

Interactive comment on “Emissions of Carbon Tetrachloride (CCl₄) from Europe” by Francesco Graziosi et al.

Anonymous Referee #2

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In this paper the authors take long-term atmospheric data records of carbon tetrachloride (CCl₄) from several European sites and use an inversion-type model to derive top-down estimates of European industrial emissions. The rationale behind this is that there is a well-known discrepancy between global bottom-up emission estimates provided by producers and recent top-down estimates derived from atmospheric measurements. Assuming that the atmospheric loss processes are understood correctly, this discrepancy implies there is a missing or unknown source of this important ozone depleting chemical, which would explain why the global atmospheric abundance of CCl₄ is not declining as fast as would be expected.

The major finding of this work is that Europe is a relatively minor contributor (4%) to global CCl₄ emissions and that there are continuing small emission sources in particular regions of Europe. Although this is not a startling discovery (indeed a European

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contribution of 4% was also calculated for the earlier period 1996-2004 by Xiao et al., 2010) this work is a useful contribution to the global jigsaw. Although I am not qualified to comment on the detailed mathematics that goes into the Bayesian statistics and inverse modelling, the uses of these methods by the authors and others have been widely published in recent years so I would expect them to be robust. The measurement and modelling teams both have an excellent reputation and I see no major flaws in their work. The manuscript is generally well written and I would recommend publication following consideration of the following minor comments.

Detailed comments

At the end of the discussion section (or in the conclusions) when putting their European emissions into a global context I wonder if the authors could summarise the current state of play regarding the CCl₄ story. It seems from the references given that recent US, European, Australian and even Chinese top-down emissions still may still not add up to the total amount required to maintain the current atmospheric abundance. Is there still a missing source, or is the budget balanced within the various levels of uncertainty?

Line 35: “European emissions correspond to 4.0% of global emissions for 2006-2012”. Do the authors mean cumulative emissions over the 7 year period or is it an average of 4% each year? Please clarify.

Line 61: “total chlorine in the troposphere”. Firstly, do the authors mean total organic chlorine? Secondly, which part of the troposphere are they referring to?

Line 88: replace of with for i.e. “being responsible for more than half”

Line 136: I am intrigued as to why the MHD data is taken from the GC-ECD instrument rather than the Medusa-GC-MS that also operates at the site? At JFJ they are using data from a similar (identical?) Medusa GC-MS instrument, but the authors choose not to report the GC-MS data from MHD. Is there a reason for this?

Line 143: delete “are” i.e. “tertiary tanks used as”

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Line 144: "at least twice" is not the same as "regularly calibrated". The working tanks are prepared at SIO and are calibrated (at SIO) at the beginning and end of the life of the tank?

Line 167: Is there a reference or web link for the E-PRTR database?

Line 185: move "also" – i.e. "During 2010-2014 data from JFJ were also used"

There are a few places in the text when the word "the" should be added:

Line 200: the emission distribution ... Line 202: the main deviations ... Line 210: during the study period ... Line 240: The maps in ... Line 259: the chloro-alkali industry ...

Line 215-216: how can emissions be negative? Please explain in the text.

Line 216: is there a reference for the UNEP production database?

Line 217-218: "Such discrepancy holds also ..." This sentence doesn't really make sense. Please explain and expand.

Line 225: change to "... contribution to the total European emissions of CCl4 from the ..."

Line 230: change to "... as the main emitter in the EGD over the entire study period ..."

Line 231: exactly 25%? Perhaps approximately 25% would be better (or give the range)

Line 233: replace "the" with "a" – i.e. "reached a maximum ..."

Line 318: delete "as reported above"

Figure 2:

What actually are the time series? Are they averaged in any way or are they individual samples?

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What is the dip seen in the CCl4 concentrations at CMN in 2006? This does not appear to be seen at MHD which suggests it is a local phenomenon. How can such a drop below the expected NH background be accounted for? The period seems to last for several months so presumably cannot be put down to a stratospheric event or southern hemisphere air. Does this period of abnormally low concentrations have any impact on the inversions? Why is the baseline signal in the middle panel so much more variable than the other 2? Perhaps this also relates back to the choice of ECD over MS at Mace Head?

Figure 4:

What do the error bars in Panel a represent? Please add an explanation in the caption.

Figure 7:

I am not entirely convinced by the regression lines in Figure 7 or the trends described in lines 280-284. Could the yellow trend line be biased by the slightly higher value in 2006 and the slightly lower value in 2012? With the very large uncertainties highlighted by the error bars can you really say there is a statistically significant difference between the blue and yellow lines? It is hard to see from this Figure as they are plotted on different y axes. Can the authors say with any certainty that European emissions were falling faster than global emissions over this period?

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