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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Re-evaluation of the lifetimes of the major CFCs and CH_3CCl_3 using atmospheric trends" *by* M. Rigby et al.

M. Rigby et al.

matt.rigby@bristol.ac.uk

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We thank the reviewer for their comments. Our responses are given in italics below. Please note that we have made some minor changes to the methodology in the revised manuscript. Principally:

- *We now solve for inter-annually varying seasonal transport parameters, removing the constraint that transport was inter-annually repeating*
- *We no longer solve for the logarithm of stratospheric loss rates and OH concentrations, to maximise the linearity of the inversion*
- *We now focus on steady-state lifetimes, rather than mean transient lifetimes (al-*

though the difference between the two is very small)

- *We discovered an error in our "emissions uncertainty" quantification, which resulted in the PDF being skewed slightly towards high lifetimes. This was because some low-lifetime (high-emissions) ensemble members were erroneously being rejected in the estimation scheme. This rejection was caused by numerical errors in the model causing the inversion to diverge for some ensemble members. The divergence is more likely for the low-lifetimes cases. To avoid this problem we now estimate the sensitivity of the inversion to small emissions perturbations using the same methodology (but using a 1% emissions perturbation), and then scaling this sensitivity up to the chosen emissions uncertainty (20% for the CFCs in the revised manuscript).*
- *We now present the discussion of the variability of the derived lifetimes in the Supplement, as we feel this section was distracting and not the central aim of the paper (indeed none of the 'variations' in lifetime were statistically significant, with all of the variability being potentially explainable by erroneous emissions). The paper now focusses on the derived time-averaged instantaneous lifetimes, which was our main aim.*
- *We have included a short section in the Supplement, in which we estimate the lifetimes of HFCs and HCFCs using our CH3CCl3-derived OH fields.*

1 Major remarks

This paper re-evaluates the lifetimes of CFC-11, CFC-12, CFC-113 and CH3CCl3 using observations of the AGAGE network and the NOAA network. To this end, a 12-box model is used in which the stratospheric lifetimes (CFC-11, CFC-12, CFC-113) and

tropospheric lifetimes (CH3CCl3) are optimized, together with inter-box diffusion parameters and initial conditions. Technically, the system seems to work and realistic lifetimes are obtained, not too much out of tune with earlier estimates. The main results are presented in figure 4. I find these results very confusing since large variations in the lifetimes are derived as a function of time.

We do not agree that the main results are presented in Figure 4. In fact, as we make clear in the text, and as shown by the uncertainties presented in Figure 4, none of the variations are statistically significant, and are likely to be driven by uncertainties in emissions. The Figure was intended to show the rationale for choosing the post-burden average as our recommended lifetime, as well as demonstrating how uncertainties due to emissions change with time. We feel that the main results of this paper are given in Table 2, and indeed it is these results which are highlighted in the abstract and conclusions, rather than any inter-annual variability. We have now expanded the discussion of the time-variability of our solution and placed it in the Supplement, so as not to distract the reader from the central aim of the paper.

For CFC-113, for instance, derived lifetimes vary between 160 and 80 years. Moreover, the derived variations are outside the envelope that is associated with the emission uncertainty. The authors report the lifetimes (and uncertainty) for the year at which the peak burden was observed. According to me, this is not the most obvious choice. Why not report the lifetimes for more recent periods, in which the influence of (uncertain) emissions is less?

As is stated in multiple places (e.g. P24485 L10, P24485 L16, P24486 L19, P24486 L25) the average lifetimes SINCE peak burden are taken, precisely for the reason stated by the reviewer. We have modified some of these lines slightly to make this a little clearer.

What is even more important is that for most of the studied compounds large variations in the atmospheric lifetimes are not expected. Lifetimes of CFC-11, CFC-12, and CFC-113 are mostly determined by the slow transport to the stratosphere, which probably does not change over time (the model optimizes yearly recurring diffusion parameters, i.e. not inter annual variations and trend, so inter-annual changes in transport are driven by changes in gradients).

We agree that the inter-annual changes in strat-trop exchange and therefore overall lifetime are likely to be small. To clarify, the changes that we derive in the lifetime, which reflect "corrections" that must be applied to the trend to make it agree with observations, are likely to be too large. This is assumption is quantitatively explored with the ensemble of inversions that were performed with perturbed emissions. To reiterate, the derived changes are not statistically significant. Our derived uncertainties are consistent with the reviewer's assessment that large changes in lifetime are not expected. To investigate the influence of inter-annual transport variations, we now allow model diffusion parameters to change inter-annually. However, this modification not substantially change our global lifetimes.

For CH3CCl3 the situation is different, since tropospheric lifetimes may change if tropospheric OH shows a trend. From figure 4 a slight positive trend in the CH3CCl3 lifetime seems to be derived. As the authors correctly state, these estimates are sensitive to the assumed emissions, but avoid much further discussion about this issue.

The reason why this slight trend was not discussed is because it is not statistically significant. Prinn et al., 2005 obtain similar results and discuss the difficulty in deriving a trend in their paper. We now present a slightly longer discussion in the Supplement.

The focus of the paper is on re-evaluation of the lifetimes, also with the aim to make future projections. Given the arguments above, it would make much more sense to optimize stratospheric and tropospheric loss-rates that are not allowed to vary inter-

annually (just as the transport parameters).

We disagree with this assertion. It is well recognized in inverse modeling studies (e.g. Thompson et al., 2011) that optimizing time-invariant parameters as the reviewer suggests leads to aggregation errors (i.e. the model is not allowed the flexibility of adjusting to potentially-real changes in the target parameter). By taking the average over the post-burden lifetimes (we remind the reviewer that this was done, rather than taking a single year as they assumed), we avoid these aggregation errors. However, we again reiterate that we do not believe that the variations that we see in CFC lifetimes are real, as is shown by the derived uncertainties.

One could argue that a poor fit with observations will be obtained since the system is given less freedom to adjust the misfits. This can be resolved by also optimizing emissions (within a pre-described uncertainty range).

This is a tempting idea, not least because emissions uncertainty could be directly included in the inversion, and indeed we have performed such inversions. As might be expected, we found that a "compromise solution" was obtained in which both emissions and lifetimes were simultaneously adjusted to match the trend. However, we ultimately decided against this approach for two reasons: 1) the derived emissions and lifetimes were strongly dependent on the assumed measurement and emissions uncertainties (these uncertainties are not well known for the emissions), 2) the derived lifetime uncertainties tended to be relatively small. For example, using the emissions uncertainties described in the text, we obtained a lifetime for CFC-11 of 58$\pm$1.5 years, whereas lifetimes outside this range can readily be obtained by scaling the emissions dataset by only a few percent. The reason for uncertainty under-estimation is, we feel, a limitation of the Bayesian approach as applied to this problem, which assumes that all uncertainties are random and not systematically biassed. As we clearly demonstrate, using our emissions-uncertainty analysis, biases in the emissions can have a very large influence on the derived lifetimes (of the order of tens to hundreds of years

during some periods for CFC-12). We feel that the setup that we use in the paper gives a fairer representation of the (significant) uncertainty in trend-based approaches to this problem.

In the current set-up the errors in emissions is translated into (unrealistic) life-time variations.

We agree and would like to re-iterate here that our emissions-uncertainty analysis quantitatively reflects the point the reviewer is trying to make. None of the variations are statistically significant.

Also, different sets of emissions can be used as prior to investigate the effect of the emissions on the final estimates (do posterior emissions and lifetimes converge to the same values?).

An alternative inversion with different prior emissions has been performed in the paper (see discussion of inversion using UNEP/TEAP emissions).

I think a lot of confusion about figure 4 can be avoided with this approach and that the main question of the paper can be addressed more adequately.

We agree that some readers may find Figure 4 confusing. In fact there was much debate about whether it should be included at all (since, as we note above, the changes are not statistically significant). However, we decided that it was instructive to see how the uncertainty changed with time and we wanted to be open about how the derived lifetimes changed with time. However, to avoid such confusion, we have decided to expand the discussion of the time-variation of the derived lifetimes and move this discussion to the Supplement in the interest of brevity.

Another modification in strategy that would strengthen the manuscript would be the

tuning of the transport by other tracers, like SF6 and 85Kr. Although the multi-tracer constraint on the transport parameters is indeed elegant, some aliasing between transport and lifetimes can still be expected.

We agree that this is also an appealing approach, and it was indeed investigated. However, we rejected the idea of using AGAGE SF6 measurements to determine transport parameters for the simple reason that AGAGE SF6 measurements are only available at all AGAGE stations from 2004 onwards, and Rigby et al., 2010 find, using a 3D model, that global SF6 emissions are likely to be too low during this period (which would therefore lead to a 'bias' in the derived transport parameters). There are no AGAGE measurements of 85Kr. We also note here that even in the "worst case scenario" that the derived transport parameters were nonsensical (which they do not appear to be), the derived overall lifetimes could still be realistic because the model trend must still agree with the observations, given the emissions.

I read between the lines that the derived seasonal variations in OH and K-diffusion parameters may be large (sometimes negative values?) and that therefore logarithmic values are optimized instead.

A stable solution can be found without using logarithmic parameters, but instead by setting the 'descent' parameter (μ) to a small value. The solution obtained using this approach does not differ significantly from the solution obtained using logarithms (whatever the formulation, the model trend must still agree with the observations, given the emissions). However, this increased the time taken to find a solution. To address the reviewer's concerns, and to increase the linearity of the problem, we have removed the logarithmic form on the stratospheric loss rates and OH concentrations.

This seems to work technically, but according to me this is a clear signal that the set-up contains several weaknesses that have to be resolved first. The basic idea of the paper is good, however, and I hope that my suggestions help to sharpen the manuscript.

As we hope we have demonstrated in our responses above, we have previously investigated many of the suggestions made by the reviewer, and have arrived at the chosen methodology after careful experimentation with a large number of approaches. There are clearly limitations to trend-based lifetimes estimates, which we feel are primarily due to uncertainties in emissions estimates, rather than the specific details of the inverse scheme or the transport model (reasonable lifetimes estimates can be derived using a 1-box model). We feel that the approach used in this paper allows us to rigorously investigate the influence of emissions uncertainties on our derived lifetimes (in addition to errors in measurements and transport model parameterisations).

2 Minor remarks

Supplement: I noticed that the advection parameters are not mass conserving. Although the transport is dominated by diffusion, I think it is important to have mass conservation.

It is important to have mass conservation and indeed our 12-box model does conserve mass. The advection equations in the model take account of the mass of air passing across each box interface and form a closed system when the values given in the table are used. Please see Cunnold et al., 1983 for a discussion of this.

page 5, maybe give units, such as kg/year, etc

We do not feel that units are required in this generic discussion.

page 12, line 11. What is the reduction in the gradient?

We have added the line "Once a minimum in the cost function is reached (which we defined as the point where the change in the value of J dropped below 0.1% of its initial value)...".

page 14, line 24. This is a strange procedure. The vertical gradient is very much determined by stratospheric loss for most compounds. Why should this gradient be a good measure for the grid-box uncertainty in the initial concentration?

We agree that this measure of initial condition uncertainty is relatively crude. However, we find that our conclusions are not significantly affected by the choice of this uncertainty. Furthermore, we now reject all lifetimes estimates for the first 5 years of every simulation to remove any influence of initial conditions on our derived lifetimes.

page 16, line 21. I had the impression that the stratospheric lifetime of CH3CCl3 was fixed

This is correct. We have removed the reference to CH3CCl3 here.

page 16, line 27. The promised material could not be found in the Supplement

This was accidentally omitted from the auxiliary material. This line has been changed to "Optimized model transport parameters are also shown in the Supplement.", and the updated transport parameters are now given.

3 Additional references

Rigby, M., Mühle, J., Miller, B. R., Prinn, R. G., Krummel, P. B., Steele, L. P., Fraser, P. J., et al. (2010). History of atmospheric SF6 from 1973 to 2008. Atmospheric Chemistry and Physics, 10(21), 10305–10320. doi:10.5194/acp-10-10305-2010

Thompson, R. L., Bousquet, P., Chevallier, F., Rayner, P. J., Ciais, P. (2011). Impact of the atmospheric sink and vertical mixing on nitrous oxide fluxes estimated using inversion methods. Journal of Geophysical Research, 116(D17), D17307. doi:10.1029/2011JD015815