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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Optimal estimation of the surface fluxes of methyl chloride using a 3-D global chemical transport model" *by* X. Xiao et al.

Anonymous Referee #1

Received and published: 19 February 2010

Scientific Significance

This paper presents a detailed inverse 3-D modeling study of the tropospheric budget of CH3Cl, a mainly naturally-produced chemical species involved in stratospheric ozone chemistry. The paper makes a significant contribution to our understanding of the CH3Cl budget by placing new constraints on its various sources, especially the source from tropical plants, which is the largest, and perhaps least well-defined. While the CH3Cl system has been subject to inverse modeling before (Yoshida et al, 2006), the current study is done on a finer resolution, and with a different inversion method. It also uses a time series of meteorological and observational inputs, rather than using an 'average year'. Finally it addresses explicitly seasonal and spatial variation in the biogenic (plant) source in a much more detailed way than has previously been

done, enabling a new quantitative estimate of this source to be made. The discussion of a linkage between model-inferred changes in the sources and global effects of the 2002-2003 El Nino episode is novel and interesting and hints at effects which might be observed under conditions of future climate change. These potential changes are worth considering in simulations of future halogen budgets, and may have implications for modeling of future stratospheric ozone recovery.

Scientific Quality

The approach taken is sound, and well-explained. The model, method, and input data are well-referenced, and the main conclusions are supportable and clearly presented. (For a few specific comments, see below).

There are two major omissions, which should be addressed before publication in ACP. First, no reference is made to the inverse model study of Yoshida et al (2006). That work should be acknowledged, and it should be explained how the present study represents an advance in knowledge since the earlier work. Second, it would be good to see a comparison to some aircraft data, particularly the TRACE-P data, which was collected during the period of the current simulation, in the tropical region. The authors mention the scarcity of tropical surface data, which inhibits a fuller understanding of the distribution of the large tropical sources. It seems that including the available airborne tropical data would be helpful.

Presentation Quality

The title and abstract are clear and adequately describe the content of the paper. The paper itself is impressively readable, with good language and logical flow and an almost complete lack of typos. The references are sufficient and appropriate (except as noted above). The figures and tables are well-presented and self-explanatory. This reviewer would not omit any of them. The equations, symbols and units are clear and appropriate. (For a few specific suggestions for improvement, see notes below). This reviewer appreciated the Appendix explaining the Kalman filter.

Specific minor points:

Pg 27697, bottom: the size of the 'missing' source added by Lee-Taylor et al ('01) was slightly lower than stated here.

Pg 27702, top: Please give values (with appropriate references) for A and E

Pg 27702, bottom: Please state explicitly whether or not you neglect stratospheric photolysis, and if it is included, give values and/or a reference for the photolysis rate expression.

Pg 27704, line 10: Should the reader understand that you perform 60 monthly pulse runs for *each* of your 8 seasonally-varying sources (ie 480 runs in total)?

Pg 27705: please include Watling and Harper (1998) as the original citation for the fungal source.

Pg 27706, lines 26-28 + Figure 3: Please clarify whether West Africa and Spain are in the Eastern or Western biomass burning region? The text and figure do not seem to agree on this.

Pg 27707, equation 1: What does T represent?

Pg 27708, line 4: What are the criteria for deciding whether the uncertainty is 30% or 50%?

Pg 27709, line 20: Surely the Eastern BB source is elevated in early 2002 as well as in early 2003?

Pg 27710, bottom: I think the Lee-Taylor ('01) extrapolation was made *from* the CH3Br observations of Shorter ('95), rather than these being two separate items.

Pg 27711: The rankings of the different sources with respect to the relative amount of error reduction do not appear to be consistent with those shown in Figure 7. For example, salt marshes and the soil sink show large relative changes but small absolute

changes in the figure. They are ranked in the text among the least-changing on a relative basis. Please clarify this section.

Page 27712, lines 10-20: The AM tropical plant source seems to show a peak in January (larger than the peak identified in March). Similarly, the AF source shows a peak in December, which is not mentioned. Adapting the plots (figure 8) to show the interpolation between December and January would assist the perception of the seasonal cycle as continuous, and allow a more accurate assessment of where the emissions peaks are. Similarly, the salt marsh peak appears to be in January, not June (pg 27714 line 15). Also, the discussion of seasons (spring, summer) is more appropriate for northern mid-latitudes than for tropical regions spanning the equator. This part of the discussion should be revised in terms of the timing of equinoxes / solstices, wet/dry seasons, and the overhead passage of the ITCZ.

Figure 2: Please use the same notation for the numbers on the two y-scales. Please put the a) and b) in the caption at the beginning of the description of the relevant panel, not after it.

Figure 8: This figure would be greatly improved by extending the lines to interpolate between December and January at the edges of the plots.

Figure 10: The NH and SH symbols look very similar, perhaps because they are so small. Please find some way of making them more visually distinct.

Very minor suggestions: Pg 26797, line 19: '.. based on a revised loss' <rate>? Pg 27712, line 4: 'process' not 'processes' Pg 27716, line 11: perhaps 'global in-soil consumption'? The soil itself is not being consumed! Figure 4: 'equal to their reference' <values>?

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 27693, 2009.