Atmos. Chem. Phys. Discuss., 12, C12110–C12115, 2013
www.atmos-chem-phys-discuss.net/12/C12110/2013/

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

Received and published: 29 January 2013

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, which estimates the lifetimes of HFCs and HCFCs using our CH3CCl3-derived OH fields.*

1 General comments

The methods used are sound and well described and overall the manuscript is well written, therefore, only a few points need clarification:

p24480: The authors state that 5-10 iterations were used, however, what criteria used to know that the cost function was at minimum? In other words, how was the number of iterations necessary determined?

We terminated the scheme when the change in J from one iteration to the next should was less than 0.1% of the initial value. The following line has been added to the manuscript: "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), ...".

p24481: The authors mention that eddy diffusion transport parameters were included in the optimization but not advective parameters. Therefore, could the authors please clarify if any of the parameter(s) for transport between the lowermost stratospheric box and the uppermost tropospheric box were optimized? The stratosphere to troposphere exchange rate would be an important parameter for the rate of loss of CFC species, which are predominantly lost in the stratosphere.

As described in the Supplementary material, no advection was assumed across the tropopause. Exchange of air between the troposphere and stratosphere is assumed to occur entirely by eddy diffusion. In the ACPD paper, the four eddy diffusion timescales between the upper tropospheric and stratospheric boxes were optimized in the inversion for every month of the year, using the Cunnold et al., 1994 estimate of 3.5 years as a priori constraints. Please note that we have now changed the inversion scheme so that eddy diffusion parameters are now optimised in every season for the entire period, allowing the possibility of inter-annual transport variations.

p24481: Due to numerical considerations, the log of the inverse lifetime was optimized. Could the authors please clarify, was this transform also applied to the observations, which are the log of the mixing ratios?

As noted in Section 3.4, the log of the mixing ratios were solved for, to maximize the

linearity of the problem. No further transforms were applied to the observations.

p24481: Regarding the transform to optimize the log of the state variables ([OH], inverse lifetime and eddy diffusion coefficients) was the sensitivity matrix H recalculated for this transformation?

The sensitivity matrix was calculated as the sensitivity of the observations (log(mole fraction)) to changes in each parameter, accounting for the transformation applied to the parameter. For example, the elements of H corresponding to the sensitivity to log([OH]) were calculated as H=d(log(mole fraction))/d(log([OH])). However, please note that in the revised manuscript, to maximise the linearity of the problem, we have dropped the use of the logarithm of the inverse stratospheric lifetimes and OH concentration. The correct transforms are still used in each case in determining the sensitivity matrix.

p24481: Again regarding the transformation to optimize the log of certain state variables, how were the uncertainty covariance matrices R, P, redefined?

Since y was in units of log(mole fraction), the uncertainty $\mathbf{R}$ can be shown to be in units of squared fractional mole fraction error.If y is some differentiable function of mole fraction (χ), the uncertainty in y is approximated by (for small uncertainties):

$$\sigma_y^2 \approx \left(\frac{\partial y}{\partial \chi}\sigma_\chi\right)^2 \tag{1}$$

So in the case that $y = ln(\chi)$, we obtain:

$$\frac{\partial y}{\partial \chi} = \frac{1}{\chi} \tag{2}$$

so:

$$\sigma_y^2 \approx \left(\frac{\sigma_\chi}{\chi}\right)^2 \quad (3)$$

The diagonal values of $\mathbf{R}$ were therefore set to the squared fractional uncertainties in mole fraction, which were calculated as described in the text. Similar considerations were given to the uncertainties applied to the diagonal elements of $\mathbf{P}$, although the functional form depended on the corresponding state vector element in that case.

p24487: Fig. 4 shows considerable inter-annual variability in the lifetime after the peak in atmospheric abundance. Is this variability within the uncertainty ranges calculated on each mean lifetime? If it is not, how does the mean lifetime depend on the time window selected?

For all species, the 1-sigma uncertainty ranges calculated for any averaging window within the period used to define the "best estimate" overlap with the "best estimate" 1-sigma uncertainty ranges. This comparison is now shown in the Supplement.

2 Technical comments

p24476, l21: “by Daniel et al (2007)”

We have removed this citation.

p24477, l13: the reviewer could not find where “TEAP” is defined, this should be defined somewhere

We have added this definition in Section 3.1.

p24484, l6: “in Sect. 3.1 do not. . .”

Corrected.

Table 2: in the caption of this table “emission uncertainties” is misleading as the table only shows the lifetimes and their associated uncertainties.

We have removed the erroneous reference to emissions uncertainties.

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