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 # 1

Comments on Scientific Quality:

C1. 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.

We agree. We have added text and a reference to Yoshida et al. (2006) in the introduction section. In the paper the authors used a Bayesian least squares inverse method in a 3-D model to constrain hemispheric and seasonal biogenic and biomass burning sources. Although the available measurements covered several years, an "average year" was generated for both the measurements and the derived sources. The estimated biogenic, biomass burning, and oceanic sources were 2500 (close to the Lee-Taylor et al. (2001) estimate), 545, and 761 (double the bottom-up estimate) Gg yr^{-1}, respectively. However, potential month-to-month and interannual variability in these sources related to climatic changes was not resolved by this approach.

C2. 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.

We agree that the TRACE-P data can provide additional constraints for the tropical region. Due to the required extensive computer capacity and time, it is impossible to repeat the inversions again with the TRACE-P data in a reasonable time. Moreover, the TRACE-P data only covered the time period February – April 2001, which is a very short time compared to the five-year period addressed in our paper. Our inversions used continuous measurements in the tropics from 4 high frequency in situ instruments and 3 low frequency

flask sites, so we do use a large amount of tropical data. We would certainly include aircraft data in future studies of CH_3Cl. Modifications and additions have been made to Sect. 6 to address these issues.

Comments on Presentation Quality:

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

Should be 2380 (2330 – 2430) Gg yr^{-1}. We corrected that in the text.

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

Done. A **is the pre-exponential factor (= 2.4×10^{-12} cm^3 $molecule^{-1}$ s^{-1}),** E **is the activation energy (J mol^{-1}) (E/R = 1250 K) (Sander et al., 2006). We have added their values in the text.**

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.

The stratospheric photolysis of CH_3Cl is neglected. We added text at the end of Sect. 3 to note and explain this.

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)?

The 8 seasonally-varying sources have been done simultaneously in each monthly pulse run as 8 separate “tracers”. Therefore we performed 60 monthly pulse runs instead of 480 runs.

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

Done.

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.

West Africa and Spain are in the Eastern biomass burning region (BB East). We have added words in the text to clarify.

Pg 27707, equation 1: What does T represent?

Description for *T* (= 11 months) has been added to the text.

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

The criterion is to allow for sufficiently large initial uncertainties while keeping the adjustment of a priori fluxes within a reasonable range. The choice of 30% or 50% is based upon an (albeit subjective) estimate of the quality of the a priori data and expected year-to-year variability. Text added to this effect.

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

We agree. Unusually high emissions from the eastern biomass burning source occurred in the late spring of both 2002 and 2003, which is supported by Balzter et al. (2005) and Simmonds et al. (2005). We have revised the text.

Pg 27710, bottom: I think the Lee-Taylor ('01) extrapolation was made *from* the CH_3Br observations of Shorter ('95), rather than these being two separate items.

We agree. Lee-Taylor et al. (2001) parameterized soil uptake of CH_3Cl by assuming proportionality to a methyl bromide (CH_3Br) soil sink extrapolation (Lee-Taylor et al., 1998). The latter approach used CH_3Br observations of Shorter et al. (1995) and assumed a microbial activity/soil temperature relationship. We have re-worded to clarify this method.

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.

Thank you for noting this discrepancy. We have re-ranked the relative uncertainty deductions in the text. Uncertainties for Trop AS plants, Oceans, and the Soil Sink decrease the most, those for Trop AF plants, BB West, and Salt Marshes have smaller reductions, while those for Trop AM plants and BB East have the least reductions, relative to their corresponding initial uncertainties.

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.

Thank you. Two emission peaks of Trop AM are in January (its reference value shows a peak in December) and in August. As noted in the paper, the variability in tropical emissions is the net of the combined influences of the variables atmospheric temperature, precipitation, and available light, and therefore does not generally reflect the annual cycle

of one of these variables alone. Emissions from tropical plants in Africa have a maximum in December and a minimum in July. Compared to the reference values, emissions from the global salt marshes are deduced to have a weak semi-annual rather than a strong annual cycle. We have revised the text and Fig. 8.

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.

Done. See below.

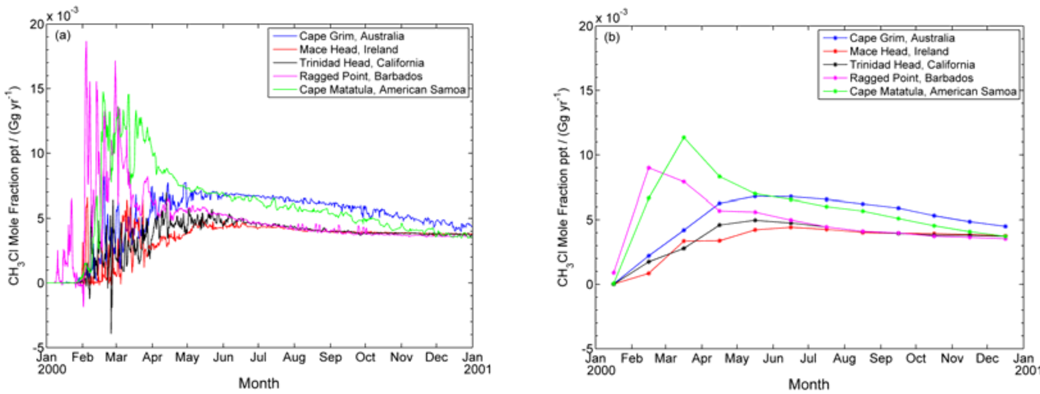

Fig. 2. MATCH-modeled sensitivities of CH_3Cl **(a)** high frequency and **(b)** monthly mean mole fractions at each AGAGE site to a January 2000 emission pulse from African tropical plants. Eventually the mole fractions reach similar values for each site consistent with a CH_3Cl emission pulse into a well-mixed atmosphere. In addition to dispersion, the sensitivities are affected by a slow CH_3Cl decrease due to reaction with the OH radical.

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

Done. See below.

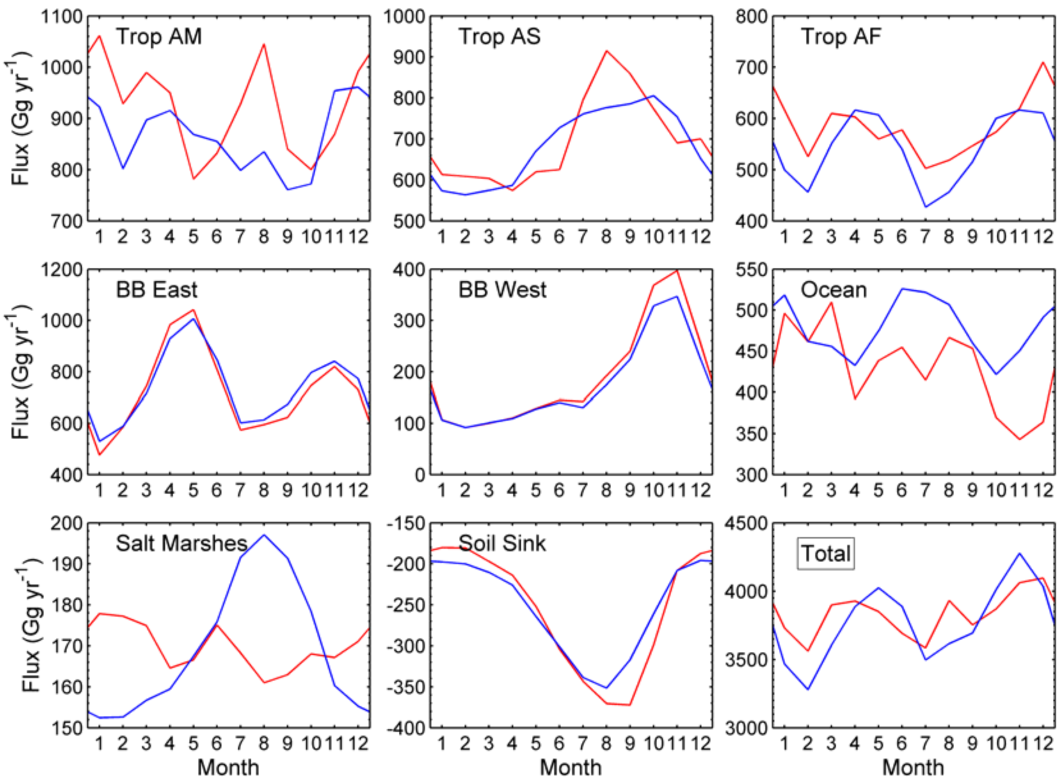

Fig. 8. Five-year averaged results for the 8 seasonally-varying emission processes of CH_3Cl. Blue lines show the reference magnitudes. Red lines show the optimized estimates.

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.

Done. See below.

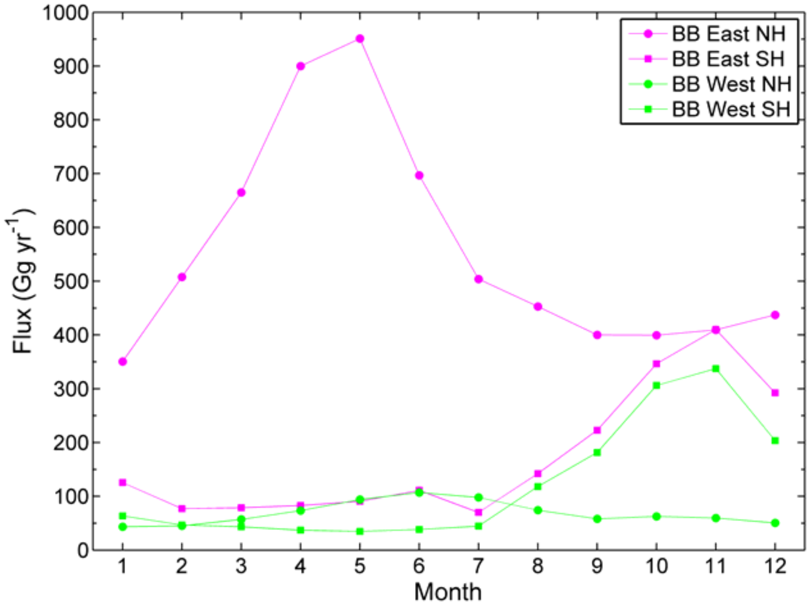

Fig. 10. Partitioning of the deduced average seasonal cycles of the Eastern and Western biomass burning sources into the Northern and Southern Hemispheres. Note the dominance of the Eastern Northern Hemispheric emissions of CH_3Cl. Also note the emission peaks of the Northern and Southern Hemispheres occur in the respective spring seasons consistent with dry conditions leading to increase biomass burning activity.

Very minor suggestions: Pg 27697, 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>?

All are done. Thank you.

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