Review of ACPD Manuscript 2026-1236, "Developing tracer interrelationships to derive stratospheric age of air…" by Castillo *et al.*

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 **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? 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' **[L23]**? The wording in general and the specific wording of 'previous counterparts' is often confusing (in the abstract and the throughout the manuscript). 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 Section 4 (Results and Discussion) and in Section 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.

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.

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.

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 Section 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.

Section 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.

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.

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...]

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 co-authors 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.

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)

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

Section 3.2.2 "SF6 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 SF6 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).

L317-325: Redundant (although Fig. 4 was never introduced).

L343-345: ???... Ray et al. data are not shown; MIPAS used here refers to…? "This comparison here shows why MIPAS was selected to compute mappings"…??? [And use of 'mappings' is ambiguous (to me) in multiple places in the manuscript.]

Section 3.3.2: Issues include The section is one sentence. The wording is ambiguous. The text refers reader to Supplemental Material S3, but that text is unintelligible and appears to be copied and pasted twice (?). Yet how and why the data are smoothed is essential for readers to understand.

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.)

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.

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…

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.

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.

Figure 6 (and Figure 7): Needs a, b, c, d labels. 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 20-30^{o} not 40-50^{o}. [See the text and Figure captions.] Further, the "extratropics" is generally considered to be ~35-90^{o} (except when there is also a polar vortex). Yet the authors use 'extratropics' to mean midlatitudes and only to 70^{o}. 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)!

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.

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 N2Oage 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.

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 SF6age:N2O relationships is to the resulting N2Oage 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.

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?

~ 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 votex 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.

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

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…

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.

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.).

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.

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.