**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 #2

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General

The paper presents new methane data (mixing ratio, d13CH4, dD-CH4) from the upper troposhere over the Western Pacific but also from the lower troposphere. The data are valuable and are interpreted following known concepts: transport of air masses, source attributions, Cl-sink.

The precision of data is sufficient to resolve the expected signal with standard deviations for CH4 mixing ratios of around 2ppb, for d13CH4 of 0.08permil and dD-CH4 of 2.2permil. However, atmospheric signals that are within these errors should not be over-interpreted (see also specific remarks). Furthermore it would be beneficial for the

weight of the paper to note regression coefficients when comparing the time series in chapter 3.

With respect, one remark considering the GC-IRMS method: In the methodological paper by the lead author (DOI:10.2151/jmsj.87.365) it is described that the isotopic results are independent of the amount of injected air (above a certain threshold) while CH4 mixing ratio and matrix are kept constant. However, in this study the amount of sample was kept constant, but the CH4 mixing ratio changes. I am aware that it is difficult to assess this issue, however I want to point to the fact that matrix effects / mixing ratio effects could influence isotope numbers which is not ruled out by the authors. I suggest to mention this issue in the paper or even better assess it experimentally.

In some parts of the manuscript structure and English language could be improved to result in a more concise paper.

Specific

p9042: Point out that the "test gas" is basically your reference that you use to calibrate d13CH4 and dD-CH4 to the international scales (VPDB and VSMOW). Give the information how the isotope values of "test gas" have been determined in the first place. This is important as the pure CO2 and H2 used as working standard gas are not suitable for referencing ("identical treatment" - principle: Werner, R. A. & Brand, W. A. Referencing strategies and techniques in stable isotope ratio analysis Rapid Communications in Mass Spectrometry, 2001, 15, 501-519) L22: Tell us which conclusions you drew following the Bock 2010 reference? Considering pyrolysis conditions you could additionally cite: Brass, M. & Röckmann, T. Continuous-flow isotope ratio mass spectrometry method for carbon and hydrogen isotope measurements on atmospheric methane Atmos. Meas. Tech., Copernicus Publications, 2010, 3, 1707-1721

P9047 L20-p9048 L2: Maybe it would be beneficial to elaborate a bit more on the mentioned seasonally varying air transport in this section.

P9051 L15-22: considering the analytical precision of 0.08permil for d13CH4 and 2.2permil for dD these numbers should be considered constant.

P9053 L26 to p9054 L 5: There is no significant difference for CH4 mixing ratios (difference = 23 and 31 ppb), d13CH4 (difference 0.12 and 0.14permil) nor dD-CH4 (difference 3.0 and 3.2 permil) from both South/East Asia compared to baseline. This should be mentioned. p9054 L6 et seqq: Consequently, the Keeling approach will give a similar result for all source areas. The end member for baseline should be given for a comparison, to make this clear to the reader.

Technical

p9044 L9: EDGAR instead of EDGER

p9064 L22: Aircraft instead of Aircrat

p9052 L24: after UT: mention (Table 1 and Figure 1).

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

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