Reply to Reviewer 1

We thank all three reviewers for their careful considerations of the manuscript and their well thought-out comments. These certainly helped to significantly improve the paper. In the following, we address all comments and questions raised (Reviewer's comments in italics). Text changes in the manuscript are highlighted in color (except minor wording changes). The main concerns of the reviewers were:
(i) 'The manuscript is not very focused.' (Reviewer 3)
(ii) 'The paper is a bit long.' (Reviewer 2)
(iii) 'The discussion is too terse in places.' (Reviewer 1)
(iv) 'The manuscript does a poor job of discussing previous studies.' (Reviewer 3)
We have taken this criticism seriously and applied several changes to the manuscript. To intensify the focus, the paper now concentrates clearly on the main question regarding the variability of lower stratospheric age spectra on seasonal to inter-annual time scales (as suggested by Reviewer 1), and on the additional aspect regarding the effects of residual circulation and mixing on the spectrum from a global perspective. We removed 3 figures (Figs. 5, 10, 15 in the submitted version), two subsections of the discussion (Sects. 6.2, 6.3 in the submitted version), and the discussion of ENSO-related variability in Sect. 5 (suggested by Reviewer 1). The parts from the old Sect. 6.2, concerning variability of the fraction of young air, are now included in Sect. 3, which presents the results regarding seasonality. The revised discussion therefore clearly focusses on the seasonal and inter-annual variability in the age spectrum and how this variability generates the multiple peaks in the spectrum. Regarding inter-annual variability, we only discuss the QBO, as this is the dominant mode of inter-annual variability in the tropical lower stratosphere and its effects on the age spectrum have not been studied in detail, so far. The discussion of QBO-related effects in Sect. 5 is enhanced (as suggested by Reviewer 2) and an additional figure (new Fig. 12) is used, presenting easterly and westerly QBO composites, to further illustrate these effects (as suggested by Reviewer 3). A by-product of these changes is that the revised manuscript now is substantially shorter (as requested by Reviewer 2).
We changed large parts of the text and included 'strategically placed clarifying phrases', as required by Reviewer 1, in order to enhance clarity of the discussion. A new figure (Fig. 2) is introduced, showing the dispersal of the winter/summer tracer pulses, and is referred to in Sect. 2 and in the discussion to illustrate the transport processes discussed. In particular, we thank Reviewer 3 to point out certain passages in the text where the submitted manuscript was not precise about the existing literature. It was definitely not our intention to mix up results of this paper with what is already known. Hence, we carefully revised the manuscript in order to cite existing literature correctly and to clearly state what is known already and what is not.

General comment:
This paper is very well written, enlightening, and concerns an important and timely aspect of atmospheric transport. My primary criticism is that the discussion is too terse in places. I suggest the addition of strategically placed clarifying phrases will help the reader understand the logic of the arguments. In the specific comments, I have noted a few examples where I had a particularly difficult time following the logic. In addition, the discussion of the relationship between ENSO and inter-annual variations of the age spectrum (Figure 10 and the last paragraph of Sec. 5) is not convincing. It should either be improved with some quantitative measures of the information content of that relationship or removed.
Thank you for your encouraging comments. As mentioned in our 'General comment' above, we carefully revised the paper in order to enhance the clarity of the discussion ('strategically placed phrases', e.g. in introduction/paragraphs 4-5, discussion Sect. 6, text referring to new Fig. 2 in Sect. 2/6). To better focus the paper, the discussion of ENSO-related variability has been removed, as suggested.

Specific comments:
P. 2, line 17: *The discussion in this paragraph could greatly enhanced if you briefly explained (1) the motivation for representing transport as a diffusion process and that for representing the age spectrum by a Greens function, (2) the assumption implicit in those representations, and (3) why the assumptions needed for such a representation are not met.*
We rephrased the respective paragraph to be more precise about these points.
P. 2, line 34: *Changing using multiple pulses to using multiple tracer pulses will help clarify what you are doing.*
Done.
P. 6, line 16–19: *Perhaps I am confused here. Shouldnt the mean age calculated from the age spectrum be identical to the value calculated from a perfectly linear clock tracer? Why is there a (small) discrepancy between the two calculations? Is it because, for ages approaching 10 years, the clock tracer does not capture the full range of source concentrations (i.e., the clock does not start early enough)?*
Thanks for pointing out this lack of clarity. Indeed, if the time span for convergence for both the clock-tracer and the age spectrum would be infinity both should be exactly equal. However, the calculations here effectively are of different lengths. We explain this point in detail in a new paragraph at the end of the appendix now (see

also our reply to the 'Minor comment' of Reviewer 3).

P. 6, line 26–27: *It took me a while to decipher the meaning of the phrase coinciding with the youngest peak from spring to fall and the second youngest peak in winter. Perhaps it would help to be more explicit, for example, the modal age as determined by the apex of the largest peak coincides with the youngest peak during spring, summer and fall and with the second peak during winter.*

The respective sentence has been rephrased according to the Reviewer's suggestion.

P. 7, line 9–12: *The description of the propagation of the peak and the argument that the peaks are due to the fact that the most efficient transport from the boundary layer to the stratosphere is too terse. Perhaps another sentence or two will clarify the argument. For example, I can understand why efficient transport during winter leads to a peak in the summertime age spectrum at 6 months. However, I dont understand why the subsequent wintertime peaks are also linked to transport efficiency during winter. It seems to me that once the air is in the stratosphere (i.e., after 6 months) then an additional boundary layer to stratosphere transport boost the following winter is irrelevant; unless an important fraction of the air released during a specified winter remains in the troposphere until the following winter when the troposphere-to-stratosphere transport window re-opens.*

We admit that the wording was not clear. We changed the paragraph, and strongly advise the reader of the discussion (Sect. 6), which has also been changed to enhance clarity (see our 'General comment' above).

P. 7, line 24–25: *Vertical velocity near the tropopause is also slower during summer than during winter (W. Randel and co-authors have written papers on this) and could be important.*

Indeed, the vertical velocity near the tropopause is slower during summer. However, particularly the reduced young air fractions in the summer hemisphere (Fig. 7 in the revised version) indicate that mixing plays an important role. Likewise, enhanced fractions of air older than 2 years directly above the tropopause (Fig. 7 c/d) can only be caused by in-mixing of old extratropical air. However, in order to better focus the manuscript we removed the respective figure (old Fig. 5) and discuss the related issues now with reference to Fig. 7, in a revised paragraph.

P. 10, line 8–20: *The connection between ENSO and inter-annual variations of the age spectra is, perhaps, believable, but is not convincingly demonstrated in Fig. 10. This could be due to the fact that, while equatorial convection patterns shift substantially as sea surface temperature patterns shift, variations of the average strength of tropical convection have relatively weak connections to ENSO. It seems the authors are trying to see make too much of patterns that appear in Fig. 10 a problem that can arise when analysis rely too heavily on a qualitative comparison and the ability of the human eye to recognize patterns in a chaotic system (whether or not the patterns are meaningful). At the very least, Fig. 10 should be changed to make it easier to discern how well the age spectra are related to ENSO. It would be better to make the analysis more quantitative. For example, how much to the inter-annual variance of the age spectra is explained by a lag-relationship with ENSO variability? It might be best to simply remove Fig. 10 and the last paragraph of Sect. 5. Fig. 11: Did you create this plot for the global release experiment? (as opposed to 15S-15N) Does it look the same for both experiments? If so, or even if not, that is an important comparison to make.*

We admit that the presented relation between age spectrum and ENSO variability was too vague. We did some further analyses on that, like investigating the global release experiment which yields very similar results to the presented reference. However, in order to shorten and better focus the paper (as suggested by all Reviewers) we followed the advise of Reviewer 1 and removed the figure and the related text. The discussion of inter-annual variability now focusses on the QBO, as the main mode of inter-annual variability in the tropical lower stratosphere (see also our 'General comment').

P. 11, line 11–12: *Does the distribution in Fig. 11b mean that the particle pulses do not always have the same mass? That the rate of particle release varies? If so, how is this justified physically? Regardless, please explain this figure a bit more.*

The pulse release rate at the surface is not varying over the year. The figure shows the distribution of source times of the air sampled in the stratosphere. This distribution shows a strong peak at NH winter source times, although the release rate at the surface was the same as in summer. We changed the respective paragraph in the discussion to be clearer about this point in the revised version.

Sect. 6.1, last 3 paragraphs: *This discussion could use elaboration. Please be sure that, each time the effect of some phenomenon on peaks is mentioned, the explanation for how (or why) the effect is carried out is clear.*

We revised parts of the discussion to enhance clarity (see also our 'General comment'). Furthermore, reference to the new Fig. 2, showing the dispersal of winter/summer pulses explicitly, should help illustrating the processes described in the text.

P. 12, line 26–7: *Regarding This flushing . . . has implications for the chemical composition . . .. Can you give an example?*

The flushing of the summertime lowermost stratosphere with young air is e.g. important for short-lived species and pollutants. However, in order to better focus and shorten the paper (suggested mainly by Reviewer 3) we removed this part of the discussion (see also our 'General comment')..

Technical details:

P. 2 L. 16: *Many studies of stratospheric age . . .*
The wording has been changed.
P. 3 L. 2: *Change spectra from seasonal . . . to specta on seasonal . . .*
Done.
P. 8 L. 19: *plays a dominant role is an over-statement without further analysis. Change dominant to important. Better yet, use plays a more important role during this season than during NH winter.*
Done.
P. 8 L. 21: *Change The isolation of tropical air through the subtropical transport barriers to imposed by the subtropical transport barriers (or some analogous change) to avoid the contradictory imagery invoked by the words isolation and through.*
The entire paragraph is changed in the revised version, and in particular the wording mentioned above.
P. 9 L. 3: *Change transit times above to transit times longer than.*
Done.

Reply to Reviewer 2 (E. Ray)

We thank all three reviewers for their careful considerations of the manuscript and their well thought-out comments. These certainly helped to significantly improve the paper. In the following, we address all comments and questions raised (Reviewer's comments in italics). Text changes in the manuscript are highlighted in color (except minor wording changes). The main concerns of the reviewers were:
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(iv) 'The manuscript does a poor job of discussing previous studies.' (Reviewer 3)
We have taken this criticism seriously and applied several changes to the manuscript. To intensify the focus, the paper now concentrates clearly on the main question regarding the variability of lower stratospheric age spectra on seasonal to inter-annual time scales (as suggested by Reviewer 1), and on the additional aspect regarding the effects of residual circulation and mixing on the spectrum from a global perspective. We removed 3 figures (Figs. 5, 10, 15 in the submitted version), two subsections of the discussion (Sects. 6.2, 6.3 in the submitted version), and the discussion of ENSO-related variability in Sect. 5 (suggested by Reviewer 1). The parts from the old Sect. 6.2, concerning variability of the fraction of young air, are now included in Sect. 3, which presents the results regarding seasonality. The revised discussion therefore clearly focusses on the seasonal and inter-annual variability in the age spectrum and how this variability generates the multiple peaks in the spectrum. Regarding inter-annual variability, we only discuss the QBO, as this is the dominant mode of inter-annual variability in the tropical lower stratosphere and its effects on the age spectrum have not been studied in detail, so far. The discussion of QBO-related effects in Sect. 5 is enhanced (as suggested by Reviewer 2) and an additional figure (new Fig. 12) is used, presenting easterly and westerly QBO composites, to further illustrate these effects (as suggested by Reviewer 3). A by-product of these changes is that the revised manuscript now is substantially shorter (as requested by Reviewer 2).
We changed large parts of the text and included 'strategically placed clarifying phrases', as required by Reviewer 1, in order to enhance clarity of the discussion. A new figure (Fig. 2) is introduced, showing the dispersal of the winter/summer tracer pulses, and is referred to in Sect. 2 and in the discussion to illustrate the transport processes discussed. In particular, we thank Reviewer 3 to point out certain passages in the text where the submitted manuscript was not precise about the existing literature. It was definitely not our intention to mix up results of this paper with what is already known. Hence, we carefully revised the manuscript in order to cite existing literature correctly and to clearly state what is known already and what is not.

General comment:
This paper uses trajectories from the CLaMS transport model driven by ERA-Interim meteorology to analyze stratospheric age of air spectra. This work provides unique and revealing insights into the stratospheric transport on various time scales and latitude regions. The authors have recently done nice work on explaining various aspects of the stratospheric mean age of air and how the residual mean circulation and isentropic mixing contribute to the observed mean age distributions. But as they mention here, the age spectra provide another level of information, in particular how the transport variability imprints on the age spectra for years afterward. The analysis is excellent and I highly recommend publication with consideration of the minor comments below. The paper is a bit long, 16 figures is quite a lot but I dont have any specific suggestions on what could be left out. Perhaps with fewer figures a bit more time could be spent explaining some of the new and unique features of the remaining plots.
Thank you for your encouraging comments. As explained in our 'General comment' above, we revised the manuscript in order to better focus and shorten it.

Minor comments:
P. 2, line 14: *awkward sentence, change to something like comes with the benefit of allowing one to separate. . .*
Done.
P. 2, line 35: . . .*and allows one to calculate the. . .*
Done.
P. 2: *Just a comment that in my Ray et al. [2014] paper figure 4 shows age spectra from the TLP model with multiple peaks with clear seasonal and QBO influence. We didnt explain what caused the peaks beyond the known variability in the MERRA transport input to the model but thought you might want to include a mention of this paper here.*
We apologize for missing this reference here. A notice is included in the introduction of the revised manuscript version.
P. 7, line 13: . . .*allows one to quantify air. . .*
Done.

P. 8, line 13–15: *The younger tropical spectrum peak in DJF in Figure 6 is hard to see. Ill take your word for it but this figure doesnt seem to back up that statement unless Im missing something.*
We admit that it is difficult to read this difference from the figure. We therefore explicitly state the numbers of the 20N/S average modal age in the text now (3.5 months during NH winter versus 4.1 months during summer).
P. 10, line 1–8: *Figure 9 is really nice and has so many features it takes some time to appreciate them all and what they mean. The propagation of the signals to older parts of the age spectra with time is reminiscent of the tropical tape recorder signal but has a different physical meaning. It's actually difficult to interpret physically whats going on with these signals because its in a different phase space than we're used to thinking about. For instance, in Figure 9c-f there are anomalous peaks and troughs in the spectra that appear to propagate from the time when an event like a QBO easterly phase occurred at ages from 1-2 years for the following 4 years out to ages of 6 years. The actual air masses that were influenced by the QBO transport anomaly move around the stratosphere and many of them actually leave the stratosphere over the following 4 years and yet at this theta level and latitude range there is a signal that remains. It's as though the air influenced by the particular QBO event circulates around and enough of it comes back through this theta and latitude region to maintain an anomalous signal. That's actually remarkable! It might be worth spending a bit more time explaining the physical meaning of this plot and the features since I dont think it's obvious and I'm not even sure I'm getting the full picture.*
We significantly enhanced the discussion of the QBO-related inter-annual age spectrum variability in Sect. 5 (including the new Fig. 12 showing QBO composites), trying to thoroughly address all points raised by the Reviewer.
Figure 5: *in the caption the hemispheres are switched for (b) and (c).*
Yes - however, we removed this figure from the revised manuscript order to shorten the paper. The corresponding discussion now refers to Fig. 7.
Figure 6: *in the caption should be show*
Corrected.
Figure 7: *In the interest of shortening the paper could be one to consider removing.*
We kept this figure, but removed other figures as explained in our 'General comments'.
Figure 9: *Really interesting as mentioned above and a lot going on here. My suggestion to be able to see the features more clearly on b,d and f primarily is to separate out the delta age, mode and RCTT that are on the right axis into their own plots. Its hard to see their oscillation around the zero line as it is and it obscures somewhat the propagation of the pdf anomalies.*
We admit that the figure includes a lot of information. However, we think it is good to have the delta age, mode and RCTT and age spectra together in one figure to be able to relate them easily, without the need to compare different figures. As the modal age anomalies for the extratropics (Fig. 13 d/f) are characterized by particularly large scatter and contain no substantial additional information we removed these from the respective panels, following the Reviewer's advice. Furthermore, we enlarged Fig. 13 a bit to enhance readibility.
Figure 14: *Is each of the 3 lines of each color an individual month within DJF?*
Each line is the average over DJF, with different age spectra of the same color representing different latitudes within the respective latitude band (corresponding to the respective color). We state this explicitly in the caption now.

Reply to Reviewer 3

We thank all three reviewers for their careful considerations of the manuscript and their well thought-out comments. These certainly helped to significantly improve the paper. In the following, we address all comments and questions raised (Reviewer's comments in italics). Text changes in the manuscript are highlighted in color (except minor wording changes). The main concerns of the reviewers were:
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We have taken this criticism seriously and applied several changes to the manuscript. To intensify the focus, the paper now concentrates clearly on the main question regarding the variability of lower stratospheric age spectra on seasonal to inter-annual time scales (as suggested by Reviewer 1), and on the additional aspect regarding the effects of residual circulation and mixing on the spectrum from a global perspective. We removed 3 figures (Figs. 5, 10, 15 in the submitted version), two subsections of the discussion (Sects. 6.2, 6.3 in the submitted version), and the discussion of ENSO-related variability in Sect. 5 (suggested by Reviewer 1). The parts from the old Sect. 6.2, concerning variability of the fraction of young air, are now included in Sect. 3, which presents the results regarding seasonality. The revised discussion therefore clearly focusses on the seasonal and inter-annual variability in the age spectrum and how this variability generates the multiple peaks in the spectrum. Regarding inter-annual variability, we only discuss the QBO, as this is the dominant mode of inter-annual variability in the tropical lower stratosphere and its effects on the age spectrum have not been studied in detail, so far. The discussion of QBO-related effects in Sect. 5 is enhanced (as suggested by Reviewer 2) and an additional figure (new Fig. 12) is used, presenting easterly and westerly QBO composites, to further illustrate these effects (as suggested by Reviewer 3). A by-product of these changes is that the revised manuscript now is substantially shorter (as requested by Reviewer 2).
We changed large parts of the text and included 'strategically placed clarifying phrases', as required by Reviewer 1, in order to enhance clarity of the discussion. A new figure (Fig. 2) is introduced, showing the dispersal of the winter/summer tracer pulses, and is referred to in Sect. 2 and in the discussion to illustrate the transport processes discussed. In particular, we thank Reviewer 3 to point out certain passages in the text where the submitted manuscript was not precise about the existing literature. It was definitely not our intention to mix up results of this paper with what is already known. Hence, we carefully revised the manuscript in order to cite existing literature correctly and to clearly state what is known already and what is not.

General comment:
This manuscript presents calculations of the seasonal and inter annual variations in the stratospheric age spectrum obtained from the CLaMS model driven by ERA-Interim winds. The manuscript contains some interesting and new results that certainly warrant publication. However, the manuscript is not very well focused, does a poor job of discussing previous studies, and some of the major conclusions (as stated in abstract or conclusions) are not new results. I think the manuscript needs to be revised to be more focused on the new aspects of their calculations (seasonal and inter annual variability, including QBO) and to put these in context of previous studies.
We appreciate the constructive criticism. As explained in our 'General comment' above, we significantly changed the paper (mainly introduction, Sects. 3, 5 and discussion) in order to better focus on seasonal and inter-annual variability, and in order to correctly cite the existing literature and to state what is known and what is new (see reply to the detailed concerns below).

Major comments:
1: *The manuscript is not very focused, and the new results are not clearly presented. This lack of focus can be seen from the title, which is very vague (and doesnt match much of the manuscript). In my opinion the relatively new and important aspect of the study is looking at seasonality and inter annual variability of the age spectrum, which has only been done in a few previous studies, and none of the previous studies have included the QBO (as the authors highlight in the Introduction). However there is actually relatively little discussion of these aspects, and there is just as much (or even more) discussion is on the age spectrum - mean age relationship, the approximation of G by inverse Gaussian, and comparisons of residual circulation with modal age. All of these latter issues could be examined using steady flows, and much of what is discussed is already known. My recommendation is to focus on the seasonal and inter annual variations (and QBO) and to minimize the discussion of the other issues.*
The manuscript is significantly changed to focus more clearly on seasonality and inter annual variability of the age spectrum, as suggested (see also our 'General comment'). We also changed the title to be more precise about that ('Seasonal and interannual variability of lower stratospheric age of air spectra'). The discussion of the approximation of G by an inverse Gaussian has been removed, and the main message condensed into the

last sentence of the discussion. The discussion subsection on the age spectrum - mean age relationship has also been removed. The seasonality of young air fractions (which was previously included in this discussion part) is now presented in the seasonality section 3.

2: *While there are references to previous studies that examined similar aspects of the age spectrum in the Introduction there is very little discussion of these when interpreting the calculations presented here. Because of this it is not clear to me how much of the results are new and how much are just reproducing earlier results with Lagrangian model driven by reanalyses. The clearest examples of lack of discussion of previous studies are Sections 3 (seasonality) and 5 (inter annual variability) where there is not a single mention of the Li et al. 2012a,b studies which examined exactly these issues. How do the results presented compare with these previous studies? What is new in what is presented (other than a slightly different approach)?*

Thank you for the constructive criticism! We have tried to improve the discussion of previous literature throughout the text. Examples are:

(i) The introduction, where we now explain in detail that most existing age spectrum studies are based on the assumption of a stationary flow, and that only a few recent ones considered time-dependent age spectra (e.g., Reithmeier et al., 2007; Diallo et al., 2012; Li et al., 2012a/b). We frequently refer to these studies in the introduction, in Sect. 2 and in the discussion. Furthermore, in the revised version we also explain at the beginning of Sect. 3 that the occurrence of multiple peaks in the age spectrum has already been pointed out in recent publications, and we discuss probable causes in the discussion (Sect. 6) in relation to the literature (e.g., Reithmeier et al., 2007; Diallo et al., 2012; Li et al., 2012a/b). In the discussion, we refer first (P13, L9ff) to the discussions of the peaks in these papers, explain that there is no common understanding, and finally discuss the CLaMS results in relation to them. The fact that in the reanalysis-driven CLaMS simulation multiple spectrum peaks emerge not only at high latitudes but almost globally is clearly different to previous studies, and thoroughly discussed now. As further discussed, we think that it is neither of the recently proposed mechanisms which causes these peaks, but a combination of them (see discussion).

In particular, our analysis is the first full transport model analysis on age spectra based on reanalysis winds including a QBO. Such inter-annual variability can be easily investigated using our modified BIER approach, as it is based on a transient multi-year simulation (Li et al., 2012a/b for instance considered only time-slice experiments). The clear effect of the QBO in modifying and even generating age spectrum peaks is clearly a new result.

Furthermore, in the introduction we now explain precisely (P3, L12ff) that in the tropics the modal age is known to be closely related to the residual circulation, but that no global investigations exist on that, to our knowledge. Hence, our global analysis on regions and seasons where modal age is controlled by the residual circulation (the tropics and the wintertime extratropical stratosphere above about 500 K) is, in our opinion, also a new result.

3: *Some of the conclusions (as stated in the abstract) are well known facts. This paper may be providing more support, but as written it appears these are new results. One example is the statement in the abstract that Interpretation of the age spectrum in terms of transport contributions due to the residual circulation and mixing is generally not straightforward. This is well known and not sure this counts as a significant enough statement for an abstract. Another example are the statements towards the end of both the abstract and conclusions regarding benefits of age spectrum calculations and need for inclusion in model inter comparisons. Again not new, and in fact age spectrum calculations were actually included in model inter-comparisons in late 1990s (Hall et al. 1999). Is this really the major take home message from this manuscript (ending abstract and conclusions with this gives this impression)?*

As stated above (see our 'General comments'), we tried to better focus the paper on seasonal and inter-annual age spectrum variations. We also removed two sentences from the abstract to be more focussed here. We kept the last part of the conclusions, although it is indeed not new that considering the age spectrum is beneficial compared to mean age (the full skewed spectrum always includes more information than just its first moment). We included citations of two existing examplary papers on that (Hall, 1999; Waugh and Hall, 2002). However, time-dependent age spectrum diagnostics have not been implemented in global models as standard transport diagnostics, hitherto. While advantages of considering the full age spectrum have certainly been stressed many times before, most studies in the last $\sim$10 years focus still on mean age, which is a quantity very dangerous to interprete. We therefore feel that it cant hurt to (re-)stress the benefits of age spectra analyses, and to conclude the paper with a related (slightly modified) paragraph.

4: *There are multiple places where I think previous papers or the current understanding are misrepresented. Examples:*

P. 1, line 24: *As written it suggests that the Hall and Plumb 1994 and Waugh and Hall 2002 papers did not appreciate that there was a range of pathways and an age spectrum, whereas the opposite is true and Hall and Plumb focused on this fact.*

This misrepresentation was certainly not intended - we fully agree about the merits of these previous studies. We placed the respective citation to a different place not to cause confusion.

P. 2, line 6: *I dont think it is correct to indicate that the apparent disagreement between observed and models changes in*

transport is due to age spectrum versus mean age differences. This disagreement occurs if you compare mean age from observations with mean age from models, so not a residual circulation vrs mean age issue.
Agreed, although the representation of mixing, and in particular corresponding long-term changes, is likely less robust across models. We have restructured this paragraph somewhat, which should emphasize the structural BDC changes more.
P. 2, line 28: *Is it a common view that modal age can be related to residual mean mass circulation? Maybe in the tropical lower stratosphere, but I dont it is common to think such a relationship extends beyond this region.*
We admit that we were imprecise with this statement. In the tropics this relation between the mode and residual circulation is known. However, globally no study on this relation exists to our knowledge. Therefore, our results confirm the common view in the tropics and further show that a similar relation holds in the wintertime extra-tropical stratosphere above about 500 K, whereas particularly in the summertime lower stratosphere mixing effects are important. We changed the text at several places in order to be precise (e.g., P3, L12ff).
P. 13, line 19: *It is not correct to say that Hall and Plumb (1994) argued that the stratospheric age spectrum may be well approximated by the Greens function for a one-dimensional diffusion process. They used the 1 diffusion model to illustrate aspects of the age spectrum not to model the actual stratospheric age spectrum (they are explicit about this: our goal at this point is not to quantify stratospheric transport, but rather to illustrate the points of the previous discussion).*
Thanks for pointing the confusing statement out! We entirely agree with the Reviewer's view and changed the text accordingly (corresponding text changes at two places in the revised manuscript: introduction/L20ff, and first paragraph of Sect. 3).
P. 14, line 7: *This process can evidently not be described by simple one-dimensional diffusion. I think this well known, not only from looking at G(t) from other threedimensional models, but also from the tropical leaky pipe model where expressions for the age spectrum have been derived (Hall 2000) (see also Waugh and Hall (2002) review).*
We agree with the Reviewer. However, the entire discussion part about age spectrum fits has been removed from the revised manuscript, in order to better focus the paper (as suggested by the Reviewer, see also out 'General comments').

Minor comments:
P. 6, line 15–19: *How long was the clock tracer run for; is it in (quasi-)steady state? It is stated that the agree is good but in polar regions the clock tracer is younger by over 0.5 yrs. Note, the paper of Hall and Haine (2002) might be relevant here. They derive the relationship between ideal age (which is an alternate clock tracer) and mean of age spectrum. In their example the clock tracer converges quicker than the mean of the age spectrum, which appears opposite to your result. However, this may because you have run calculations for different length.*
Thanks for pointing out this lack of clarity. Indeed, the calculations for the two quantities effectively have a different length. The clock-tracer was subject to a 10 year spin-up (repeating 1979 conditions) and another 10 years of transient simulation (1979-1988) before taking into consideration, whereas the age spectrum has explicitly been calculated over 10 years of transit time (1979-1988 simulation) but with the tail fitted to infinite transit times. Hence, the effective calculation length is longer for the corrected age spectrum than for the 'clock-tracer', and there is no contradiction to the results of (Hall and Holzer, 2002). This is explained in detail now in a new paragraph at the end of the appendix (see also our reply to Reviewer 1).
Eq. 7: *Why reference Bonisch et al 2009? As you have just mentioned the G(t) for 1D advection diffusion was used in Hall and Plumb, and this same expression was given in the Waugh and Hall (2002) review paper. Furthermore, this is not a parameterization, this is the exact G(t) just expressed in terms of the mean and width (rather than flow velocity and diffusion).*
We changed the text according to the Reviewer's suggestion, and refer to Waugh and Hall (2002) in the revised paper.
Section 5: *Is it possible to make composites on east and west phase of the QBO?*
Thanks for this suggestion. We present composites for easterly and westerly QBO phases now in the new Fig. 12 at the beginning of Sect. 5 (inter-annual variability). These composites clearly illustrate how the fraction of young air is increased during QBO easterly phase, due to anomalously strong and fast tropical upwelling, and how the spectrum tail becomes more pronounced during QBO westerly phase. Remakably, secondary peaks develop in the tropical age spectrum at older transit times during QBO westerly phase, indicating an increased impact of in-mixing of old extratropical air on tropical composition. We discuss these findings in Sect. 5, and enhanced the discussion about the QBO-related age spectrum variability.
P. 11, line 16: *Are the results in Orbe et al. (2014) relevant for this discussion?*
Indeed, the fact that pulses released during summer are more efficiently dispersed meridionally before they reach the tropical pipe and therefore are less likely to undergo recirculation in the stratosphere would be consistent with a stronger return flux into the troposphere for air entering the stratosphere in July compared to January, as found by Orbe et al. (2014). We mention this in the revised discussion version now. However, it is not clear how much this comparison really tells because of the different pulse tracer settings (this paper:

released at the surface every other month; Orbe et al.: released at the thermal tropopause in January and July)

Seasonal and inter-annual variability of lower stratospheric age of air spectra

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Abstract. Trace gas transport in the lower stratosphere is investigated by analysing seasonal and inter-annual variations of the age of air spectrum – the probability distribution of stratospheric transit times. Age spectra are obtained using the Lagrangian transport model CLaMS driven by ERA-Interim winds and total diabatic heating rates, and using a time-evolving boundary-impulse-response (BIER) method based on multiple tracer pulses. Seasonal age spectra show large deviations from an idealized stationary uni-modal shape. Multiple modes emerge in the spectrum throughout the stratosphere, strongest at high latitudes, caused by the interplay of seasonally varying tropical upward mass flux, stratospheric transport barriers and recirculation. ~~The annual mean spectrum, on the other hand, is found to be well described by an idealized stationary spectrum.~~ Inter-annual variations in transport (e.g., QBO) cause significant modulations of the age spectrum shape. In fact, one particular QBO phase may determine the spectrum's mode during the following 2-3 years. Interpretation of the age spectrum in terms of transport contributions due to the residual circulation and mixing is generally not straightforward. Advection by the residual circulation turns out to represent the dominant pathway in the deep tropics and in the winter hemisphere extratropics above 500K, controlling the modal age in these regions. In contrast, in the summer hemisphere, particularly in the lowermost stratosphere, mixing represents the most probable pathway controlling the modal age. ~~Analysis of the full age spectrum compared to mean age is highly beneficial for separating the effects of different transport processes, and is strongly recommended as a diagnostic for model inter-comparisons.~~

1 Introduction

The composition of the lower stratosphere includes radiatively active trace gases such as water vapour and ozone, which strongly affect the Earth's radiation budget and surface temperatures (e.g., Riese et al., 2012; Solomon et al., 2010). The trace gas distribution in this region is strongly shaped by the global-scale Brewer-Dobson circulation, which may be separated into a residual mean meridional mass circulation and additional eddy mixing (e.g., Holton et al., 1995; Butchart, 2014). Both the residual circulation and mixing are largely driven by breaking Rossby waves and, to some extent, gravity waves (e.g., Haynes et al., 1991; Haynes and Shuckburgh, 2000).

A commonly used diagnostic to study transport in the lower stratosphere is the mean age of air, the average transit time of a stratospheric air parcel since entering the stratosphere. However, stratospheric air parcels are affected by various mixing pro-

cesses along their pathways. Hence, an air parcel consists of a mixture of air with different transit times and is more fully characterized by a transit time distribution, commonly referred to as the *age spectrum* (e.g., Hall and Plumb, 1994; Waugh and Hall, 2002). Mean age of air (the first moment of the age spectrum) only provides a succinct description of stratospheric transport for narrow and nearly symmetric age spectra. Generally, the effects of different transport processes (e.g., residual mean mass transport and mixing) cannot be distinguished with a single measure such as mean age and may lead to puzzling results (e.g., Waugh and Hall, 2002).

One such puzzling result concerns potential changes in stratospheric transport in a changing climate. Climate models indicate a strengthening residual circulation causing decreasing mean age during recent decades and into the future (e.g., Butchart et al., 2010), whereas observations indicate weakly increasing or statistically insignificant mean age trends, depending on the region and length of the data set (e.g., Engel et al., 2009; Stiller et al., 2012; Haenel et al., 2015). Recently, progress has been made to reconcile these apparently contradictory results. When dividing the Brewer-Dobson circulation into a shallow and a deep branch, evidence for a strengthening circulation is found for the shallow branch from both models (e.g., Garny et al., 2011) and in-situ observations (Bönisch et al., 2011; *Ray et al.*, 2010; Ray et al., 2014). Reanalyses indicate a long-term strengthening of the residual circulation (Abalos et al., 2015), which causes decreasing mean age. However, strong decadal variations and, in particular, changes in eddy mixing have been found to mask these long-term residual circulation driven age changes (Ploeger et al., 2015a).

Consideration of the full age spectrum compared to just the mean age comes with the benefit of allowing one to separate the effects of different transport processes and may ultimately lead to an improved understanding of stratospheric transport and its long-term changes. Most existing studies on stratospheric age spectra are based on the assumption of stationary atmospheric flow and approximate the age spectrum by the Green's function for the diffusion process, an inverse Gaussian distribution (e.g., Andrews et al., 2001; Schoeberl et al., 2005; Bönisch et al., 2009). Such an approximation is based on the fact that for tracers of long enough chemical lifetimes stationary stratospheric transport reduces to a 1D flux-gradient relationship (Plumb and Ko, 1992). The 1D diffusion Green's function has been used in the pioneering work of Hall and Plumb (1994) to illustrate the main characteristics of stratospheric age spectra. These studies lead to insights into a number of characteristics of stratospheric transport, including the relationship between mean age and the age spectrum. Consideration of the age spectrum has furthermore aided model intercomparisons of stratospheric transport (Hall, 1999). However, assuming stationary flow is a strong simplification as stratospheric transport shows variations on multiple time scales (e.g., seasonal, inter-annual) and is clearly non-stationary (e.g., Haine et al., 2008).

A few recent studies calculated time-dependent age spectra for non-stationary flow from Lagrangian back trajectories (Reithmeier et al., 2007; Diallo et al., 2012). All of these above mentioned studies based on back trajectories, however, did not incorporate the effects of small-scale mixing on transport (i.e., mixing of air masses between trajectories). The only published analyses, to our knowledge, of non-stationary age spectra for complete 3D transport (including mixing) are Li et al. (2012a), focussing on seasonality, and Li et al. (2012b), focussing on long-term trends. These studies found significant differences of seasonal age spectra from the idealized stationary shape, with the age spectrum within particular regions even showing multiple peaks (modes). However, their explanations for these seasonal variations and the multiple peaks are different, involv-

ing seasonality in tropical upward mass flux, the strength of the polar vortex transport barrier, or the existence of different circulation branches. A common understanding of the intricate age spectrum characteristics appears to be lacking. Moreover, regarding inter-annual age spectrum variations the only published results (Li et al., 2012a), to our knowledge, are based on a model simulation without a quasi-biennial oscillation (QBO). Hence, age spectrum modulations by the QBO have not been studied in detail hitherto, although an indication of such modulation is contained in Ray et al. (2014, their Fig. 4).

Figure 1 illustrates several of the above mentioned age spectrum characteristics, highlighting additional insights that can be gained from consideration of the full age spectrum. The figure presents climatological wintertime age spectra along a given mean age contour of 2.5 years, calculated with the CLaMS model as described in Sect. 2. In general, stratospheric age spectra are characterized by skewed distributions with a long tail at old ages. As age spectra here are calculated for time-dependent flow, without any stationarity assumptions, these spectra show clear multi-modal shapes, with a strong regional dependence (as discussed in Sect. 3 and Sect. 6). Clearly, the spectrum shape significantly varies for the same mean, with a narrow spectrum at low latitudes and substantially broader spectra at higher latitudes. Quantifying the amount of young air and its variability and changes is important for understanding the transport of short-lived chemical species and pollution into the stratosphere (e.g., Schoeberl et al., 2005). This information requires evaluation of the full age spectrum and cannot be estimated from the mean age alone (e.g., Waugh and Hall, 2002).

The modal age, defined as the transit time of the age spectrum peak, by definition corresponds to the most probable transit time. The occurrence of multiple peaks (potentially of similar size) in time-varying age spectra indicates the challenging nature of interpreting the modal age correctly. Within the tropics, modal age is known to be closely related to the residual mean mass circulation (e.g., Waugh and Hall, 2002; Li et al., 2012b). In other regions of the stratosphere, where mixing processes have a predominant effect on transport (e.g., in the NH lower stratosphere during summer, see Konopka et al., 2015), this relation is expected to break down. However, no global analysis exists to our knowledge which systematically investigates the relationship between modal age, residual circulation strength, and mixing.

In this paper, we present a method to calculate stratospheric age spectra in the Chemical Lagrangian Model of the Stratosphere (CLaMS), a state-of-the-art reanalysis-driven chemistry transport model. This method is a further development of the approach by Li et al. (2012a), based on boundary impulse responses using multiple tracer pulses launched at the Earth's surface in the deep tropics, and allows one to calculate the age spectrum in a transient simulation without additional assumptions such as the smallness of inter-annual variability. The analyses focus on the following questions: (i) How large is the variability of lower stratospheric age spectra on ~~from~~ seasonal to inter-annual time scales and how do multiple spectral peaks (modes) develop? (ii) How do residual circulation and mixing affect the age spectrum globally? ~~(iii) How accurate are commonly used approximations of the age spectrum shape?~~

We describe our methodology in Sect. 2 and consider seasonal age spectrum variations in Sect. 3. The effects of residual circulation transport and mixing are investigated in Sect. 4. Inter-annual variability is discussed in Sect. 5. A discussion of the development of multiple modes in the spectrum ~~, the benefit of consideration of the full spectrum compared to mean age alone, and the goodness of commonly used age spectrum fits~~ is presented in Sect. 6. The final section concludes the paper.

2 Methodology

2.1 The CLaMS model simulation

The model used in this study is the Chemical Lagrangian model of the Stratosphere (CLaMS). CLaMS is a Lagrangian transport model with trace gas transport based on the motion of three-dimensional forward trajectories and an additional parametrization of small-scale atmospheric mixing (e.g., McKenna et al., 2002; Konopka et al., 2004). This mixing parametrization induces strong mixing in regions of large flow deformations. Vertical transport in CLaMS is based on a hybrid σ–θ coordinate, which transforms smoothly from an orography-following coordinate ($\sigma = p/p_s$, with p pressure and p_s surface pressure) near the surface into potential temperature θ (see also Mahowald et al., 2002). Above $\sigma = 0.3$, hence throughout the stratosphere and the TTL, the hybrid vertical coordinate exactly equals potential temperature and the vertical velocity is determined by the total diabatic heating rate. Further details about this particular model set-up are described in Pommrich et al. (2014).

For this study, we carried out a simulation for the 1979–2013 period, with model transport driven by European Centre for Medium-Range Weather Forecasts (ECMWF) interim Reanalysis (ERA-Interim) winds (e.g., Dee et al., 2011). Furthermore, we implemented a method to calculate the age of air spectrum within the model, which will be described in detail next.

2.2 Age spectrum calculation

The solution to the continuity equation for a conserved and passive tracer with mixing ratio χ at location r and time t may be expressed as (e.g., Waugh and Hall, 2002):

$$\chi(r,t) = \int_0^\infty \mathrm{d}\tau\, \chi(\Omega, t-\tau) G(r,t\,|\,\Omega, t-\tau)\,. \tag{1}$$

The kernel $G(r,t\,|\,\Omega, t-\tau)$ is called the *boundary propagator*. This propagator is related to the transport operator's Green's function (see Holzer and Hall, 2000), and relates the tracer mixing ratio at (r,t) to the mixing ratio in the boundary source region Ω (chosen to be the tropical boundary layer, in the following) a time $\tau = t - t'$ ago. Here, t is called the *field time* when the tracer mixing ratio is sampled, whereas t' is called the *source time* when the tracer had last contact with Ω. Interpreting Eq. (1), $G(r,t\,|\,\Omega, t-\tau)\mathrm{d}\tau$ is the mass fraction of air at (r,t) that had last been in contact with Ω between τ and $\tau + \mathrm{d}\tau$ ago. For this reason, $G(r,t\,|\,\Omega, t-\tau)$ is a transit time distribution and has been termed the *age spectrum* by Hall and Plumb (1994). Note that the propagator G can be interpreted as a joint air mass origin and transit time distribution, such that integration over τ yields the mass fraction of air originating from Ω (e.g., Orbe et al., 2013).

For an inert tracer with a pulse in Ω at source time $t' = t'_0$ the source time history is given by $\chi(\Omega, t') = \delta(t' - t'_0)$, and Eq. (1) reduces to (recalling $t' = t - \tau$)

$$\chi(r, \tau + t'_0) = G(r, \tau + t'_0\,|\,\Omega, t'_0)\,. \tag{2}$$

Here, $G(r, \tau + t'_0\,|\,\Omega, t'_0)$ is the *boundary impulse response* (BIR), the time-evolving response at location r to a δ-distribution boundary condition in Ω at time t'_0. In general, as a function of transit time τ the BIR is not equal to the age spectrum, because

the τ-dependency occurs in different arguments of G. Only for stationary flow the boundary propagator is time translation invariant and depends only on the transit time τ, such that the age spectrum and BIR are equal (e.g., Haine et al., 2008).

Equation (2) provides a method to calculate the boundary propagator G by using N different pulse tracers χ_i ($i = 1, \ldots, N$), with pulses in Ω at times t_i' (e.g., Haine et al., 2008). The age spectrum, as a function of τ, may be constructed from these N pulse tracers at each field time t and location r as

$$G(r, t \,|\, \Omega, t - \tau_i) = \chi_i(r, t)\,. \tag{3}$$

Hence, from N pulse tracers N pieces of information on the age spectrum at the discrete transit times $\tau_i = t - t_i'$ may be deduced. Recently, this BIR-method was used to investigate the seasonality (Li et al., 2012a) and long-term behaviour (Li et al., 2012b) of stratospheric age spectra in the Goddard Earth Observing System Chemistry-Climate Model (GEOSCCM).

Here, we use a total of $N = 60$ different boundary pulse tracers, with pulses released in the lowest model layer (orography following) in the tropics between 15°S–15°N, constituting the source region Ω. This source region has been chosen similar to that of Li et al. (2012a) for ease of comparison. Variations of the source region (e.g., 10°S–10°N, entire global surface layer) lead to qualitatively the same results. Note that another common choice, particularly for observationally based age spectrum estimates, is to define Ω as the tropical tropopause. Related differences for transit times are expected to be a few weeks to months, which is the time scale for transport from the surface to the tropopause. For each pulse, the particular tracer mixing ratio is set to unity in Ω for 30 days, and is set to zero in Ω otherwise. Pulses are launched every other month. Consequently, for the considered period 1979–2013 the first tracer pulse has source times in January 1979, the second tracer pulse in March 1979, and so on.

Figure 2 shows the evolution of the tracer mixing ratio in the meridional plane during the months following the pulse release. The dispersal of the tracer plume illustrates the known features of the global scale stratospheric Brewer-Dobson circulation. During the first months after release, the tracer–tagged air disperses throughout the troposphere and slowly ascends across the tropical tropopause into the stratosphere, with this tropical upwelling being stronger in NH winter than summer. Above the tropopause, the air is mixed rapidly (to high latitudes), with this mixing being stronger in the summer hemisphere (particularly for NH summer). In the winter hemisphere, meridional mixing to high latitudes is suppressed due to the existence of the polar vortex transport barrier. One year after release, the tracer has largely been washed out in the extratropical lower stratosphere via downward transport into the troposphere. In the tropics, on the other hand, the pulse air further ascends slowly within the tropical pipe (above about 450 K), well isolated from further mixing with the extratropics. For air masses leaving the surface during winter, a substantially higher fraction reaches the stratosphere compared to air released in summer (see two right columns in Fig. 2). In particular, after 3 years almost twice as much winter surface air is found in the stratosphere compared to summer surface air. We will further discuss this seasonal difference in Sect. 6.

The age spectrum calculation method in the transport model CLaMS is further illustrated in Fig. 3, which shows the BIR map at 400 K and 60°N, i.e. the propagator G versus field time and source time. Following the initial pulse at source time t', each pulse tracer mixing ratio evolves with field time t producing a vertical section in the BIR map. The set of all tracers comprises the BIR map. Therefore, a vertical cut through the BIR map provides the boundary impulse response at a given

source time t' as a function of field time t (e.g., the vertical dashed arrow shows the BIR for January 1995). A horizontal cut through the BIR map provides the age spectrum at a given field time as a function of source time, or equivalently of transit time by noting that $\tau = t - t'$ (e.g., the horizontal arrow shows the age spectrum for June 2002).

Due to the finite number of tracers used here ($N = 60$) initialized every other month in source time (i.e., 6 pulses per year for 10 years), the calculated age spectrum is truncated to a maximum transit time of ten years. We used results of a ten year long perpetual simulation (repeating 1979 winds) as initialization for the transient simulation starting at 1 January 1979. Therefore, values in the BIR map at source times before 1 January 1979 (and field times before 1989) will be influenced by this initialization procedure. For this reason, we do not consider age spectra before 1989 in this paper, and calculate climatologies for the 1989–2013 period. Once all sixty tracers have been used (after the first ten years) the first tracer is reset to zero and then used for the pulse at source time January 1989, and so on for consecutive pulses. Hence, we obtain time-dependent age spectra at every day of the transient 1979–2013 simulation, with a resolution along the transit time axis of 2 months. Li et al. (2012a) showed that a 2-month resolution of the pulses is suitable for resolving the seasonal age spectrum variations, and it is therefore sufficient for the results presented in this paper.

Our implementation of the age spectrum method in CLaMS differs from the BIR calculation by Li et al. (2012a) and Li et al. (2012b), who considered time slice simulations and used each tracer only once. To emphasize the fact that the modified BIR tracer setting in CLaMS evolves with time in a transient simulation, we denote the age spectrum calculation as implemented here the *Boundary Impulse Evolving Response* (BIER) approach.

Figure 3b shows the annual mean age spectrum at 400 K in NH mid-latitudes at 60°N, the horizontal cut through the BIR map in Fig. 3a. ~~In general, stratospheric age spectra are characterized by skewed distributions with a long tail at old ages.~~ The maximum of the spectrum (*modal age*, here highlighted as vertical black dashed line) corresponds to the most probable transit time and likely to the most probable pathway (to be discussed further below). The *mean age* (vertical black solid line) is defined as the first moment of the age spectrum

$$\Gamma(r,t) = \int_0^\infty \mathrm{d}\tau\, \tau\, G(r,t\,|\,\Omega, t-\tau) \tag{4}$$

and strongly depends on the tail of the distribution. Another quantity characterizing the distribution is the age spectrum width

$$\Delta(r,t) = \sqrt{\frac{1}{2}\int_0^\infty \mathrm{d}\tau\, [\tau - \Gamma(r,t)]^2\, G(r,t\,|\,\Omega, t-\tau)}\,. \tag{5}$$

In addition, the red dashed line shows the residual circulation transit time (RCTT), the hypothetical transit time of an air parcel if it was advected by the residual circulation only (e.g., Rosenlof, 1995; Birner and Bönisch, 2011). RCTT's and their relation to mean age of air have recently been discussed by Garny et al. (2014). Here, RCTT's are calculated from 2D CLaMS backward trajectories in the latitude-potential temperature plane, driven by the ERA-Interim residual circulation in isentropic coordinates $(\overline{\mathrm{v}}^*, \overline{\mathrm{Q}}^*)$, as described by Ploeger et al. (2015b). In this isentropic zonal mean formulation $\overline{\mathrm{v}}^* = (\overline{\sigma v})/\overline{\sigma}$ and $\overline{\mathrm{Q}}^* = (\overline{\sigma Q})/\overline{\sigma}$ are the mass-weighted zonal mean meridional and vertical velocities, where $\dot{\theta} = \mathrm{Q}$ is the cross-isentropic vertical velocity and $\sigma = -g^{-1}\partial_\theta p$ is the density in isentropic coordinates, with p pressure and g the acceleration due to gravity (e.g., Andrews et al.,

1987, Chapter 9). The RCTTs are calculated with respect to the 340 K surface. This causes a weak young bias of the RCTTs compared to age of air, corresponding to the transit time from the Earth's surface to 340 K. The relation between the age spectrum and the RCTT will be studied in Sect. 4.

A limitation of the described approach to calculate the age spectrum originates from the limited number of pulse tracers in the model, such that only the first ten years of the spectrum are calculated explicitly here. Because the mean of the distribution strongly depends on the spectrum's tail this leads to an underestimation of mean age. However, for transit times above about 4-5 years age spectra are generally found to decay roughly exponentially (cf. Fig. 3c). The corresponding decay rate is related to the exponential decay of the mixing ratio of a conserved tracer in the stratosphere (e.g., Ehhalt et al., 2004). To estimate the age spectrum's tail for transit times larger than ten years, the spectrum may therefore be extrapolated using an exponential fit based on the values at transit times between 5-10 years (red dashed line in Fig. 3c), as explained in detail in Appendix A. A similar tail correction procedure was applied by Diallo et al. (2012). Throughout this paper we present the tail-corrected mean age.

Global mean age distributions for NH winter (DJF) and summer (JJA), as obtained from age spectra, are shown in Fig. 4, confirming well-known characteristics of the stratospheric Brewer-Dobson circulation. A stronger circulation during NH winter causes younger ages in the tropics, compared to the boreal summer season. Mean age is older in the winter than in the summer extratropics, due to stronger Brewer-Dobson circulation downwelling in the winter hemisphere and weaker transport barriers in the summer hemisphere. Particularly young mean age is found in the NH lower stratosphere during summer as a result of strong quasi-horizontal mixing during this season (Konopka et al., 2015). The effect of the correction for the finite spectrum tail is an increase of mean age of about half a year compared to the uncorrected ages, mainly at high altitudes and latitudes (see Appendix A for details). Comparison to mean age calculated from a model 'clock-tracer', an inert tracer with a linear increase in the boundary layer (see also Hall and Plumb, 1994), shows good agreement with the spectrum-based mean age and therefore affirms the internal consistency of the age spectrum calculation in the model (see Fig. 4 c/d).

3 Seasonality of age spectra

The climatological annual mean age spectrum in the NH lower stratosphere (here 400 K, 60°N) from the CLaMS simulation (black line in Fig. 5) is characterized by a skewed distribution with a maximum around half a year. This annual mean distribution is well approximated by the idealized age spectrum for stationary flow, as represented by an inverse Gaussian distribution, the Green's function for 1D diffusion (grey shading). For long-lived tracers stratospheric transport may be approximated by a one-dimensional flux-gradient relationship (Plumb and Ko, 1992). Motivated by this fact, Hall and Plumb (1994) discussed properties of the age spectrum by considering the Green's function for a one-dimensional diffusion process. Such a consideration requires the assumption of stationary flow (see Holzer and Hall, 2000, for a more general discussion). The 1D diffusion Green's function can be expressed in terms of the mean age and spectrum width as (e.g., Waugh and Hall, 2002, Eq. 9)

$$G^{\text{diff}}(\tau) = \sqrt{\frac{\Gamma^3}{4\pi\Delta^2\tau^3}} \cdot \exp\left(-\frac{\Gamma(t-\Gamma)^2}{4\Delta^2 t}\right) \tag{6}$$

and is frequently used to approximate the age spectrum shape for deducing age of air information from observed tracers (e.g., Schoeberl et al., 2005).

The seasonal age spectra, however, show a more complicated structure with multiple peaks along the transit time axis (coloured lines in Fig. 5). The occurrence of multiple peaks in seasonal age spectra has already been noted and discussed in recent papers (e.g., Reithmeier et al., 2007; Diallo et al., 2012; Li et al., 2012a). Here, we simply present the CLaMS based findings and relate to published results in Sect. 6. In Fig. 5 the modal age, as determined by the apex of the largest peak in the seasonal age distribution, coincides with the youngest peak during spring, summer and fall and with the second peak during winter.~~The location of the global maximum in the seasonal age distribution (the modal age) depends on the season, coinciding with the youngest peak from spring to fall and with the second youngest peak in winter.~~ Furthermore, the location of the mode strongly depends on the region (e.g., Sect. 4).

The occurrence of multiple peaks in the seasonal age spectrum (Fig. 5) reflects the seasonality of transport. The highest fraction of young air in the NH lower stratosphere at 400 K is found in NH summer (red line in Fig. 5). The BIR map in Fig. 3a shows that these air masses had left the boundary layer about half a year earlier, in the previous winter (source time of the peak corresponds to NH winter). ~~Hence, the highest fraction of young air in the NH lower stratosphere during summer is caused by particularly efficient and fast transport from the boundary layer into the stratosphere during NH winter and spring, consistent with the results of (Reithmeier et al., 2008). This efficient transport is related to strongest tropical upward mass flux in boreal winter and weakening subtropical and polar transport barriers in spring and summer.~~ Figure 5 further shows how the summertime peak of young air (transit times of about half a year) ages throughout the course of the year. The coloured crosses in the figure illustrate the position of the peak during the following seasons: at about 0.75 years during the following fall, ~ 1 year during the following winter, ~ 1.25 years during the following spring, with a ~ 1-year offset from the original summer peak during the following year's summer, and so on. The evolution of the age spectrum peaks becomes even clearer in a presentation versus season and transit time in Fig. 6. An exceptionally high fraction of young air arrives in the extratropical NH lower stratosphere during spring and summer, causing the strong summertime peak at young transit times. This peak propagates to older transit times during the following months, and occurs around transit times of one year during next winter. A more detailed discussion of the processes affecting the age spectrum peaks will be presented in Sect. 6. ~~The occurrence of multiple peaks in the age spectrum is therefore directly linked to the seasonality of transport, with most efficient transport from the boundary layer into the stratosphere during NH winter. Further discussion of the spectrum peaks will be presented in Sect. 6.~~

Consideration of the full age spectrum allows one to quantify air mass fractions corresponding to certain transit times. Of particular importance for short-lived chemical species in the lower stratosphere is the fraction of very young air with transit times below some threshold τ^* (denoted *fresh-air-fraction* F_{τ^*} in the following), which can be deduced from the age spectrum by integration over transit time

$$\mathrm{F}_{\tau^*}(r,t) = \int_0^{\tau^*} \mathrm{d}\tau \, G(r,t\,|\,\Omega, t-\tau)\,. \tag{7}$$

This fresh-air-fraction is an important quantity for understanding the abundances of short-lived trace constituents, and we will discuss its seasonal variability in the following.

Figure 7a–b shows the fresh-air-fraction with transit times younger than six months (F_6) for NH winter and summer. In the tropical lower stratosphere, the F_6 is larger in NH winter than summer, consistent with the seasonality in tropical upwelling which maximizes in NH winter (e.g., Rosenlof, 1995). Although the strongest upwelling is displaced into the respective summer hemisphere (e.g., Seviour et al., 2011; Abalos et al., 2015), the corresponding F_6 in this region is lower than in the winter hemisphere tropics and subtropics (Fig. 7a–b). This indicates a significant impact of in-mixing of old air masses from the extratropics on the tropical composition, maximum during NH summer. Such mixing increases the amount of old air at low latitudes, consistent with an enhanced mass fraction of air older than two years in this region (Fig. 7c–d).

The two hemispheres show opposite annual cycles in F_6, with the largest extratropical F_6 values in the lower stratosphere found during summer in each hemisphere. A striking hemispheric asymmetry exists in the fresh air fraction below about 450 K (Fig. 7a–b). In particular the NH lowest stratosphere is flushed with young air during summer (e.g., Hegglin and Shepherd, 2007; Bönisch et al., 2009), increasing F_6 there to about 20%. This flushing with young air has implications for the chemical composition of this region, for instance by enhancing the amount of short-lived species and pollutants. At levels closer to the extratropical tropopause the maximum in NH F_6 appears lagged by 1-2 months compared to the levels above (not shown). The timing of maximum youngest air in the extratropics during hemispheric summer to fall is consistent with strongest horizontal transport from the tropics during each hemisphere's summer. Furthermore, the minimum tropical F_6 during NH summer is consistent with stronger horizontal eddy mixing in the NH than in the SH (e.g., Konopka et al., 2015).

Geographical distributions of F_6 and mean age at 400 K are shown for winter and summer in Fig. 8. Clearly, the largest F_6 values occur during NH winter directly above the tropopause over the Western Pacific, the Indian Ocean, and Central Africa. During NH summer, the largest F_6 occur in a narrow band south of the equator and within the Asian monsoon anticyclone (from the Tibetan plateau and North India to the Middle East). The enhanced young air mass fractions in the Asian monsoon likely have implications for pollution transport into the lower stratosphere (see also Randel et al., 2010), as the Asian monsoon system is close to the geographical source regions of highest anthropogenic pollution in India and China. Both the fresh air fraction and mean age show a clear planetary wave signature at middle and high latitudes during NH winter.

The additional information included in the spectrum as compared to mean age is illustrated by comparing mean age with the fractions of either young or old air masses. The difference between mean age and F_6 increases with altitude and latitude (Fig. 7a–b). As mean age is strongly affected by the age spectrum's tail, for broader age spectra at higher altitudes and latitudes its structure more closely resembles that of the fraction of old air with transit time larger than two years (Fig. 7c–d). Especially the Southern polar vortex during June–August shows very old mean age and an almost vanishing fresh air fraction F_6 (Fig. 7b/d). The large-scale geographical patterns of F_6 at 400 K largely resembles the geographical variability of the mean age distributions (Fig. 8c–d). On smaller spatial scales, however, the young air fraction and mean age patterns may differ significantly (e.g., compare the tongue of old air south of the Asian monsoon anticyclone in Fig. 8d in both diagnostics).

4 Residual circulation and mixing effects on age spectra

From a conceptual point of view, zonal mean stratospheric transport may be separated into net mass transport (given by the residual mean meridional circulation) and additional two-way mixing due to eddies (e.g., Andrews et al., 1987, chapter 9). The transit time corresponding to the residual circulation, the RCTT (see Sect. 2), describes the pure effect of residual circulation transport (e.g., Birner and Bönisch, 2011). Therefore, the difference between the RCTT and the "real" atmospheric transport time scale (the age of air) provides a measure of the effect of eddy mixing (Garny et al., 2014). The peak of the age spectrum (modal age) is related to the most probable transport time scale, likely corresponding to the most probable transport pathway. Comparison between the modal age and the RCTT therefore allows an analysis of the regions and seasons where either the residual circulation or eddy mixing dominates stratospheric transport.

Figure 9 presents the age spectra at 400 K and 600 K for all latitudes during winter and summer, together with the corresponding modal ages, mean ages, and RCTTs. In the tropics, the age spectrum generally shows one distinct peak, hence a well-defined modal age, at time scales close to the RCTT. From the subtropics to high latitudes, however, the spectrum shape is characterized by multiple peaks of similar strength, and therefore the modal age is ill-defined. The annual cycle in tropical upwelling, with faster upwelling in NH winter compared to summer, is reflected in a younger tropical spectrum peak during winter (the 20°N/S average modal age is 3.5 months during NH winter versus 4.1 months during summer). The small difference between the modal age and the RCTT in the lower tropical stratosphere at 400 K, with the modal age slightly older, shows an effect of in-mixing of old extratropical air into the tropics just above the tropopause.

At 600 K there is a clear separation of youngest air (around 1 year) in the tropics during both seasons, consistent with a tropical pipe model of stratospheric transport (Plumb, 1996). Interestingly, the modal age during NH summer appears at somewhat older transit times, suggesting that recirculation of older air from the extratropics plays a plays a more important role during this season than during NH winter ~~dominant role during this season~~.

At lower levels (here 400 K), steep latitudinal gradients in the fraction of youngest air (F_6) exist in mid-latitudes only during winter. These latitudinal gradients are significantly weaker during summer, in particular in the NH lower stratosphere where the high fraction of young air at high latitudes indicates strong horizontal transport from the tropics. This is consistent with studies showing that the isolation of tropical air imposed by the subtropical transport barriers does not extend down to 400 K (e.g., Volk et al., 1996). The large difference between modal age and RCTT in the lower extratropical stratosphere (particularly poleward of about 50°N during NH summer) further indicates that transport in this region is predominantly related to mixing. On the other hand, equatorward of about 50°N modal age and RCTT agree well at both levels and seasons, which points to a more dominant role of residual circulation transport. Therefore, the age spectrum analysis is consistent with recently published results relating the summertime "flushing" of the NH extratropics with young air to the combined effect of residual circulation transport equatorward of about 50°N, and quasi-horizontal mixing poleward of about 50°N (Bönisch et al., 2009; Konopka et al., 2015). In general, the existence of multiple spectral peaks in the extratropics questions the value of modal age as an reliable descriptor for age of air.

Profiles of modal age and RCTT at different latitude bands for winter and summer are compared in Fig. 10. Throughout the tropics, the modal age agrees well with the RCTT and consequently transport is largely related to the residual circulation. The figure also includes a simple proxy for the time scale of tropical upward transport (only panels a and d) based on the definition of cross-isentropic vertical velocity $\dot{\theta} = \Delta\theta/\Delta t$. Note that this simple proxy significantly differs from both the modal age and the RCTT during NH summer above about 650 K. This is related to seasonal changes in the structure of tropical upwelling and apparently emerges at transit times longer than ~~above~~ about 1.5 years. The wintertime high-latitude stratosphere above about 500 K (55–85°N during DJF, and 85–55°S during JJA) represents another case where modal age and RCTT closely match and transport appears to be well described by the residual circulation. For all other regions and seasons modal age and RCTT differ, at least partly, and eddy mixing has a significant effect on stratospheric transport. The clearest difference between modal age and RCTT and therefore the strongest mixing effect emerges in the high-latitude (55–85°N/S) lower stratosphere below about 500 K, in particular during summertime. However, apart from this exception the RCTT represents a good approximation to modal age, but not mean age.

The comparison between modal age and RCTT is summarized in Fig. 11, which shows the percentage difference between RCTT and modal age $(\mathrm{RCTT} - \mathrm{mode})/\mathrm{RCTT}$ for winter and summer. Small differences indicate a strong effect of the residual circulation on transport, as found throughout the deep tropics and in the respective winter hemisphere. The dipole pattern in each hemisphere below about 500 K, with RCTT smaller than the modal age in the tropics and larger than the modal age poleward of about 50°, is related to the strong effect of eddy mixing at these levels. As eddies mix air quasi-horizontally between the tropics and the extratropics, relatively old air is transported into the tropics while young air is transported into the extratropics. Therefore, in the tropics mixing causes an older modal age as compared to the pure residual circulation effect, reflected in the negative values in that region (Fig. 11). In the extratropics, mixing causes the modal age to be younger than resulting from pure residual circulation advection, and hence positive differences.

5 Inter-annual variability of the age spectrum related to the Quasi-Biennial Oscillation (QBO)

The Quasi-Biennial Oscillation QBO (e.g., Baldwin et al., 2001) represents the dominant mode of inter-annual variability in the tropical lower stratosphere. The GCMs used in recent studies (e.g., Reithmeier et al., 2007; Li et al., 2012a, b) did not include a QBO. Existing trajectory calculations based on reanalysis data (e.g., Diallo et al., 2012; Ray et al., 2014), while in principle including the QBO, have not analysed such effects in detail. Here, we discuss the effect of the QBO on the age spectrum. QBO-related inter-annual variability of mean age has recently been shown to be well represented in the ERA-Interim driven CLaMS simulation used here, if compared to Michelson Interferometer for Passive Atmospheric Sounding (MIPAS) observations (Ploeger et al., 2015b, Fig. 1).

The QBO is known to induce a secondary meridional circulation with anomalous upwelling in the tropics during easterly shear phase and anomalous downwelling during westerly shear phase (e.g., Baldwin et al., 2001). Figure 12 shows composites of the tropical age spectrum at 600 K, calculated by averaging all age spectra in that region for 1989–2013 during periods when the corresponding zonal mean zonal wind was greater than 10 m/s versus less than −10 m/s (defining westerly/easterly

QBO composites, respectively). The limit of ± 10 m/s has been chosen to enhance the clarity of the differences between the two composites. Relaxing the limit to 0 m/s results in qualitatively the same conclusions. Consistent with faster QBO-induced upwelling, the easterly QBO composite results in an age spectrum skewed towards shorter transit times, with modal age and mean age about 0.5 years younger as compared to the westerly QBO composite. Likewise, RCTT's are shorter during easterly QBO phase by ≈ 0.3 yr. Furthermore the spectrum shape differs between the two composites. During easterly QBO phase the tropical age spectrum shows a mono-modal shape, whereas secondary peaks develop during westerly QBO phase. These secondary peaks at older transit times (here 2.5, 3.5, 4.5 years) indicate an increased impact of in-mixing of old extratropical air on the tropical composition, consistent with enhanced subtropical mixing during westerly QBO phase (e.g., Shuckburgh et al., 2001; Abalos et al., 2016). This enhanced mixing results from shifts in the critical lines, allowing Rossby waves to propagate further equatorward and into the tropics.

Figure 13 (left panels) shows the time series of simulated age spectra in the lower stratosphere at 600 K for the tropics (a) and extratropics of both hemispheres (c/e). The tropical spectra (Fig. 13a–b) at 600 K show the strong QBO-related inter-annual variability, as discussed above. Comparing the respective deseasonalized age spectrum anomalies (constructed by subtracting the mean annual cycle at each transit time) to tropical zonal mean zonal wind (red bars highlight easterly wind periods) confirms that, indeed, the clearest mode of inter-annual variability is related to the QBO, with increased fractions of young air during QBO easterly phases, and opposite behaviour during QBO westerly phases. Furthermore, the modal age (white diamonds) varies between one and two years, with inter-annual variations closely matching the variability in RCTT (red line). This highlights the dominant influence of the residual circulation on transport variations in the tropical lower stratosphere on inter-annual time scales. Even the inter-annual variations of mean age (black line) closely follow those of the RCTT (Fig. 13b). A discontinuity in the tropical age spectrum time series at the end of 1993 (most strongly evident in panel b) could hint at problems with the reanalysis data, but we were not able to relate it to specific changes in the assimilation system.

Also the extratropical age spectra show strong QBO-related inter-annual variations in both hemispheres (Fig. 13c–f). The fraction of young air increases at the end of QBO easterly phases, lagging the tropical signal by a few months. Remarkably, these young air peaks originating in QBO easterly phases propagate to older transit times and determine the modal age during the following 2–3 years. For example, the modal age during 1996–1999 is set in the easterly QBO phase in 1996 (see Fig. 13c). Analogous behaviour results from QBO westerly phases, with anomalously old air originating in the tropics and being transported poleward. Clearly, the tropical air affected by anomalous transport during a specific QBO phase subsequently circulates around and may leave the stratosphere. However, the propagation of the signal shows the remarkable fact that enough of the air recirculates back to preserve the memory of the original signal over the 2-3 following years. The propagation of the modal age along the transit time axis clearly shows how the QBO affects the composition of the lower stratosphere on a global scale. Inter-annual variability on the 400 K isentropic surface implies modulations due to the El Niño Southern Oscillation (ENSO, not shown), although not as conclusively as the modulations due to the QBO.

~~In the lower stratosphere (at 400 K, Fig. 10) time series of age spectra are dominated by the annual cycle of transport in this region (see Sect. 3). Modal age and RCTT show a general offset of about 1–2 months, which points to a significant effect of mixing in this region (see also Sect. 4). However, the deseasonalized time series (Fig. 10, right column), show remaining~~

~~variability, which particularly in the tropics appears to co-vary partly with the multivariate El Niño–Southern Oscillation (ENSO) index MEI. El Niño phases are indicated in Fig. 10 by highlighting periods with a sufficiently positive MEI (here > 0.5). The fraction of young air masses increases during El Niño phases (positive multivariate ENSO index), consistent with enhanced tropical upwelling during these periods. Analogous behaviour with oppositely signed anomalies is found for La Niña phases. The age spectrum therefore shifts to younger transit times during El Niño and to older transit times during La Niña. This is especially evident during strong events, such as the 1997/1998 El Niño followed by two strong La Niña years. These co-variations between the age spectrum time series and ENSO at 400 K are however generally not as succinct as the co-variations with the QBO at 600 K (see Fig. 10). Nevertheless, some of the tropical variability at 400 K emerges at middle and high latitudes (e.g., after 1998 in Fig. 10d/f), indicating a global impact of ENSO on the inter-annual variability of lower stratospheric transport.~~

6 Discussion

As mentioned already in the introduction, different explanations have been proposed for the occurrence of multiple peaks in seasonal age spectra. Bönisch et al. (2009) refined the work by Andrews et al. (2001) and assumed that age spectra in the lowermost stratosphere (below about 380 K) result from the superposition of two single peak spectra, related to a fast (quasi-horizontal mixing and shallow circulation branch) and a slow pathway (deep circulation branch). They noted that this superposition does not necessarily cause a bimodal spectrum shape for sufficiently strong overlap of the individual spectra. Reithmeier et al. (2007) found several peaks in their age spectra, but only at polar latitudes. They explained these peaks as resulting from the annual cycle in tropical upward mass flux into the stratosphere, maximizing during NH winter, and the seasonal variation in the polar vortex transport barrier, allowing horizontal transport to high latitudes only in spring and summer. This results in the superposition of single mode spectra from low latitudes once per year. Li et al. (2012a) also found multi-peak age spectra in their model simulation at polar latitudes, but conversely to Reithmeier et al. (2007) argued that these peaks form mainly due to the fact that air masses leaving the boundary layer during NH summer would have the highest probability to recirculate into the polar stratosphere.

As indicated in Fig. 9, we find multiple peaks as a generic feature throughout the lower stratosphere, although these peaks are most pronounced at high latitudes. Closer inspection of the peaks shows that they correspond to NH winter pulse release times (cf. peaks in the BIR map in Fig. 3a). To further highlight this point, Fig. 14a presents the distribution of the source times of the age spectrum peaks, hence the time when the air corresponding to the peaks has left the boundary source region at the Earth's surface. The clear maximum at NH winter source times shows that the air corresponding to the peaks has left the surface layer during NH winter. Note that the figure shows the distribution of surface source times for air parcels sampled in the stratosphere – the pulse release rate at the surface is not varying. Figure 14b further shows that also for the full age spectrum (not only the peaks) air leaving the boundary surface during NH winter is most likely. For example, about twice as much stratospheric air has left the surface in January compared to May. The link of the age spectrum peaks to an enhanced probability of stratospheric

air to originate at the surface during NH winter explains also the occurrence of the peaks at approximately the same transit time independent of latitude and level (Fig. 9), and is consistent with the interpretation by Reithmeier et al. (2007).

One explanation for the enhanced occurrence of NH winter surface air in the spectrum is that air parcels preferentially enter the stratosphere during NH winter (cf. argument in Reithmeier et al., 2007). Another possible explanation is that vertical lofting in the tropical stratosphere only occurs efficiently during NH winter when tropical upwelling is strongest and the tropical stratosphere is well isolated. Some of this air then reaches the region of interest through a more or less direct pathway. Some of it recirculates and reaches the region in following years. Pulses released during NH spring and summer, on the other hand, are efficiently dispersed meridionally before they reach the tropical pipe and therefore are less likely to undergo recirculation in the stratosphere. This is consistent with a stronger return flux into the troposphere for air entering the stratosphere in July compared to January (Orbe et al., 2014). A sensitivity calculation with pulses released at the tropical tropopause (not shown) also results in age spectra with annually repeating peaks, however weaker. Therefore, a combination of all transport processes described above likely plays a role.

At first glance, the aging of the peaks by 1 month per month at a given location in the stratosphere (e.g. Fig. 6) would be consistent with air simply staying at that location. This is possible in the extratropical middle and upper stratosphere during summer where the circulation essentially shuts down. But it is inconsistent with the aging of the peaks throughout the year - during winter the air is expected to sink, for example. Hence, the propagation of the peaks to older transit times (Fig. 6) results from the interplay of various processes.

Figure 9 shows that maximum tropical upward mass flux during NH winter causes a strong peak at short transit times in the tropical lower stratosphere age spectrum. This tropical young air is transported rapidly by isentropic mixing and the shallow circulation branch to middle and high latitudes during spring and summer when the subtropical jet transport barrier is weak and the polar vortex transport barrier is absent. Therefore, the young air peak extends from the tropics to the summer pole (see Fig. 9a–b), particularly in the NH. A fraction of the tropical air, however, is transported through the deep circulation branch and arrives at higher latitudes later (e.g., during the following winters, when the deep circulation branch is most active). The comparison between air originating at the surface during winter with air originating at the surface during summer after 3 years of being transported through the atmosphere in Fig. 2 (right panels) shows that particularly at higher levels in the stratosphere winter surface air is more prevalent. As this air corresponds to the same winter source time as the air which was transported directly to higher latitudes at lower levels, it effectively creates the age spectrum peak at older transit times and compensates the diluting effect of downwelling. Some of the extratropical air recirculates into the tropics and amplifies the peaks in the spectrum tail. As the deep circulation branch is strongest during NH winter, the older spectrum peaks appear stronger in winter than summer (e.g., Fig. 9a–b). Mixing causes attenuation of the peaks.

Other modes of variability in transport, such as the QBO, may also modify the shape of the age spectrum and the occurrence of multiple peaks (see Fig. 13 and related discussion). Remarkably, the age spectrum mode may remain attached to one specific QBO phase for several years, with similar behaviour also possible for other modes of inter-annual variability. The existence of a shallow and fast transport pathway further affects the shape of age spectra in the lowermost stratosphere. Transport by the

deep circulation branch is likely responsible for the amplification of the older peaks at high latitudes (Fig. 9), but is not creating an additional peak, in agreement with Bönisch et al. (2009).

In contrast to Reithmeier et al. (2007) and Li et al. (2012a), the CLaMS age spectrum peaks are more pronounced, with multiple peaks occurring also in the subtropics and mid-latitudes, and even weakly so in the tropics (e.g., Figs. 9, 12). Problems with representing the subtropical transport barrier in ECHAM4 (see Reithmeier et al., 2007) likely caused a masking of the multiple peaks at these latitudes in their study. Li et al. (2012a) used a climate model (GEOSCCM) that likely contained stronger numerical diffusion compared to the Lagrangian transport model used here. On the other hand, CLaMS is driven by reanalysis winds, which are known to be overly dispersive (Schoeberl et al., 2003). A detailed understanding of the differences in the occurrence of multiple spectral peaks at lower latitudes in different models requires further work.

In summary, multiple peaks are a generic characteristic of age spectra in the lower stratosphere and are caused by seasonality and inter-annual variability in transport. Hence, an accurate fit of age spectra in the lower stratosphere requires the superposition of a number of 1D diffusion Green's functions, with this number equal to the number of distinct peaks in the spectrum. We tested such a fitting procedure and found that it works well.

6.1 ~~Relation between age spectrum characteristics and mean age~~

6.2 ~~Age spectrum approximation with 1D diffusion Green's function~~

7 Conclusions

We presented seasonally and inter-annually varying age spectra in the lower stratosphere calculated with the Lagrangian transport model CLaMS driven by ERA-Interim winds for the period 1979-2013. Our approach is based on the boundary impulse response (BIR) method (e.g., Haine et al., 2008; Li et al., 2012a), generalized to transient simulations using quasi-observational winds, and is therefore more appropriately termed the Boundary Impulse (time-)Evolving Response (BIER) method.

Seasonal age spectra in the lower stratosphere show large deviations from an idealized stationary uni-modal shape. Multiple peaks emerge in the age spectra throughout the stratosphere (strongest at high latitudes), caused by the interplay of seasonally varying tropical upwelling, stratospheric transport barriers and recirculation. These multiple peaks are largely related to the fact that air entering the stratosphere during NH winter makes up the biggest fraction throughout the stratosphere. While the annual mean spectrum is in large parts well described by a Green's function representative of idealized stationary transport, seasonal age spectra with multiple peaks can only be well approximated by a superposition of such functions. In addition to seasonality, inter-annual variations in transport (e.g., QBO) cause significant age spectrum modulations. Stronger upwelling during easterly QBO phase increases the fraction of young air in the spectrum. We found that one specific QBO phase may determine the modal age (age spectrum maximum) for up to 3 years across a wide range of latitudes.

Interpretation of the age spectrum in terms of residual circulation and mixing is not straightforward, with different effects dominating in different atmospheric regions. We found residual circulation trajectories, which are much more easily obtained than age spectra or even mean age, to represent a good approximation of the dominant pathway in the deep tropics and in the

winter extratropics above about 500 K, as represented by the modal age in these regions. In contrast, eddy mixing strongly modifies the modal age in summer, particularly in the lowermost stratosphere.

~~Mean age appears to be a reliable proxy for spatial variations in the age spectrum just above the tropopause, but less so throughout most of the stratosphere.~~ Analysis of the full age spectrum compared to mean age is advantageous for separating the effects of different transport processes, as has been already emphasized in earlier studies (e.g., Hall, 1999; Waugh and Hall, 2002). In this sense, knowledge of the exact fractions of air masses with certain transit times, as included in the full age spectrum, is highly beneficial for a more detailed understanding of stratospheric chemistry and composition. Including time-dependent age spectrum diagnostics in state-of-the-art atmospheric climate and transport models would help to avoid ambiguities in model inter-comparisons of stratospheric transport.

Appendix A: Correction for finite age spectrum tail

The tail of the age spectrum $G(\tau)$ in the lower stratosphere generally decreases exponentially at transit times larger than about 4-5 years (e.g, Fig. 3c), as noted by several authors (e.g., Reithmeier et al., 2007; Diallo et al., 2012; Li et al., 2012a). At large transit times $\tau > \tau^*$, larger than some threshold τ^*, the spectrum tail may therefore be approximated by an exponential function to define a *corrected age spectrum* by

$$G_{\mathrm{corr}}(\tau) = \begin{cases} G(\tau) & \text{for } \tau \leq \tau^* \\ G(\tau^*)\, e^{-\frac{\tau}{\xi}} & \text{for } \tau > \tau^* \,. \end{cases} \tag{A1}$$

As our multi-pulse set-up in CLaMS with 60 pulse tracers every two months (see Sect. 2.2) allows calculating the age spectrum for transit times up to ten years, we chose $\tau^* = 10$ years and determine the decay time scale ξ by fitting the exponential function for transit times $5\,\mathrm{yr} < \tau < 10\,\mathrm{yr}$.

Integration of the corrected age spectrum (Eq. A1) over transit time (from zero to infinity) yields the corrected norm and the first moment of the age spectrum (the corrected mean age) (see also Diallo et al., 2012, Eq. 2)

$$N_{\mathrm{corr}} = N + G(\tau^*)\,\xi \,, \tag{A2}$$

$$\Gamma_{\mathrm{corr}} = \Gamma^* + G(\tau^*)\,\xi\,(t^* + \xi) \,. \tag{A3}$$

Figure 15 shows the reference (uncorrected) mean age from the CLaMS climatological age spectrum for winter and summer and the difference to the corrected mean age (corrected minus reference). As expected, the correction of the spectrum tail up to infinite transit times causes older mean age globally. The differences increase with increasing latitude and altitude, remaining less than about 6 months throughout large parts of the lower stratosphere, but increasing to about 1 year inside the polar vortex (particularly in the SH).

An interesting side note concerns the comparison with the 'clock-tracer' age, which is younger than the corrected (but older than the uncorrected) age spectrum mean age (Fig. 4). From theoretical considerations based on stationary flow (Hall and Haine, 2002, Eq. 9) showed that 'ideal age' (an alternate for the 'clock-tracer' used here) converges faster to a steady-state value than

the spectrum mean age, which seems at first glance contradictory to the comparison between 'clock-tracer' and corrected spectrum mean age presented here. However, there is no contradiction as the time span for convergence is not directly comparable. The 'clock-tracer' was subject to a 10 year spin-up (repeating 1979 conditions) plus 10 years of transient simulation (1979-1988) before taking it into consideration, whereas the age spectrum has explicitly been calculated over 10 years of transit time (1979-1988 simulation) but with the tail fitted to infinite transit times, as explained above. Hence, the effective calculation length is longer for the corrected age spectrum than for the 'clock-tracer'.

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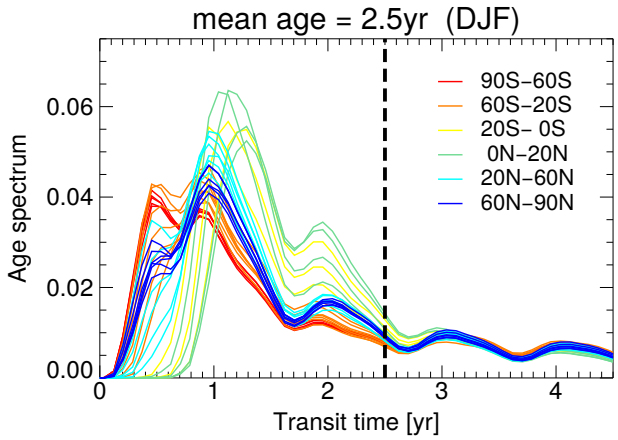

Figure 1. Climatological age spectra for the same mean age of 2.5 years but at different latitude ranges, for December–February. Each line is the average over DJF, with different age spectra of the same color representing different latitudes within the respective latitude band. The black dashed line highlights the mean age of 2.5 years.

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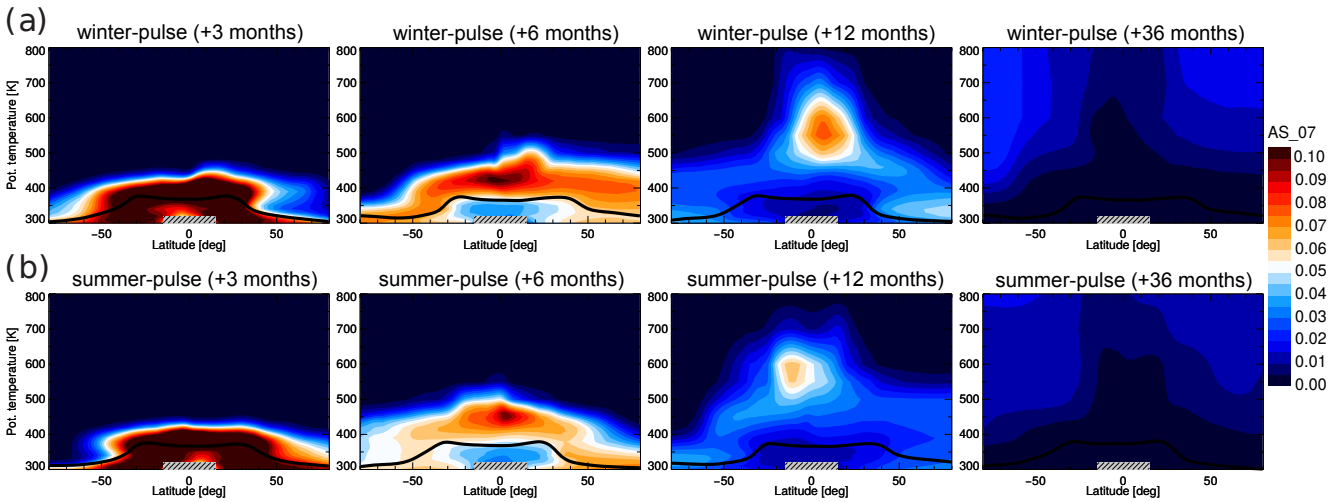

Figure 2. Evolution of tracer pulses released in the tropical boundary layer during NH winter (a) and summer (b) during the following months (3, 6, 12, 36 months after pulse release). The winter pulse was released during January 2000, the summer pulse during July 2000. The black line shows the climatological tropopause, the grey hatched line at the tropical surface the pulse release region.

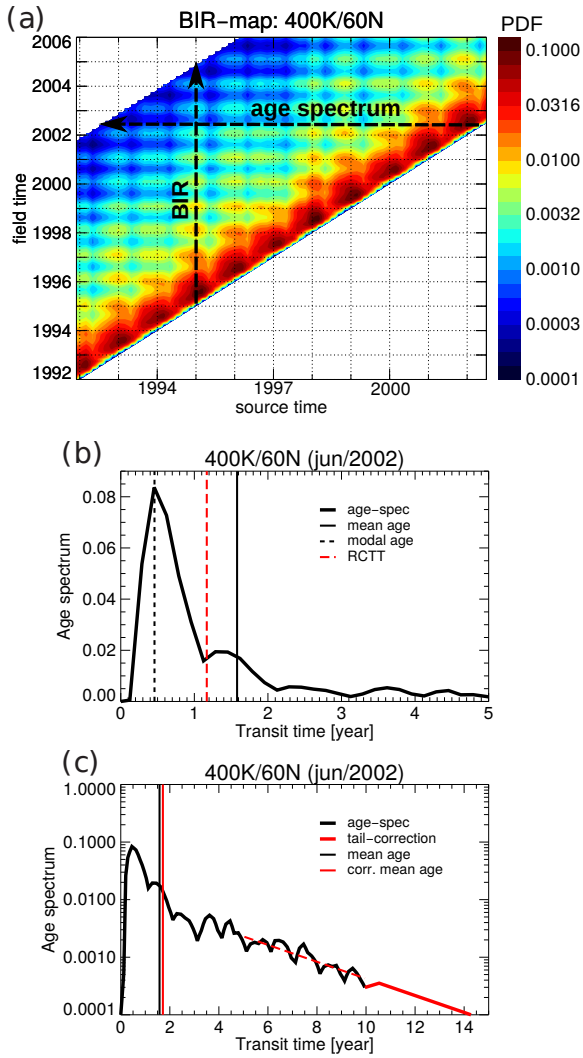

Figure 3. (a) BIR map from CLaMS pulse tracers at 400 K and 60°N, plotted only for part of the full simulation period. Arrows highlight vertical and horizontal cuts through the BIR map representing BIR (vertical) and age spectrum (horizontal), respectively (cf. Haine et al., 2008). (b) The age spectrum for June 2002, corresponding to the horizontal cut in (a). Vertical lines show mean age (solid), modal age (black dashed) and the residual circulation transit time (red dashed, see text). (c) Same as (b), but including a correction for the finite spectrum tail using an exponential fit (see text). The red vertical line shows mean age for the tail-corrected spectrum, the black line for the uncorrected case (Note the larger transit time range and logarithmic y-axis).

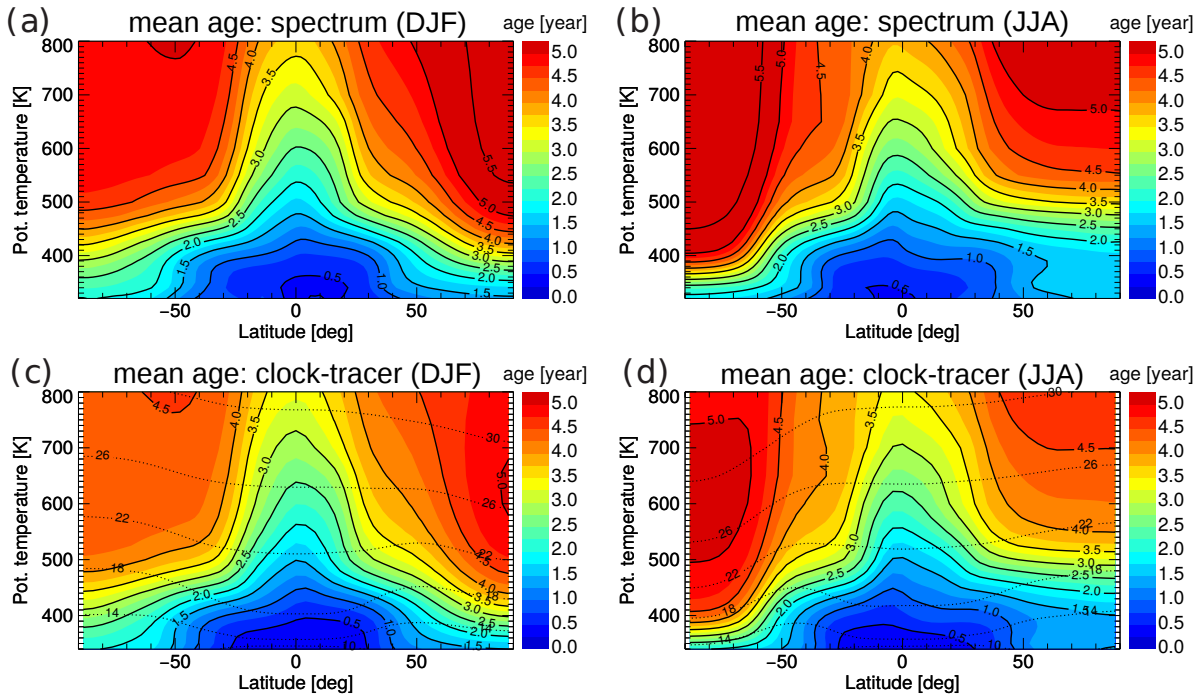

Figure 4. Mean age from the 1989–2013 CLaMS age spectrum climatology for (a) December–February and (b) June–August. Corresponding mean ages calculated from the model's 'clock-tracer' are shown in (c) and (d) for comparison. Black dotted lines in (c/d) show altitude levels in km for reference.

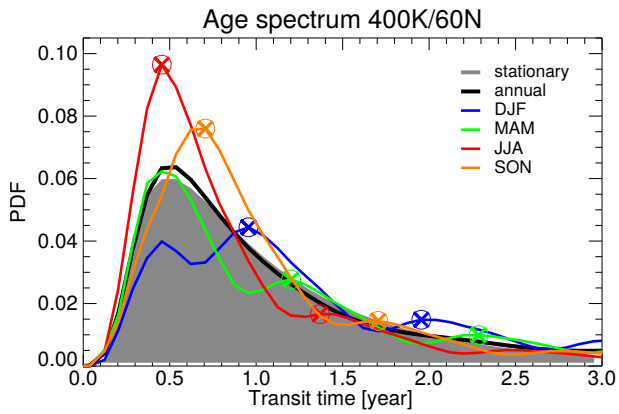

Figure 5. Seasonality of the climatological age spectrum at 400 K in NH mid-latitudes (60°N). The annual mean spectrum is shown as a black thick line, the idealized stationary shape (see text) as grey shading, and different seasons as coloured lines. Symbols illustrate the evolution of the peak (see text for details).

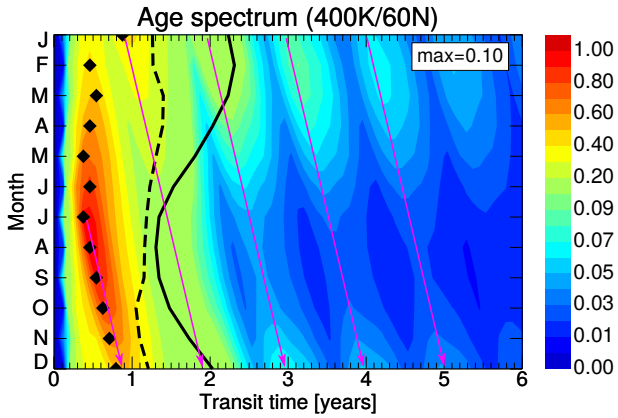

Figure 6. Seasonal cycle of the climatological age spectrum at 400 K in the NH extratropics (60°N). Mean age is shown as black thick line, modal age as black symbols, and RCTT as black dashed line. The pink arrows illustrate the time evolution of the spectrum peaks. (Note the non-linear colour scale).

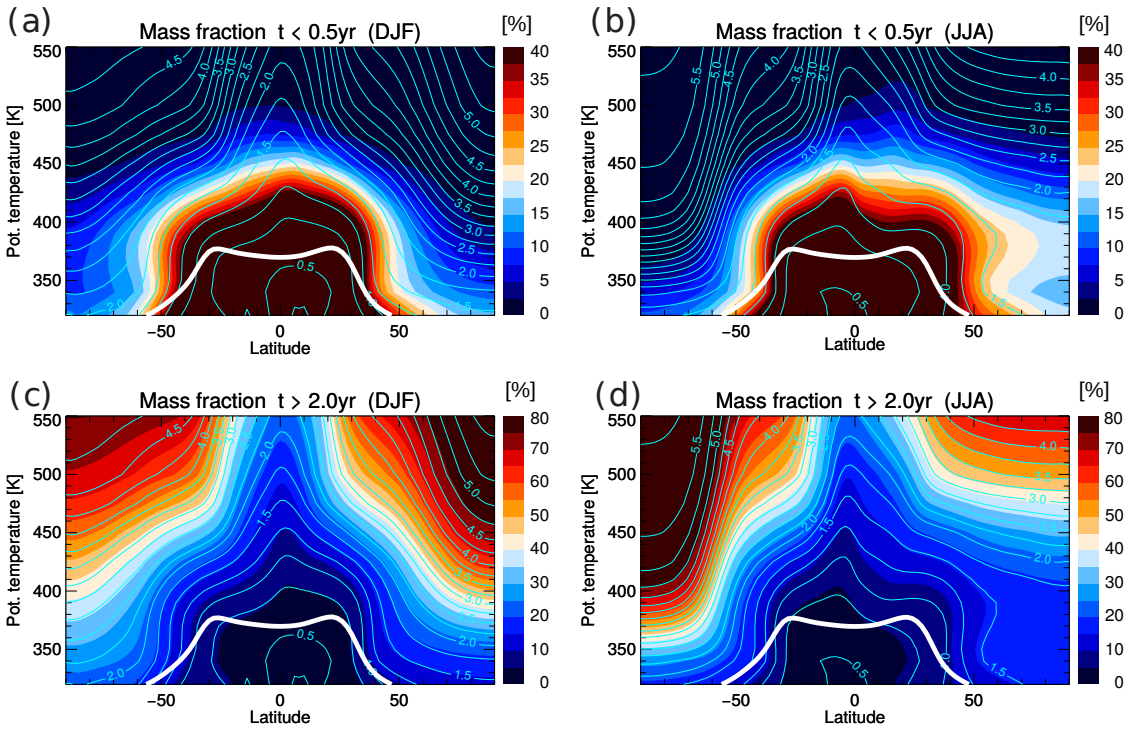

Figure 7. Fresh air fraction F_6 (transit time below 6 months) during (a) December–February, and (b) June–August. (c) and (d) show the fraction of old air (transit time above 2 years). The white contour shows the thermal tropopause, cyan contours show mean age.

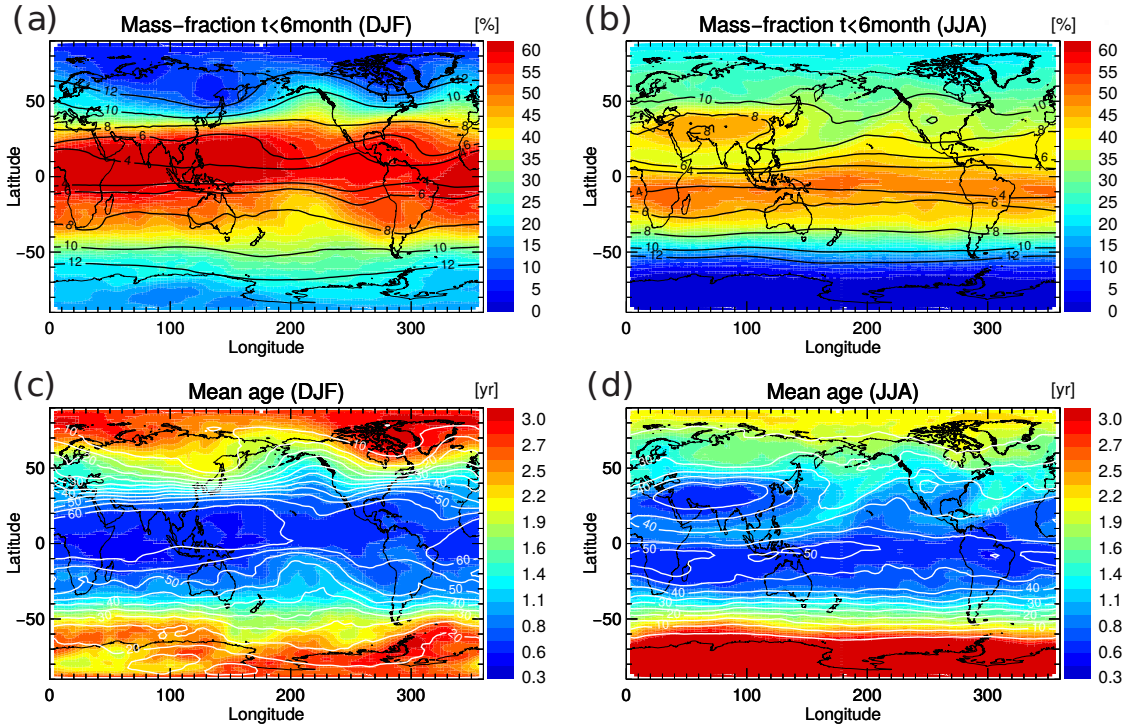

Figure 8. Fresh air fraction F_6 (transit time below 6 months) at 400 K during (a) December–February, and (b) June–August. Black contours show potential vorticity from ERA-Interim (±4, ±6, ±8, ±10, ±12 PVU). (c) Mean age at 400 K during December–February, and (d) June–August (white contours illustrate the fresh air fractions from a/b).

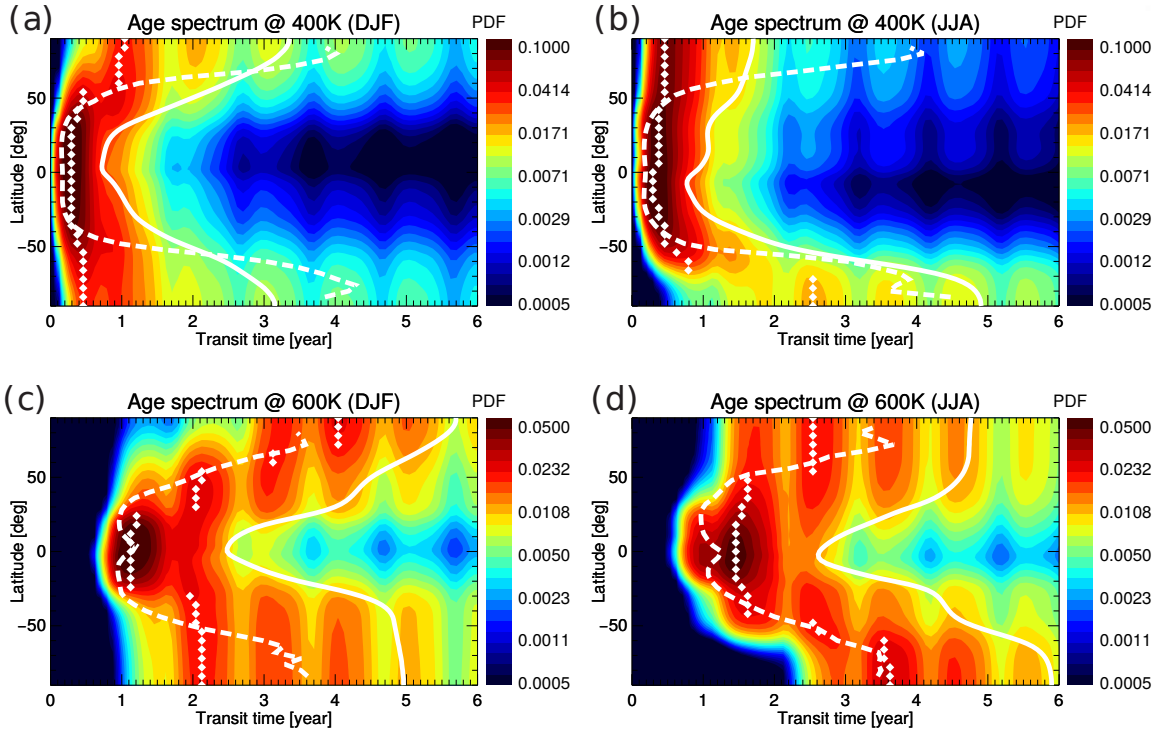

Figure 9. Age spectra at 400 K for (a) December–February and (b) June–August. (c) and (d) show the same but at 600 K. White lines show mean age (solid) and the residual circulation transit time (dashed). White diamonds show modal age.

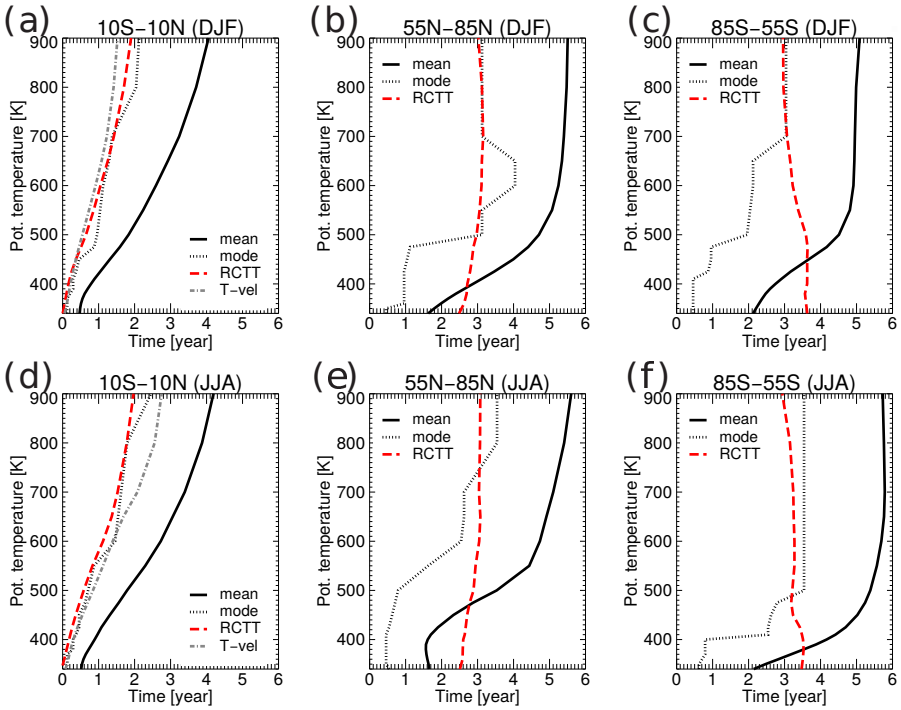

Figure 10. Profiles of mean age, modal age and RCTT for DJF (top) and JJA (bottom) at the equator, in the NH between 55°N–85°N, and in the SH between 55°S–85°S (from left to right). The grey dashed line in (a) and (d) shows the mean tropical upwelling time scale estimated from the zonal mean vertical velocity.

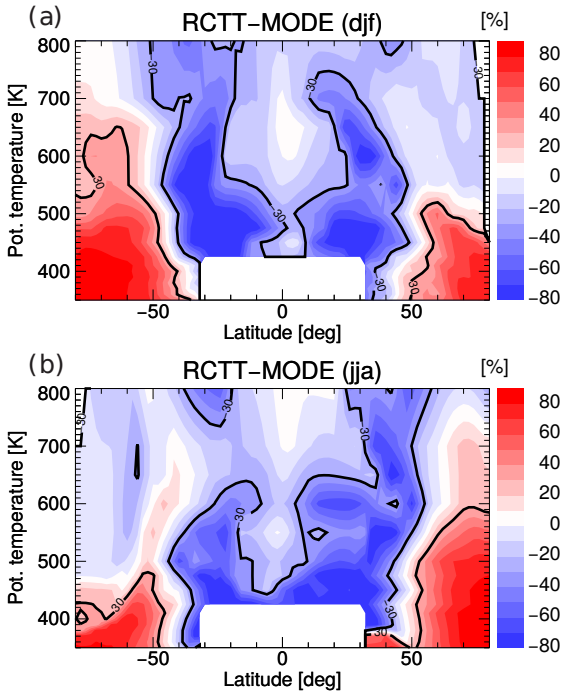

Figure 11. Difference between RCTT and modal age (RCTT − mode)/RCTT in percent for (a) December–February, and (b) June–August. The black line highlights the 30% contour. The tropical lowest stratosphere (below 420 K) is left white as relative differences become arbitrarily large there due to small transit times.

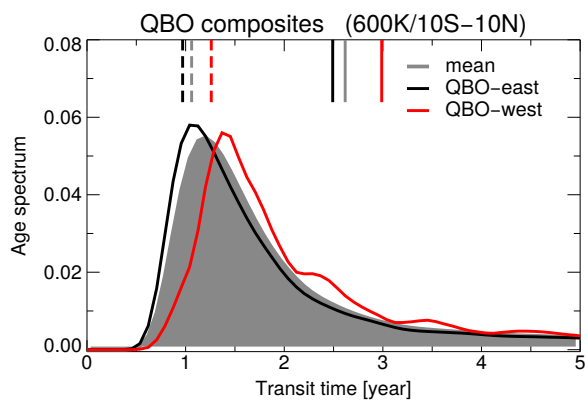

Figure 12. Age spectrum composites for easterly (black) and westerly (red) QBO phases, calculated from tropical (10°S–10°N) CLaMS age spectra at 600 K. Mean ages are shown as solid vertical lines, RCTTs as dashed lines. The climatological mean spectrum, mean age and RCTT are shown in grey. (For better clarity of the differences, the westerly/easterly composites have been defined using the condition that ERA-Interim tropical mean zonal winds at 600 K go above/below ±10m/s).

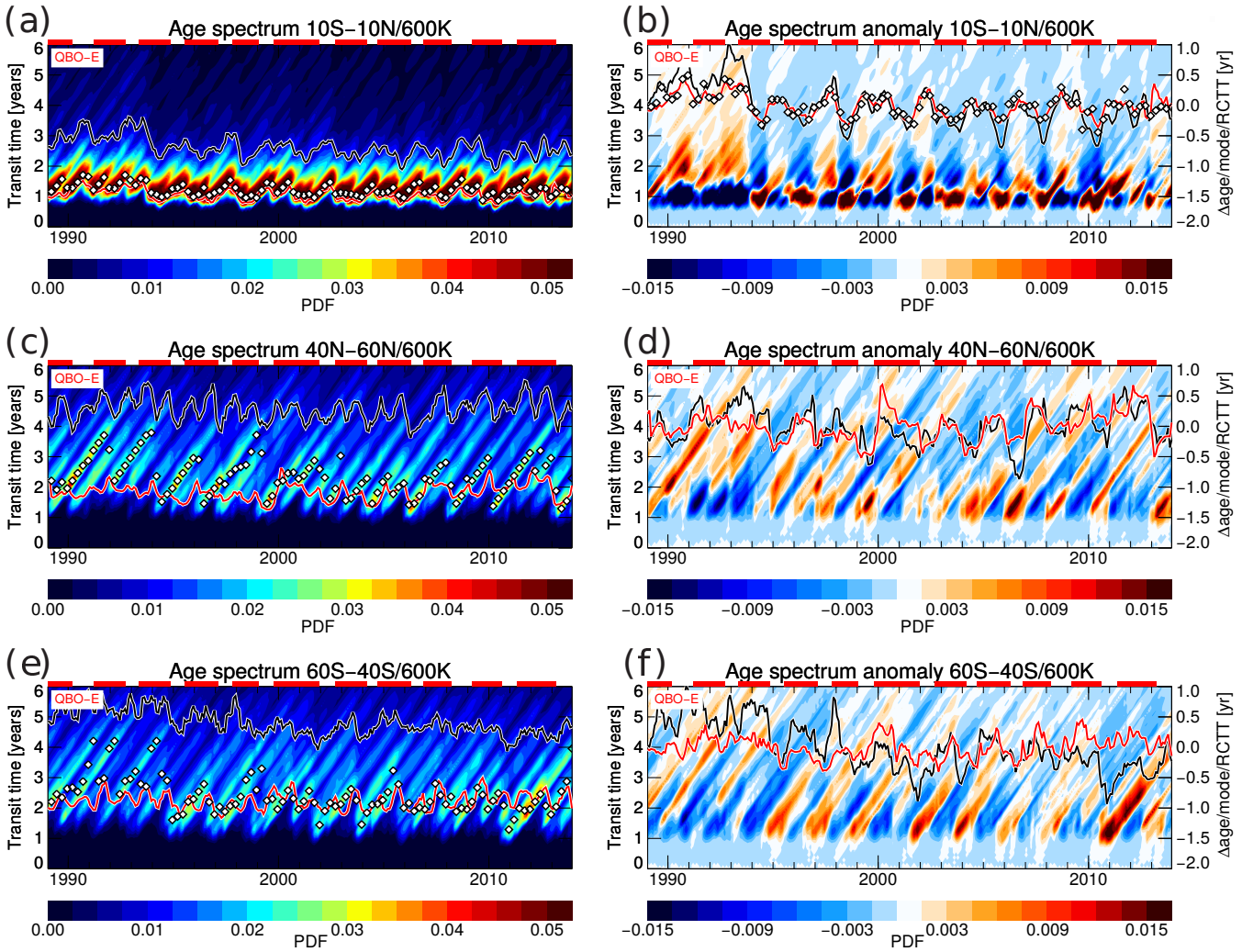

Figure 13. Age spectrum time series at 600 K and 10°S–10°N (top), 40°N–60°N (middle), and 60°S–40°S (bottom). Shown are the spectra (left) and their deseasonalized anomalies (right). Black lines show mean age, symbols modal age, and red lines show RCTT (full values on the left and their respective deseasonalized anomalies on the right). Red horizontal bars highlight easterly QBO phases.

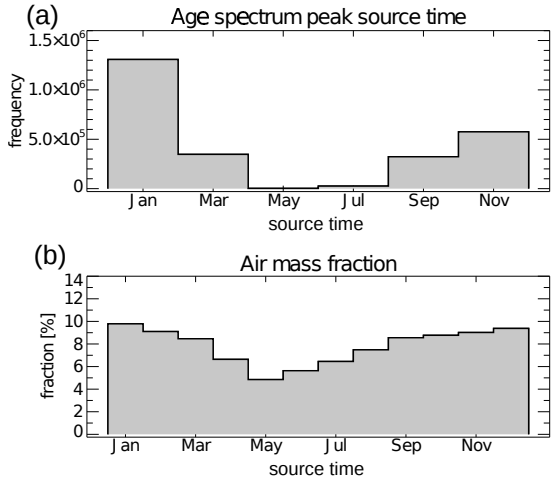

Figure 14. (a) Distribution of time of last contact with the boundary surface (source time) for age spectrum peak air. (b) Seasonality of time of last contact with the boundary surface for all stratospheric air masses. The distributions have been calculated from all CLaMS age spectra in the 380–1000 K potential temperature layer during the 1989–2013 period.

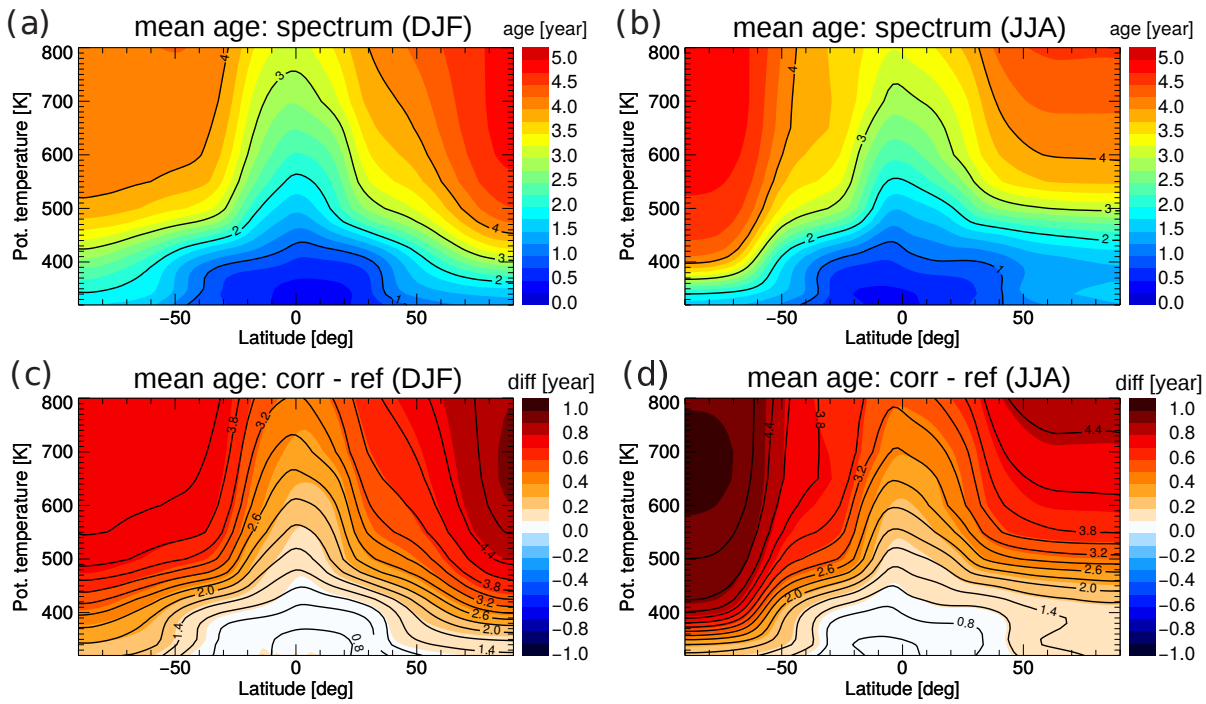

Figure 15. Climatological mean age from CLaMS age spectrum for (a) December–February and (b) June–August, without applying the tail-correction (see text). (c) and (d) show the difference of age spectrum mean ages with correcting for the finite spectrum tail to this reference. Black lines highlight particular mean age contours.