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Interactive Comment

Interactive comment on "Stratospheric lifetimes of CFC-12, CCl_4, CH_4, CH_3Cl and N_2O from measurements made by the Atmospheric Chemistry Experiment-Fourier Transform Spectrometer (ACE-FTS)" *by* A. T. Brown et al.

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1. Page 6ff Please update the reference for the Laube et al. Paper: Laube, J. C., Keil, A., Bönisch, H., Engel, A., Röckmann, T., Volk, C. M., and Sturges, W. T.: Observation based assessment of stratospheric fractional release, lifetimes, and ozone depletion potentials of ten important source gases, Atmos. Chem. Phys., 13, 2779-2791, doi:10.5194/acp-13-2779-2013, 2013.

This reference has been changed

2. Page 6, Line 171 "This value has been scaled by the effective linear growth rate (0) of CFC-11 during this time (the values for which can be seen in table 2)." What does this means exactly? Looking at table 1 the gradient dCFC-11/dAOA (AoA: mean age of air) is monotonically increasing from 2005 to 2010 while the effective linear growth rate of CFC-11 does not show the same behaviour.

These values were calculated using the Laube et al. value as a base value. This value was then scaled by the effective linear growth rate relative to 2009 (the date for which Laube's work was calculated). The VMR of CFC-11 has decreased during this time. This has the effect of making the slope of the CFC-11 vs AoA less negative.

3. Page 6, Line 171 For the sake of traceability and because it is an important part of this kind of lifetime calculation, it would help, if you can specify precisely, how you calculate the effective linear growth rate. I assume that you use equation (A13) of Volk et al. (1997) – further on V97. If so, it will still be interesting over which time interval and how you fit the tropospheric time series (probably eq. (A12) in V97). Especially, this is true for tracers which could not well be characterised by a quadratic function.

The paper has been changed:

"The effective linear growth rate ($\gamma 0$ – equation 5) was calculated using monthly global means from the Advanced Global Atmospheric Gases Experiment (AGAGE) network (Prinn et al., 2001;Prinn et al., 2000) for CFC-11 (Cunnold et al., 2002), CFC-12 (Cunnold et al., 1997), CCl4 (Simmonds et al., 1998), CH4 (Cunnold et al., 2002;Rigby et al., 2008), CH3Cl (Simmonds et al., 2004;Cox et al., 2003) and N2O (Prinn et al., 1990), following the method laid out in Volk et al. (1997) using equations 3 and 4. For this study a quadratic function was fitted to 5 years of AGAGE data prior to each year between 2005 and 2010. For CH3Cl AGAGE data from between 2004 and 2010 was used. Strong seasonality was removed from the data using a sinusoidal term."

4. Page 6ff Why you are using only 2 seasons? If the seasonality has a significant impact on the lifetime estimation then it will be better to calculate gradients and

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subsequently lifetimes for all 4 seasons. It is a fact that temporal (and also spatial) coverage is the big advantage of the satellite remote sensors in comparison to high-resolution/precision but sparse in-situ measurements. Therefore you should fully resolve the seasonality - if possible with the given amount of ACE satellite occultations during each season. This would help to interpret the seasonal influence for specific localised in-situ measurements.

The paper has been changed to clarify these points:

“The decision to use 6 month periods of analysis was made to increase the sample size used in this work. If seasonality has an effect on the calculated lifetime, dividing the data into two periods would be sufficient to see this difference.”

5. Page 7, Line 203ff “In addition measurements made within the vortex were filtered out when the outliers of the data were removed.” How you filtered out vortex data? Is the criterion of 2.5 times the median absolute deviation sufficient enough to remove vortex data – especially in the northern hemisphere where the elongation of the vortex to lower latitudes is much more prominent than in the southern hemisphere?

Previous work on stratospheric chlorine and fluorine inventories (in preparation) has shown that 2.5 MAD is sufficient to remove vortex measurements between 70°N/S.

6. Page 7, Line 195-197 “Tropical correlations thus reflect local rather global sources and sinks and are thus unrelated to stratospheric lifetimes (Plumb, 1996).” This is only true for the tropical pipe model (Plumb, 1996), but not for the global diffuser model (Plumb,1992) and not for the more up to date and realistic tropical leaky pipe model (Neu and Plumb, 1999). However, it is not the scope of this paper to discuss the nature of tropical tracer-tracer correlations, but it is definitely not true or at least misleading to say that tropical correlations are unrelated to stratospheric lifetimes. The tropical tracertracer correlations are at minimum related to the photochemical decay in the tropical stratosphere and therefore these correlations are related to the overall stratospheric lifetime even though they do not reflect and cannot be used to calculate the overall

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stratospheric lifetime. My suggestion would be to change the sentence to: "Tropical correlations thus reflect more local rather than global sources and sinks (Neu and Plumb, 1999; Plumb, 2007) and are therefore not suited to derive global stratospheric lifetimes."

The paper has been changed to reflect this comment:

"Tropical correlations thus reflect local rather global sources and sinks, and are therefore not suited to derive global stratospheric lifetimes (Plumb, 1996; Neu and Plumb, 1999)."

7. Page 7-8, Line 224f and 244f You stated out that CFC-11 is on the y- and the other correlating species is on the x-axis. In all of your Figures (1,4-24) it is the other way round.

The paper has been amended to reflect this

8. Page 8, Line 228-234 It is not clear to me why the derived slopes are irregularly spaced – fewer points for all correlated species in the range of 140 to 200 ppt CFC-11 (see left side of Figure 1). Following your description how you calculate the slopes, the yielded data points should be evenly spaced when using an 80 ppt CFC-11 window and a regular step size of 5 ppt. If it is true that the density of points (each representing a tracer-tracer slope over the range of 80 ppt CFC-11) is distributed like it is shown in Figure 1 and all other appended correlations (Figures B1 to B21) then the the polynomial fit to this data is biased towards the higher (>200 ppt CFC-11) and the lower (<140 ppt) mixing ratios. Especially the latter is problematic because one must ask why this part of the correlation far away from the tropopause has a higher weight the data closer to the tropopause on the estimation (extrapolation) of the slope at the tropopause.

At both ends of the data the windows contain areas with no data in them. The extent is larger than the data (data less than 130 ppt removed as it is too far from the tropopause). In this case the middle of the data range is not the centre of the window.

Instead, the centre of the window is amended to reflect the centre of the data. The same affect occurs at the tropopause. This produces a series of windows which become smaller and smaller the closer they get to the tropopause and the "lower limit" of the data (100 ppt). The fit to this data does not include data below 120 ppt so as to avoid any bias in the fit towards these higher altitudes.

9. Page 10, Line 318-321 "Since neither the season or the hemisphere appears to have a significant effect on our results it is possible to calculate a total weighted mean." This finding is beside the calculated lifetimes itself the core of this paper. However, the conclusion is questionable. First, the error analysis has to be revised (see comments above), because realistic error estimation is the basis for such a conclusion. Second, it is not correct to draw the conclusion from the seasonal and/or hemispheric averaged lifetimes shown in Table 4. The universality of the tracer-tracer correlation must hold for each hemisphere and season separately. That means the non-averaged values shown in Table 3 must be the basis for the analysis. Inside the given error bars the specific correlations are definitely not universal. As an example, N2O has a lifetime of 66 (+12) years for SHS and 185 (-35) years for NHS in 2009. This is more than twice of the estimated lifetime in SHS and clearly outside the given error bars.

This line has been removed from the paper. The fact that these values are different makes the calculation of weighted means more important so that random errors are removed.

10. Page 24ff, Appendix A The references in the text referring to the Tables and Figures in the Appendix are often incorrect. Also the nomenclature and numbering of the Tables and Figures in the Appendix A is inconsistent. Further, I would suggest moving the Table A4 (Mean VMRs at the tropopause) and Table A6 (Mean atmospheric VMRs) to the main section and discussing the results – see remarks above.

Tables and figure references have been changed

11. Page 29 and 41, Figure 1 and Figure B9 There is missing northern hemisphere

winter 2009 correlations in the supplementary. Instead Figure 1 and supplementary Figure B9 both showing the same data - northern hemisphere winter 2008. The latter should show 2009 winter correlations.

As can be seen in the result tables there are no results for northern hemisphere winter 2009. This is due to a failure in the retrieval program for CFC-11 for this data bin.

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 4221, 2013.