

Dear Editor,

Thank you for giving us the opportunity to submit a revised version of our manuscript. We have addressed both Referees' comments thoroughly and our point-by-point responses to the Referees' comments have already been submitted on the dedicated EGU sphere platform. However, they will also be included in this document.

The aim of this study was to present the suitability of AirCore datasets to retrieve stratospheric lifetime estimates of COS and CH₄, given their tight relationship with N₂O, employing two well-established methods (Plumb and Ko, 1992; Volk et al., 1997). These methods were also applied to ACE-FTS averaged data. The retrieved lifetime estimates were then compared with each other and with results reported in previous studies. We found no significant differences between AirCore and ACE-FTS results. Moreover, no significant trends emerge from the lifetime estimates reported in this study and the results reported literature, dating back to the early 1990s.

Following the comments of the Referees, parts of the manuscript have been modified. The new version of the manuscript will be uploaded with the revisions marked as Microsoft Word mark-up text. In particular, the revision comprised:

1. **Revision of Structure and improvement of Figures.** Following agreeing comments made by both Referees, the number of tables and figures in the manuscript has been reduced significantly. Part of them have been moved to the Supplement, while occasionally multiple figures have been merged in single ones, to improve the readability of the paper.
2. **Clarification of crucial aspects of the Manuscript.** An important aspect highlighted by Referee #2 regarded the relevance of the dynamical context of the retrieved AirCore profiles. In fact, while averaged ACE-FTS data can well represent long-term transport, AirCore profiles are instead representative of specific air masses. Following the Referee's comment, we have now introduced an equivalent latitude analysis (Nash et al., 1996) within the discussion, to corroborate the comparison of the obtained results reported in this paper. The equivalent latitude was calculated from the ERA5 hourly potential vorticity data (Hersbach et al., 2023) for every flight, providing further insights regarding the origin of air samples collected with AirCore. In particular, this analysis showed that stratospheric air collected during the KRN and SOD campaigns is typically below polar latitudes, which may explain the larger difference (although still not significant) between AirCore and ACE-FTS lifetime estimates obtained for CH₄ with the Volk et al. (1997) method.

On top of this, some references have been introduced or adjusted and some sentences were rephrased for clarity, where stressed by the Referees. Our point-to-point response includes detailed clarifications.

With these, we believe that we have addressed the major concerns raised by the reviewers within the scope of this study, hoping that our manuscript could now be considered for publication in the Journal of Atmospheric Chemistry and Physics.

With kind regards, also on behalf of the coauthors,

Alessandro Zanchetta and Huilin Chen

Reply to Referee #1

The study by Alessandro Zanchetta and coauthors deduces stratospheric lifetimes from vertical profiles of carbonyl sulfide and methane from AirCore measurements and ACE-FTS satellite observations. The analysis is sound and rather comprehensive, comparing slightly variant methodologies of estimating lifetimes and pointing out caveats with respect to, e.g., regional sampling and observation altitude. The manuscript is generally well presented and well written and fits into the scope of ACP.

Authors' response (AR): *We thank the Referee for the positive comments and for the generous effort in providing this detailed feedback. We provide our point-by-point responses below.*

One major aspect that, in my opinion, should be improved is the length of the paper and the amount of information presented. Specifically, I wonder if all 13 figures and 11 tables are needed. In that context, please consider the following:

- Table 2 contains important information, but it is not essential for the main message of the paper and could be shifted to a supplement.
- Table 3 also contains information that should be provided by all means, but the paper would not suffer from this being included in a supplement rather than the main body of the paper.
- Figure 4 could also be shifted to a supplement. It is important to have this information, but how these profiles lead to the actual burden numbers is not visible in the figure, so it is not essential for the main paper.
- Figures 5 – 7 and 8 – 10 take up a lot of space. I wonder if it is not possible to combine these into two figures (one for COS and one for CH₄) without overloading them.
- It is good to show all results and important parameters for individual flights and analyses in Tables 4 – 6 for COS and 7 – 9 for CH₄. But the important pieces of information are given and discussed in the text and these long and detailed tables would be better off in a supplement. Also, the information displayed in Figures 11 and 12 is already given in Tables 5/6 and 8/9 for COS and CH₄ respectively.
- Does Figure 13 add new information compared to Table 10? In my opinion, the graphical representation of these data has limited added value, so showing just the table would be sufficient. But if you want to keep the figure, then the table with the more detailed information could be moved to a supplement.

AR: *We agree that many of these tables and figures are not essential additions to this manuscript. Consequently, the following modifications have been implemented:*

Table 2 is now Table S1 in the Supplement.

Table 3 is now Table S2 in the Supplement.

Tables 4-6 and 7-9 are now Tables S3-S5 and Tables S6-S8 in the Supplement, respectively.

Figure 4 is now Figure S19 in the Supplement.

Figures 5-7 have been incorporated in one multi-panel figure, now Figure 4 (a-c).

Figures 8-10 have also been incorporated in a single figure, now Figure 5 (a-c).

We thank the Referee for this suggestion and agree that Table 10 can be moved to the Supplement, where it can be found now as Table S9. We would nevertheless like to retain Figure 8 (previously Figure 13) in the main text, as it offers an intuitive visual summary of a core finding, while the table provides the detailed numerical support in the Supplement.

Besides the number of figures and tables, there is quite a bit of redundancy in the text as well. Essentially the same numbers for COS and CH₄ lifetime and stratospheric sink estimates from the two different methods (Plumb and Ko, 1992 vs. Volk et al., 1997) and for polar vs. mid latitudes are given in a similar way in Section 3 (lines 297 – 310 and 339 – 347), Section 4 (lines 380 – 382 + Section 4.1 and Section 4.2) and Section 5 (lines 506 – 516). While this information does fit into the flow in each individual Section, it may be worth avoiding some of this redundancy by cross referencing.

AR: We agree that cross referencing helps improve the readability of the manuscript. We now present the results in Sect. 3.2.2 for COS and Sect. 3.3.2 for CH₄. Throughout Sect. 4 (and its subsections) the lists of results have been replaced with references to their respective Results sections. We have kept the numerical values in Section 5 (Conclusion), to keep them readily accessible for readers and to keep a direct link with the Abstract and the Introduction.

Note that these suggestions are not meant to push the paper below a certain page or word limit because shorter is always better. But I had the impression that the take-home messages of the study are somewhat hidden underneath the wealth of information shown.

Besides the general recommendations to make the paper more concise, I have a few specific suggestions and technical corrections:

Introduction:

The introduction almost exclusively focuses on COS, while in the title and the rest of the paper, CH₄ is treated in parallel. I suggest adding one or two paragraphs on CH₄ in the introduction as well.

AR: We agree that the introduction lacked a discussion of CH₄. Accordingly, we have added the following paragraph to the introduction (Lines 48-59):

Methane (CH₄) is the second most important anthropogenic greenhouse gas in terms of radiative forcing, after carbon dioxide (CO₂) (Myhre et al., 2014). Its current tropospheric abundance is over 1900 ppb and has been constantly increasing in the past 20 years (Thoning et al., 2022). CH₄ is produced by both natural and anthropogenic sources. Among these, some of the major natural sources are wetlands, fresh waters and oceans, while the anthropogenic include livestock farming, agriculture, landfills and fossil fuels (Saunois et al., 2025). Its major sink is oxidation by OH• in the troposphere (Lelieveld et al., 1998; Saunois et al., 2025). With an average atmospheric lifetime of about 9 years, CH₄ can enter the stratosphere (Rohs et al., 2006; Saunois et al., 2025), which acts as a CH₄ sink. There, this gas reacts with stratospheric-abundant oxidizing species such as excited atomic oxygen, O(¹D), radicals such as OH•, or undergoes photolysis (Rohs et al., 2006). Moreover, stratospheric CH₄ chemistry affects ozone (O₃) concentrations and the oxidation of stratospheric CH₄ makes it the main sources of stratospheric water vapor (Han et al., 2025; Rohs et al., 2006). However, uncertainties in the magnitude of chemical loss of stratospheric CH₄ remain large due to variability in atmospheric transport and chemistry (Lelieveld et al., 1998; Portmann et al., 2012; Saunois et al., 2025).

Lines 39/40: an explicit reference for the COS tropospheric lifetime would be appropriate. And for COS being able to reach the stratosphere, a more original reference such as Crutzen (1976) would be more appropriate than the ones cited.

AR: We thank the referee for noticing the missing reference to the original study. The reference to Crutzen (1976) has been added.

Lines 43 – 45: Again, the chosen references appear a bit arbitrary. For example, Vernier et al. (2011) don't explicitly discuss the COS contribution but rather the effects of cumulative sulfate injection from volcanic eruptions. I suggest the Kremser et al. (2016) review paper, where the tropospheric budget and stratospheric sink of COS is discussed in detail and relevant literature is cited.

AR: We acknowledge the mismatch between the used reference and the presented information. We have replaced the reference to Vernier et al. (2011) with Kremser et al. (2016) in the revised version.

Methods:

Lines 100 – 105: It would be nice to show a comparison between the actual AirCore and ACE-FTS profiles. It's not essential to actually show this in the paper, so such a figure could go into the supplemental material.

AR: We agree that a comparison between AirCore and ACE-FTS could be an interesting addition. However, since this has already been presented and extensively discussed in Zanchetta et al. (2026 – see Figs. 9 and 10), we would prefer to refer to the existing literature and have therefore included the following sentence in the revised version:

The resulting 502 mid-latitude and 1681 polar latitude ACE-FTS profiles were averaged for each latitudinal interval. The resulting two averaged profiles (see Figs. 9 and 10 in Zanchetta et al., 2026) were then treated identically to the AirCore-derived profiles [...]

Line 114: I tend to agree that this is probably a fair assumption, but maybe you can add one sentence to explain why it is justified for the gases considered.

AR: the sentence has been modified as follows:

For simplicity, **given the smaller stratospheric abundances of the species when compared to the tropospheric portion of their profiles**, we assume that the ratio of the atmospheric mean mole fractions can be approximated by the ratio of representative tropospheric mean mole fractions.

Line 132 – 134: I find this explanation a bit confusing. Did you use the Prather et al. estimate of 116 years for the full period, or did you apply a 2.1 % per year correction, even though you state that the trend is not significant? Please clarify!

AR: the sentence has been modified as follows:

Moreover, the N₂O lifetime has been reported to depend on solar activity, which causes a variability of approximately 7 % (Prather et al., 2015). We therefore chose to infer N₂O lifetime in 2019, 2021 and 2023 from the 116 ± 9 years estimate of Prather et al. (2015), given the small -2.1 ± 1.2 % per decade decreasing trend (Prather et al., 2023) is not significant over such a short time span **and that any variability due to solar activity lies within the reported uncertainty.**

Lines 163 – 170: In my opinion, limiting the analysis to reasonably low N₂O mole fractions so that a tropospheric influence is minimized is important. This is a fundamental difference to the study by Karu et al., who analyzed only data with N₂O > 317 ppb due to the altitude limitation. This may explain why that study compares least well to your analysis and other COS lifetime estimates in the literature, and it is worth highlighting that here or in one of the later sections.

AR: We agree that this would be indeed a major cause of discrepancies between this study and the one presented in Karu et al. (2023). This has been addressed in the Discussion section (see further comments, later in this document).

Results:

Lines 287 – 288: Either here or in Section 2.3, please add the actual number of ACE-FTS profiles that were averaged.

AR: this information was already present in Sect. 2.3 (now Lines 127-128):

The resulting 502 mid-latitude and 1681 polar latitude ACE-FTS profiles were averaged for each latitudinal interval.

Line 329: For CH₄, why are the uncertainties for satellite data as large as for campaigns, different than for COS?

AR: We thank the referee for pointing out this difference between the two species. We speculate that one reason behind this difference may be found in the QCLS precision, which is relatively lower for COS than for CH₄. Therefore, when comparing the averaged ACE-FTS profiles, the effects of instrumental precision are probably more noticeable in the COS results than in the CH₄ ones. Moreover, we observe that the COS-N₂O local slopes obtained with ACE-FTS “flatten” already at N₂O > 200 ppb, while for most AirCore profiles this happens at higher N₂O levels. On the other hand, ACE-FTS and AirCore profiles exhibit more similar behaviors for CH₄, which may be reflected in the obtained uncertainties.

Discussion:

Lines 393 – 397: I think that the statement that Karu et al. (2023) “introduced” another point of uncertainty is misleading here. They did not introduce this but, due to the very limited altitude range of commercial aircraft, had to use data with a very limited altitude (and consequentially N₂O, see my comment above) range. In my opinion, this warrants more discussion and explains why their lifetime estimate differs from your and other studies.

AR: Yes, we have tried to make this difference clearer as follows:

A further discrepancy between the studies arises from the different altitudinal ranges covered by the observations. Our profiles cover altitudes roughly between 15 and 22 km, comparable with the highest altitudes observed by Krysztofiak et al. (2015) and Barkley et al. (2008). While Toon (1991) and Toon et al. (2018) measured COS vertical profiles up to 40 km, **Karu et al. (2023)’s observations were limited to between 10 and 12 km altitude. At such low altitudes, the mixing of lower stratospheric with tropospheric air is very likely to bias the tracer-tracer correlations and the consequent lifetime estimates.**

Lines 404 – 405 and Table 10: I suggest including stratospheric sink estimates from the important modeling studies by Brühl et al., (2012) of 35 Gg S/yr and Sheng et al. (2015) of 40.7 Gg S/yr. These papers do not explicitly state stratospheric lifetimes, but that also goes for some of the cited papers (e.g. Weisenstein, 1997).

AR: The suggested references have been added to the Table.

Lines 439 – 440: Please discuss this in more detail (cf. my previous comments on the comparison with the Karu et al. estimate)!

AR: We have highlighted the discrepancies with the study of Karu et al. (2023):

In the case of Krysztofiak et al. (2015) and Karu et al. (2023), the slopes of the tracer-tracer regressions were reported, and it was possible to recalculate the stratospheric lifetime and sink of COS using new estimates of the COS and N₂O tropospheric burdens. **It is**

important to notice that in the case of Karu et al. (2023), due to the limited altitudinal range of the observations, the tracer-tracer regression may have been biased by mixing with tropospheric air, which has probably affected the final lifetime estimates. The newly calculated lifetime results obtained with our method [...]

Technical corrections:

Line 206: it should be “advocate” instead of “advocates”

Line 233: Please replace the term “evident criticalities” (e.g. by “atypical behavior” of “distinctive behavior”)

Line 252: This reference is meant to be Figures 5 – 6 and 8 – 9, correct?

Line 290: Figures and tables should be referenced in order of appearance. So please refer to Table 5 prior to Table 6 or swap the numbering of these tables!

Lines 326 – 327: There seems to be a grammar issue. I suggest: “The application of the Plumb and Ko (1992) method leads to an average slope at mid latitudes of 0.2537 ± 0.0022 , while at polar latitudes it is 0.2381 ± 0.0063 .”

Line 492: It should be “applied in this study”.

AR: All the technical corrections have been implemented.

Reply to Referee #2

Review of: Estimates of carbonyl sulfide and methane stratospheric lifetimes based on AirCore profiles

The authors estimate stratospheric lifetimes of OCS and CH₄ using ten AirCore profiles and an age-of-air analysis following a methodology similar to that presented by Karu et al. for the IAGOS-CARIBIC aircraft dataset. The resulting OCS and CH₄ lifetime estimates are generally consistent, within their uncertainties, with previous assessments. I particularly appreciated the historical comparisons with other datasets presented in Figures 1 and 13. However, I encourage the authors to also consider the more comprehensive historical analysis of OCS observations presented in:

Gurganus, C., Bednarz, E. M., Rollins, A., Waxman, E., Ray, E., Hintsa, E., et al. (2026). Constraining the stratospheric sulfate budget in global models: Insights from in situ OCS measurements during 2023 SABRE and comparison with satellite, balloon and surface data. *Geophysical Research Letters*, **53**, e2025GL117405.
<https://doi.org/10.1029/2025GL117405>

The AirCore dataset analyzed here was comprehensively described in the accompanying Zanchetta AMT publication, so I believe the concise methodology section in the present manuscript is appropriate. Overall, the manuscript presents a useful application of existing age-of-air techniques to a high-quality AirCore dataset. However, I recommend a substantial reduction in the number of figures and tables, many of which add little to the scientific discussion and could be consolidated or moved to the Supplement. The principal conclusions do not substantially alter the current understanding of stratospheric lifetimes or age of air for either species. Nevertheless, the manuscript provides a useful discussion of the uncertainties associated with estimating age of air from different assumptions regarding tropospheric boundary conditions, particularly for species such as CH₄ and N₂O with long-term increasing trends.

Authors' response (AR): We thank the referee for the insightful comments and the positive feedback. *We provide our point-by-point responses below.*

We have now added and discussed Gurganus et al. (2026) throughout the manuscript. Most of the Figures and Tables presented in the original submission have been moved to the Supplement, following the suggestion of both Referees:

Table 2 is now Table S1 in the Supplement.

Table 3 is now Table S2 in the Supplement.

Tables 4-6 and 7-9 are now Tables S3-S5 and Tables S6-S8 in the Supplement, respectively.

Figure 4 is now Figure S19 in the Supplement.

Figures 5-7 have been incorporated in one multi-panel figure, now Figure 4 (a-c).

Figures 8-10 have also been incorporated in a single figure, now Figure 5 (a-c).

Major Comments

1. Comparison with ACE-FTS climatology

The manuscript compares a relatively small number of AirCore profiles with more than a decade of ACE-FTS observations. While the ACE-FTS climatology provides a reasonable representation of long-term, large-scale stratospheric transport, it is difficult to directly compare such a climatological average with individual AirCore profiles, which sample specific air masses. I encourage the authors to provide additional dynamical context for the AirCore observations. For example, trajectory analyses or plotting the measurements as a function of equivalent latitude would help determine whether differences arise from transport variability rather than methodological differences.

AR: We thank the referee for raising this important point. We agree that direct comparison between ACE-FTS climatology and individual AirCore profiles requires dynamical context. In the revised manuscript, we have therefore included equivalent-latitude coordinates (following Nash et al., 1996) for a more dynamically meaningful comparison. Contextually, we introduced Sect. 4.3 as follows:

4.3 Equivalent Latitude of AirCore observations

To further contextualize the AirCore results and their comparison with ACE-FTS averaged data, the equivalent latitude (Nash et al., 1996) was calculated from the ERA5 hourly potential vorticity data (Hersbach et al., 2023) for every flight. Equivalent latitude, in fact, represents the transport history of air parcels more accurately than geographic latitude, particularly in the upper troposphere/lower stratosphere region. The computed equivalent latitudes for each pressure level of the retrieved AirCore profiles are shown in Sect. S5 (Fig. S20-S28) in the Supplement. From these results, we computed the stratospheric partial columns of COS, CH₄, and N₂O for both AirCore profiles and their respective ACE-FTS profiles between the tropopause and 12 km above it, and investigated the difference between them. Figures 9-11 show the resulting differences against the average stratospheric equivalent latitude. TRN4 is not shown due to missing data that prevented the calculation of the stratospheric partial column of COS. Overall, it is evident that most of the sampled air had mid-latitude origins and that the partial columns obtained from TRN samples underestimate all tracers when compared to those obtained from ACE-FTS mid-latitude averaged profiles. The opposite is true for the AirCore samples collected in SOD. From the principle of AirCore sampling, we see no reason that biases could differ at the two different sites. Therefore, these opposing differences likely indicate that the AirCore profiles capture localized, latitudinally dependent air masses that are not fully represented by the zonally averaged ACE-FTS. Notably, the average equivalent latitude of the samples collected in KRN and SOD is well below polar latitudes, which may partially explain the larger discrepancies (though not significant) observed between the CH₄ lifetimes obtained with the Volk et al. (1997) method (see Fig. 7).

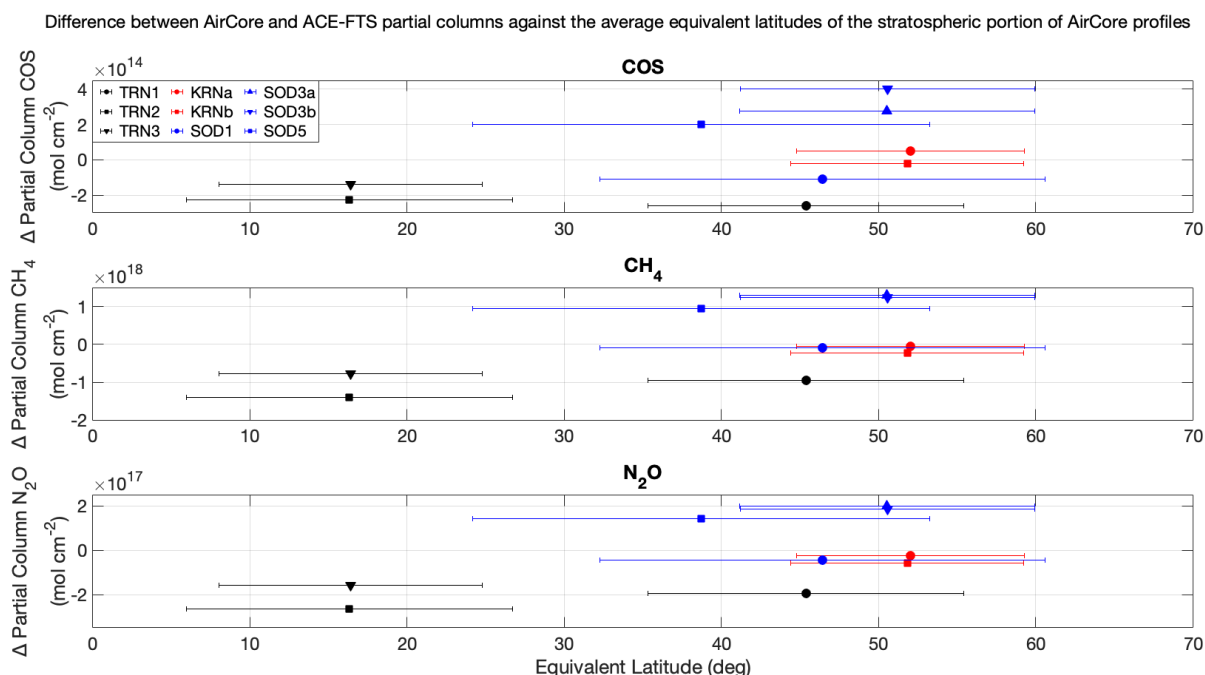


Figure 9: retrieved differences between stratospheric partial columns of AirCore and ACE-FTS profiles, against the average stratospheric equivalent latitude of AirCore profiles. The partial columns are reported in molecules cm⁻².

Moreover, Section S5 in the Supplement now reports the plots of all tracers against the retrieved equivalent latitude over the retrieved profiles.

2. Comparison between the Volk and Plumb & Ko methodologies

The manuscript devotes considerable discussion to differences between the Volk and Plumb approaches for calculating age of air. However, both methods ultimately use the same Equation (2); the primary distinction lies in how the tropospheric "burden" (i.e., the representative mean tropospheric concentration) is calculated.

Based on the values presented in Section 3.1, the difference in the OCS/N₂O burden ratio for 2023 is less than approximately 3% (487/336.8 using Plumb & Ko versus 457/324 using Volk). This difference is comparable to, or smaller than, the measurement uncertainty of the AirCore OCS observations (approximately 4% assuming a 20 ppt precision for QCLS). Consequently, the small differences in derived age of air between the two methodologies are expected to remain well within the observational uncertainty, consistent with the overlap shown in Figures 11 and 12. I therefore recommend shortening this discussion and emphasizing that, for the present dataset, the methodological differences are small relative to the measurement uncertainties.

AR: We thank the referee for this careful and quantitative assessment. We agree that the both methods ultimately rely on the same underlying mathematical framework and the differences in the derived burden ratio are indeed small. We also acknowledge that this difference is comparable to the measurement uncertainty of the AirCore OCS observations and that the methodological differences unlikely cause a significant impact on the derived lifetimes.

However, we would like to clarify that the major difference between the two methods not only lies in the calculation of the tropospheric burden, but also resides in the method employed to obtain the tracer-tracer slope. While Plumb and Ko (1992) infers this slope by a tracer-tracer regression (e.g., against N_2O) over altitude, Volk et al. (1997) refines this approach and extrapolates the tracer-tracer local slope at the tropopause, which is formally more correct, as the slope over altitude (and, therefore, time) may change due to different stratospheric transport and chemistry patterns.

Within the selected altitudinal range of our profiles, the differences between the local slopes obtained by the two methods are generally small. For COS, they average around 5%, but show peaks up to 12%. Similarly, for CH_4 the average difference is around 5.5%, with peaks of 14%.

To address the referee's suggestion, we have shortened and focused the discussion in Sect. 4.2. To clarify the referee's point, we added the following sentences (lines 477-481):

Overall, within the methods applied in this study, we believe that COS and CH_4 lifetime and sink calculations share the same sources of uncertainty. **The differences between burdens and local tracer-tracer slopes obtained with the two employed methods remained moderate within the selected altitudinal range. Since the two studies are based on a similar mathematical construction, where Volk et al. (1997) is more formally correct, these results suggest that Plumb and Ko (1992) provides an acceptable approximation for estimating the lifetime of COS and CH_4 within these altitudes.**

Specific Comments

- **Table 3:** This table primarily summarizes basic data analysis and does not appear to provide information that substantially advances the discussion. I recommend removing it or moving it to the Supplement.
- **Figure 4:** Presenting a "global average" ACE-FTS profile is potentially misleading because the ACE-FTS sampling is strongly biased toward middle and high latitudes, with relatively sparse tropical coverage. This limitation should be acknowledged more clearly, or the figure should be reconsidered.
- **Figures 5–10:** I do not see a strong scientific justification for including these figures in the main manuscript. They would be more appropriate in the Supplement, where readers can examine the individual profiles if desired.
- **Tables 4–6:** The OCS results would be much easier to interpret if they were consolidated into a single table, allowing readers to directly compare the results obtained using the Volk and Plumb & Ko methodologies.
- **Tables 7–9:** Similarly, the CH_4 results should be combined into a single summary table to facilitate direct comparison between the two age-of-air methods.

AR: We have implemented most of these suggestions (see above). The tables reporting the data for each methodology have been kept separated in the Supplement, mostly to avoid an overloaded single table.

Overall, I believe this is a worthwhile contribution that demonstrates the application of established age-of-air methods to a valuable AirCore dataset. The principal conclusions are generally consistent with previous work, but the manuscript would benefit substantially from a more concise presentation, consolidation of figures and tables, and a more focused discussion of the practical significance of the differences between the two age-of-air methodologies.

AR: We thank the referee for the insightful discussion and we modified the manuscript considerably following the suggestions.