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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Halogenated organic species over the tropical rainforest" *by* S. Gebhardt et al.

S. Gebhardt et al.

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We thank the reviewer for the very helpful comments on our paper. The comments and our replies are listed below; to keep the response in an adequate length specific comments were not repeated.

Main comment: "The manuscript lacks a clear, up-front answer to the question: Does this paper suggest significant changes to the tropical source/sink budgets of the three gases? I would like to see this question answered in the abstract, the answer developed throughout the paper, and the impacts addressed in the conclusions."

Reply: In order to address this comment, parts of the manuscript were rearranged and several sections of new text were added (see below). The last paragraph of the abstract was revised in order to better highlight the findings in the abstract. "The global budget

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of methyl chloride contains large uncertainties, in particular with regard to a possible source from tropical vegetation. Our measurements are used in a large-scale approach to determine the net flux from a tropical ecosystem to the planetary boundary layer. The obtained global net flux of 1.5 (+-0.6 2sigma) Tg yr-1 for methyl chloride is at the lower end of current estimates for a tropical vegetation sources, which helps to constrain the range of tropical sources and sinks (0.82 to 8.2 Tg yr-1 from tropical plants, 0.03 to 2.5 Tg yr-1 from senescent/dead leaves and a sink of 0.1 to 1.6 Tg yr-1 by soil uptake). Nevertheless, these results show that the contribution of the rainforest ecosystem is the major source in the global budget of methyl chloride. For chloroform, the extrapolated global net flux from tropical ecosystems is 56 (+-23 2sigma) Gg yr-1, which is of minor importance compared to the total global sources and might be already contained in the soil emission term." Within the "Results and Discussion " chapter the net fluxes obtained are evaluated in the context of the global budget source terms. From our point of view these measurements are very valuable. Since the derived numbers are based on a large scale approach, local phenomena are leveled out and a more representative average value is obtained. The studies which led to the actual numbers used for the global budget tropical source terms (Yokouchi et al. (2002) and Hamilton et al. (2003)) are restricted to glass house or laboratory measurements. Therefore the large scale approach of this study might be more suitable to assess the global emission strength. The following paragraphs have been added to the manuscript. "The calculated flux of 1.5 (+-0.6 2sigma) Tg CH3Cl yr-1 from the tropical forest would account for half of the additional source postulated by the models (Lee-Taylor et al., 2001; Yoshida et al., 2006). In comparison with the current best estimates of global tropical source terms, it is at the lower end of the range reported (0.82 to 8.2 Tg yr-1 from tropical plants, 0.03 to 2.5 Tg yr-1 from senescent/dead leaves and a sink of 0.1 to 1.6 Tg yr-1 by soil uptake). Nevertheless an emission of this order of magnitude would be the main global source for CH3Cl, exceeding the ocean, biomass burning and anthropogenic sources. It should be noted that the studies providing the basis for current estimates (Yokouchi et al. (2002) and Hamilton et al. (2003)) were restricted to the small scale

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(greenhouse or laboratory measurements), and therefore the large-scale approach applied here should be a more reliable assessment of the real global net flux from this ecosystem. When the CHCl3 flux results are extrapolated to all tropical ecosystems, the result is a net flux of 56 (+-23 2 sigma) Gg yr-1. This is between 5 and 10 % of the total sources, and as previously mentioned, it could be already incorporated in the soil source term. The global extrapolation of the lowest detectable CH3Br flux yields 17 (2 sigma) Gg yr-1. A source of this size would be a non-negligible contribution to the global budget, supplying between 6 and 22% of the total global sources. Once again it is important to recall that the global fluxes presented in this study are net sources. To compare them with the numbers listed in Table 1 all possible sinks and sources within the measurement area have to be combined." To further emphasize the impact of this study the following sentences were added to the conclusions: "These fluxes can be incorporated in global models to provide an overall net source strength of the rainforest ecosystem. Since these numbers are based on a large scale approach local phenomena tend to be averaged out and a more representative value is obtained. "

Comment: "The authors use the term "emission fluxes " in this paper, which in reality examines net ecosystem fluxes from the tropical rainforest. These net fluxes probably result from many competing sources and sinks within the ecosystem. This work estimates the net amounts of these gases exported from the forest canopy to the PBL above the canopy, not their "emission fluxes".

Reply: To prevent any misleading interpretation the term "emission flux " was changed throughout the paper to "net flux ".

Comment: "In view of the previous comment, I would prefer to see all the background information about rainforest and other competing (biomass burning, coastal ecosystem) sources and sinks in the introduction. For example, discussions of leaf and fungi and plant emissions (pg 1173-1174) should appear earlier to emphasize that the ecosystem flux for each gas is comprised of multiple sources/sinks. The reader would like to know early on what the potential processes are that might influence the

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net fluxes. In other words, tell the reader what we know (and how well we know it), what we don’t know that your study can explore, then what you learned from the study and how it impacts what we think we already know. ”

Reply: We are somewhat surprised by this comment since the introduction does contains a comprehensive summary of the average mixing ratios, long term trends, and global budgets of the investigated compounds. On several occasions in the introduction the reader is referred to Table 1, which lists all sources and sinks currently considered for species investigated. For succinctness and readability we summarize these various sources and numbers in table form within the introduction, and then discuss them in detail in the results/discussion section. The ranges given in the table tell the reader directly ”how well we know it ”. A detailed discussion of the possible production pathways is reserved for section 3.4, since it is relevant then to follow the argumentation regarding the different production mechanisms. For clarity we added the following sentence: ”This list compiled from the latest available summary papers (Lobert et al. (1999); McCulloch (2003); WMO (2003) and (WMO, 2007b)) presents the global budget of the investigated compounds. It includes all established sources and sink terms. The range of these numbers is an indication of how well these terms are currently determined. The net effect on the atmosphere is given by the sum of all these sources and sinks. ”

Comment: ”The terminology ”missing source(s) ” needs clarification. The budgets of MeCl and MeBr are not out of balance if the very large uncertainties are considered. The premise of a ”need ” for additional sources (or sinks) in specific latitudinal ranges (e.g., tropics) arises from inverse models. The estimates for many sources/sinks cover such a wide range of values that they can be increased or decreased until agreement with the models is reached. So, realistically, there may be sources/sinks that we currently don’t know about (”missing ” from the budgets), but more likely the sources/sinks already identified simply need adjustments.”

Reply: Reviewer 1 makes a very valid point here, and we agree that the term ”missing

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source " is misleading in this context. We have therefore removed it throughout the text. It was replaced by "additional source".

Comment: "I’m curious as to what the uncertainties are in the time over land (TOL) calculations. If relatively large (as suspected given the uncertainties of back trajectories), and accounted for in the linear fits (Figure 5), these can significantly alter the slopes of fit lines. Standard linear regression accounts only for uncertainties in y-variables; adding uncertainties to the x-variables and performing orthogonal distance regressions (also called "generalized least squares ") instead may considerably change the slope values (fluxes)."

Reply: Regarding the uncertainty of the TOL calculation the reviewer raises a very valid point concerning the uncertainty in the trajectories. The uncertainty associated with trajectories of this sort is highly dependent on the meteorological situation. As already mentioned in the text the meteorological conditions experienced in this campaign showed little day-to-day variability. The region is under the influence of very stable trade winds, as evidenced by the relatively small ranges of wind speed and direction shown in Figure 2. This would certainly not be the case if the region had been heavily influenced by synoptic scale meteorological events (passage of fronts) as is the case at more temperate latitudes or if deep convection was important factor. However, for the period of this study the meteorology was rather invariant and as a result there is very little divergence in the path of the trajectories starting off the flight track (see Fig. 3). There is also little evidence for vertical motion in the trajectories over the past 10 days. Generally the uncertainty of trajectories grows with increasing distance from the starting point. The TOL values used here, however, do not exceed 48 h. For the reasons stated above we feel that the trajectories can be used in this case to estimate our TOL with reasonable accuracy. Furthermore we consider this trajectory based method to be the best currently available to determine the time the air parcel spent over land. Nevertheless, the trajectories do have an associated uncertainty and we have estimated this to be ś2 h for the TOL data used. This uncertainty was incorporated as weighting

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parameter for the orthogonal distance regression analysis suggested by reviewer 1. A full comparison with other trajectories obtained for example from NCEP data set is beyond the scope of this study.

To address this point we have added the following paragraph to the text: ”For this procedure we used 10-day back trajectories, provided by P. van Velthoven, Royal Netherlands Meteorological Institute (KNMI), De Bilt, Netherlands. Trajectories were calculated using the KNMI trajectory model TRAJKS based on 6 hourly ECMWF three-dimensional meteorological wind fields, interpolated to a 1° Œ 1° lat–lon grid. The uncertainty associated with the trajectories is highly dependent on the employed meteorological parameters. Since the general weather situation in the region is quite invariant (trade wind zone) and not more than the first 48 h of the calculated trajectories are considered for TOL calculation. Nevertheless, to account for the uncertainty in the trajectories we have applied an error of +-2 h for TOL. For the reasons stated above we feel that the trajectories can be used in this case to estimate our TOL with reasonable accuracy. Furthermore we consider this trajectory based method to be the best currently available to determine the time the air parcel spent over land. ”

Comment: ”It is of interest to know if low-altitude samples collected off the coast were similar in composition to all those collected at the coast (TOL=0). In other words, were there significant marine and/or coastal source influences on samples that set the boundary conditions (TOL=0) for determining additional mixing ratio enhancements by the rainforest? This is especially important because you are fitting absolute mixing ratios against TOL, not the changes from the boundary conditions for each flight against TOL. For example, if near-coast mixing ratios of MeCl are elevated in some samples (due to coastal sources) but not in others (Figure 5), the TOL=0 boundary conditions are very different for those trajectories. I think this can be addressed by showing that off-coast mixing ratios are the same as TOL=0 mixing ratios (see next comment for Figure 5).”

Reply: Figure 5 shows both the ocean AND coastline samples at 0 h TOL. Owing to

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the small number of datapoints available we cannot resolve ”Time Before Land ” and therefore consider all of the samples taking over the ocean to be TOL=0, i.e. there is no negative TOL. Comparing the actual values of the contemplable samples, there is no significant bias between the average coastline and ocean samples. To emphasis this point the following paragraph was added: ”Samples taken above the ocean and coastline (TOL = 0 h) consisted of marine air and are used to define the boundary conditions, e.g. start mixing ratio prior to accumulation of compounds emitted from the forest. Mixed layer samples at TOL = 0h are encircled in black in Fig. 4. In general there is no significant difference between average ocean and coastline mixing ratios. ”

Comment: ”I find Figure 2 to be superfluous because the data can be described in a simple way in the text. Instead I would like to see the CO and CH3CN vertical profiles added to Figure 4, the actual data from 99 samples presented in Figure 4 instead of vertical bin means and medians and boxes and whiskers, and ML data for TOL $<$ 0 (ocean) added to Figure 5 (fits should still cover the range TOL $>$= 0). It needs to be clarified what is plotted against TOL in Figure 5 - individual sample data or some statistical bin averages?”

Reply. We find figure 2 to be useful to give an impression of the general meteorological conditions and helpful to distinguish between different layers of air used in Fig. 5. Furthermore, we also refer to it when addressing the uncertainty of trajectories issue, raised above by reviewer 1. Therefore we do not regard it as superfluous. Accepting the arguments of reviewer 3 the detailed interpretation of the vertical profiles was removed. The manuscript was revised pointing on a general trend deducible from the vertical profiles, but no longer stating a detailed interpretation regarding possible influences of biomass burning. We examined the vertical profiles of CO and acetonitrile. However, in light of the changes in the interpretation of section ”3.2 Vertical distribution ” it no longer appears necessary to add them to Fig. 4. Figure 4 was changed as requested by the reviewer; we now present the single data points, including their individual error bars. Finally the mixed layer samples at TOL=0 have been circled. The figure caption

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was changed to: ”Figure 4: Vertical distribution of CH3Cl, CH3Br and CHCl3 (black circles mark the mixed layer samples at TOL = 0h). The horizontal dashed lines mark the divisions referred to in the text as mixed layer (ML), lower free troposphere (LFT) and free troposphere (FT). BB marks the sample influenced by biomass burning. ”

Figure 5 already contains the data sampled over the ocean (see reply above). It shows all samples collected in the mixed layer without any averaging. We try to clarify this point by changing the figure caption to ”Figure 5: Mixing ratios of CH3Cl, CH3Br and CHCl3 vs. TOL. Dots represent the mixed layer samples with their individual error bars. The black line indicates the regression line. Fit coefficients including their uncertainty are given in the boxes; black asterisks mark samples excluded from the calculation. ”

Specific comments:

P1160 L5: Reply: Reviewer 1 is quite correct. We removed this term from the text.

P1160 L12: Reply: The term ”long dry season ” is a meteorological phrase used in Suriname and the surrounding area. It describes the dry period from August to November in between the two rainy seasons. The dry period from to February to mid April is called in contrast the ”short dry season”. But the reviewer is right, we should not generally extrapolate our measurements to the whole long dry season, although weather condition are usually very stable throughout one period. Therefore we have changed that phrase in the text to ”for one week within the long dry season”.

P1160 L19: Reply: Indeed it was not our intention to point out a seasonal variation. That’s why we called it ”ecosystem variation ” and stated in the following sentence that it is apparently not due to meteorological parameters. In order to restructure the manuscript, as suggested by reviewer 1 in his/her main comment section, the second paragraph was completely revised and this sentence is no longer included. To prevent any misinterpretation regarding seasonal variation between the two field campaigns, the following was pointed out in section ”3.4 Net fluxes”: ”In the case of CHCl3 there is only one comparable flux reported in literature. Scheeren et al. (2003) obtained

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a value approximately three times higher in March 1998 than the one measured in our study during October 2005. There are two wet seasons in Suriname associated with the passage of the ITCZ over the country. These wet seasons are bracketed by two dry seasons, one from February to April termed the short dry season, and one from August to November - the long dry season. Thus both available fluxes were determined in dry seasons and therefore only small differences in the meteorological conditions were observed between the March and October field campaigns. The precipitation and therefore presumably soil moisture were very similar (70.3 mm during March 1998, 65.6 mm in October 2005), whereas the maximum temperature was on average higher by 1.6 °C in October 2005 (33.4 °C), while the minimum temperature was similar (23.6 °C March 1998, 23.8 °C October 2005) (C. Becker, Meteorological Service Suriname, personal communication). Meteorological parameters therefore do not provide a good explanation for the discrepancy between the two measurements. ”

P1161 L4: Reply: Lifetimes originating from the latest WMO Assesment were added - ”(methyl chloride 1.0 yr, methyl bromide 0.7 yr, chloroform 0.41 yr ((WMO, 2007b))) ”

P1161 L6: Reply: Historical paper citations were added as requested - ”(Stolarski and Cicerone, 1974, Wofsy et al., 1975; Levine et al., 2007).”

P1162 L4: Reply: This sentence was expanded to ”Although 50-60 times less abundant than CH3Cl, CH3Br concentrations are of great interest. It has a 25 times higher ozone depletion potential than CH3Cl, caused by the fact that on per atom basis bromine is 60 times more effective in destroying ozone than chlorine (WMO, 2007a).

P1162 L10: Reply: We agree with reviewer 1. The sentence was therefore changed to ”Although balanced within the uncertainties, sinks seem to outweigh the sources, pointing on an underestimated or yet unknown source term. ” Indeed it was not our intention to identify a ”missing ” source; rather we use our measurements to check if the assumptions fed into the inverse models agree with field campaign observations.

P1163 L5-8: Reply: We have made this sentence clearer by changing it to: ”Fur-

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thermore Lee-Taylor et al. (1998) find that the annual observations of CH3Br are best represented if an additional terrestrial net source of 89–104 Gg yr-1 is assumed, which is located for 50 to 71% in the southern hemisphere. " As shown in this model assessment, and in that of Warwick et al. (2006), additional or enhanced sources are needed to reproduce the global observations. In Table 1 the total sources are generally lower than total sinks, although they might be balanced within the range of uncertainty.

P1164: Reply: As suggested by reviewer 1, the statement regarding the sample stability in the canisters was moved to paragraph "2.1 Sample collection " and the information on sample collection and GC parameters was added in paragraph 2.1 and 2.2, respectively. "Canisters were filled sequentially at 10-minute intervals without any in line drying agent." "Approximately 450 mL of the compressed air sample was introduced into the sampling inlet, dried by flushing through a magnesium perchlorate filled tube heated to 100 °C and prefocussed by a cryo-concentrator unit (1/16 " ss line filled with glass beads, cooled to –70°C)." "Storage tests on similar canisters (Colomb et al., 2006) have indicated that the investigated halocarbons are stable over 60 days under dry and humid conditions. " "The temperature profile of the GC was ramped (35 °C for 1 min, heating at 8°C min-1 to 120 °C, hold 1 min, further heating at 70 °C min-1 to 230 °C, hold 2 min, hold 1 min at 200 °C). "

P1165 L5-6: Reply: Information concerning the precision has been added to the paragraph addressing the total uncertainty thus: "The overall uncertainty was calculated based on the accuracy of the calibration standard (5%) and its precision (CH3Cl 3.2%, CHCl3 6.3%, CH3Br 6.0%) and resulted in 5.9% for CH3Cl, 8% for CHCl3 and 7.8% for CH3Br. "

P1165 L17: Reply: Information on the range used for the linearity check is added to paragraph 2.3 Calibration: "(CH3Cl 1.9-1900 pmol mol-1, CHCl3 0.14-470 pmol mol-1, CH3Br 0.5-25 pmol mol-1). It should be noted that the CHCl3 mixing ratio in the calibration gas was significantly higher than in the ambient air samples ".

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P1166 L8: Reply: We agree with the reviewer it is indeed very difficult to determine the boundary layer height. In order to determine the convective boundary layer height over the rainforest as a function of time of day we have used inversions (fluctuations) in the static air temperature measured on-board from the climbing aircraft. The lowest identified inversion was considered to be the top of the mixing layer, and the values obtained in this way are in good agreement with previously reported heights. The method is described in detail by Eerdekens et al. (2008), (paper in preparation for submission to ACPD - GABRIEL Special Section). In order to obtain a tractable estimation of the uncertainty of the averaged mixed layer height in this study, the standard deviation (2 sigma) was used. This approach results in a slightly higher number +-260 m instead of +-100 m used in the original calculations. The resulting higher flux uncertainty is partly outweighed by the ”new ” lower mixing ratios uncertainty. The end result is that there is no big change in the overall uncertainty of the calculated fluxes. To clarify this we modified the last paragraph of chapter ”3.1 Meteorological conditions”: ”Often there are a number of inversions present in the vertical profiles, and for this study we assumed that the lowest identifiable inversion delineates the top of the boundary layer. The derived boundary layer grew from around 600 m around 9:30 local time (UTC-3 h) to 1200 m at 12:30 stabilizing around 1400 m in the afternoon. The empirically determined mixed layer height seems to be in agreement with the investigations by Krejci et al. (2005), who reported mixing layer heights from the same area in 1998. Because most of our measurements took place during midday or early afternoon, we considered an average boundary layer height of 1400 m. The variability in the boundary layer height determinations between 12:00 and 16:00 was used to derive an uncertainty in the boundary layer height, which was determined to be +-260 m (2 sigma). ”

P1166 L21: Reply: The sentence was removed from the text and relocated to the figure caption of Figure 2.

P1166 L23-24: Reply: We agree with the reviewer that this sentence is misleading. We changed that paragraph to: ”Maritime air masses were advected from the Atlantic

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Ocean and then over approximately 1000 km of pristine rainforest. The 10-day back trajectories of the boundary layer samples are shown in Fig. 3. These indicate that air arriving at the coast of French Guyana and Suriname was transported for the most part within the Southern hemisphere above the Atlantic Ocean. ”

P1167 L16: Reply: This statement was added to the text. ”The vertical distribution of CH3Br was quite scattered, which is in part due to the 7.8 % precision error. ”

P1167 L22: Reply: On reflection, we agree, the word strong was removed.

P1168 L24: Reply: The reviewer makes a good point and accordingly we changed the term ”ambient background ” to ”boundary conditions ”. Moreover this paragraph was shortened for clarity, in that we now only use the CH3Cl/CO ratio to exclude recent biomass burning events. This reduction does not change the overall sense but makes it easier to understand.

P1169 L9-18: Reply: Both reviewer 1 and 3 have questioned the interpretation of the vertical profile given in this section. It is clear from the new Fig. 4, in which the individual points are shown with error bars, that there are insufficient measurements in the vertical to support the detailed interpretation of vertical structure presented previously. We agree with the reviewer that the structure at 2.3 km is not statistically significant from the mixed layer. Therefore this interpretation of the vertical structure was removed from the script. As a consequence our interpretation of possible entrainment from an upper layer to the mixed layer needs to be changed. We removed the paragraph regarding entrainment from the text and replaced it with: ”Since the vertical profile showed no statistically significant mixing ratio changes in the transition zone between ML and the influence of entrainment from upper layers of air is not taken into account in the following calculations. ” To be able to investigate the vertical structure and the influence of entrainment/boundary layer ventilation thoroughly, we recommend collecting a higher density of samples particular in the 2 to 4 km region for future projects in this region’.

P1170: Reply: The paragraph regarding the TOL calculation was moved forward and

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complemented by some extra information on the trajectories and their uncertainty. See reply above on TOL/trajectory uncertainty.

P1170 L18: Reply: That’s why they were excluded from further calculation. ”Ambient background ” was changed to ”boundary conditions ” as discussed previously.

P1171 L13-14 Reply: We thank the reviewer for spotting this typo. It was changed to pmol mol-1. The temperature and pressure averages were calculated from all data points within the mixed layer. Since the top height was changed to 1660 m (including the uncertainty) the averages and their uncertainties changed. We added the information on the number of samples used to clarify this point: ”Where F is the flux (μg m-2 h-1), ΔMR/ΔTOL the linear regression slope (pmol mol-1 h-1), HML the mean mixing layer height (1400+-260 m (2 sigma) (n=11)), p the mean air pressure below 1660 m (941+-71 hPa (2 sigma) (n=34710)), M the molar weight (μg), R the gas constant (0.08314 hPa m3 K-1) and T the mean air temperature below 1660 m (296+-15 K (2 sigma) (n=33596)). ”

P1172 L22: Reply: We agree that this paragraph is not helpful for the argumentation. The discussion regarding the differences between the two kinds of detection is inconclusive. We therefore removed it from the manuscript. Nevertheless we don’t want to keep this information back from the reader; therefore we decided to give it in the sub-clause: ”although these results were obtained using a different measurement technique (GC-ECD). ”

P1173 L3-4: Reply: The reviewer is right. The Li study was conducted in a different region, but is mentioned for the sake of completeness. To address this point the following sentence was added: ”However, the latter value was obtained in a coastal area and might be influenced by coastal/marine production. It is included here for completeness, but is not expected to match the other vegetation focused studies. ”

P1173 L5-7: Reply: At that point of the text our intention was not to show a quantitative comparison, but to point to the general agreement on the existence of a ”land

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based, constant source in the tropics ". To point that out more clearly the sentence was changed to: "These findings support previous model results (Lee-Taylor et al., 2001; Yoshida et al., 2004 and Yoshida et al., 2006), which have postulated a land based, constant source of CH3Cl all over the tropics. "

P1175 L15: Reply: "Outweighed " was replaced by "reduced ".

P1175 L16-19:Reply: This statement is illustrated in a recent paper of A. Kerkweg, accepted for publication in ACPD (ACPD-2008-0095). The results of this study imply a 50% longer lifetime for CH3Br, i.e. 386 days instead of 255 days referring to the latest Scientific Assessment of Ozone Depletion (WMO, 2007). This "new " -by a factor of 1.5- longer lifetime implies that the sinks are also smaller by a factor of 1.5 compared to prior estimated. Applying this factor to the CH3Br global sinks estimates of 129 to 387 Tg/yr (WMO, 2003) results in a range of 86 to 258 Tg/yr. This is much closer to the given source range of 77 to 293 Tg/yr (WMO,2003).

P1175 L22: Reply: This paragraph applies to the different possible production pathways of the investigated halocarbons. Mentioning this detailed information in the introduction would rather distract the reader. Potential sources and sinks that might influence the net fluxes, as requested by reviewer 1, can be seen in Table 1. Therefore we prefer to leave this paragraph in the "Results and Discussion " chapter to maintain the context.

P1176 L8-12: Reply: We thank the reviewer for this valid point. We added the comparison to the "Results and Discussion " chapter as follows: "In comparison with the current best estimates of global tropical source terms, it is at the lower end of the range reported (0.82 to 8.2 Tg yr-1 from tropical plants, 0.03 to 2.5 Tg yr-1 from senescent/dead leaves and a sink of 0.1 to 1.6 Tg yr-1 by soil uptake). Nevertheless an emission of this order of magnitude would be the main global source for CH3Cl, exceeding the ocean, biomass burning and anthropogenic sources. It should be noted that the studies providing the basis for current estimates (Yokouchi et al. (2002) and Hamilton et al.

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(2003)) were restricted to the small scale (greenhouse or laboratory measurements), and therefore the large-scale approach applied here should be a more reliable assessment of the real global net flux from this ecosystem. When the CHCl3 flux results are extrapolated to all tropical ecosystems, the result is a net flux of 56 (+-23 2 sigma) Gg yr-1. This is between 5 and 10 % of the total sources, and as previously mentioned, it could be already incorporated in the soil source term. The global extrapolation of the lowest detectable CH3Br flux yields 17 (2 sigma) Gg yr-1. A source of this size would be a non-negligible contribution to the global budget, supplying between 6 and 22% of the total global sources.

P1176 L24: Reply: The inverse models referred to in the text assume an additional tropical source of 2.3-2.4 Tg/yr (Lee-Taylor et al., 2001) and 2.5 Tg/yr (Yoshida et al., 2006) to reproduce the global CH3Cl mixing ratios. Our calculated flux of 1.5 Tg/yr would account for more than half of it. In comparison with the very uncertain terms for the global tropical sources it is at the lower end of the range of reported fluxes (see reply above). This point was also addressed by changing ”missing ” into ”additional ” source, see above.

P1177 L5: Reply: The phrase was changed to ”airborne measurements ”.

P1177 L15-19: Reply: We thank the reviewer for this point. The respective paragraph in was changed to: ”No significant trend for CH3Br could be determined from these measurements. The tropical vegetation gross source postulated by an inverse model prediction (Warwick et al., 2006) is most probably reduced to a very small net flux by concomitant sinks like soil uptake and photolysis. Our measurements over the South American rainforest agree with global observations, and vertical gradients between the boundary layer and free troposphere appear to be small or absent. This supports the conclusion that tropical forest ecosystem is not a significant net global CH3Br source.”

Typing, grammatical and wording recommendations were obeyed.

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 1159, 2008.