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Atmospheric Chemistry and Physics Discussions

ACPD

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Interactive Comment

Interactive comment on "Global carbon tetrachloride distributions obtained from the Atmospheric Chemistry Experiment (ACE)" *by* N. D. C. Allen et al.

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We thank the reviewer for his/her helpful comments.

Comment: This is a fairly short paper that describes the distribution of carbon tetrachloride within the upper troposphere and stratosphere as observed by the ACE satellite instrument. Generally the paper is well written but I think a little more attention needs to be paid to (a) the ACE and MkIV balloon comparisons and (b) the derivation of the carbon tetrachloride lifetime. Once these issues and the minor items below are corrected the manuscript should be worthy of publication in ACP mostly since the carbontetrachloride distributions are the first observed by satellite.

Specific Issues: Abstract: Make it clearer that the ACE carbon tetrachloride distributions are within the upper troposphere and stratosphere. In addition, briefly state the results of the satellite and model comparison. A sentence has been added summarising the comparison between ACE and the models. It has also been clarified in the abstract that measurements were taken in the upper troposphere and lower stratosphere

Page 13300 Line 25: How much does carbon tetrachloride contribute to the global chlorine budget (include a percentage estimate and a reference)?

The contribution of carbon tetrachloride to the total chlorine budget in the troposphere (11% using WMO report 2007 for reference) and the stratosphere (3% calculated using the budget derived from ACE by Nassar et al.) has been included.

Page 13301 Lines 7: Please provide the basic reactions of carbon tetrachloride within the troposphere/stratosphere.

Reactions 1-3 have been provided to show the chemical pathways for the destruction of carbon tetrachloride. An addition has been made to the modelling section to explain which reactions, and their rates, were used in the models.

Page 13301 Lines 9-11: This sentence is not very clear. Do you mean the cumulative loss into the oceans? Also, the lifetime of carbon tetrachloride was reduced to 26 years; reduced from what? Please clarify.

The loss of carbon tetrachloride caused by ocean uptake and hydrolysis reduced the estimated atmospheric lifetime of carbon tetrachloride from 35 to 26 years.

Page 13302 Line 1: What measurement technique was used to make the first atmospheric observations?

Lovelock and Maggs used gas chromatography to make their measurements, which were the first atmospheric observations of carbon tetrachloride.

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Page 13304 Line 9: Grammar. I'm sure a comma should be inserted after the Nassar et al. (2005) reference.

Comma added.

Page 13306 Lines 19-20: Style, for example: '…cloud tops to the balloon (35-40 km)…' - am I to believe a balloon is as big as 40 km? Please rephrase this sentence to make it clearer.

This sentence has been expanded to explain that the MkIV balloon retrieves from the cloud tops (retrievals cannot be made through the opaque atmosphere) to the float altitude for the balloon payload typically 35-40 km.

Page 13307 Line 4: There is no mention of the location to which Figure 6 refers.

Figure 6 refers to measurements taken over Fairbanks in Alaska and this information has been added to the paper.

Page 13307 Lines 14-15: Apart, from Fairbanks the ACE and MkIV profiles agree fairly well in the troposphere. Please mention this.

A sentence added to acknowledge this and a further one has been added to comment on the poor agreement with the balloon profile on 8 July 1997.

Comment: With reference to the balloon comparisons, I think it would be beneficial to also compare ACE monthly or seasonal averages with the MkIV data rather than just averages over the entire 2004-07 ACE dataset. For example, compare ACE observations averaged over Sept 2004-07 with the balloon flights at Ft. Sumner or ACE observations averaged over March-April-May 2004-07 at Esrange.

Seasonal ACE average lat bin plots were reintroduced into the paper, these were removed due to concerns of over cluttering the figures with the addition of error bars. Therefore the averaged profiles have been included in figures 5-7 without the error bars to give a clearer overview. For New Mexico the seasonal SON (September-

October-November) latitude bin from the ACE mission between February 2004 and August 2007 is displayed; for Fairbanks the average profiles for the MAM (March-April-May) and JJA (June-July-August) seasons are also included between February 2004 and August 2007. The average seasonal profile for MAM (March-April-May) is included on the Esrange figure over the 60-70°N bins solely for 2004. Analysis of the seasonal profiles has been added.

Page 13307 Line 23: The global atmospheric lifetime of carbon tetrachloride is given as 26 years, is the tropospheric lifetime the same?

The tropospheric lifetime of carbon tetrachloride is different from the 26 years given in the WMO reports, which is the estimated atmospheric lifetime.

Page 13308 last paragraph: Whilst the fall in carbon tetrachloride mixing ratios with altitude is the about same between the model data and ACE observations there is a clear offset between them, both in the stratosphere and troposphere, which is not discussed. Is this offset attributed to the carbon tetrachloride surface observations which are used to constrain the models? Are the models lacking any carbon tetrachloride sources or sinks, or chlorine photochemistry? More discussion is required here.

A paragraph was written to try and explain possible reasons for the differences: The discrepancies observed in the troposphere are due to the imposed surface boundary concentration values used to constrain the models, thus values will be no greater than the ground input value. The models assume a uniform surface boundary condition with no latitudinal or longitudinal variation, due to the longevity of carbon tetrachloride, however asymmetric emissions could lead to some fluctuations. In all three models the sources and sinks are the same. They include loss of carbon tetrachloride by photolysis and reaction with O(1D) but no other sink mechanisms, such as ocean uptake. Differences in the modelled vertical profiles are due to different rates of stratospheric transport (Brewer-Dobson circulation).

Page 13309 Line 19: Please define ‘OCS’ as carbonyl sulphide. This was rectified.

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Page 13309 Equation 1: Please add that the VMRs are representative of concentrations in the troposphere not stratosphere. I also think a slightly more detailed analysis is required here, for example: (a) What tropospheric mixing ratio of carbon tetrachloride was used in the calculation of its stratospheric lifetime? What uncertainty was assigned to this value? (b) Does the curvature in Figure 10 arise from observations within different latitude bands? (c) How does the carbon tetrachloride lifetime vary with latitude? (d) How does the carbon tetrachloride lifetime change if newer estimates of CFC-11 and CFC-12 lifetimes are used (e.g., WMO 2006 report)?

We are not sure what the referee has in mind here: we simply took all observed values from the ACE data in both the stratosphere and troposphere and made our plot (Figure 10). The slope was taken for CCl4 values that lie between about 20 and 100 ppt, i.e., these values are mostly in the stratosphere. Using newer estimates from the WMO report is likely to have a linear effect reducing the lifetime of carbon tetrachloride. The lifetime of carbon tetrachloride does not seem to vary significantly with latitude.

Page 13310 Line 7: Please fully reference any previous estimates of the carbon tetrachloride lifetime.

Early estimates for the lifetime of carbon tetrachloride were between 30-50 years (Singh et al, 1976; Simmonds et al. 1988) before Montzka et al. (1999) and the 1998 WMO report (1999) reduced this to 35 years. Inclusion of an ocean sink (Yvon-Lewis and Butler, 2002) for carbon tetrachloride in the 2002 WMO report (2003) further reduced the lifetime to 26 years and suggestions of a soil sink (Happell and Roche, 2003) give a lifetime of only 20 years.

Page 13310 Line 10: What were the estimates of the carbon tetrachloride lifetime from the GMI and AER-2D models?

These were not particularly informative and gave lifetimes for carbon tetrachloride just over 40 years.

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Summary and Conclusions: The authors have shown some nice carbon tetrachloride retrievals from the ACE satellite but it is unclear to me what the 'take home message' of the paper is. For example, how do the ACE retrievals improve our understanding of carbon tetrachloride and chlorine chemistry within the atmosphere? Are there any plans to assimilate the ACE observations into one of the chemistry-transport models? I think the 'take home message' needs to be more clearly stated in this section (and abstract accordingly).

There are no plans at this time to assimilate ACE data with Chemical Transport Models. The WMO report (2007) suggests that the atmospheric lifetime and budget of carbon tetrachloride are still poorly understood. Global satellite measurements, such as these profile retrievals from ACE, provide a dataset that will lead to an improved understanding of the carbon tetrachloride budget.

Figures: Figure 2: State where the average tropopause height is determined from.

The average tropopause height is taken from DMPs averaged over five degree latitude bins like ACE data.

Figure 4: The orange line for 45-60 S did not clearly print (after using two different printers). Please consider using a different colour.

The line colour was altered from orange to dark green for clarity.

Figures 5-7: Please remove the plot-titles from these figures as it is misleading (the ACE observations are not made from a balloon platform).

Titles were removed.

Typographic Issues: Page 13300 Line 10 and elsewhere: 'equator' should be capitalized.

Does not seem common to capitalise equator but have changed this regardless.

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Interactive comment on Atmos. Chem. Phys. Discuss., 9, 13299, 2009.

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