

Interactive comment on “Stratospheric lifetimes of CFC-12, CCl₄, CH₄, CH₃Cl and N₂O from measurements made by the Atmospheric Chemistry Experiment-Fourier Transform Spectrometer (ACE-FTS)” by A. T. Brown et al.

Anonymous Referee #2

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General:

The paper presents a study on the lifetimes of several atmospheric tracers on basis of space-borne measurements of their atmospheric abundance by the ACE-FTS instrument. The methods for deriving the lifetimes are outlined, and the steps towards the derivation of the lifetimes are presented in much detail. Finally, the derived lifetimes are compared to values in the literature. I found the paper hard to read over some chapters (esp. chapters 3 and 4) since a lot of details are given which sometimes distract from the logical thread of the paper. On the other hand, some underlying assumptions like

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the fact that the measurements need to be free from altitude-dependent biases, have not been discussed at all. The error estimation refers solely to the standard error of the averaged ensembles ignoring any other errors relevant to the method applied. I recommend publication of the paper after these and the more specific comments outlined below have been considered.

Specific comments:

p4223, l2-3, 5-6: The regions where the Cl and F containing molecules are much wider, e.g. CFC-12 absorbs around 920 cm⁻¹ and many other molecules like CFC-142a/b etc. have absorption bands in the 1400 cm⁻¹ wavenumber region. The statements here should be corrected for this.

p4224, l22-27: Is a vertical resolution of about 2-3 km as provided by ACE-FTS (p4225, l17) really sufficient to resolve the tropopause? I think this issue needs further discussion.

p4227, l9 ff: The concept of correction for a non-linear tropospheric increase by calculating an effective linear growth rate has not become clear to me. Some more details on this methods should be provided.

p4227, l20: a Λ factor seems rather high to me, other publications refer to Λ of 0.7 (Waugh and Hall, 2002) or 0.8 (Garcia et al., 2011). How would the results of your study alter with other values for Λ ? A discussion on this question should be added.

p4229, l1: Does the scarcity of ACE-FTS data not allow to perform a seasonal analysis? What was the reason for restricting the analysis to periods of 6 months? I think a seasonal analysis would be much more appropriate to the problem, in particular if the paper aims at excluding any seasonality of the calculated lifetimes, as stated on p4235, l 22.

p4229, l8 ff: This section is a bit difficult to understand and the arguments should be re-ordered. Why not starting with the statement that the analysis has been limited

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to the mid-latitudinal well-mixed region and provide the arguments for excluding the tropics and the polar region afterwards? In this context the statement “unrelated to stratospheric lifetimes” (I12) seems a bit too radical to me. Further, the term “MAD filter” is already used at page 4230, I1, but introduced at p4230, I12 only. The description of this filter is a bit vague to my opinion.

p4230, section 4.1: As I understand the method, it fully relies on the assumption that the profiles of the species which go into the correlations don't have biases which change with altitude; since any altitude-dependent bias would affect the slope of the correlation and spoil the analysis. Has it been ensured previously to this study that this pre-condition is valid, i.e. have the data products been validated carefully and could any biases be excluded? These biases will affect the results of this study much more significantly than just leading “to a small underestimation in error on the results” (p2431, I19). I think a thorough discussion on this issue is necessary.

Technical and minor comments:

p4223, I23: Wouldn't be Solomon et al., 2010 the more appropriate reference?

p4225, I12: A more appropriate reference for the ATMOS missions which should at least be added is probably Gunson et al., 1996.

p4225, I22: Actually it's almost 10 years by now.

p4233, I20/21: Improve the wording of this sentence: “The lifetimes calculated for CH₃Cl and CH₄ show significant variation between the calculated lifetimes.”

Figures:

X-axis labels and descriptions for the figures A1 to A3 are missing. Axis descriptions, axis labels and figure titles are too small throughout the paper, but in particular for Figures A1 to A3.

Tables:

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Table A2 and A4: There are no entries NHW etc. in the tables the table caption refers to.

References:

Gunson, M.R., M.M. Abbas, M.C. Abrams, M. Allen, L.R. Brown, T.L. Brown, A.Y. Chang, A. Goldman, F.W. Irion, L.L. Lowes, E. Mahieu, G.L. Manney, H.A. Michelsen, M.J. Newchurch, C.P. Rinsland, R.J. Salawitch, G.P. Stiller, G.C. Toon, Y.L. Yung, and R. Zander, The Atmospheric Trace Molecule Spectroscopy (ATMOS) experiment: Deployment on the ATLAS Space Shuttle missions, *Geophys. Res. Lett.*, 23, 2333-2336, 1996.

Waugh, D. W. and Hall, T. M.: Age of stratospheric air: theory, observations, and models, *Rev. Geophys.*, 40, 1010, doi:10.1029/2000RG000101, 2002.

Garcia, R. R., Randel, W. J., and Kinnison, D. E.: On the determination of age of air trends from atmospheric trace species, *J. Atmos. Sci.*, 68, 139–154, 2011.

Solomon, S., K.H. Rosenlof, R.W. Portmann, J.S. Daniel, S.M. Davis, T.J. Sanford, G.-K. Plattner, Contribution of stratospheric Water Vapor to Decadal Changes in the Rate of Global Warming, *Science*, 327, 1219 (2010), doi: 10.1126/science.1182488.

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