

## ***Interactive comment on “New insight into the atmospheric chloromethane budget gained using stable carbon isotope ratios” by F. Keppler et al.***

### **Anonymous Referee #1**

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Review of "New insight into the atmospheric chloromethane budget gained using stable carbon isotope ratios" by F. Keppler et al., paper ACPD-2005-0094

### General Comments

This paper investigates the budget of chloromethane in the atmosphere using an isotopic mass balance approach, incorporating new measurements of kinetic isotope effects associated with several chloromethane sinks. The results suggest the existence of a large chloromethane source with a highly-depleted isotopic signature. The authors identify this with chloromethane emissions from senescent plants and leaf litter in the

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tropics and sub-tropics.

I consider this to be an interesting paper with potentially important conclusions regarding sources of atmospheric chloromethane. It is well-written and clearly presented, with good citation of related work. I found the analysis and results quite convincing. I think that this paper should be published in Atmospheric Physics and Chemistry.

Specific Comments (page numbers in the following sections refer to the PDF print version)

1. On pages 3904-3906 and later in paper: I was confused by the sign of the quantity Delta. In the first paragraph of page 3904, depletion relative to unburnt fuel was quoted as a negative quantity, whereas in the next paragraph, depletion relative to bulk biomass was a positive quantity. Are these consistent?

2. Page 3910, 2 lines before equation (5): based on Table 1,  $\delta^{13}\text{C}_{\text{known}}$  sources = -52.5 per mil only if the wetlands and rice sources are taken as having isotopic signatures of 0 per mil. Is this what the authors intended? If these two sources are completely omitted, or if they are assigned a more likely isotopic signature (say -50), the weighted sum is more like -53.3. The authors should clarify this. Would a value of -53.3 make any difference to the conclusions?

3. Page 3910, equation (5): Deriving this equation requires that  $\delta^{13}\text{C}_{\text{known}}$  sources is weighted versus only the known sources, while  $\Phi_{\text{mis}}$  is normalized by the sum of all sources, not just the known sources. I think it would be worth mentioning this.

4. In Figure 1, how is it possible to assign a fairly accurate isotope signature to "Unknown processes" when the known sources "Marine bacteria" and "Soil reactions" only have question marks?

Technical Corrections

1. Abstract line 22: instead of "...the bulk fraction of atmospheric  $\text{CH}_3\text{Cl}$ ..." I suggest

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- "...a major fraction of the source of atmospheric CH<sub>3</sub>Cl...".
2. Page 3902 lines 2-3: I suggest replacing "...atmospheric chloromethane..." with "...the source of atmospheric chloromethane...".
  3. Page 3902 lines 14: I suggest deleting the second occurrence of "for atmospheric CH<sub>3</sub>Cl".
  4. Page 3902 lines 16-22 and elsewhere in paper: Is it necessary to describe sources in terms of both Tg/yr and Gg/yr? For example, 370 Gg/yr is 0.37 Tg/yr, and a few lines later 0.18 Tg/yr is used.
  5. Page 3903 line 23: Again I suggest "major" rather than "bulk".
  6. Page 3904 line 18: Is "experimentally" perhaps a better term than "empirically"?
  7. Page 3906, line 10: I suggest replacing "89.9" by "90" as the other values are quoted to the nearest unit.
  8. Page 3910 line 7: "52,5" should be "52.5".
  9. Page 3910, line 18: "an highly" should be "a highly"
  10. Page 3911, line 1: "used in for" should be "used in"
  11. References: The Simmonds et al. (2004) citation in the text is not in the reference list.
  12. References: The Kalin et al. (2001), Dimmer et al. (2001), and Goldstein et al. (2003) references in the reference list are not cited in the text.
  13. Table 1: Is the range in delta\_13C a full range or an uncertainty magnitude, e.g. does the value "12" mean "+/-6" or "+/-12"?
  14. Table 3: The Scenarios should be labeled "A B C", not "1 2 3"

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