

Response to Reviewers – “Developing tracer interrelationships to derive stratospheric age of air from satellite observations of nitrous oxide” by Ariana Elena Castillo et al.

We thank the reviewers for their time and insight, and we appreciate their thoughtful comments. The reviewers' comments are given below in **bold** with our responses in **red**.

Reviewer 1:

This manuscript presents a methodology for calculating age of air (AoA) using nitrous oxide observations from MLS and ACE-FTS, supplemented by SF6-derived relationships from MIPAS. While the study provides a valuable framework for extending the stratospheric age record, several methodological choices regarding data presentation and normalisation require further clarification and sensitivity testing. Overall, I am happy to accept the manuscript with minor corrections if authors can address the technical points raised below.

The study successfully demonstrates that tracer interrelationships between sink-corrected SF6-age and normalised nitrous oxide are highly compact and display a clear meridional dependence. A significant finding is the identification of a distinct inflection point at a normalised nitrous oxide value of 0.7, where photochemical loss begins to dominate over transport, particularly in the deep tropics. Below this threshold, typically corresponding to ages between 3.0 and 3.5 years, the nitrous oxide abundance decreases rapidly with only minimal further aging. Furthermore, the authors confirm interhemispheric asymmetries in AoA seen in chemical models, noting that at a constant normalised nitrous oxide abundance, mean ages in the Southern Hemisphere are older than those in the Northern Hemisphere between values of 0.7 and 0.3.

The comparison between the newly derived MLS and ACE-FTS nitrous oxide-AoA products and existing SF6-age records confirms the broad structure of the Brewer-Dobson circulation while highlighting specific instrumental biases. For instance, MIPAS SF6-age remains older than the MLS nitrous oxide based AoA estimates in the lower stratosphere, likely due to known high biases in tropical MIPAS retrievals. These latitudinally-resolved relationships provide a more subtle comparison between the hemispheres and highlight the relative importance of mixing versus chemical loss in different regions. To further strengthen the validation of these products, especially considering the reported instrumental drifts in MLS after 2010, I would suggest comparing the results with the TCOM-N2O dataset (Dhomse et al., <https://zenodo.org/records/18197444>). This record utilises machine learning and TOMCAT simulations to provide an observationally constrained, gap-free nitrous oxide profile that could serve as a robust independent reference.

Thank you for this comment and suggestion. This is a valuable dataset, and we look forward to considering it in future work using a longer time series to explore long-term variability and trends. We note that the MLS drift bias becomes more apparent after 2015, which is beyond the scope of this manuscript.

Regarding the chemical mechanisms, it is important to elaborate on the pathways of nitrous oxide destruction to provide clearer context for the observed interrelationships. The loss occurs via two primary pathways: one that results in the formation of inert molecular nitrogen (N₂) and another that produces reactive species within the NO_y family. Adding specific chemical branching ratios and the fact that these yields are heavily influenced by the incoming UV radiation — which is itself regulated by the thickness of the overhead ozone column — are details from broader stratospheric science which would be useful to the readers.

We thank the reviewer for their insightful comments about loss mechanisms. We agree that noting N₂O loss strengthens the manuscript and the argument for non-linearities in Age:N₂O relationships. We have made the following change below:

L396-399 previously read as:

“In addition to insights of stratospheric mixing and transport, the relationships are non-linear, and the presence of curvature is not surprising given N₂O loss throughout the stratosphere. When air enters the stratosphere through the tropopause, it rises gradually and is exposed to N₂O sinks in the middle to upper stratosphere. In the context of tracer interrelationships, air ages with little N₂O loss, and then N₂O_{norm} decreases more rapidly as it ages.”

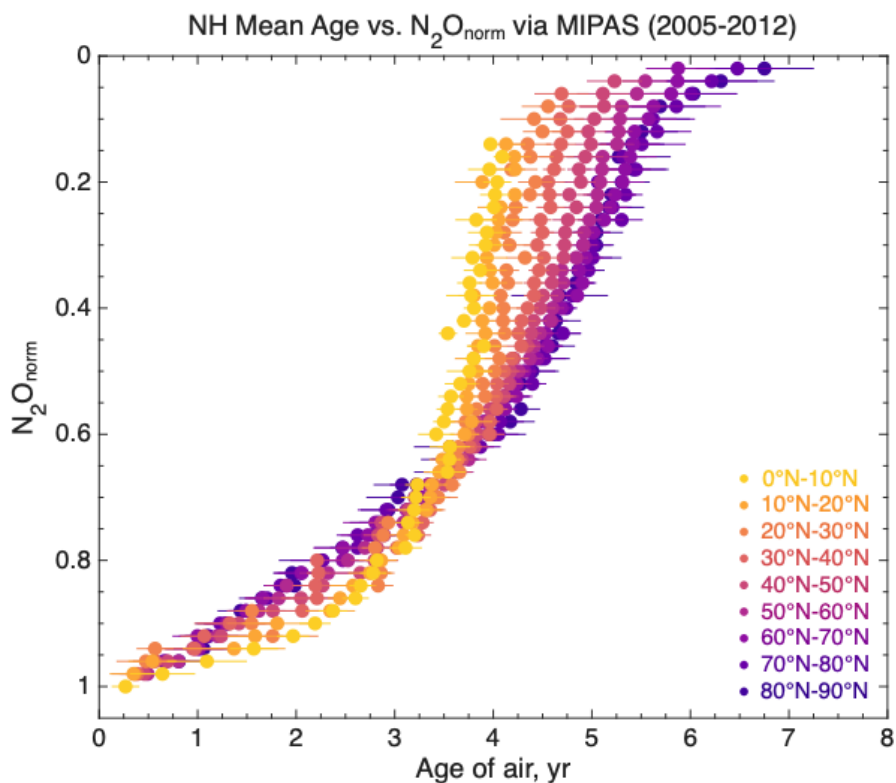
It now reads as:

L317-323: “The latitude dependence of mean-Age:N₂O interrelationships can be conceptualized with a cross section of the BDC in the NH in Fig. 4b. When air enters the stratosphere through the tropopause, it rises gradually and is exposed to higher energy UV radiation in the middle to upper stratosphere, especially above the local ozone maximum, and this UV exposure drives more rapid N₂O loss via photochemical oxidation. In the context of tracer interrelationships, as air in the tropical lower stratosphere increases in age, N₂O decreases more gradually, so the slope of the correlation is relatively shallow. As air ascends, N₂O_{norm} loss rapidly increases while air constantly ages, and the slope of the correlation becomes nearly vertical as the N₂O loss happens much faster than the aging.”

The significance of the inflection points at a normalised value of 0.7 deserves further emphasis in the text as a marker of shifting dynamics. At values above 0.7, which correspond to the lower stratosphere, the tracer distribution is primarily governed by the horizontal mixing of young tropical air with more aged air from the extratropics. Once air parcels reach the 0.7 threshold, they have transitioned into a region where local photochemical sinks become the dominant influence, explaining why the tropical relationship exhibits a rapid decrease in nitrous oxide for a relatively small increase in age

beyond this point. The emergence of hemispheric asymmetry at this same threshold suggests it is a robust indicator for the onset of significant chemical depletion within the stratospheric circulation.

We thank the reviewer for the suggestion to further emphasize the importance of the inflection points at $N_2O_{norm} = 0.7$ as denoted in the original figure below (previously Fig. 5):



In addition, this reviewer also encourages the emphasis of the inflection point as an indicator for shifting dynamics. We have incorporated your suggestions in the text in Sect. 4.1. To facilitate our improved discussion of the inflection point we have updated the mean Age: N_2O (was Fig. 5, now Fig. 4) relationship figure with approximated isentropes overlaid (now Fig. 4a), and we added a zonal cross section. of the circulation to the right (now Fig. 4b).

The new figure and the corresponding caption is below:

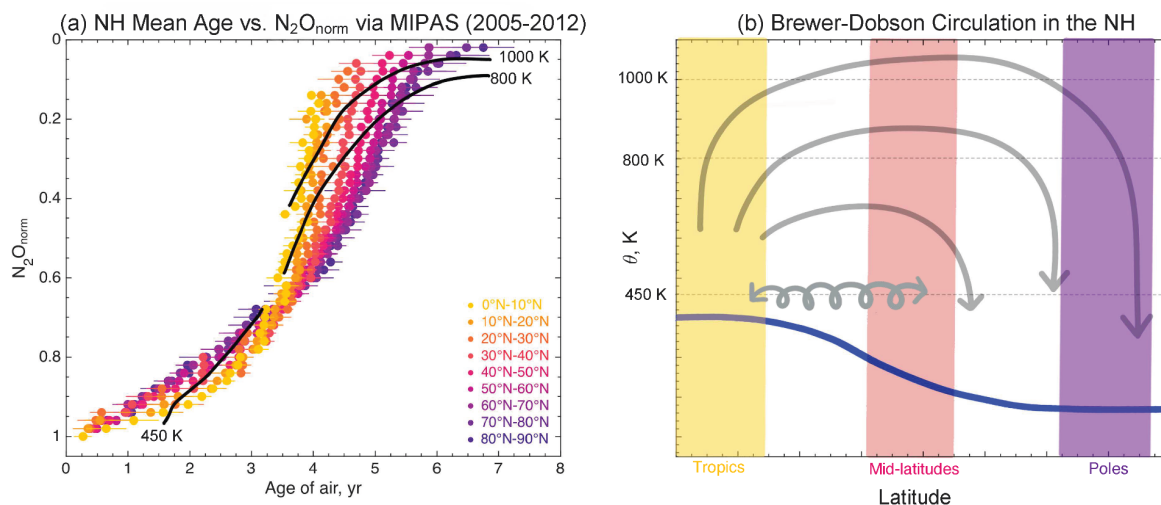


Figure 4: Mean SF_6 -age versus normalized N_2O with a latitude resolution of 10° in the NH averaged over 2005-2012. The solid circles represent the mean age for 0.02 bins of normalized N_2O , with error bars indicating 1 standard deviation. Lighter, warmer colors (yellow and orange) represent lower latitudes, while darker, cooler colors (magenta and purple) represent higher latitudes. Data are from MIPAS V5R_224 and V5R_225 N_2O and SF_6 observations. The black solid lines are approximate isentropic surfaces corresponding to the empirical relationships. (b) A zonal cross section of the BDC with the tropopause is shown by the blue line. The solid arrows denote the approximate zonal flow of the circulation. The yellow, pink and purple bands denote the tropics (0° N- 10° N), midlatitudes (30° N- 60° N) and poles (70° N- 90° N) respectively.

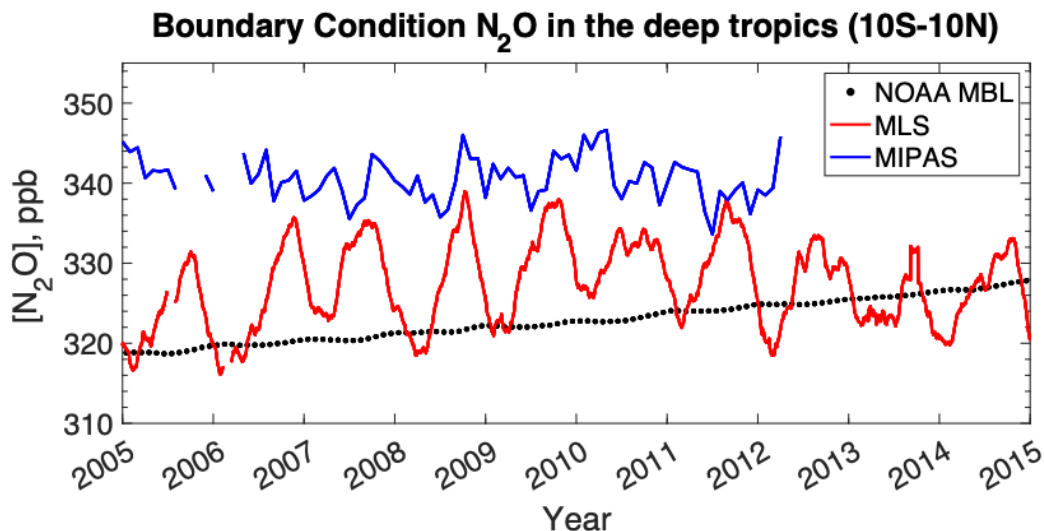
The latitude dependence can be conceptualized in Fig. 4b: air is upwelled throughout the deep tropics (yellow) on average, and then it is entrained into the extratropics (i.e., magenta and dark purple), where air is rapidly mixed on a θ surface and the mean flow is downward. Despite being relatively isolated, tropical air is advected at all levels, as denoted by the squiggly line. We observe mixing of younger, recently upwelled air with older, N_2O -depleted air because of subsidence and poleward transport denoted by the arrows in Fig. 4b. In the context of the Age: N_2O relationships, we see the along isentropic mixing in the middle to upper stratosphere, where air ages and mixes with older N_2O -depleted air as it moves along θ surfaces (i.e., 800K and 1000K), which results in unique Age: N_2O mappings in the extratropics – denoted by subtropics (orange; 10° N- 30° N), midlatitudes (magenta; 30° N- 60° N) and poles (dark purple; 60° N- 90° N).

Minor Comments:

Figure 1: The caption should contain all the relevant information required for reproducibility. Specifically, it should define what is meant by the lower stratosphere in this context and explain how the data has been screened for quality. Furthermore, it is unclear why ACE-FTS data is not plotted alongside MIPAS and MLS in this figure, as it would provide a useful third-party validation for the tropical entry values.

You make excellent points in this comment. We have revised Figure 1 and its caption as the following:

The former figure has only MLS, MIPAS and NOAA MBL observations.



The updated figure includes ACE-FTS:

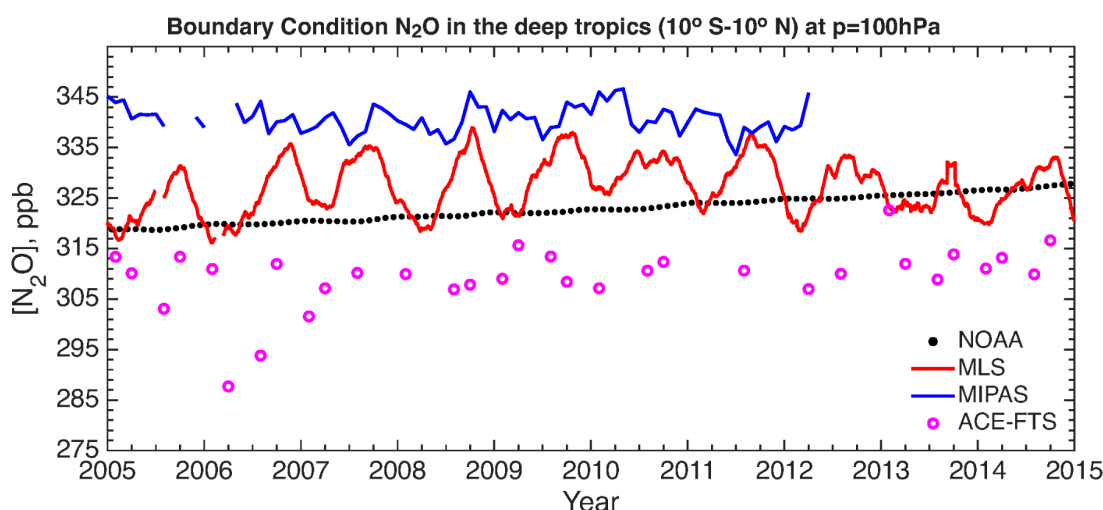


Figure 1: A time series of maximum N_2O concentrations measured by satellites in the deep tropics ($10^\circ \text{ S}-10^\circ \text{ N}$) at 100 hPa in the lower stratosphere (the boundary condition) and tropical NOAA Marine Boundary Layer (MBL) reference N_2O from 2005 to 2012. NOAA MBL reference N_2O is black. The monthly mean N_2O from MIPAS is blue. The 30-day running mean MLS v5 N_2O is red. The monthly mean N_2O measurements from ACE-FTS are shown as the pink circles. Data are from NOAA MBL N_2O , MIPAS V5R_224 and V5R_225 N_2O , MLS v5 N_2O and ACE-FTS v3.5/v3.6 N_2O .

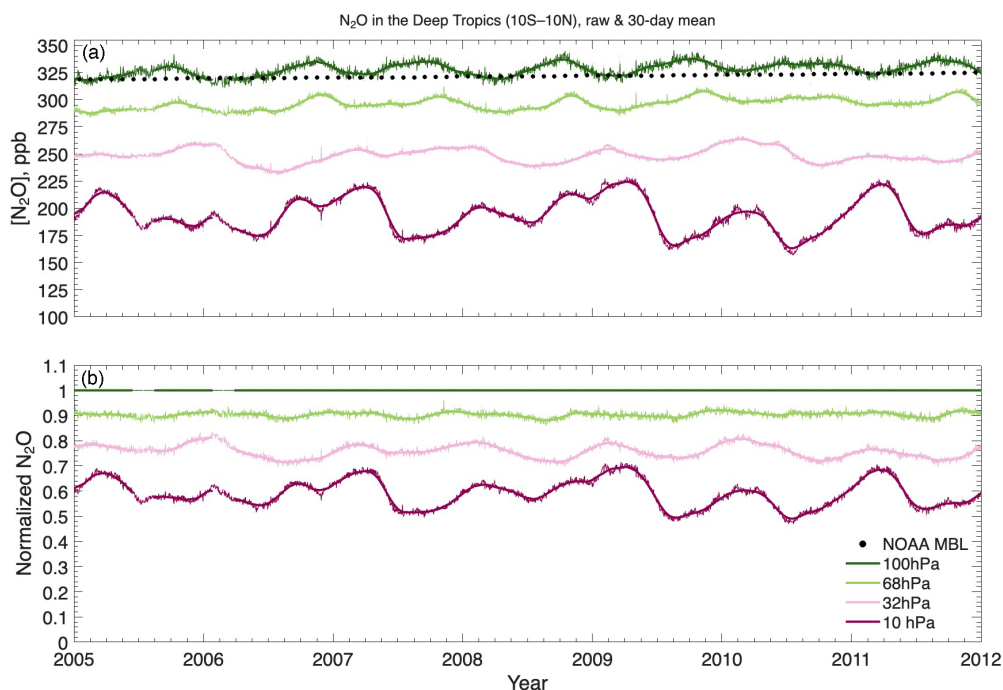
Here we have incorporated ACE-FTS maximum N_2O values at $p=100$ hPa in the deep tropics. We have defined the boundary condition as ($10^\circ \text{ S}-10^\circ \text{ N}$) at 100 hPa in the figure title.

Including this provides more perspective on the biased signatures of all three satellites in the tropics. This addition also strengthens the rationale for using MIPAS observations to create our SF6-age: N_2O because ACE-FTS sampling is sparse – most observations are collected at high latitudes. Meanwhile, ACE-FTS boundary conditions are lower when compared to NOAA MBL observation points.

Line 207: The normalisation procedure is somewhat confusing. Using the maximum VMR at each time step as the denominator may inadvertently skew the distribution. I note that Supplementary Figure S6 shows the time variation of maximum nitrous oxide concentrations in absolute terms, but it does not include the equivalent normalised values. To better assess the impact of the normalisation choice, I suggest modifying S6 to be a two-panel figure: one panel showing the absolute VMR (as currently shown) and a second panel showing the corresponding normalised VMR values. Each panel should include two lines for mean and maximum VMR (solid/dashed) for different pressure levels and an extra line NOAA MBL measurements at the surface), and second panel should show normalised value estimates using both the mean VMR and the maximum VMR as the reference.

We appreciate your concern regarding the normalization process. We have incorporated this insightful suggestion into the supplemental material. In addition, we have moved S6 to S1 per Reviewer 2's suggestion and created a two-panel figure as follows:

The normalization process (Fig. S1b) accounts for the variation and the drift at the tropopause level in the deep tropics (as seen in Fig. 1 and S1a), while conserving the seasonal and interannual cycles at higher pressure levels (re: light green and light magenta, and dark magenta).



S1: Time series of MLS (a) N₂O VMR in ppb and (b) Normalized N₂O in the deep tropics (10°S-10° N) on four pressure surfaces: 100 hPa (dark green), 68 hPa (light green), 32 hPa (light pink), and 10 hPa (magenta). The smoothed line represents the 30-day running mean of maximum N₂O (a) and normalized N₂O (b). The smaller, jagged lines are the raw daily N₂O VMR (a) and normalized N₂O (b). The time series spans from 2005 to 2012. Data are from MLS v5 N₂O.

Finally, I would like to recommend that authors use the TCOM-N₂O dataset (Dhomse and Chipperfield, 2023; 2026) to assist in quantifying the impacts of MLS instrumental drifts.

We thank the reviewer for this suggestion. We agree that incorporating the TCOM-N₂O dataset would be useful for quantifying MLS drift impacts (in addition to the N₂O SWOOSH dataset) in future work.

Reviewer 2:

The main goal of this manuscript is to create robust new global stratospheric age-of-air datasets derived from satellite observations that ultimately may provide the basis for studying the strength and structure of the Brewer-Dobson circulation and how this may be changing on decadal timescales. The authors do this in this work by combining – in what in what I perceive at least (see below) to be an informed and novel way – the best characteristics of satellite observations of the age tracer SF6 and its correlation with N2O from MIPAS (with its high spatial resolution) and ACE-FTS (with its high SF6 precision and a long time series but poor spatial resolution) and to map these relationships onto high precision global MLS N2O observations. One important new aspect of this work is that the authors convincingly show that the age:N2O relationships vary significantly with latitude – as expected but not so convincingly demonstrated previously as is accomplished in this work, to my knowledge – and that they can quantitatively demonstrate (and which they also interpret) in this new study. While the work presented here seems informed and may make important contributions to age of air studies, the manuscript is still in what I would call “rough draft” form. The extent of its roughness means that it was difficult to follow the specifics of their methodology, the explanations for (or lack thereof) and implications of instrument drift and various systematic biases of the different satellite measurements on their study, and even the derivation of as well as distinctions and comparisons between their age-product time series. While I give some specific comments below (with the hope that comments can help guide the substantive edits that need to be made before this manuscript is acceptable), I will generalize here that the current version of the manuscript was difficult and time-consuming to read due to vague and/or imprecise wording throughout (at the phrase, sentence and even paragraph levels), a significant number of typo issues (including errors and/or inconsistencies in figures and text), major redundancies throughout which can be drastically reduced (thus making the manuscript significantly shorter to read through, as well as copyedit!), an unorthodox supplementary material file and its callout in the main text, and a lack of focus on and precise language regarding the most important and unique aspects of the work. In short, I think the work is interesting and important, and the authors’ hard work is apparent. But the manuscript as currently written is unacceptably rough, making it difficult for me to evaluate the technical details underlying it and the implications of this for their overall results and conclusions and, perhaps even more importantly, making it difficult for someone in the future to reproduce their analyses or improve upon them as new satellite data becomes available. I would welcome reviewing the manuscript again after substantial edits are made to improve how the work is written up and explained.

In the spirit of constructive feedback, which I hope is how the authors will perceive them, I provide some notes, questions, and suggestions below.

L12-26 (Abstract): Some additional clarity and specifics would be helpful to have in the abstract (without increasing the length). The extra clarity in the abstract can then be used as a template for the main text. For example, for the main gist of the work starting in ...

We thank the reviewer for spending time on details of the abstract. Please refer to the responses below as we address each point.

L18: “We compute tracer relationships of age of air and N₂O” – from WHAT observations here? In a phrase, clarify that they are from which specific observations. (Indeed, on the first read-through the word ‘compute’ made me think they might be model simulations, even though the sentence before includes the word “observations.” People will be reading the abstract quickly, and the more specific one can be, the better, especially in the abstract.) Here [L18], the authors mean the mapping of the SF₆- age:N₂O relationships from MIPAS (or ACE-FTS) onto the global MLS N₂O observations?

We recognize that this did not draw enough attention to the use of observations. Without overwhelming with details, we have emphasized this by saying “using **three separate satellite datasets** of N₂O”

Also, the authors refer to ‘a’ timeseries [L19], and yet several are shown in, e.g., Fig. 10, at least for comparison. Or alternatively, the other time series in Fig. 10 are either from ‘previous studies’ or ‘previous counterparts’

You’re right! We present a few new time series, so we removed the word ‘a’ and made the rest of the sentence plural.

[L23]? The wording in general and the specific wording of ‘previous counterparts’ is often confusing (in the abstract and the throughout the manuscript).

We have removed this phrasing in the manuscript. In addition, we changed this phrasing to “previous work” on L24.

Finally, L20- L26 – what specifically has been learned from this study – would also benefit from more precise language and better framing of the unique contributions of the results of this work overall, rather than a string of seemingly unrelated sentences that are fairly general and vague [e.g., L21: “a better understanding of the structure and seasonality of the BDC” is obtained]. Why and how specifically is the new N₂O-age product improved? What about our understanding of the BDC is improved with the improved N₂O-age product? This clarity and specificity is also lacking in Sect. 4 (Results and Discussion) and in Sect. 5 (Summary), so fixing it here in the abstract will also aid in editing these sections in the main text to help bring the work of the authors into focus for the reader.

We have edited the abstract to better represent the new contributions of the work.

The text (L20-25) now reads as:

“The tracer interrelationships and their variability with latitude provide a better understanding of the structure and seasonality of the BDC; in particular, latitudinally-resolved Age:N₂O relationships reflect the relative importance of photochemical loss of N₂O in different regions and enable hemispheric structural comparisons. The N₂O-age product has more extensive spatial coverage than previous work. Additionally, N₂O and SF₆-age agree, and Age:N₂O relationships are robust on seasonal and interannual time scales. Although our new time series is only 7 years long, this manuscript lays the groundwork for calculating a longer record of N₂O-age to understand long-term variability and shifts of the BDC.”

Suggestions and comments beyond the abstract:

L56-L65: The authors explain in some detail that overturning mass fluxes and horizontal mixing – critical aspects of the BDC —can be derived from observations of gradients in age of air observations. Yet they never return to this rather extensive introductory setup in the Discussion or Summary; it would strengthen the work of the authors bookended their manuscript with the ultimate use of the N₂O-age time series they work hard to robustly derive and analyze.

We thank the reviewer for this suggestion.

We have modified lines 59 through 67 for conciseness (which is now L52-58, as we have significantly edited the Introduction. The detail about circulation strength and mixing now reads as:

L52-58: “Gradients of age can be qualitatively related to both components of the circulation (Linz et al., 2016; Linz et al., 2021) and used to calculate mean circulation strength using observations of long-lived tracers (Linz et al., 2017). Specifically, the strength of the overturning circulation can be calculated by taking the difference of age between upwelling and downwelling latitudes on a constant potential temperature (θ) contour, and the strength of the circulation has an inverse relationship with the age difference. In order to evaluate shifts in circulation strength using this method, a long-term record of age is necessary for providing a quantitative measure of the stratospheric circulation.”

In addition, we have added what was previously S1 to the main manuscript as Figure 8, where we compare N₂O-age computed by our Age:N₂O latitude-dependent (lat-varying) relationships with N₂O-age derived by a single relationship (lat-fixed) from Andrews et al., (2001). Here, we added a sub-section in the results section as Sect.. 4.2.2 and titled as “ Implications of deriving N₂O-age using latitude-dependent Age:N₂O relationships.” This new section explains the context of the overturning circulation strength and horizontal mixing that is mentioned in the summary all while strengthening the argument of implementing latitude-varying relationships to derive age.

We have added a new figure (now Fig. 8) with the following caption below:

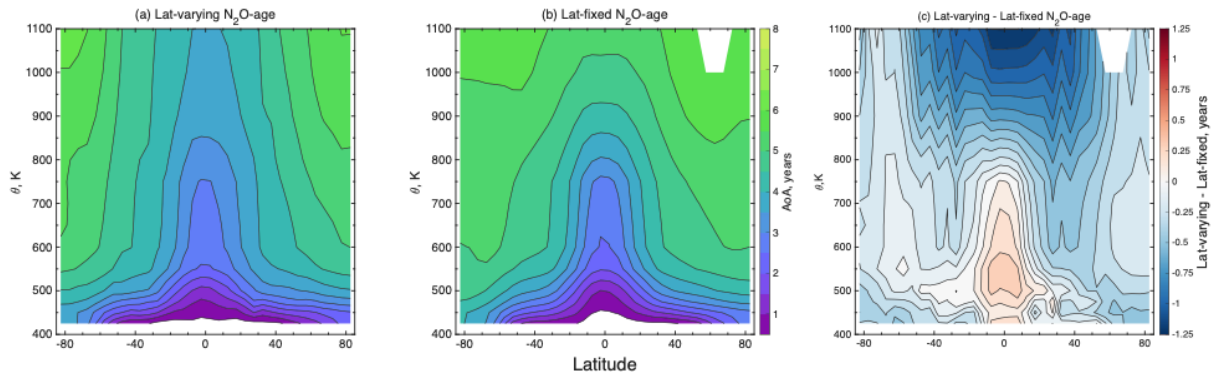


Figure 8: Mean age as a function of latitude and θ averaged from 2005 to 2012 (a) Latitude-varying N₂O-age from MIPAS SF6-Age:N₂O relationships from Fig. 4 (b) latitude-fixed N₂O-age from Andrews et al., (2001) Age:N₂O relationship (or Andrews-age) (c) Actual age difference between latitude-varying age (a) and latitude-fixed age (b) latitude-varying – latitude-fixed (a) and (b) contours are in increments of 0.5 years from 0.5 to 8 years, and (c) contours are in increments of 0.1 years from -1.25 to 1.25 years. Data are from MLS v5 N₂O observations. The Andrews et al., (2001) relationship is based on in situ observations from the 1990s. N₂O measurements are from ATLAS and mean age is from ER-2 derived by CO₂ measurements ranging from 30°N to 70°N .

The introduction in general, especially L28-55 and L66~100: A lot of background material is presented about the BDC. I find the authors' implementation is in general good, but then a lot of this information is extensively rehashed again – again and again in some cases –in later sections, and in ways that do not enhance reader understanding and which in effect add more imprecise language for the reader to muddle through. In many cases, less is more, and I suggest eliminating as much redundancy as possible throughout the manuscript (but while, at the same time, providing enough concise and complete information about which satellite dataset or ‘computation’ resulting from it is being discussed at the moment!). By keeping this in mind throughout the paper – eliminating redundancies at the sentence and paragraph level but inserting critical qualifiers where needed – will enhance readers' understanding and improve appreciation of the authors' work as well as enable others to reproduce the authors' approach and apply it to future datasets.

We have made edits throughout the introduction to reduce redundancies, improve clarity, and add appropriate qualifiers. Thank you for these suggestions.

Putting addressing redundancies into context: While this is one example, we have removed what was Sect. 3.2.2 (approximately 1 page long). The original text was approximately 26 pages and is now 23 pages after removing Sect. 3.2.2 all while including a new section (Sect. 4.2.2).

L109-L121: Both the flow of information as well as clarity of what is being accomplished with which datasets needs to be better focused here. Like the abstract, this is what readers will be looking for in the rest of the manuscript. Some details can be eliminated (e.g., no need to state what will be covered in Sect. 2.2 at L108) while others are important to note here (are monthly averages being taken over, e.g., for all months between for the years 2005 to 2012 for MIPAS observations?). This critical section of the introduction setting up the work in the rest of the manuscript is difficult to follow, thus detracting from the work.

Thank you for both of these suggestions. We have made edits throughout the introduction to reduce redundancies, improve clarity, and add appropriate qualifiers.

Specifically, we have removed the sentence on L108. We have changed L102-104 to specify the resolution of the dataset we use for methods described in L116-119.

L98-107 read as:

“In addition, previous work calculated age from SF₆ observations, with the first vertically resolved global age product using SF₆ observations from MIPAS during the 2002-2012 period (Stiller et al., 2008, 2012; Haenel et al., 2015). This spatially and temporally-resolved dataset created a foundation for age of air studies; however, the short time series is only 10 years, which poses a challenge for circulation trend analysis (see Fig. 1 and discussion below). The measurement qualitatively changed after the first few years, making the continuous record even shorter. This limitation motivated more recent work to develop a longer timeseries of global SF₆-derived age (henceforth SF₆-age) with a sink correction applied to MIPAS and ACE-FTS SF₆ described in Sect. 2 (Saunders et al., 2025; Garny et al., 2024a). Saunders et al. (2025) calculated a new global, satellite-based SF₆-age dataset using ACE-FTS SF₆ observations, including a mesospheric sink correction from Garny et al. (2024a). This dataset spans from February 2004 to February 2021, representing the longest available age of air timeseries from SF₆ profiles

It is now L87-95, and it reads as:

“In addition, previous work calculated age from SF₆ observations, with the first global age product created using SF₆ observations from MIPAS during the 2002-2012 period (Stiller et al., 2008, 2012; Haenel et al., 2015). This zonal-mean, monthly-mean dataset created a foundation for age of air studies; however, the time series is only 10 years, which poses a challenge for circulation trend analysis (see Fig. 1 and discussion below). The measurement qualitatively changed after the first few years, making the continuous record even shorter. This limitation motivated more recent work to develop a longer time series of global SF₆-derived age (henceforth SF₆-age). Saunders et al. (2025) calculated a new global, satellite-based SF₆-age dataset using ACE-FTS SF₆ observations, including a mesospheric sink correction from Garny et al. (2024a). This zonal-mean, monthly-mean dataset spans from February 2004 to February 2021, representing the longest available age of air time series from SF₆ profiles.”

To further clarify what is being accomplished with each dataset we have changed L113-121 from:

“In this study, we present two new data products. (1) We derive latitude-dependent relationships between sink-corrected SF₆-age and N₂O based on MIPAS observations from 2005-2012. We then apply the relationships from (1) to MLS and ACE-FTS N₂O observations to derive a (2) timeseries of N₂O-age from 2005 to 2012. While these timeseries are shorter than previous counterparts, this paper lays the groundwork for an eventual long record of N₂O-age for future long-term variability and trend calculations. The following section describes the data, and Sect. 3 covers the methods used in developing the products. In Sect. 4, the content of both products—MIPAS Age:N₂O and N₂O-derived age from MLS and ACE-FTS will be discussed in detail. Caveats and biases, followed by the reasoning for a shorter timeseries will then be addressed. Finally, we summarize the results and age comparisons.”

To what is now L96-107:

“This work uses results from these recent studies to introduce a new time series of stratospheric age of air derived from N₂O measurements from National Aeronautics and Space Administration’s (NASA) MLS and Canadian Space Agency’s ACE-FTS over the MIPAS time period: 2005 to 2012. MLS N₂O-age has both the high spatial and temporal resolution and long temporal coverage that MIPAS and ACE-FTS age products provide separately. We (1) derive latitude-dependent relationships between sink-corrected SF₆-age and N₂O based on MIPAS observations averaged over 2005 to 2012 and (2) time series of monthly-mean, zonal-mean N₂O-age from 2005 to 2012 by applying the relationships from (1) to MLS and ACE-FTS N₂O observations. While these time series are shorter than previous studies, this paper lays the groundwork for a long record of N₂O-age for future long-term variability and trend calculations.”

Sect. 2 on Satellite Datasets, especially 2.1 and 2.3: Although such ‘methods’ sections should always contain a lot of specific information, it should still be readable, and I found these sections difficult to parse, even though they contain some critical information about systematic errors, instrument drifts, and how specifically these were dealt with. This information needs to be understandable in order to help evaluate the authors’ approach. I found several major sections vague or just confusing For example, L141-150: what specific data are being used, which are being ‘re-used’ or re-calculated, which are being created or re-created, which are zonal means in a time series, which are monthly means or spatial (i.e., meridionally-resolved) and are they monthly means over every year in a given time period, or...? In short, how the data are gridded and averaged and what is new to this study and which are ‘previous products’ is unclear. While I don’t need to follow every detail,

with the lack of clarity in the abstract and introduction and further lack of clarity here, it is difficult to just ‘trust’ that the approach is sound or that others will be able to reproduce it in future work.

This comment is much appreciated. As a result, this section has been drastically revised. Although we address some changes to L141 to 150 as the following:

For 2.1 (L124-150), the previous text reads as:

“As seen in Fig. 1, the NOAA Marine Boundary Layer (MBL) reference time series demonstrates that the global average N_2O increases in the troposphere. In addition, MBL N_2O (Lan et al., 2026) is plotted to compare with the maximum values observed in the tropics by satellite observations. In this work, we use MIPAS versions V5R_224 and V5R_225 as used in Garny et al. (2024b) (Haenel et al., 2015). SF_6 and N_2O (Plieninger et al., 2015; 2016) retrievals are biased in the deep tropics, with N_2O VMR larger than those from NOAA MBL observations. In contrast to N_2O , SF_6 retrievals are biased lower, thus resulting in older SF_6 -age in the tropics compared to that of ACE-FTS SF_6 -age and in situ CO_2 and SF_6 -age (Garny et al., 2024b, Saunders et al., 2025).

We use SF_6 -age of air (with and without a sink correction) created by Saunders et al. (2025) and N_2O to compute a corresponding N_2O product specifically for this study. The monthly zonal mean N_2O and SF_6 -age from 2005-2012 on both altitude (km) and θ surfaces were obtained from MIPAS V5R_224 and V5R_225 products. The monthly zonal mean values from the provided dataset are gridded in 5° bins ranging from 90° S to 90° N. In addition, we also evaluate the effects of using SF_6 -age with a SF_6 sink correction applied from Garny et al. (2024a) in our analysis.”

For 2.1 (L110-136), the new text reads as:

“MIPAS was a Fourier-transform spectrometer launched into a sun-synchronous orbit on Envisat on March 1, 2002. It had a high spatial resolution and a high spectral resolution ($415\text{-}14.6\text{ }\mu\text{m}$), and it was designed to measure temperature and trace gas abundances, including 20 trace gases (Fischer et al., 2008). Vertical profiles of tracers were measured over an altitude range of 6 to 150 km (depending on the gas) on a sun-synchronous orbit, crossing the equator at 10:00 h (morning) and 22:00 h (evening). MIPAS operated at its original full-spectral resolution until March 2004. There was an instrumental issue resulting in a gap from March 2004 to January 2005 before resuming operation, where the spectral resolution decreased from 0.05 cm^{-1} to 0.125 cm^{-1} , although vertical sampling increased by about 20%. After the switch of operation mode in 2004, MIPAS was operated on a reduced duty cycle during 2005 and 2006, before it resumed full operation again. Therefore, some gaps in the time series during these two years occur. MIPAS collected approximately 1300 individual profiles per day, covering latitudes ranging from 87° S to 89° N with an inclination angle of 98.55° . Profile measurements were retrieved on a uniform vertical grid with an altitude range from 6-70 km.

In this work, we use MIPAS versions V5R_224 and V5R_225, specifically N_2O and SF_6 from 2005 to 2012 (Plieninger et al., 2015; 2016; Haenel et al., 2015). We also use the monthly-mean, zonal-mean sink-corrected SF_6 -age of air from Saunders et al. (2025) and N_2O from Garny et al. (2024b). These datasets are provided in altitude (km) coordinates, and we also created new versions

on isentropic (K) coordinates. All zonal-mean datasets are averaged into 5° bins ranging from 90° S-90° N. We use sink-corrected SF₆-age because the correction provides a more accurate representation of age of air, and we observe these impacts in S2 and provide additional context for the sink correction in the Supplement.

Previous work has shown that MIPAS SF₆ and N₂O retrievals are biased in the deep tropics at altitudes below ~25km (Pleninger et al., 2015; 2016). In Fig. 1, MIPAS monthly mean N₂O data are compared with measurements from the NOAA Marine Boundary Layer (MBL) N₂O reference time series (Lan et al., 2026). MIPAS measurements are biased high. In contrast, MIPAS SF₆ retrievals are biased low, which would result in older SF₆-age estimates in the tropics compared to that of ACE-FTS SF₆-age and in situ CO₂ and SF₆-age (Garny et al., 2024b, Saunders et al., 2025).”

The comment on 2.3 is much appreciated. Upon rereading, we have rewritten the subsection as follows:

For 2.3 (L169-193), the previous text reads as:

“MLS (Waters et al., 2006) is a NASA satellite on the Aura mission (Schoeberl et al., 2006), which that was launched on 15 July 2004 and is in a sun-synchronous, near-polar orbit at approximately 705 km with an inclination of 98°. In addition, MLS observes thermal microwave emissions, 190 GHz in the context of N₂O and 18 and 240 GHz for temperature. MLS makes approximately 14 orbits and ~3500 scans per day, to provide daily near-global spatial coverage spanning a latitude range of 82° S to 82° N. This spatial and temporal coverage make MLS unique and particularly useful for our applications. The instrument measures vertical profiles of tracers and chemical constituents. In this study, we use level 2 (swathed), version 5 (v5) of MLS from vertical ranges of 100 hPa to 0.46 hPa, with a vertical resolution of approximately 3km. In particular, we use N₂O (Lambert et al., 2021) and temperature (Schwartz et al., 2021) to calculate θ so both N₂O and N₂O-age are expressed in isentropic coordinates.

Here we use the simplified assumption of the tropical tropopause boundary – the maximum N₂O value at 100hPa between 20° S and 20° N – to address the unphysical seasonality signature from retrievals and to normalize correctly. MLS N₂O increases until around 2010, while after this date, a decrease magnifies over time. This drift becomes apparent in the time series around 2012, but it is not prominently visible in Fig. 1. This drift is height dependent, where it decreases with height (Livesey et al., 2021), and so the drift is not constant with altitude.

In this study, we use MLS N₂O over the same time period as MIPAS (2005-2012) to ensure the most accurate N₂O-derived age that considers both the drift and possible shifts in of SF₆-Age:N₂O relationships that could result in biases in computed N₂O-age over time. We further elaborate on this in Sect. 3.1

Corrections were applied to account for the drift of decreasing N₂O concentrations in v5 retrievals, but the data still departs from tropospheric observations over time. The seasonal cycle is unrealistically strong in the lowermost tropical stratosphere in v5 MLS, which is not present at all heights and can be seen in Fig. S6. Attempts to account for this spurious seasonal cycle will be

discussed in Sect. 3.1. In addition to the MLS drift and seasonal cycle, both MIPAS and MLS have biased higher N₂O VMR in the tropics (e.g., Fig. 1) that could result in biased younger ages. These biases need to be taken into consideration and will further be addressed in Sect. 3.1 as well.”

For 2.3 (L156-187), the text now reads as:

“MLS (Waters et al., 2006) is a NASA satellite on the Aura mission (Schoeberl et al., 2006), which was launched on 15 July 2004 and is in a sun-synchronous, near-polar orbit at approximately 705 km with an inclination of 98°. MLS observes thermal microwave emissions, 190 GHz in the context of N₂O and 118 and 240 GHz for temperature. The instrument measures vertical profiles of tracers and chemical constituents – making approximately 14 orbits and ~3500 scans per day – to provide daily near-global spatial coverage from 2004 to present day while also spanning a latitude range of 82° S to 82° N. This spatial and temporal coverage make MLS unique and particularly useful for our applications to create a longer age time series in future work.

In this study, we use level 2 (swath), version 5 (v5) of MLS data, and these datasets are provided on pressure surfaces ranging from 100 hPa to 0.46 hPa, with a vertical resolution of approximately 3 km. In particular, we use daily N₂O (Lambert et al., 2021) and daily temperature (Schwartz et al., 2021) from 2005 to 2012. Both N₂O and temperature are indexed on pressure surfaces ranging from 100 hPa to 1 hPa to best reflect the stratosphere.

To use long-lived trace gases and age of air as metrics to diagnose stratospheric transport, we transform the height coordinates of N₂O from pressure surfaces to isentropic coordinates. Potential temperature is calculated at each pressure level and daily retrieval using corresponding temperature dataset. Given the horizontal resolution of MIPAS and ACE-FTS N₂O, we re-grid the swath N₂O and θ observations by binning data at each pressure level into 5° bins ranging from 90° S to 90° N. Then we calculate zonal-mean of the binned values at each daily timestep from 2005 to 2012, and so both variables are functions of latitude, pressure and time. Using the zonal-mean θ as the vertical coordinate, we interpolate the zonal-mean N₂O on a fixed set of 13 θ surfaces where θ ranges from ranging from 425 K to 1000 K. As a result, we use daily zonal-mean MLS N₂O as a function of latitude, θ , and time.

It is important to note that MLS N₂O has a drift. MLS N₂O increases until around 2010, while after this date, a decrease magnifies over time. This drift becomes apparent in the time series around 2012, but it is not prominently visible in Fig. 1. This drift is height dependent, where it decreases with height (Livesey et al., 2021), and so the drift is not constant with altitude. The seasonal cycle is unrealistically strong in the lowermost tropical stratosphere, which is not present at all heights and can be seen in Fig. S1a and b. Corrections were applied to account for the drift of decreasing N₂O concentrations in v5 retrievals (Sect. 3.1), but the data still departs from tropospheric observations over time (refer to Fig. S1b).

Here we use the simplified assumption of the tropical tropopause boundary – the maximum N₂O value at 100 hPa between 20° S and 20° N – to address the unphysical seasonality signature from retrievals. Attempts to account for this spurious seasonal cycle will be discussed in Sect. 3.1. In addition to the MLS drift and seasonal cycle, both MIPAS and MLS have biased higher N₂O VMR

in the tropics (e.g., Fig. 1) that could result in biased younger ages. These biases need to be taken into consideration and will further be addressed in Sect. 3.1 as well. We use MLS N₂O over the same time period as MIPAS (2005-2012) to ensure the most accurate N₂O-derived age; and this considers both the drift and possible shifts in of SF₆-Age:N₂O relationships that could result in biases in computed N₂O-age over time.”

Unorthodox Supplementary Material: The first mention of supplementary material is for “Fig. S6” on L188. Figure S1 is not called out until the Summary (Section 5) – although I argue that that is such an important and motivating figure that it actually ought to be in the main text! My suggestions: First major issue with the SM: The supplementary figures and/or text should be called out in order in the text. More importantly, the second major issue is that the figure captions and associated text nearby in the SM should be understandable. I found much of the text in the supplementary material to be unintelligible and not even rising to the level of a rough draft. Your comment is appreciated. We have moved S6 to the S1 placement in the order of the text to improve the flow of the manuscript. And we have noted this in the text (see responses above). Thank you for suggesting this!

We have since edited the captions of each supplemental figure per your suggestion. In addition, we have removed S5, as it is not relevant to the text material as suggested.

Methodology (Section 3): This long multiple sub-headings section is crucial for documenting the approach and yet needs to be edited for clarity. Furthermore, some of the material in Section 2 (satellite data) would fit better into Section 3. For example, there is mention of various biases, drifts, or artifacts in Section 2, only to be repeated in Section 3 when discussing how they dealt with it. A brief mention of it would be ok – an acknowledgement in the satellite data section that there may be an issue. But to bring it up without discussing it clearly in Section 2 until later in Section 3 serves to undermine confidence versus if it were covered once, and thoroughly, in the methodology section. [It would also shorten the sections overall, I argue, and therefore require less editing to make it easily understandable and less reading to better see the forest...]

Agreed. See response to comment referring to Sect. 2. We extensively mention the drift in Sect. 2.3 as to not take away from Sect. 3.

At this point, having read Sections 1-3, I think that a good copyeditor may be needed to correct grammar, word usage issues, typos, coherence, flow and connection of ideas, and so on, if the coauthors do not have time to contribute. For example, in L224-L246, the text is rambling, oddly colloquial, unconvincing or not understandable in places, or naively expressed, which all undermine the authors’ important approach that they are trying to express here... Except where something has made a scientific conclusion questionable, I will not mention these in my comments below.

We thank the reviewer for this observation. In addition to the specific items below, we have undertaken to improve Sect. 3.1 (L195-246) through significant rewriting.”

L224-L246 previously read as:

“Normalizing N₂O makes it easier to compute stable tracer interrelationships over an extended period of time because this transforms observations on to the same scale and takes changing

boundary conditions into account. In addition, any bias between instruments is inherently removed during this step in the lower stratosphere, assuming that the bias is constant over altitude and latitude. This is not the case with MIPAS, where the bias is limited to altitudes below $\sim 25\text{km}$, and MLS, where the impact of the drift decreases with increasing altitude. The biases and variability are demonstrated in Fig. 1, where MLS has a seasonal cycle that is not physically observed in the stratosphere, and this is a signal unique to MLS. In addition, the variability and drift impacts are not constant in time (e.g., Fig. S6), but normalizing to daily maximum values in the deep tropics maps variability on the same scale, although it is not perfect. Nonetheless, normalizing N_2O accounts for the biases of tropical measurements, as seen in Fig. 1, and allows for comparisons using multiple years of observations, thereby enabling the derivation of climatological relationships. The method is also useful because it avoids the limitations of tropopause height – it accounts for the possible shifts in thermal structure over time, potentially allowing for comparisons over multiple decades.

The general shape of the distribution is illustrated in Fig. 2, where we have the zonal mean of MLS v5 N_2O observations as a function of height, or potential temperature (θ), from January 2005 to December 2012. Fig. 2a shows MLS zonal mean VMR, and Fig. 2b shows the zonal mean $\text{N}_2\text{O}_{\text{norm}}$. In Fig. 2a, the maximum zonal mean N_2O is centered in the deep tropics at $\theta = 425\text{K}$, where N_2O abundant air is upwelled into the stratosphere. In this context, the boundary condition is defined as zero age, or $\Gamma = 0$ and $\text{N}_2\text{O}_{\text{norm}} = 1.0$, where air enters the stratosphere. As air moves poleward, it disperses and mixes with air from higher altitudes, that has been subjected to transport through and chemical sinks regions and subsequent photolytic loss of the tracer. Consequently, $\text{N}_2\text{O}_{\text{norm}}$ decreases with latitude and altitude. For example, $\text{N}_2\text{O}_{\text{norm}} = 1$ denotes new or $\text{N}_2\text{O}_{\text{norm}}$ abundant air (100% remaining), and $\text{N}_2\text{O}_{\text{norm}} = 0.02$ indicates the air is almost entirely depleted (2% remaining). The shape and distribution of normalized N_2O VMR and $\text{N}_2\text{O}_{\text{norm}}$ is nearly the same, with a bell-shaped structure. Therefore, normalized N_2O also reflects the structure of the stratospheric circulation, making it an excellent scaled metric to better understand the BDC.”

It now reads as (L209-218):

“Normalizing N_2O makes it easier to compute stable tracer interrelationships over an extended period of time because it takes changing boundary conditions into account. In addition, any systematic bias between instruments is removed during this step in the lower stratosphere. Although biases generally vary over altitude and latitude (e.g., Fig. S1), normalization maps N_2O variability on the same scale, although it is not perfect. The method is not fixed to a particular pressure level or tropopause height; thus, it accounts for shifts in thermal structure, allowing for comparisons over multiple decades.

Fig. 2 shows the zonal-mean distributions of N_2O (a) and $\text{N}_2\text{O}_{\text{norm}}$ (b) as a function of potential temperature, based on MLS v5 measurements taken over the 2005 to 2012 time period. The maximum zonal-mean N_2O is located in the tropics at $\theta = 425\text{ K}$. In this context, we define the boundary condition as zero age, or $\Gamma = 0$ and $\text{N}_2\text{O}_{\text{norm}} = 1.0$, where air enters the stratosphere. As air moves poleward, it mixes with air from higher altitudes that has been subjected to photolysis and subsequent loss of the tracer. Consequently, N_2O and $\text{N}_2\text{O}_{\text{norm}}$ decrease with latitude and altitude, as expected.”

L250-270: This is largely redundant with the introduction on the BDC, its effect on tracer distributions, the fact that Plumb and Holton and Mahlman recognized it decades ago, etc. While you might need some discussion/context here, it repeats quite a bit of the intro (and this happens several more times in other sections (see below when I call them out again))

This comment is appreciated. We addressed this by removing some of the text in the introduction.

L250-270 previously read as:

“Since all tracers are transported by the same circulation, they are all affected by stratospheric transport. Therefore, the distribution of long-lived tracers is primarily determined by transport as well as the chemical or photolytic loss of each tracer (Holton 1986, Mahlman 1986). Because tracer isopleths are closely aligned, if one were to plot one long-lived tracer against the other (henceforth referred to as tracer interrelationships or tracer:tracer relationship, e.g., CH₄:N₂O relationship), this results in compact correlations, meaning that they have a clear mapping with each other (Plumb, 2007). Compactness arises for two reasons: (1) tracers are long-lived, meaning that their stratospheric lifetimes are longer than the timescale of the stratospheric circulation, and (2) tracers are transported by the same circulation (e.g., Fig. 2). The simplest example of tracer interrelationships for two conserved tracers with constant boundary conditions. These ‘clock’ tracers would scale linearly, so one conserved tracer can be mapped or plotted on to the other with a constant linear slope. In reality, sinks and boundary conditions vary over time for different tracers. These variables introduce the presence of nonlinearities – or curvature – in tracer interrelationships. Fig. 3 demonstrates this, with SF₆-age and N₂O_{norm} plotted against each other. There is a clear non-linear mapping with similar curvature based on satellite observations of MIPAS and ACE-FTS, where mean age increases as N₂O decreases in abundance.

Plumb (2007) demonstrated that tracer relationships also vary meridionally using in situ observations, and we show this in Fig. 3. Mixing between two different regions of the stratosphere can cause spread or variability around the curves at given latitudes. This is because the tropics are relatively isolated from the extratropics (outside of 20° S and 20° N), resulting in a so-called tropical pipe (Neu and Plumb, 1999). This is evident in Fig. 2, where there is a weak vertical gradient above the peak N₂O and $\theta=550$ K. Tropical relationships also differ from the extratropics because vertical velocities are smaller than mixing along isentropes, also known as quasi-horizontal mixing. The polar vortex isolates air from the midlatitudes, thus resulting in different relationships in the higher latitudes. Further discussion on latitude varying tracer interrelationships will be in Sect. 4.”

It now reads as (L224-243):

“As previously discussed, the distribution of long-lived tracers is primarily determined by transport, as well as the chemical or photolytic loss of each tracer (Holton 1986, Mahlman 1986). Because tracer isopleths are closely aligned, plotting one long-lived tracer against the other – henceforth referred to as tracer interrelationships or tracer:tracer relationship (e.g., CH₄:N₂O relationship) – results in compact correlations, meaning that they have a clear mapping with each other (Plumb, 2007). This compactness arises for two reasons: (1) tracers are long-lived, meaning their chemical lifetimes exceed the timescale of the stratospheric circulation, and (2) tracers are transported by the same circulation (e.g., Fig. 2). The simplest example of tracer interrelationships is two conserved

tracers with constant boundary conditions: these tracers would scale linearly, so one conserved tracer could be mapped or plotted onto the other with a constant linear slope.

In reality, sinks and boundary conditions vary over time for different tracers, which introduces the presence of non-linearities – or curvature – in tracer interrelationships. Fig. 3 demonstrates this, with SF_6 -age and $\text{N}_2\text{O}_{\text{norm}}$ plotted against each other. There is a clear mapping with similar curvature based on satellite observations of MIPAS and ACE-FTS, where mean age increases as N_2O abundance decreases.

Plumb (2007) demonstrated that tracer relationships also vary meridionally using in situ observations, and we show this in Fig. 3. Mixing between two different regions of the stratosphere can cause spread or variability around the curves at given latitudes. This is because the tropics are relatively isolated from the extratropics (outside of 20° S and 20° N), the so-called ‘tropical pipe’ (Neu and Plumb, 1999). This is evident in Fig. 2, where there is a weak vertical gradient above the peak N_2O and $\theta=550 \text{ K}$. Tropical relationships also differ from the extratropics because vertical velocities are smaller than velocities associated with mixing along isentropes. The polar vortex isolates air from the midlatitudes from the air downwelling from the mesosphere above, thus resulting in different relationships in the higher latitudes. Further discussion on latitude-varying tracer interrelationships will be in Sect. 4.”

We would like to note that, while the Sect. 3.2 Tracer Interrelationships revisits some aspects stated in the introduction, it provides context and motivation for using MIPAS for computing our latitude-varying Age: N_2O relationships in Sect. 3.3. (e.g., explaining the spread and variability of ACE-FTS relationships in comparison to MIPAS) and discussing the relevance of both chemistry and mixing described in Sect. 4.1.

L260: I only see MIPAS data in Figure 3; Figure 4 (not mentioned here) shows ACE-FTS data.

We thank the reviewer for catching this discrepancy. This is an error on our part. We have since reorganized the figures accordingly – what was Figure 4 (MIPAS Age: N_2O with ACE-FTS relationship overlay) is now Figure 3. We provide further detail on these changes in our response to the following comment below.

We have also made the distinction that ACE-FTS relationships are in Fig. 3 in L250-252 as explanation for why MIPAS data was used over ACE-FTS.

“This study provides a mapping of Age: N_2O using satellite data spanning both hemispheres. MIPAS SF_6 -age and $\text{N}_2\text{O}_{\text{norm}}$ were used to examine latitudinal relationships in the SF_6 -age: N_2O relationship averaged over the 2005 to 2012 time period because MIPAS sampling is much denser than ACE-FTS (e.g., Fig. 3).”

Section 3.2.2 “ SF_6 sink correction”: This section is long and convoluted, with grammar and organization issues. Detail is also being repeated from Section 2. I understand that the authors are now showing the satellite observations for SF_6 both corrected and not-corrected for the mesospheric sink, but all the background does not need to be repeated here (which would therefore

save space and time for editing it to be readable). Instead, I advise the authors to aim for conciseness and clarity and focus on what is needed for readers to understand and future scientists to reproduce their method (for these or newer data).

Thank you for this suggestion. We agree that this level of background detail is not essential for the main text and narrative of this manuscript, as it takes away from the motivations and details of this study. We have adjusted and moved Sect. 3.2.2 and Figure 3 to the Supplemental on pages 2 and 3.

Section 3.4: If I weren't already familiar with this idea of mapping the mean age:N₂O relationships onto global scale MLS measurements, I would not be able to follow this critical section. It is vague, consists of 'sentence dumps' (e.g., includes applications and conclusions as well as some methodological descriptions in a seemingly random order.)

We thank the reviewer for this excellent point. This is very helpful. Some details about the data used were repeated in this section, introducing redundancies (e.g., L455-458). The details now appear in Sect. 2.3, as to not take away from the methodology of deriving age described in Sec. 3.4. To be more straightforward, we have entirely rewritten this section for readability and for replication for age to be derived from the data itself.

Because we go into more details about the data and drift in 2.3, we have shortened Sect. 3.1. We discuss not only the data sources and types, but also the processes of gridding and interpolating data from pressure coordinates to potential temperature coordinates.

Section 4 (Results and Discussion): This section begins with the sentence “We want to emphasize which data are used in which parts of this study.” I would not recommend starting the entire Results section out with a statement like this if the authors want to highlight the important aspects and conclusions of their work.

Agreed. This sentence has been removed.

Figure 5: A critical figure for the work! And the clear inflection point is very cool and NEW! The authors need to hype this figure up more! Shorten up and clarify the methodology and set up in order to get readers to focus on this figure and the discussion of it. A few suggestions: the authors frequently write in text and figure captions “[some property] from 2005-2012”; this is ambiguous – is it an average over the years? At least say “[some property] for the period 2005- 2012.” Or, if I'm mistaken, then that should be informative on how it should be clarified/fixed. Note also that the authors use ‘we demonstrate’ or ‘we illustrate’ in an odd way here and elsewhere. Explain first, then conclude that these observations, or this analysis of the observations demonstrates or illustrates something...

We thank the reviewer for this comment! We added some clarity for each instance of “[insert variable here] from 2005-2012” throughout the manuscript.

For example in the Fig. 4 caption that reads as “Mean SF₆-age versus normalized N₂O with a latitude resolution of 10° in the NH averaged from 2005 to 2012 time period.”

In addition, we have since removed the specific ‘demonstrate’ and ‘illustrate’ throughout the manuscript when appropriate, as we recognize that some of these instances can take away from the results shown here.

Regarding the important, final sentence:

We explain characteristics of Tracer Interrelationships in Sect. 3.1, and then we make conclusions in 4.1. In addition, we have altered components of Sect. 4.1. Since the tracer:tracer space is not always intuitive, providing illustrations provides context of the theory in relation to the physical processes in the stratosphere. As a result, we are able to describe the drivers of the inflection point and so on. In light of explaining and then showing, we have altered Fig. 5 (now Fig. 4 because we moved the sink correction section to the Supplement) to be two panels. We have updated Fig. 4 as a 2-panel figure with the Age:N₂O relationships with different colors representing a relationship at specific latitude bins with overlaying isentropes in Fig. 4a. In Fig. 4b, we show zonal structure of the circulation with colored latitude bands corresponding to a specific relationship, where the zonal structure is both a visual reference and explanation of Fig. 4a throughout the text.

L413-L414: The sentence “Overall...”: Do the authors really mean that the overall take-home message for this section is that “shifts in relationships are a useful metric to evaluate perturbations in mixing”? Also, if this is being introduced as a new metric, it needs to be discussed first and/or not be used as the main conclusion for this whole section, as I believe there are also other important takeaways here.

We thank the reviewer for this notable comment. Based on this comment and the previous suggestion, we agree – there is much more to take away from this section (and this final sentence, which has since been removed)

Both reviewers have emphasized the importance of this section, and so we have provided additional insight into the mechanisms that drive the shifts in relationships and how they relate to chemistry and mixing in Sect. 4.1, specifically altering Fig. 4 in the discussion.

L420: “stratospheric structure” without referring to dynamics or transport and the BDC seems really odd to me. I suggest including a few words of this context.

This is a good point. We will briefly describe what drives the midlatitude characteristics.

The new sentence now reads as:

L364: “Conversely, tracer interrelationships capture stratospheric structure in both hemispheres, where there is more horizontal mixing as a result of wave-breaking and stronger circulation overall in the NH. In addition, these relationships demonstrate hemispheric asymmetry in the mixing regions from 30° to 40° N and S in Fig. 5b and midlatitudes from 50° to 60° N and S in Fig. 5c.”

Figure 6 (and Figure 7): Needs a, b, c, d labels.

Since panel labels are already included in each subfigure title, we are unsure what is needed here.

Also, the wording in the main text and in the figure caption are not consistent with each other, e.g., the ‘mixing region’ is usually 20-30o not 40-50o . [See the text and Figure captions.] Further, the

“extratropics” is generally considered to be ~35-90° (except when there is also a polar vortex). Yet the authors use ‘extratropics’ to mean midlatitudes and only to 70° . I can be consistent but we are clear about our definitions.

We define ‘extratropics’ in Line 238 as follows:

“This is because the tropics are relatively isolated from the extratropics (outside of 20° S and 20° N)...”

We believe this usage is consistent with our stated definition. However, we do recognize the importance of using terminology consistently. Given the definition of the ‘extratropics’ above, we use this defined term throughout the text. In addition, we have also changed “mixing regions” to “midlatitudes” throughout the manuscript (e.g., L421)

Also in text and Figure captions: “absolute age difference” – I do not think the authors mean this. The word ‘absolute’ modifying a difference refers to the absolute value or the magnitude of the age difference but the authors mean the actual difference, and it is important whether that difference is positive or negative (as is clear in their discussion)!

We thank the reviewer for pointing out the incorrect terminology. We have addressed this in the figure caption to read as: “(c) Age comparison between MLS N₂O-age (a) and MIPAS SF₆-age (b) (MLS N₂O-age – MIPAS SF₆-age).” Instead of “absolute age difference,” we now use the term “actual age difference” throughout the manuscript (e.g., captions in Figs. 7, 8, 9).”

Figure 8: There are errors in the text, the figure caption, and/or the figure labels. The figures appear to be MLS minus MIPAS in 8c. This is mixed up in various places and needs to be fixed. This comment is appreciated. We corrected errors in the figure captions. See response above.

Figure missing in here somewhere and in the line of reasoning and presentation in the paper, IMO: Fig S1: I think an important case to be made and to be discussed is how important using the latitude dependent SF₆-age:N₂O relationships from MIPAS are on the resulting N₂O age product. Figure S1 (not introduced until Section 5/summary, and in passing) shows this, and could be highlighted more. Other figures could be relegated to supplemental material, and Fig S1 elevated to the main text (or, if clear, to the SM but only if there is clear text regarding it). A substantial discussion is also needed in comparison. This is an important part of the work that is not addressed or highlighted sufficiently, IMO.

We thank the reviewer for this comment, which closely aligns with the previous comments on L56-65. As noted in a previous comment, we have elevated Fig. S1 to the main text as Fig. 8, where we provide a discussion on the significance and impact of using MIPAS latitude-dependent tracer interrelationships to derive N₂O-age which addresses the reviewer’s concern of this in-passing mention in the Summary. For this section, refer to L432.

Section 4.2: This section is a crux of the paper but is not written with clarity nor a view towards what novel insights or applications this new study provides, or at least lays the groundwork to do. And in the discussion, I am confused as to how important the new work showing the latitude dependent SF₆-age:N₂O relationships is to the resulting N₂O age product, as well as what exactly the authors refer to [L494] as an “N₂O signature”...Presumably the N₂O biases, but this is not clear nor the implications for their work.

We thank the reviewer for this insightful comment and suggestion. In line with the reviewer's overarching suggestions to discuss age gradients and their relationship to circulation strength and mixing, and to clarify the significance of implementing the satellite-observed relationships to derive age, we have restructured Sect. 4.2 (MLS N₂O-age product) to two subsections:

4.2.1 – to discuss validation of the N₂O-age product with an independent age product via MIPAS

4.2.2 – to address why the use of latitude-dependent relationships is relevant and the implications on N₂O-age

These are changes that have also been discussed above, in response to the comment about L56-L65.

L494: We agree that the “N₂O signature” term is vague. The sentence previously read as:

“Any additional differences in these plots are due to N₂O signatures from MLS and MIPAS observations (i.e., MIPAS N₂O used for SF₆-age:N₂O relationships and MLS N₂O).”

It now reads as:

L423-424: “Any additional differences in these plots are due to N₂O measurement characteristics specific to each satellite (i.e., MIPAS N₂O used for SF₆-age:N₂O relationships and MLS N₂O).”

Section 4.3: This section is also critical and needs to be rewritten for clarity and focus. Also, L504-505: “These SF₆- and N₂O-age products confirm the previously inferred structure of the stratospheric circulation.” What’s the purpose of this statement? To be the devil’s advocate, don’t simple satellite observations of CFC-11 and CFC-12 also show this structure, more or less? Rather, what is important the authors’ work and new contributions, or why might they have expected a mess in all they did to normalize and smooth out age tracers and N₂O and yet wind up with such a nice product? And what have we learned?

We have removed this sentence on L504-505 and edited this section. We agree that other long-lived tracers also show this structure (e.g., CFCs and CH₄). We have made the distinction between ‘why’ ACE-FTS N₂O-age is relevant in this study by adding this paragraph.

L487-493: “Despite these discrepancies, the qualitative agreement between ACE-FTS N₂O-age (Fig. 9b) and MLS N₂O-age (Fig. 7a) demonstrates that MIPAS Age:N₂O relationships can be used to derive age of air from N₂O datasets across different instruments in the same time period. When we normalize N₂O to a common scale and apply the smoothed relationships to more sporadic retrievals of ACE-FTS N₂O, this results in smoother N₂O-age isopleths in Fig. 9b compared to SF₆-age in Fig. 9c, which demonstrates that the methodology filters variability. This result suggests that the methodology here can be used on more sporadic N₂O observations, thus supporting its use for future ACE-FTS N₂O retrievals and additional sporadic N₂O datasets.”

~ L516, SF₆ correction discussion: By the way, ACE-FTS observations (specifically N₂O:CH₄ correlations) have shown that a significant amount of N₂O can be produced in the mesosphere and thermosphere by high energy events (plasma chemistry), and this can be transported into the polar vortex (see papers by Bernath). This chemistry might change the SF₆:N₂O relationship in the mesospheric air making it into the stratospheric polar vortex which in turn might also affect how the

SF6 correction is made as a function of N2O. Have the authors (or anyone else) considered this? It's potentially relevant to their discussion here.

This is an excellent point and is appreciated. You are right! Using tracer correlations, Sheese et al. (2016) did show that N₂O produced in the lower thermosphere and mesosphere can descend as low as ~ 40 - 45 km in the polar vortex related to Sudden Stratospheric Warming events. However, this has varied from year-to-year (refer to their Fig. 3).

The Saunders et al. (2025) ACE-FTS SF6-age and gridded N₂O height range ($z = 8 - 38\text{km}$) is below the altitude of upper atmosphere N₂O influence.

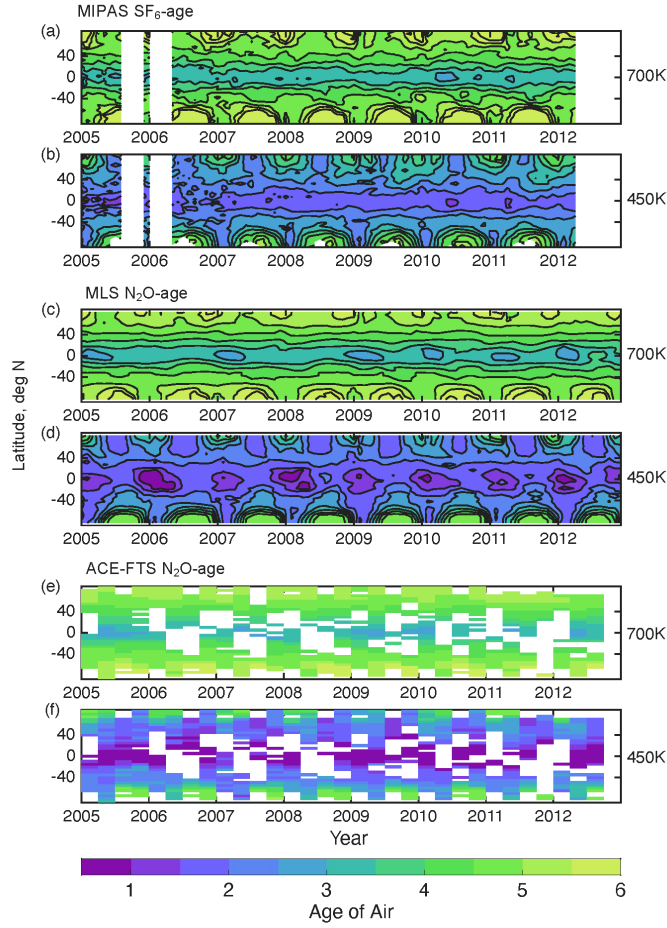
L522-535: Redundant, this has been said at least two times before in the text. More redundancy in L540-545.

You are correct. We have removed those lines and have addressed redundancies throughout the text.

Figure 10: It would be helpful to put additional text next to each satellite name (e.g., “MIPAS” “MIPAS SF6-age monthly mean values”.... Also, why not swap (a) with (b) and so on? Then the high altitude/higher theta data will appear ABOVE the lower altitude/lower theta data. That would be conceptually and aesthetically pleasing (and quicker to process for readers like me). Figure caption: L551 observations of what specifically (so I don't have to go look it up), also in alignment with my suggestion above...

We thank the reviewer for this comment. We have changed the caption for Figure 10. In addition, we have added satellite names and their respective age products (i.e., the distinction of SF6 vs. N2O age). You are right in that the ordering of altitudes used for these time series is not intuitive and confusing, and so we have rearranged the data such that the higher altitudes are the top and the lower altitudes are on the bottom.

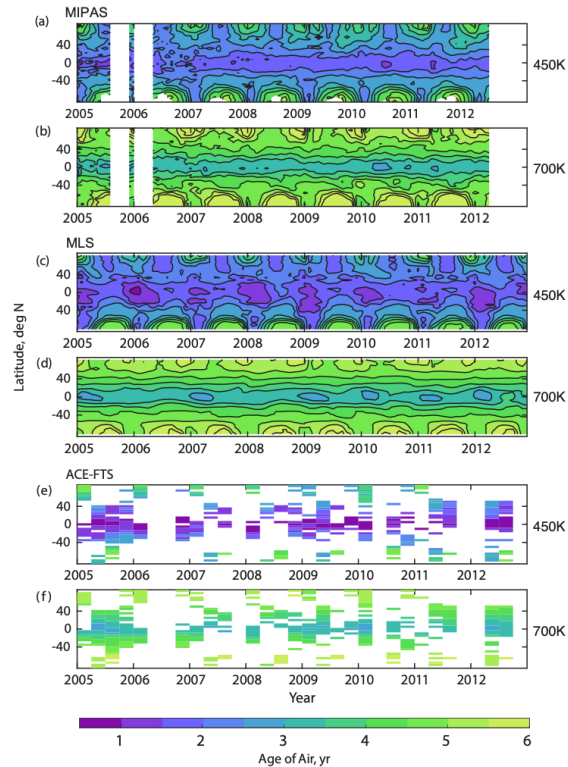
We have modified the Fig. 10 to reflect these changes as shown below:



The caption now reads as:

Figure 10: Time series of age as a function of latitude (deg N) and θ on constant θ surfaces from 2005 to 2012. (a) Monthly-mean MIPAS SF_6 -age on $\theta = 700$ K surface. (b) Monthly-mean MIPAS SF_6 -age on $\theta = 450$ K surface. (c) Monthly-mean MLS N_2O -age on $\theta = 700$ K surface. (d) Monthly-mean MLS N_2O -age on $\theta = 450$ K surface. (e) 3-month mean ACE-FTS N_2O -age on $\theta = 700$ K surface. (f) 3-month mean ACE-FTS N_2O -age on $\theta = 450$ K surface. SF_6 -age data of (a) and (b) are from MIPAS V5R_224 and V5R_225. N_2O data (c) and (d) are from MLS v5. N_2O data of (e) and (f) are from ACE-FTS v3.5/3.6 observations. Contours are in increments of half a year from 0.5 to 6 years.

The former figure for reference:



L 568-575: This text at this point in the section should be relating the gist of the study , but it is not expressing really what one gets out of all the work the authors did. As in the abstract, this needs to be clearer here. The chosen plots alone are not showing me this, readers need it spelled out explicitly. And doesn't this work do more than "add credibility" (L571) to the MLS N2O-age derivations? Or, if that actually IS the point, and this is laying the groundwork for future observations and even longer term records, that should be expressed explicitly/more clearly here. Thank you for this insightful comment. We have edited this section to emphasize that we have laid the groundwork for future work.

Lines 568-575 previously read as:

"The results shown in Fig. 10 illustrate the potential of applying latitude-varying Age:N2O to derive age from N2O observations. The structure of the MIPAS and ACE-derived age qualitatively agrees with the MLS-derived age. Despite biased MLS N2O observations, having stratospheric age products from three satellites, is valuable because the addition of the MLS N2O-age product fills the gaps that ACE-FTS cannot, while MIPAS SF6-age adds credibility to MLS N2O-age derivations in this study. The consistency in the spatial structure of both SF6 and N2O-age products further demonstrates the effectiveness of normalizing MLS N2O to address biases and shifting boundary conditions and using latitude-varying Age:N2O to calculate N2O-age, thus indicating the robustness of the Age:N2O product over the 2005-2012 period."

It now reads as (L532-536):

“The results shown in Fig. 10 highlight the potential of applying latitude-varying Age:N₂O to derive age from N₂O observations. The structure of the MIPAS and ACE-derived age qualitatively agrees with the MLS-derived age. Having stratospheric age products from three satellites is valuable because the addition of the MLS N₂O-age product fills the gaps that ACE-FTS cannot, and once the time-dependence of tracer interrelationships is determined and a corrected N₂O product has been created, this will enable a decades-long, spatially resolved age product from MLS. Both are the subject of ongoing work.”

Summary: L580: As noted above, S1 is only first referred to here, in the summary. (See my remarks above how the figure and its discussion in the text is likely important enough to at least be called out earlier in the results/discussion, even if the figure is not put in the main text.).

As noted above, Figure S1 has been moved to the main text and the figure order in the Supplement has been rearranged

Usually whole new ideas are not introduced in the summary but in the discussion section (even if they are separate from the results in a combined results/discussion section, as in this manuscript). The summary as written is long and rambling and would benefit from deciding what is crucial to include about the main points of this work and their usefulness and applications. And Equation (1) was discussed in the introduction but never returned to anywhere in the Results/Discussion nor the Summary. I recommend incorporating a discussion of that, as that would tie in the motivation raised in the introduction about how observations of the vertical and horizontal age gradients can be used to derive quantitative information on the strength of the BDC and horizontal mixing, as well as the structure of the BDC with respect to the lower branch, upper branch, and other altitude and latitude regimes and their seasonality. It doesn't make sense to introduce it then never return to it.

We appreciate these suggestions and we have implemented them throughout the text as previously described.

Figs S2, S4, and S5: The figures and their associated text are never called out in the main text. Why are they needed? Addressing this question might help focus the authors decide where the info actually needs to be called out and discussed – and in turn this may also add clarity to the text and figures in the main text as well.

We thank the reviewer for their concern over the Supplemental Material. We have since updated in order of the figures and their detail in the text. We have since moved S1 to the text for further discussion per previous comments. In addition, we have moved S6 to S1. We have reordered S4 to be S5 and call the figure in the updated text in Sect. 3.3.2. Also related, S2 is now S4, and we call to it in Sect. 3.3.2 for further discussion in S5 and the respective description. All excellent points! This makes the flow of the work much better.