

Responses to interactive comments on “Model Sensitivity Studies of the Decrease in Atmospheric Carbon Tetrachloride” by Chipperfield et al.

We thank the reviewers for their comments. These are reproduced below, followed by our responses in *red italics*.

Anonymous Referee #1

General remarks: I am in favor of publishing the paper after following points have been carefully considered.

1. L 48 ... and large uncertainty range (157 to ... Better mention that this is the lifetime as before you speak of losses... and large lifetime uncertainty range (157 to ...

OK. 'lifetime' has been added.

2. L 87 it was 2014.

OK. The year has been corrected.

3. L 115/223/441 numbers for Butler et al. are not identical. However, I just read that they have actually revised their number between the ACPD and the now accepted revision to ACP by 15%. See: response to referee 2: <http://www.atmos-chem-physdiscuss.net/acp-2016-311/> L 48/448 range... see just above I am also a little bit unsure what to propose here. Either authors stay with the old number from Butler et al (ACPD) which is also part of the SPARC report or they redo their analysis by asking Butler et al. what their “final-final” number and uncertainty will be in the ACP paper (which will be published soon).

We have revised the paper to include both sets of results. We have redone the model runs based on the actual published values of Butler et al., which should be considered the 'final-final' values. To maintain the link with the SPARC report we also include results from 3 tracers (CTC1s, CTC2s, CTC3s) which use the SPARC values for the ocean loss. This run is plotted in Figure 8a, which links to a figure in the SPARC report.

4. Line 403 ...show more variability than the AGAGE... should be... show more variability than the AGAGE...

OK, word corrected.

5. Line 664, Figure 6: x-axis should be ppt, not ppb.

OK, corrected.

6. Line 676, Figure 7, header: more accurate? CCl₄ partial photolysis lifetime

OK, we have added 'photolysis'.

Anonymous Referee #2

This manuscript reports on the impact of uncertainties in the removal rates of CCl₄ by photolysis, ocean uptake, and degradation in soils on the 3D chemical transport modelled decay rate of CCl₄ in the atmosphere. Overall, the manuscript is very well written and offers an update on the long standing issue of discrepancies between modelled and measured CCl₄ based on updated information on the removal processes.

The manuscript offers an important contribution to understanding the model/measurement discrepancies in CCl₄. It doesn't provide a final answer but does show that the discrepancies can be minimized by adjusting the rate of loss processes within their respective uncertainties or by increasing emissions in the model. The manuscript also points to the large uncertainties in the recently calculated ocean loss by Butler et al. as a significant contributor to the overall model uncertainty, more so than the photolytic or soil losses. Another factor that may contribute to the discrepancy is the modeled radiative transfer and transport from the troposphere to the stratosphere.

The authors use a significant amount of data for their comparisons, including ground based monitoring networks for in-situ and column data, aircraft data, and satellite data. This data, together with use of the most recent information on losses, strengthens the authors' comparison and conclusions. The figures and references are appropriate, however the units for Figure 6 should be ppt.

The units of Fig 6 have been corrected.

I recommend publishing the manuscript after addressing the clarification comments below.

1. In section 2.2 ACE; there should be a reference for the last sentence in the first paragraph, especially since both references in this section are still in press. This assumes that a previous reference would have the high bias discussed.

Although both references are in press (the final versions are available on the website, without assigned page numbers etc), they are part of a special JQSRT issue, so we have to wait until all the other contributions are ready.

At the end of this paragraph we have added "(Allen et al., 2009; Brown et al., 2011; Harrison et al., 2016).", including the two new references:

Allen, N. D. C., Bernath, P. F., Boone, C. D., Chipperfield, M. P., Fu, D., Manney, G. L., Oram, D. E., Toon, G. C., and Weisenstein, D. K.: Global carbon tetrachloride distributions obtained from the Atmospheric Chemistry Experiment (ACE), Atmos. Chem. Phys., 9, 7449-7459, doi:10.5194/acp-9-7449-2009, 2009.

Brown, A. T., Chipperfield, M. P., Boone, C., Wilson, C., Walker, K. A., and Bernath, P. F.: Trends in atmospheric halogen containing gases since 2004, J. Quant. Spectrosc. Ra., 112, 2552–2566, doi:10.1016/j.jqsrt.2011.07.005, 2011.

2. In section 2.5 TOMCAT 3-D Chemical Transport Model; do the authors have an estimate of the range of uncertainty in the modeled photolysis rates associated with errors in the model radiative transfer code, ozone distribution, etc. Could this overwhelm all other uncertainties?

There will be some uncertainty due to model radiative transfer scheme, but this is not easy to quantify. We would note that TOMCAT/SLIMCAT photolysis scheme performed well when

compared with other models in the SPARC CCMVal report (2010) and in the recent paper by Sukhodolov et al. (JGR,2016, 10.1002/2015JD024277), especially for the species which do not depend on specific parameterisations. This suggests that the model radiative transfer code is accurate. We therefore argue that other uncertainties would not dominate the uncertainty due to the cross sections and quantum yields. We have added a discussion into the paper.

Also in this section it should be stated how the ocean and soil sinks are distributed and if there are temporal variations. It is an important factor and should be included in the description.

The soil and ocean sinks were included in a similar way to Liang et al. (2014) (see their supplementary material). The sinks are treated as spatially uniform over all land or ocean grid points, respectively. There is no time variation. Text stating this has been added to the paper.

3. In section 3.3 Impacts of uncertainties in sinks; the authors state that “resolving the issue of the absolute difference in the concentrations reported by the two networks will be important...”. Is there a plan to do this?

The NOAA and AGAGE communities are in regular contact and are aware of how their observations intercompare for all species studied. They have been studying this issue for CCl₄ for some time and continue to do so. It is not possible to go into any detail in this modelling paper, or speculate on what might be the possible causes. In the revised paper we have added the text ‘and that work is ongoing’.

Also in this section, the authors indicate the overall lifetime variability associated with model transport can be significant because of meteorological variability. The authors should consider acknowledging this point in the introduction and emphasize that the focus of this manuscript is to evaluate uncertainties in the loss processes in the model on atmospheric CCl₄ under the model conditions specified. Also include, as stated in the conclusions, studies with multiple 3D models could be used to address this in a separate study.

The Introduction already made the point that there was a range in modelled lifetimes in the SPARC report due to different circulation rates. In the final paragraph of the Introduction we have added ‘using the framework of a particular 3-D model’. The impact of meteorological variability on the lifetime is a result of this paper. Therefore, we think that the comment on the need for multiple 3D model studies belongs in the conclusions.

4. In section 3.3 Interhemispheric gradient; the authors state that the 2006-2009 observations track each other and display a similar seasonal cycle. I only see this for parts of each year, but not enough to justify saying they track each other and have a similar seasonal cycle. The timing in their cycles is often different. The similarity between the two groups is that the magnitude of the IHG values are more similar in this time section than the others.

OK, that sentence has been deleted and text on the lack of agreement added. The information on the similar value for the IHG in 2006-2009 has been added to a previous sentence on this point.