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Atmospheric Chemistry and Physics Discussions

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

Anonymous Referee #3

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Review of "Halogenated organic species over the tropical rainforest" by Gebhardt et al.

General Comments: This paper uses 99 airborne measurements of methyl chloride (CH3Cl), chloroform (CHCl3) and methyl bromide (CH3Br) over South America to estimate the emission fluxes of these species from the rainforest ecosystem. The authors find that CH3Cl and CHCl3 accumulate in air traversing the rainforest, and they estimate that emissions from the global rainforest ecosystem are 1.5 +/- 0.6 Tg CH3Cl yr-1 and 55 +/- 22 Gg CHCl3 yr-1. The CH3Cl emissions may account for more than half of the current missing CH3Cl source. By contrast, CH3Br emissions from the rainforest ecosystem could not be detected. This paper is a substantial and relevant contribution to the literature. Emissions from rainforests are an area of controversy and interest for many trace gases, and measurements from this region are a particularly

useful addition to the literature. Here the authors have performed a generally careful and thoughtful analysis of 99 airborne whole air samples that were collected above Suriname, French Guyana and the Atlantic Ocean to arrive at the above conclusions. In general uncertainties have been nicely handled and assumptions have been clearly indicated. Overall the quality of the paper is very good, though several major and minor issues need to be addressed before the paper can be published, as well as other clarifications and grammatical issues to improve the paper's quality. These are listed below.

Specific Major Comments:

1162 (11) and 1175 (17): The statement about the methyl bromide's lifetime impacting the source-sink imbalance needs to be made quantitatively. I personally do not see that there is enough wiggle-room in methyl bromide's lifetime to be able to support a large budget closure (e.g. see J. J. Colman et al., 2001). If you believe this to be the case, please show the % deviation in methyl bromide's lifetime that you believe can occur, and then relate this to the quantitative amount by which it would then close the budget.

1167 (11): This interpretation uses words like "seemed" and "appears". It is important to know whether or not the variations with altitude are real, because they are the basis of the regional and global flux calculations that are presented. Use your statistics to show whether or not the trends are significant. Do the results change on a flight-by-flight basis? I would also like to see a figure that shows each individual data point vs. altitude, so we can see the scatter and what went into the Figure 4 plots. This will be especially useful for clarifying the LFT peaks for CH3Cl (e.g. 1167 (27)).

1169 (9-23): The arguments here need to be tightened. First, as stated above, it needs to be shown that the enhancement at 1-2 km was statistically significant. Next, how was it determined that the ML was influenced by entrainment of advected biomass burning air rather than another source? If biomass burning is the source, by how much

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would CHCl3 have been expected to increase? By a detectable amount? What other sources may have caused CH3Cl but not CHCl3 to increase? Do the back trajectories at this altitude give any clues? Also, the statement that "in this case the methyl chloride concentration at 2-3 km was higher than at the top of the boundary layer" does not make sense to me, based on Figure 4.

1176 (9-12): A more in-depth discussion of the CHCl3 results is needed. How do your results change our understanding of the contribution of CHCl3 to global Cl?

Specific Minor Comments:

Title: To more accurately reflect the scope of your paper and where the measurements were made, change the paper's title to "Halogenated organic species over the South American tropical rainforest".

In the conclusions you state very nicely and clearly that the derived fluxes are a net flux (1177, 8-10) and that they apply to the rainforest ecosystem (1177, 23-25). However this is not clear in the abstract and other parts of the text. These are important distinctions that need to be incorporated throughout the paper. For example, on 1160 (23), "global extrapolation" could mean all global forests, tropical forests or tropical rainforests. Be specific throughout the paper.

1160 (9): The definition of "transport times" as 1-2 days is too broad. There are many transport times in the atmosphere, especially through a vertical column like you are measuring. Specify what you mean.

1162 (20): When was the use of CHCl3 in pulp and paper production reduced (what year?).

1163 (14): Reference the statement that there is regionally prevalent deep convection. You may wish to investigate what the results from the July 2007 TC4 experiment (conducted in a similar region) have shown.

1164 (10): Is there a technical paper about the sampling system that you can refer the

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reader to? If not, discuss how you have tested the cans for leaks, artifacts, etc.

1166 (16): I think you have mis-used the word "stable". During the daytime the meteorological conditions will be unstable, so "stable" is confusing. Use "constant" or some similar word.

1166 (20): Why would surface friction cause the wind to veer?

1167 (27): Do you really mean the 2-3 km peak, or the maybe more pronounced 1-2 km peak? (see p. 1169). Please clarify.

1168 (4): It would be interesting to know where the back trajectories originated from for the biomass burning sample.

1168 (18): You might consider changing "no" to "almost no".

1170 (7-14): Because CH3Cl has a strong latitudinal gradient, your results should be directly compared to the lower latitudes (apples and apples) and not the global average (apples and oranges). In other words, it is not surprising that your values are higher than the global average, and so the presentation of this comparison should better reflect this. The same reasoning applies to CHCl3.

1170 (26): Rather than stating "probably influenced" by anthropogenic emissions, do you have other urban tracers that you measured that would confirm this?

1171 (21-22): There are too many significant figures for the mean pressure and temperature. The same comment applies to some of the linear fits cited in Figure 5.

1171 (26): Add a comment on how this lowest detectable CH3Br flux compares to global emissions.

1172 (1-3): It seems that this statement of "few measurement studies" has been applied to studies that determined local fluxes, but your measurements could also be compared to other South American studies, e.g. the vertical profiles and global flux estimates of Blake et al. (1996) that were cited earlier.

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1176 (1): Reference the statement that tropical forests account for nearly half of the global total.

1177 (5): An airborne flux measurement is something different than what you did (you took airborne mixing ratios, from which fluxes were subsequently derived). Change to "airborne measurements".

Table 1: A much more descriptive caption is needed for Table 1. Also, change "subtotal" to "total" throughout.

Technical Corrections:

There are several issues related to grammar and clarity that need to be addressed.

Once a chemical (e.g. "methyl chloride") has been introduced by its chemical formula (e.g. "CH3Cl"), use the chemical formula throughout the paper (e.g.1162 (5), etc.). The same applies to ITCZ.

Throughout, organize your references chronologically then alphabetically (e.g. 1168 (11); 1169 (2); 1172 (2-3); 1173 (29); 1174 (3-4)).

1162 (15): Papers published in 2003 are not really "recent" anymore.

Section 3.2: The verb tense switches back and forth from past to present in this section.

1171 (11-12): The use of dashes here is confusing because the first dash reads like a minus sign. Use brackets instead.

1171 (19): It's a small point, but try using "ug" instead of "g" and "pmol mol-1" instead of "mol mol-1", to be consistent with the rest of the paper.

1176 (15-19): This is a very awkward and confusing sentence. Try: "Taking into account the measurements of Scheeren et al. (2003) - which were carried out in the same region - and using the average flux of both studies, the..."

Please make the following changes to improve grammar and clarity:

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1161 (12): "and are the most";

1161 (16): "by 2050";

1161 (17): "to stratospheric";

1161 (25): "was its main source (WMO, 1999). However,";

1162 (4): "abundant than CH3Cl,";

1162 (10): "thusfar";

1162 (25): "1990, versus 25-29% in 2001, would";

1162 (29): "the addition"

1163 (1): "and the reduction of";

1163 (2): "a seasonally";

1163 (12): "Intertropical Convergence Zone (ITCZ)". Do not re-define on 1165 (24);

1164 (4): "inlet";

1164 (23): Replace "*" by "x";

1166 (23): "ARTINC429. During..." (so that Figure 3 isn't introduced twice);

1167 (4): "predominantly" (add an "n");

1168 (15): "showed that" (no comma);

1169 (4): "and 0.57" (no comma);

1169 (5): "...1996). The ratio measured here (2.77 (+/-0.64 2sigma) x 10-3) lies clearly..." (so that the ratio isn't introduced twice);

1169 (15): "mixed layer";

1170 (9): "mean value";

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1170 (16): For clarity change "data points" to "individual measurements";

1170 (19): "with black asterisks" (since there are four of them);

1173 (26): "CH3Cl as well as CHCl3 are";

1174 (5): "pectin is";

1174 (26): "forests";

1174 (26): "emissions of up to";

1174 (28): "report" (because "et al." is plural);

1175 (14): "uptake CH3Br (Table 1),";

1176 (3): "that are covered";

1176 (9): Omit "for methyl chloride and chloroform, respectively" (it's redundant);

1176 (15): "as a lower limit";

1177 (5): "and" (not "und");

1178 (4): "threefold";

1178 (18): I think you mean "top off". You could also try "enhance";

Figure 1 caption: "and black";

Throughout the paper many sentences lack commas, which makes the intent of the sentence difficult to understand on first reading. Please add commas after the following words:

1161 (12): troposphere,

1161 (17): loss,

1162 (2): sinks,

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1162 (10): "missing source",

1167 (2): height,

1168 (7): above,

1170 (25): French Guyana, Cayenne,

1171 (10): parameters,

1177 (16): observations,

1177 (26): yr-1,

1178 (13): species,

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Interactive Comment