

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.

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The following section has been added to the paper:

“An analysis of the altitude dependent systematic errors in ACE-FTS retrievals has not been carried out at this time. However, ACE-FTS occultations have been compared to data from other instruments such as the MK-IV and FIRS-2 balloon borne spectrometers (e.g. Mahieu et al. 2008). Previous validation papers for N₂O, CH₄, CFC-11 and CFC-12 have not shown significant altitude dependent errors for the altitude range

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used in this study (Mahieu et al. 2008; Velazco et al. 2011). In addition to these comparisons, the profiles of CFC-11 and CFC-12 were compared to those from the SLIMCAT 3-D Chemical Transform Model (Brown et al., 2011). The profiles used in this work showed that, whilst there were differences in the VMR from ACE-FTS and from SLIMCAT the overall shapes of the profiles were extremely similar. The differences between VMRs from ACE-FTS and other instruments (mentioned previously) can be used as a proxy for the systematic error, due to the fact that the full systematic errors associated with ACE-FTS retrievals are not known at this time. The methods described in this chapter have been repeated using ACE-FTS VMRs which have been modified by the differences calculated in previous validation work. The values used to modify the VMRs were + 10% for CFC-11 & CFC-12 from the validation work of Mahieu et al. (2008). Work by Velazco et al. (2011) also showed differences of + 10% for CH₄ and N₂O. The results of the reanalysis using these errors were combined with the statistical error. The final mean lifetimes were 113 +(-) 28 (21) for CFC-12, 123 +(-) 59 (39) for N₂O and 195 +(-) 88 (62) for CH₄.

The remaining species, CH₃Cl and CCl₄, are more problematic than the other species. Previous validations of these species have shown large differences between ACE-FTS retrievals and the retrievals from other species. For example comparisons between ACE-FTS and the MK-IV instrument (Velazco et al., 2011) found differences of 30% in the VMR retrievals of CH₃Cl. The errors on ACE-FTS retrievals of CCl₄ are estimated to be between 20 and 30% (Allen et al., 2009). The estimation of systematic errors for the CCl₄ retrieval is complicated by the position of the spectral feature used to retrieve CCl₄ VMR. There is an interfering Q-branch of CO₂, the line mixing of which is not properly accounted for in the forward model. Similarly the Q-branch of CH₃Cl suffers from line mixing which is not properly included in the forward model. The effects of line mixing on both of these retrievals are most serious in the troposphere, where the density of the atmosphere is at its greatest. In the stratosphere, where the density of the atmosphere is lower, line mixing becomes less of a problem within the retrieval. Quantifying the effects of line mixing on the retrieved VMR is a research project in and

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of itself. In this work we have approximated the systematic errors to be - 30% for CH₃Cl (Velazco et al., 2011) and + 20% for CCl₄ (Allen et al., 2009). Once more lifetimes were calculated using VMR which had been modified by the corresponding systematic error. The final mean lifetimes for these species were 35 +/- 13 (10) for CCl₄ and 69 +/- 71 (37) for CH₃Cl. These errors represent the best attempt to quantify the effect of systematic errors on the lifetimes of CCl₄ and CH₃Cl, however, due to the reasons outlined previously these errors may be different to those quoted here.”

An additional couple of sentences have also been added to the conclusion:

“The altitude dependent systematic errors in ACE-FTS retrievals are not currently known at this time. An attempt has been made to estimate the effect of systematic errors on the calculated lifetimes. Lifetimes were recalculated using VMRs which had been modified to reflect differences between ACE-FTS retrieved VMRs and those from other instruments. The mean lifetimes were unaffected by these calculations, which affected the error ranges of these calculations. The results of these calculations were as follows: 113 +/- 28 (21) for CFC-12, 123 +/- 59 (39) for N₂O, 195 +/- 88 (62) for CH₄, 35 +/- 13 (10) for CCl₄ and 69 +/- 71 (37) for CH₃Cl.”

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