Atmos. Chem. Phys. Discuss., 9, C12124–C12127, 2010
www.atmos-chem-phys-discuss.net/9/C12124/2010/

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 #2

Received and published: 15 April 2010

This manuscript presents the results of a data inversion calculation to arrive at an "optimal" estimate of methyl chloride emission fluxes from major sources. This manuscript uses a combination of high quality data and a well-described and documented modeling procedure to ultimately show the dominance of a tropical vegetation source in the global methyl chloride budget. Source emissions from biomass burning, oceans, salt marshes, and fungal activity are also estimated. The data sources are from surface sites of the AGAGE, NOAA/ESRL, SOGE, and NIES networks. The 3-D global transport model is the Model of Atmospheric Chemistry and Transport (MATCH). The paper is clearly written (though I can't say that I personally understand the details of the computation), and the methods of calculation and the results are clearly discussed. An interesting aspect of the calculation relates to anomalous results during the 2002/2003 time period when drought conditions seemed to have widespread impact on global

methyl chloride emission. This feature is discussed with reference to possible changes in methyl chloride fluxes in a changing climate. The paper builds on earlier estimates of a global methyl chloride fluxes and produces interesting results.

Based on past successful applications of the inversion technique, and my own limited knowledge of the details of the computation, I have to assume that the procedure was appropriately carried out and that the results are, indeed, the optimal fluxes given the underlying assumptions. While it seems clear from this manuscript and earlier studies that there are large sources of methyl chloride in the tropics, I have some question about how the tropical source was parameterized here and how much effect this has on the inversion. As far as I can tell, the methyl chloride source was distributed as follows: 1) methyl chloride increases were found with alpha-pinene increases (Yokouchi ref.); 2) alpha-pinene emissions are related to foliar density, light, and temperature; and 3) foliar density is proportional to NPP, so 4) methyl chloride flux is distributed with NPP. While I understand that some global emission grid must be constructed, this seems to be a pretty loose connection for the major emission component. The fact that a-pinene and methyl chloride were observed to increase under a shallow boundary layer does not indicate that the same factors are involved in controlling emission rates for methyl chloride and a-pinene. Further, the emission studies of Yokouchi et al. identified only a subset of particular species that were major emitters of methyl chloride, and the emissions could be highly variable even among individual species. I would like some discussion of how uncertainties in the distribution of emitters in the different regions impacts the final results.

I also have some question about the station data and how it is interpreted. For example, I looked at the time series at American Samoa for 2007, with data from both NOAA and AGAGE instruments (Figure 1). Though 2007 was not used in the analysis here, I was interested in the number of significant short-term increases in methyl chloride. Some were observed by both instruments; some by only one or the other. The NOAA record that I could see shows that these excursions are common in Samoa. I haven't gone to

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look at all of the other data. Do these sorts of excursions have an effect on the data used in the analysis? Is it clear that these are not due to local vegetation? If not, what is the interpretation?

I would also find useful one or several plots that shows the contribution of the individual sources and sinks to the annual cycle at selected sites. While there is a plot that shows the errors between the forward model and station data, I would find it more instructive to see how the timing and intensity of different sources impacts the temporal variations at different sites (esp. tropical). Other than the data presented by Yokouchi and colleagues near or in tropical sources of methyl chloride, most increases in methyl chloride in the remote atmosphere have been interpreted in terms of biomass burning emissions, and related tracers seem to support that interpretation. Presumably, the biomass burning source is intermingled with the tropical plant source of methyl chloride. Thus, it would be instructive to see how the variable timing of the different sources contributes to the ambient levels of methyl chloride at the monitoring sites.

Finally, given the uncertainties in parameterizing the tropical plant sources, the results would be more convincing if multiple tracers could be used in the inversion to support the calculated distributions and timing of emissions. Is there any option for this, given the range of trace gases measured at the different monitoring sites?

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

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Interactive Comment

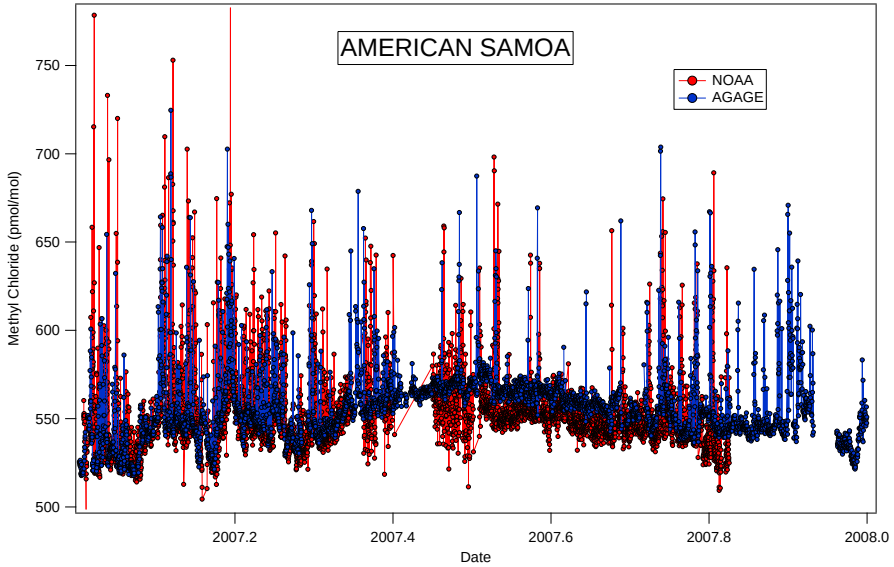

Fig. 1. Time series of methyl chloride at American Samoa 2007

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