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Atmospheric Chemistry and Physics Discussions



# *Interactive comment on* "Carbon and hydrogen isotopic ratios of atmospheric methane in the upper troposphere over the Western Pacific" *by* T. Umezawa et al.

**Anonymous Referee #1**

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Review of the paper by Umezawa et al. entitled: Carbon and hydrogen isotopic ratios of atmospheric methane in the upper troposphere over the Western Pacific

This paper by Umezawa et al. presents and analyses an original dataset of methane and its stable isotopes ($\delta$13C and $\delta$D) acquired through commercial aircraft flights over the Western Pacific between 2005 and 2010. Such data are rather rare and can be useful to distinguish the different source types of atmospheric methane, the second anthropogenic greenhouse gas and a precursor of Ozone. In this sense the existence and the reporting of these data is highly valuable for the reduction of the uncertainties on the regional methane emissions and sinks, especially downwind of East Asia where

very few long-term surface measurements are available. An extra step, if the paper gets accepted in ACP, would be to make these data available for the research community as many experimental groups do now worldwide.

General comments

-At many places I find that the text lacks precisions and quantitative statements (see also specific comments). The introduction should be fed with more quantitative information (mean atmospheric CH4 concentration, global mean source, ...). The authors compare different time series of concentrations (CH4, $\delta$13C and $\delta$D) but hardly give any precise correlation coefficients except at the end of the paper for Killing plots. I strongly suggest to go a bit further than XX shows weak/tight negative/positive correlation with YY and calculate correlation coefficients. The isotopic signatures also require more attention and details. Ranges of methane emissions from the different regions quoted should also appear somewhere.

-The CH4+Cl reaction in the marine boundary layer is still a matter of debate. The fact that this reaction has little impact on $\delta$D and CH4 but stronger impact on $\delta$13C is very interesting. I suggest the authors push more the analyses on this topic. Can you give an order of magnitude of the global sink from Allen 2007 ? For the Pacific ? Do the authors see time periods or latitudinal zones where this reaction is detected in the reported data ? It is not clear why the lack of tropical intercept is an element consistent with the CH4+Cl reaction, please clarify. More (and more clear) explanations on this topic would be an improvement for the paper

Specific comments

P9036-l23-26 : please give mean concentration and ranges of global emissions.

P9037-l19 : “wide area” I suggest to add : ... at the cost of less precise individual measurements in regards to the high precision of surface observations”

P9038-l21 : “substantial amount...” : please be more precise here and give ranges of



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emissions from the different Asian regions from the litterature.

P9039-l3 : “characteristic signature”: please use isotopic signatures instead.

P9039-l3-5 : There are ranges for isotopic signatures. Please give ranges and not only one value per type of source.

P9039-l20 “almost” : Well looking at figure 1 almost is not always that close. What is the range of horizontally projected distance between UT and LT observations? It maybe more precise to quote this range instead of only almost. . .

P9040 : “about 1.7l” : what is the pressure of the filled flasks ? It may worth writing it in the text.

P9041-l2-5 : Why the authors did not use the international NOAA04 scale ? It would make comparison much easier. The authors should explain why there are still on their own scales.

P9041-l12-17 : It seems to me there is a contradiction here. NIES and TU agree with each other but round robin found different differences between TU & WMO and between NIES and WMO. Pleas clarify this confusing paragraph.

P9042-l15-18 : The reported drift is confusing to me. Is it a slow drift? Did it happen between two measurement periods? Please clarify this part.

P9043-l18 : “observed seasonal cycles. . .” how did you do that ? What stations/dataset did you use to make the choice of latitudinal bands for grouping your data ? Please be more precise here.

P9044-l4: suggested : original GLOBAL flux field

P9044-l9 : EDGAR P9044-l9: Which version of EDAGAR has been used? Is it EDGAR3.2?

P9044-l11 : I would use “prescribed” instead of “adopted”.



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P9044-l18 How the interpolating/extrapolating has been done for the years 2005-2010? The text is not precise enough on this.

P9044-l25 : at "32°-15°" : this phrasing is not clear. Maybe write For time series between 15 and 32 °N, . . .

P9045-l7 : weak negative correlation : please give a correlation coefficient

P9045-l7-9 : Which latitudes are you talking about ? Please clarify.

P9046-l21 : "opposite phase" : what is the correlation coefficient ? There are not always in phase opposition (e.g. second half 2006, eary 2008, ..). Please be more precise.

P9047-l8-9 : UT and LT phase opposition : it is not clear from the figure, please give a correlation coefficient and modify the text according to the computed value.

P9047-l16 : almost in phase : please give a correlation coefficient

P9048–l23 : negative correlation : please give a correlation coefficient

P9049-l1 : see general comment on CH4+Cl . Can you see the impact of this reaction in your data ?

P9049-l16-18 : see general comment on CH4+Cl. Please clarify why CH4+Cl is consistent with a profile without intersection between LT and UT.

P9052-l5-14: at this end of this paragraph quoting literature, please say whether your results are consistent with the reported studies.

P9053-l11-13 : It seems that the concentrations are underestimated after 2008. What do you mean by temporal change ? How do your global emissions change from one year to the next. I suggest that emissions may be underestimated in your emission scenario. Can you provide the global emissions used ? P9054-l10-11 : Not only biogenic source are depleted compared to the atmosphere. I suggest : These values



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suggest a mix of source with substantial contributions from 13C depleted sources such as biogenic emissions from rice paddies and livestock, . . .

P9055-l7-8 : Please explain why Stratospheric CH4 is isotopically depleted as compared to tropospheric one.

P9056-l2 : idem as P9054-l10-11

P9056 : Conclusion : I would mention a potential revolution in the field of methane isotope measurements which is the Quantum Cascade Laser instruments which may deliver continuous CH4, $\delta$13C and $\delta$D measurements in a near future. This would enhance the potential of isotopic observations to reduce the uncertainties on the present methane cycle.

Figures : figure 2 is a bit small. It is sometimes hard to see variations.

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Interactive comment on Atmos. Chem. Phys. Discuss., 12, 9035, 2012.