

## **Responses to comments on “Optimal estimation of the surface fluxes of methyl chloride using a 3-D global chemical transport model”**

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

### **Anonymous Referee # 2**

**C1.** I would like some discussion of how uncertainties in the distribution of emitters in the different regions impacts the final results.

**The current assumptions about the distributions of biogenic and biomass burning emissions of CH<sub>3</sub>Cl within each region are based on significant prior knowledge and are as reasonable as available. Also, because we use monthly average observations that largely omit pollution events in our inversion, our large-scale regional emission estimates are not readily biased, for example, by errors in the distribution of emissions at grid points close to observing stations that produce these pollution events. Text has been added in Sect. 6 to address this point. If we had a reasonable basis to justify alternative distributions we could run the inversion again with those; but that is not the case.**

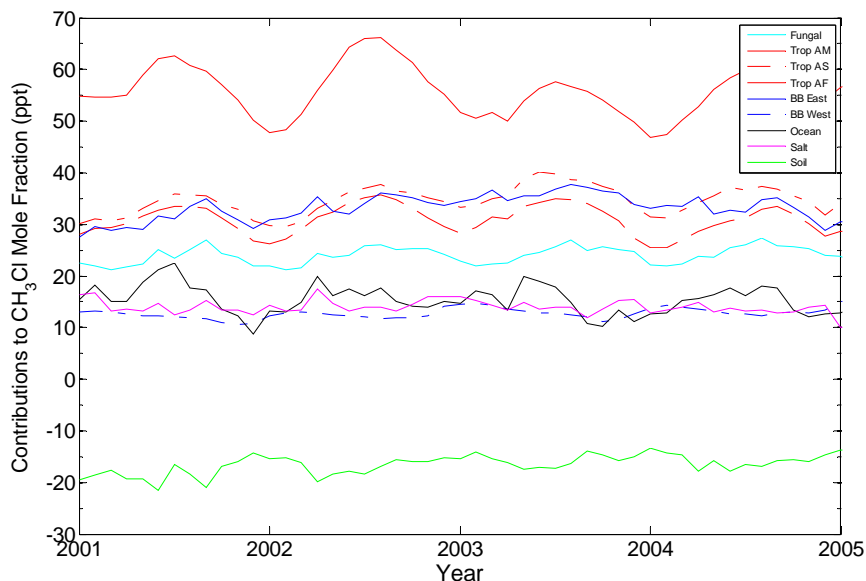
**C2.** Do these sorts of excursions have an effect on the data used in the analysis? Is it clear that these are not due to local vegetation? If not, what is the interpretation?

**The high frequency variability shown by the reviewer at Samoa has relatively short-lived peaks that may or may not be of local origin (the prevailing winds at this site are off the ocean). But these peaks are sufficiently infrequent that they do not markedly contribute to the monthly means included in the inversions (e.g., for Samoa AGAGE data in 2006-2009 the monthly means including these peaks are on average only 0.8% higher than those without the peaks; compare this to the overall observational errors of 1.5 – 2.4% and intercalibration error of 1.1% considered in the inversions).**

**C3.** 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.

**The contributions from each individual emission source or sink at each site can be computed by using the sensitivities in the H matrices and the corresponding optimized emissions/sinks. For example, the multi-year average contributions to the CH<sub>3</sub>Cl mole fraction at Cape Grim, Tasmania are 5%, 11%, 7%, 6%, 6%, 2%, 3%, 3%, and -3% from**

global fungi, tropical plants in America, Asia, and Africa, biomass burning in the eastern and western regions, global oceans, salt marshes, and soil uptake, with the remaining contribution coming from the OH sink, other sources (industry, wetlands) and the evolving global background  $\text{CH}_3\text{Cl}$ . See Fig. X below. We have added text along these lines in Sect. 5.3 (last paragraph), and anyone wanting this information for any of the 27 sites in our study could request it from us.



**Fig. X.** Contributions to  $\text{CH}_3\text{Cl}$  mole fraction at Cape Grim, Tasmania from different sources/regions.

**C4.** 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?

Although this is way beyond the scope of this paper, we agree that it is worthwhile in future work to try using multiple tracers to infer emissions of gases with similar source processes. Methyl bromide ( $\text{CH}_3\text{Br}$ ) could be such a tracer that is usually emitted with  $\text{CH}_3\text{Cl}$  from tropical plants, biomass burning, and is degraded in soils. Tropical plants are suggested as potential important sources of natural  $\text{CH}_3\text{Br}$  (Lee-Taylor et al., 1998; Yokouchi et al., 2000b; Saito and Yokouchi, 2006). Saito and Yokouchi (2006) measured foliar emission rates of  $\text{CH}_3\text{Cl}$  and  $\text{CH}_3\text{Br}$  from tropical ferns, and found that the diurnal variations in  $\text{CH}_3\text{Cl}$  and  $\text{CH}_3\text{Br}$  emissions were correlated with each other, suggesting these two are produced by a similar mechanism. Aqueous chloride and bromide ions can be methylated by enzymes in plants (Rhew et al., 2003). Besides the tropical ecosystems, there are other common sources of  $\text{CH}_3\text{Cl}$  and  $\text{CH}_3\text{Br}$ . There is a strong correlation between the fluxes of  $\text{CH}_3\text{Br}$  and those of  $\text{CH}_3\text{Cl}$  from coastal salt marshes (Rhew et al., 2000). Lee-Taylor et al. (2001) parameterized the  $\text{CH}_3\text{Cl}$  soil sink by using  $\text{CH}_3\text{Br}$  as a proxy.

## References

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