

Response to comments on “Atmospheric three-dimensional inverse modeling of regional industrial emissions and global oceanic uptake of carbon tetrachloride”

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Please note: we provide our replies in bold font after each Referee’s comments.

Anonymous Referee #2

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Comments: The authors have used an inverse modeling approach based on a Kalman filter to deduce industrial emissions for carbon tetrachloride and its geographic sources. The inversion uses the 3D MATCH model as the forward model and makes comparisons by ground based measurements taken at stations distributed globally. The magnitude of the global oceanic sink is also estimated.

The paper highlights that strong emissions across Asia, particularly the South and Southeast, lead to a much greater industrial source for carbon tetrachloride than had previous been suggested. As a result global carbon tetrachloride concentrations are in decline at a slower rate due to these fugitive emissions.

General Comment: The paper is precise and the data and MATCH model appear to give good results. The manuscript only needs a few minor changes before publishing in ACP. Here are a few minor issues that I think should be addressed:

Montzka et al. 2003 has been referenced repeatedly, whilst undoubtedly a good paper you should find other literature to cite to help bolster your case.

More references have been added to the text (e.g., Page 3, Line 7; Page 3, Lines 13-14; Page 3, Line 18; Page 4, Lines 8-9).

The use of the two phrases industrial and fugitive emissions need defining as it is unclear what is exactly meant by each term. Do these phrases encompass all anthropogenic emissions? I think it is also important that you emphasise that all of the measurements taken are background concentrations, away from large anthropogenic sources.

The two terms are equivalent. Clarifying text added on Page 3, Lines 9-10 and Page 4, Line 9. Fugitive emissions of CCl₄ from industrial uses encompass all anthropogenic emissions. As mentioned in the text (Page 7, Line 13; Page 11, Lines 13-15), obvious pollution events in the observations have been excluded to avoid impact from large anthropogenic sources.

More details about the magnitude of the J values and the constant global factors used would be helpful. How do the J values used compare with the rates in the JPL Chemical Kinetics and Photochemical Data handbook (<http://jpldataeval.jpl.nasa.gov/>)?

Due to the large number of the figures in the paper, a reference has been provided on Page 9, Lines 8-11 (http://globalchange.mit.edu/files/document/Xiao_PhD_08.pdf, see Fig. 3.1 and Sect. 3.2.2) for more details about the J values. The absorption cross sections of CCl₄ in the JPL handbook are used in the 3-D model for the stratosphere (Golombek and Prinn, 1986, 1989, 1993, updated here with the latest kinetic and photolysis data) to calculate the J values of CCl₄.

I think it is important to detail what the industrial sales data mentioned in the paper cover. Is there no sales data that has been published? No values for the emission based on sales are mentioned anywhere in the text to give an idea of their estimated magnitude. For example if you have emission sales for Asia it would be helpful to show the contrast between these values and the estimated fugitive emissions from the model. Can the increased emissions over South and Southeast Asia be solely attributed to fugitive emissions?

Industrial sales data on production and consumption of CCl₄ reported by the Parties to the Montreal Protocol on Substances that Deplete the Ozone Layer are summarized by UNEP (http://www.unep.org/ozone/publications/Production_and_consumption2005.pdf, 2005) and are used by McCulloch (private communication, 2009) to estimate regional and global industrial fugitive emissions of CCl₄ (Page 4, Lines 4-9). Annual global industrial emission estimates from industrial sales data are compared to 2-D and 3-D inversion results in Fig. 2. The comparison is mentioned in the text to indicate the much larger decrease trend seen in the sales-based emission estimates. One column regarding the regional emissions based on industrial sales data is added to Table 2. The 9-year average estimate of the emissions over South and Southeast Asia based on sales data is 37.6 Gg yr⁻¹ (Table 2, McCulloch, private communication, 2009), which is significantly smaller than our 3-D inversion result (56.2 Gg yr⁻¹, Table 2). See added text on Page 20, Lines 18-21.

To help the reader put the papers results into context I think that it is important to compare and validate the oceanic sink estimate and the total global industrial emissions with other literature values. Is there good agreement?

Our finding of prior underestimation of emissions in Asia is consistent with the research results of Palmer et al. (2003), and relevant text has been added on Page 20, Lines 20-21.

Figures – It may be helpful to use different symbols and lines (i.e. dashed, dotted etc) just in case people print in black and white.

Suggestion adopted. Figs. 2, 4, 6, 7, 8, 9, 10, and 11 have been revised.

Specific Points:

p. 12227, line 4: Please change to CCl₄.

Typo corrected (Page 2, Line 4).

p. 12227, line 11: Do you need to state what the abbreviation NOAA stands for?

The full name is now provided (Page 2, Line 12).

p. 12227, line 23: Please reference The Montreal Protocol and Subsequent Amendments.

Reference provided (Page 3, Lines 3-4; Page 26, Lines 30-32).

p. 12228, line 8: I think it should read ‘emission reductions’ and ‘The’ Montreal Protocol (please capitalise throughout).

Done (Page 3, Line 15).

p. 12228, lines 8-12: You may want to explain that due to the long lifetime of CCl₄ you would expect there to be little asymmetry without significant emissions or sources.

The explanation has been added to the text (Page 3, Lines 18-20).

p. 12229, line 23: It may be an idea to explain briefly what a discrete Kalman filter is and how you can use the estimates produced.

Brief description has been added and the reference is provided (Page 5, Lines 19-22).

p. 12229, line 24: Please be consistent with the use of hyphens. You have hyphenated intra-annual but not inter-annual, whereas inter-hemispheric is. Please rectify so that all references in the text to both words are all hyphenated.

Hyphens in intra-annual, inter-hemispheric, inter-comparison, etc. have been removed.

p. 12230, lines 3-8: It might be a good idea to explain what exactly you mean by high and low frequency stations – does this refer to the rates of sampling?

Details about the high and low frequency sampling rates have been added (Page 6, Lines 8-9; Line 13).

p. 12230, line 4: You should state, however obvious, what the abbreviation NASA stands for.

Done (Page 6, Line 9).

p. 12231, line 13: I think it would be helpful if you stated what range of pressures or altitudes these 28 layers cover.

The range of pressures (1000 – 2.9 mb) is provided (Page 8, Lines 6-7).

p. 12231, line 16 (and other places): O(1D)

Corrected.

p. 12233, line 6 and 7: You may want to say/reference where you got your population statistics from, or at least state the estimates that you used. It would also be helpful to do this for the GDP data and state which year or years these GDP values are from.

Data resources provided (Page 10, Lines 15-17).

p. 12239, line 7 and 8: Gg yr⁻¹ per year – can you clarify the two different uses?

The unit has been changed to Gg yr⁻² for the derivative with respect to time of the emission rate (in Gg yr⁻¹), and clarifying text added (Page 18, Lines 7-8).

p. 12239, line 19 and 20: I think you should have your definition of semi-hemisphere before mentioning Fig. 10 in the text.

Definition provided (Page 18, Lines 20-21).

p. 12240, line 23 and 24: I think you need to emphasize in this sentence that this is the global industrial emission of CCl₄.

Text clarified (Page 20, Line 12).

Fig. 7. Could this be split into 7 a and b, as although color bars are the same their value are clearly different, the captions will be easier too. Also, I think the heading for diagram one should be Industrial Emissions rather than just Industry.

Subtitle is revised and the caption of Fig. 7 has been re-arranged.

Fig. 8. This is a good way to highlight changes. However, some of the text is slightly obscured and you might like to put a line from each continent to the piece of pie that represents it.

Done.

Fig. 9. I think to avoid confusion it would be better to put the net loss for the CCl₄ budget separately.

Figure 9 now revised. The net loss term is put on a separate subplot.