25 (not to ERA-Interim) in the inter-comparison by Krol et al. (2018), the results need to be interpreted carefully. They found that ACTM showed the strongest convective mixing in the tropics and the young air at the high-altitude poles among the models participated in the comparison. This is stated in Sect. 2.3.

P4, L5: *"several years". Please be precise, for the sake of reproducibility. Did you use ERA-I data of year 2004 for that and repeat that year for ? years?*

Our simulation has been conducted for the period from 1 January 2000 to March 2015 by nudging horizontal winds and temperature to ERA-Interim data. The first five years (January 2000 to December 2004) are regarded as the spin period. This information is given in Sect. 2.1.

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*Sects. 2.3 and 2.4 are just the evaluation of the model. Firstly, this should be reflected in the section headers, and secondly, these sections should be considerably shortened and/or moved partly to the supplement. I suggest to reduce number of figures from 5 to 2.*

Thank you for your suggestions. The whole Sect. 2 has been rewritten by reducing the number of figures to 2. Fig. 4 is moved to Appendix for the sake of readability. Sect. 2.2 is entitled "Evaluation of the model performance" following the suggestion.

*These abbreviations, particularly "AF" and "AN" do not make much sense to me.*

We are sorry for the confusion but these follow our precedent use in BAMS papers. For clarity and readability, those abbreviations have been changed to ACTFREE (for AF) and ACTM-NUDG (for AN). Additionally, "EI" is also changed to "ERA-Interim".

*Fig. 2: I do not fancy that the streamfunction is shown again here, it was shown in Fig. 1 already and does not help much, instead, it disturbs the view on tendencies. I suggest to remove it.*

Thank you for the comment. As suggested by the reviewer, this figure has been removed.

P5, L15: *What do you mean by "selected tracers"?*

The term "selected tracers" was intended to identify pulse tracers released at a specific month such as January. The term is no longer used in the revised manuscript: "The transport features described above are limited to those at a specific time in the Northern winter." (Sect. 2.2)

P7, L1: *...temperature move upward over time.*

The sentence has been removed associated with the revision of the paragraph

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P7, L6: *backward*

Done.

P7, L15–17: *Unclear and awkward phrasing, please rephrase. Plus, what is meant “not simple”?*

The sentence has been rephrased to the following without using “not simple”: “Therefore, it is necessary to address the difference in the definition of the residence time from which age counting is started before making direct comparisons between the two. As is evident from Fig. 1, the excursion of the tropospheric air into the stratosphere depends on tropospheric transport features, including isentropic mixing with the air in the extratropical LS, and thus the mean age counted from the tropical surface is not always a sum of the tropospheric residence time and the mean age counted from the TTL.”

P7, L25–26: *Remove parentheses around dates*

Thank you for the comment. The parentheses are used to identify two-dimensional coordinates on the BIR map in the form (source time, field time). Thus, we would like to retain the parentheses around dates with the following modification: “at  $(t', t) = (\text{March 2007, October 2007})$  and  $(\text{November 2009, June 2010})$ .”

P7, L28: *change “drives the tracers upward” to “intensifies the upward tracer transport”*

This sentence is moved to Appendix B and rephrased to: “the upward tracer transport driven by extratropical pumping is intensified by the secondary circulation”

P7, L31: *“The vertical axis is ....” What is that sentence supposed to mean? It makes no sense to me.*

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This sentence has been removed.

P7, L32: *What is a latitudinal split? Please explain clearly what you talk about. Moreover, why is that important now, you explained the QBO topic already before.*

Again, we are sorry for the confusion. The sentence has been rephrased to: “The deformation of the contours at 3.0, 3.5, and 4.0 years showing wavy structures at the tropics are due to the downward motion associated with the westerly shear of the QBO (Appendix B).” (Sect. 3.1)

P8, L5: *...the spread of the transit times ( $\Delta$ )...*

“( $\Delta$ )” has been inserted as suggested.

P8, L9: *A bundle! Please be more quantitative.*

The sentence has been revised referring to Table 1 for the details of the model parameters of trajectory calculations.

P8, L9: *the spectra of AoA, CO<sub>2</sub> and SF<sub>6</sub> mole fractions? You are mixing up something here, please be specific.*

In this work, the trajectories are used to estimate not only the spectra of stratospheric AoA, but also the CO<sub>2</sub> and SF<sub>6</sub> mole fractions as well as the water vapor mixing ratio by tracking the position of air parcels advected by the 3D wind. The sentence and panels (c) and (d) of Fig. 9 are deleted following the revisions of the manuscript.

P8, L11: *Can you still give a very brief description of the method of analysis please*

Some descriptions are made: “In the Lagrangian method, the age spectra are estimated by counting the transit time  $\tau$  during the advection along each kinematic trajectory since the last passage through the top of the troposphere ( $\text{Tr}_{\text{to}}$ ) (Sect. 2.3) and “The present study tries to resolve disagreements between

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estimates from trajectory calculations and CUBE/Biak observations by increasing the number of trajectories and extending the integration period (Table 1) (Sect. 3.2).

P8, L12–14: *I do not understand what this sentence is supposed to mean. Please rephrase it and sharpen the message.*

It is an important advantage of using the ACTM and is rephrased to: “In addition to using the ERA-Interim analysis directly as 6-hour interval snapshots, the assimilated meteorological field created by nudging its horizontal winds and temperature to the ACTM are also used for trajectory calculations. In this case, one-hour averaged values are used at one-hour intervals (ACTM-NUDG).”

P8, L14: *Where do these additional levels come from?*

Additional pressure levels are set to better represent the Lagrangian cold-point temperature that controls the water transport to the stratosphere. All pressure level data, not restricted to the additional levels, are interpolated from model level data.

P8, L21: *What is CONTRAIL data? Please describe!*

The citation of CONTRAIL data is deleted as the name is not absolutely important. The description is revised to: “The tropospheric reference was derived from direct measurements of air samples collected onboard commercial airliners during the cruise within the area 5° S–5° N and 142° E–150° E at an altitude of 10 km”.

*Fig. 9 Water “vapour”! Or is ice included too? (Throughout the paper!)*

Water vapour is an important constituent to describe the ascending motion in the equatorial stratosphere, although total hydrogen ( $=\text{H}_2\text{O} + 2\text{CH}_4$ ) is a better quantity (Waugh and Hall, 2002, Rev. Geophys.). Trajectory calculations

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frequently used to estimate stratospheric water variations (e.g., Fueglistaler and Haynes, 2005, JGR). The top right panel for water vapour is retained as the right panel of Fig. 5 with description in Sect. 3.2. We have never mentioned ice.

P9, L2 and L5: *You already defined these abbreviations above.*

Thank you for the comment. We have deleted the redundant information.

P9, L3: *But what were the problems in H18? Can you provide a quick introductory paragraph? Without that, it is almost impossible to follow.*

The following descriptions are given in Sect. 3.2: “The present study tries to resolve disagreements between the estimates from trajectory calculations and CUBE/Biak observations by increasing the number of trajectories and extending the integration period (Table 1). Improvements are not limited to these simulation settings. In addition to using the ERA-Interim analysis directly as 6-hour interval snapshots, the assimilated meteorological field created by nudging its horizontal winds and temperature to the ACTM are also used for trajectory calculations. In this case, one-hour averaged values are used at one-hour intervals (ACTM-NUDG).”

P9, L18: *“It is interesting”. Does that mean the other results are not interesting?*

Thank you for the comment. Figure 11 is replaced by a movie provided as Supplementary Material. Related descriptions are made in Sect. 4: “The time evolution of the meridional location of air parcels corresponding to Sample 8 is visualized as a movie in Supplementary Material. We can see that air parcels gradually descend in reverse time sequence, and stay mostly inside the “tropospheric pipe” without appreciable latitudinal dispersion during the Northern winter (January and February 2015). The vertical advection is fastest in ACTM-FREE and slowest in ACTM-NUDG. By June 2014, an appreciable number of ACTM-FREE air parcels has descended back into the troposphere. In contrast, scarcely a

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air parcels are found in the troposphere in ACTM-NUDG. ERA-Interim shows features intermediate between the two.”

P9, L18–32: *Using these abbreviations this way make it almost impossible to follow*

The abbreviations “EI”, “AN”, and “AF” follow the use in our project paper (H1). Now we have changed AN to ACTM-NUDG, AF to ACTM-FREE and EI to ERA-Interim.

P9, L25: *How is that question linked with the general idea of the paper?*

Our purpose is to attain a better understanding on the vertical profiles of  $C_2F_6$  and  $SF_6$  ages obtained by cryogenic air sampling in CUBE/Biak campaign. We tried to reproduce the observed age profiles by applying the BIR and Lagrangian methods to the meteorological fields simulated by ACTM. The question, “What is the advection velocity in EI and AN different in spite of nudging?” is linked to the reproducibility of the observations. It is natural for us to expect that ACTM-NUDG gives similar results to EI (ERA-Interim), as AN is nudged to EI. The related descriptions are made in Sect. 4 as described in our reply to your comment on P9, L18.

P10, L5–9: *Even more abbreviations that totally disturb readability.*

Following this comment, we stopped using  $\Gamma^*$  and  $\Gamma_{adj}$  in the revised manuscript. However, we want to retain the use of  $\Gamma_{corr}$ ,  $\Gamma_{bir}$ ,  $\Gamma_{trj}$ ,  $\Gamma_{Cobs}$ , and  $\Gamma_{Sobs}$  for better readability of the manuscript. We don't think it wise to write, for example, “mean age estimated by BIR method having been applied the tail correction and source-region adjustment” every time for  $\Gamma_{bir}$ .