RE-EVALUATION OF THE LIFETIMES OF THE MAJOR CFCS AND CH_3CCL_3 USING ATMOSPHERIC TRENDS

TIME VARIATION OF DERIVED LIFETIMES

M. RIGBY ET AL.

Figure 1 shows the sensitivity of the inversion to changes in emissions (see section 3.6 for methodology). Overall, the lifetime change for a given percentage emissions change decreases as emissions decline. Since errors due to unaccounted-for emissions can be seen to plateau soon after peak emissions for each gas, we chose to make our lifetime recommendations in the main text using average lifetimes since peak burden. Averages were therefore taken from 1994, 2002, 1997 and 1992 until 2011 for CFC-11, -12, -113 and CH_3CCl_3, respectively.

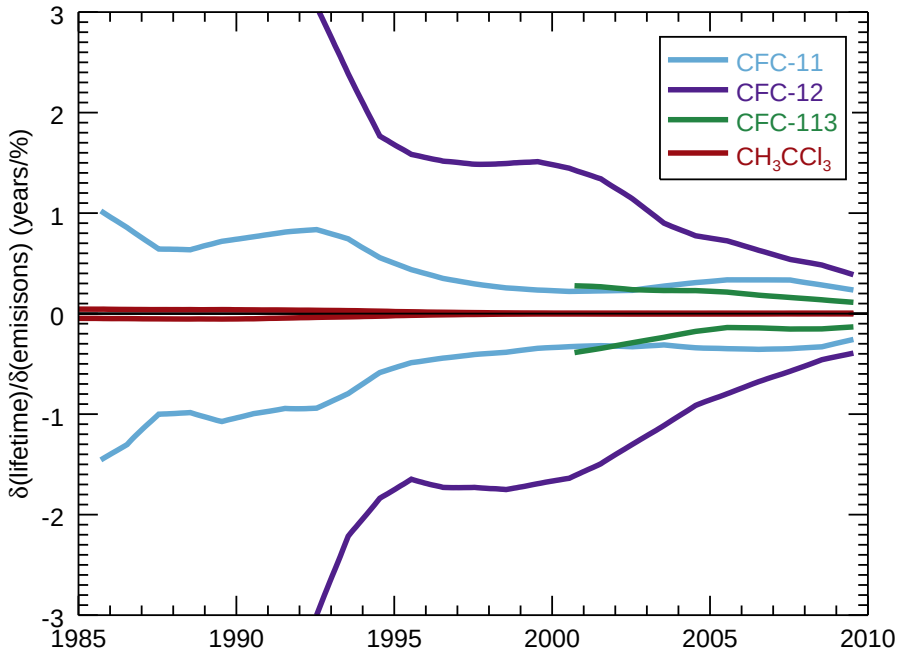

FIGURE 1. The sensitivity of the derived lifetime to changes in emissions. The figure shows the 1-sigma range of derived lifetimes from an ensemble of inversions in which emissions were perturbed by ±1% for each species, as described in Section 3.6 of the main text.

Running-mean transient lifetimes derived using AGAGE and NOAA observations are shown in Figure 2. Two sets of 1-sigma uncertainty bounds are shown in the figure, one showing the uncertainties derived in the inversion, which includes the influence of measurement and model uncertainties, and a second showing the influence of emissions uncertainties. Lifetimes derived in the first five years of each investigation were ignored, to remove the influence of spin-up errors and correlations of initial conditions with stratospheric lifetimes. The lifetimes derived using the two networks are not statistically different from one another for all gases at all times.

The maximum likelihood lifetime of CFC-11 can be seen to remain relatively stable throughout the inversion with a mean value of close to 50 years over the entire time series. The average lifetime from peak burden onwards is not statistically different from that which would be derived during any shorter averaging period since peak burden.

The estimated lifetime of CFC-12 shows relatively large, but not statistically significant, changes over timescales on the order of decades. The magnitude of the change in derived CFC-12 lifetime is unlikely to be physically realistic (e.g. the ∼50 year decrease during the 1990s). Since the same changes are seen for both networks, they are likely caused by errors in the emissions estimates that our lifetime derivation relies upon. Our emissions uncertainty estimates appear to account for these changes. Similarly to CFC-11, our recommended lifetime is not statistically different to the running mean lifetime for any period since peak burden.

For CFC-113, no significant deviation from our recommended estimates are seen during the period investigated, although relatively large lifetime "changes" are derived. Our uncertainty analysis shows that this is likely due to uncertainties in the inversion since none of these variations are statistically significant when the uncertainty is taken into account.

We find a similar timing to the inter-annual fluctuations and trends in CH_3CCl_3 lifetimes to those found by Prinn (2005) from 1980 to 2003 also using AGAGE data. These changes will be due to changes in OH concentration and/or measurement and emissions errors. The standard deviation of the annual mean, relative to the overall mean lifetime is around 4.2% in our estimates from 1980 - 2003, compared to 5.3% in Prinn (2005). The Prinn (2005) estimates used a similar model to that used here and the same observations. However, the reason for the slightly smaller variability is likely to be due to the smaller number of degrees of freedom used in the Prinn (2005) estimates, compared to ours (one OH concentration was solved for per year, compared to 4 latitudinal bands per season here). Since more parameters were solved for, our inversion should suffer from smaller 'aggregation error' than the Prinn (2005) inversion. Similarly to Prinn et al. (2001); Prinn (2005), no statistically significant trend in CH_3CCl_3 lifetime could be inferred in this work. From 1992 to 2010, we derive inter-annual fluctuations of 3% and 2.5% when AGAGE or NOAA data are used, respectively. This relatively small inter-annual variability agrees with the findings of Montzka et al. (2011). The derived mass-weighted global average OH concentration is shown in Figure 3 and is tabulated in a .csv file in this supplement.

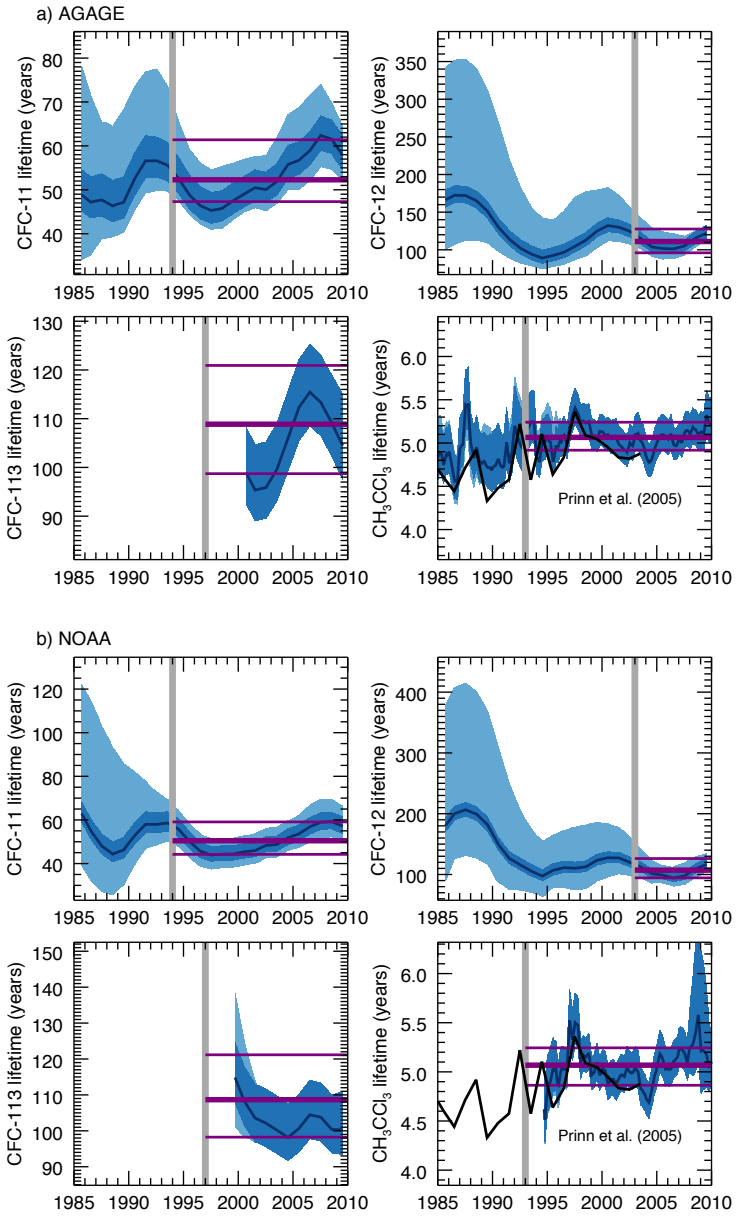

FIGURE 2. Running-mean transient lifetimes as derived in inversions using a) AGAGE and b) NOAA observations (CFC-11, -12, and -113 are shown as 5-year running means and CH_3CCl_3 is shown as a 1-year running mean). 1-sigma uncertainties that include the influence of measurement and modelling errors, but not emissions errors are shown in the boldly shaded areas. The more faintly shaded areas show the 1-sigma range of derived lifetimes from the ensemble of inversions run with perturbed emissions. The grey line indicates the time of maximum atmospheric burden of each gas. Purple lines show the maximum likelihood and 1-sigma ranges for the derived average transient lifetimes since peak burden. The black line in the CH_3CCl_3 plots shows the CH_3CCl_3 lifetime derived by Prinn (2005).

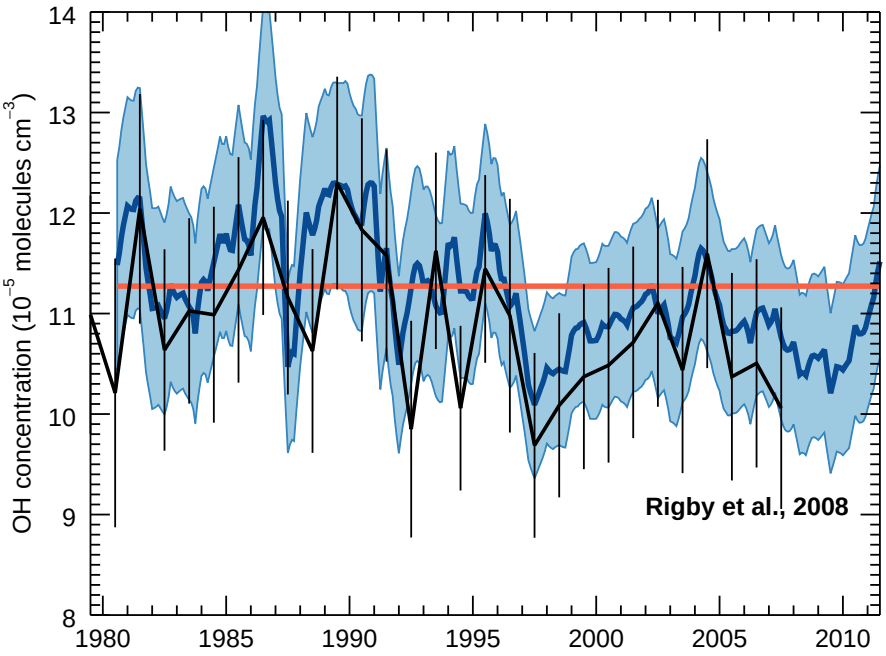

FIGURE 3. Mass-weighted global 12-month running mean OH concentration derived from the AGAGE inversion. Shaded areas show the 1-sigma uncertainty range. The red line shows the 1-year running global mean *a priori* OH concentration. The black line shows OH concentrations from Rigby et al. (2008) who presented updated estimates using the Prinn (2005) methodology.

REFERENCES

S. A. Montzka, M. Krol, E. Dlugokencky, B. Hall, P. Jockel, and J. Lelieveld. Small Interannual Variability of Global Atmospheric Hydroxyl. *Science*, 331(6013):67–69, January 2011.

R. G. Prinn. Evidence for variability of atmospheric hydroxyl radicals over the past quarter century. *Geophysical Research Letters*, 32(7):L07809, 2005.

R. G. Prinn, J. Huang, R. F. Weiss, D. M. Cunnold, P. J. Fraser, P. G. Simmonds, A. McCulloch, C. Harth, P. Salameh, S. O'Doherty, R. H. Wang, L. Porter, and B. R. Miller. Evidence for substantial variations of atmospheric hydroxyl radicals in the past two decades. *Science*, 292: 1882–8, 2001.

M. Rigby, R. G. Prinn, P. J. Fraser, P. G. Simmonds, R. L. Langenfelds, J. Huang, D. M. Cunnold, L. P. Steele, P. B. Krummel, R. F. Weiss, S. O'Doherty, P. K. Salameh, H. J. Wang, C. M. Harth, J. Mühle, and L. W. Porter. Renewed growth of atmospheric methane. *Geophysical Research Letters*, 35:L22805, 2008.