Atmos. Chem. Phys. Discuss., 12, C9344–C9346, 2012
www.atmos-chem-phys-discuss.net/12/C9344/2012/

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.

Anonymous Referee #2

Received and published: 14 November 2012

This paper re-evaluates the lifetimes of CFC-11, CFC-12, CFC-113 and CH_3CCl_3 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 (CH_3CCl_3) 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.

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?

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). For CH_3CCl_3 the situation is different, since tropospheric lifetimes may change if tropospheric OH shows a trend. From figure 4 a slight positive trend in the CH_3CCl_3 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 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). 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). In the current set-up the errors in emissions is translated into (unrealistic) lifetime variations. 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?). 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.

Another modification in strategy that would strengthen the manuscript would be the tuning of the transport by other tracers, like SF_6 and ^{85}Kr. Although the multi-tracer constraint on the transport parameters is indeed elegant, some aliasing between transport

and lifetimes can still be expected. 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. 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.

1 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.
- page 5, maybe give units, such as kg/year, etc
- page 12, line 11. What is the reduction in the gradient?
- 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?
- page 16, line 21. I had the impression that the stratospheric lifetime of CH_3CCl_3 was fixed
- page 16, line 27. The promised material could not be found in the Supplement

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Interactive comment on Atmos. Chem. Phys. Discuss., 12, 24469, 2012.