

***Interactive comment on “Interpreting methane variations in the past two decades using measurements of CH<sub>4</sub> mixing ratio and isotopic composition” by G. Monteil et al.***

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**1 Answers to major comments:**

1.1 Chlorine Sink:

Keith Lassey suggests that we take into account a possible chlorine marine boundary layer sink in our model.

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As explained in answer to referee 1 comments, a stronger KIE in the global average CH<sub>4</sub> sink would imply a different initial budget (possibly a stronger geological source and a weaker wetland source), but our conclusions on the temporal trends would remain the same. This is illustrated in Figure 1 of this document, where we varied the base KIE of the CH<sub>4</sub>+OH reaction in the discussion scenario RP2 (see answer to referee 2 comment for details about the RP2 scenario) from 1.0054 (blue), to 1.0154 (red). This results in an offset of the simulated  $\delta^{13}\text{C-CH}_4$  time series, but the temporal trends are not affected. Discussion scenario RP2 was chosen because it is the most extreme scenario, in terms of variations of the apparent KIE of the methane sink.

As explained in answer to referee 2, we purposely chose a KIE on the high end of the reported range for the CH<sub>4</sub>+OH reaction, in order to minimize the difference with the apparent KIE reported by Allan et al. (2005).

## 1.2 Geological emissions:

Keith Lassey is questioning our implementation of the geological sink. He is in particular suggesting that the emissions from terrestrial mud volcanoes should be higher.

The source strengths for our geological emissions used in our study (mud volcanoes + ocean source) falls well within the range proposed by Etiope and Klusman (2002), although the distribution between terrestrial (mud volcanoes + micro-seepages), on shore and deep water mud volcanoes is different. There are a lot of uncertainties associated with the isotopic signatures of the geologic source (Etiope et al. (2007) reports  $\delta^{13}\text{C}$  ranges from -73‰ to -34‰, for terrestrial mud volcanoes, in Italy), therefore, although it is hard to tell whether it is optimal, we think that our implementation of the geologic source is realistic. We updated the reference from Etiope and Klusman (2002), to Etiope et al. (2008), and added the reference to Etiope et al. (2007) for isotopic signature of mud volcanoes.

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### 1.3 Neef et al. (2010) paper:

Keith Lassey is asking why we didn't cite the paper of Neef et al. (2010).

This paper mostly deals with CH<sub>4</sub> on different time scales than our study. We added a citation in the introduction of the revised manuscript.

## 2 Answers to minor comments:

- Page 6773, lines 10-11: The reference was changed.
- Page 6779, lines 4-10: What we meant here is that the wetland and biomass burning sources were used to close the CH<sub>4</sub> and d13C-CH<sub>4</sub> budgets: in other words, their global emissions don't come from published inventories, but follow from the other terms in our budget. The values of 182 TgCH<sub>4</sub>/year for wetlands and 30 TgCH<sub>4</sub>/year for biomass burning are the values used in our model. The paragraph was modified to be more clear.
- Page 6780, line 20: The reference was modified to take the comment into account
- page 6782: the value of the multiplier H described in the paper is very model-specific. It depends in particular on the model resolution. Therefore we didn't find necessary to mention its value (2.781 TgCH<sub>4</sub>/ppb) in the manuscript.
- page 6783, lines 10-11: The paragraph was slightly modified to take the remark into account.

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## References

- Allan, W., Lowe, D. C., Gomez, A. J., Struthers, H., and Brailsford, G. W.: Interannual variation of  $^{13}\text{C}$  in tropospheric methane: Implications for a possible atomic chlorine sink in the marine boundary layer, *J. Geophys. Res.*, 110, doi:10.1029/2004JD005650, 2005.
- Etiopie, G. and Klusman, R. W.: Geologic emissions of methane to the atmosphere, *Chemosphere*, 49, 777–789, 2002.
- Etiopie, G. and Milkov, A. V.: A new estimate of global methane flux from onshore and shallow submarine mud volcanoes to the atmosphere, *Environmental Geology*, 46, 997–1002, doi: 10.1007/s00254-004-1085-1, 2004.
- Etiopie, G., Martinelli, G., Caracausi, A., and Italiano, F.: Methane seeps and mud volcanoes in Italy: Gas origin, fractionation and emission to the atmosphere, *Geophys. Res. Lett.*, 34, doi:10.1029/2007GL030341, 2007.
- Etiopie, G., Lassey, K. R., Klusman, R. W., and Boschi, E.: Reappraisal of the fossil methane budget and related emission from geologic sources, *Geophysical Research Letters*, 35, 1–5, doi:10.1029/2008GL033623, <http://www.agu.org/pubs/crossref/2008/2008GL033623.shtml>, 2008.
- Neef, L., van Weele, M., and van Velthoven, P.: Optimal estimation of the present-day global methane budget, *Global Biogeochemical Cycles*, 24, doi:10.1029/2009GB003661, <http://www.agu.org/pubs/crossref/2010/2009GB003661.shtml>, 2010.

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Interactive comment on *Atmos. Chem. Phys. Discuss.*, 11, 6771, 2011.

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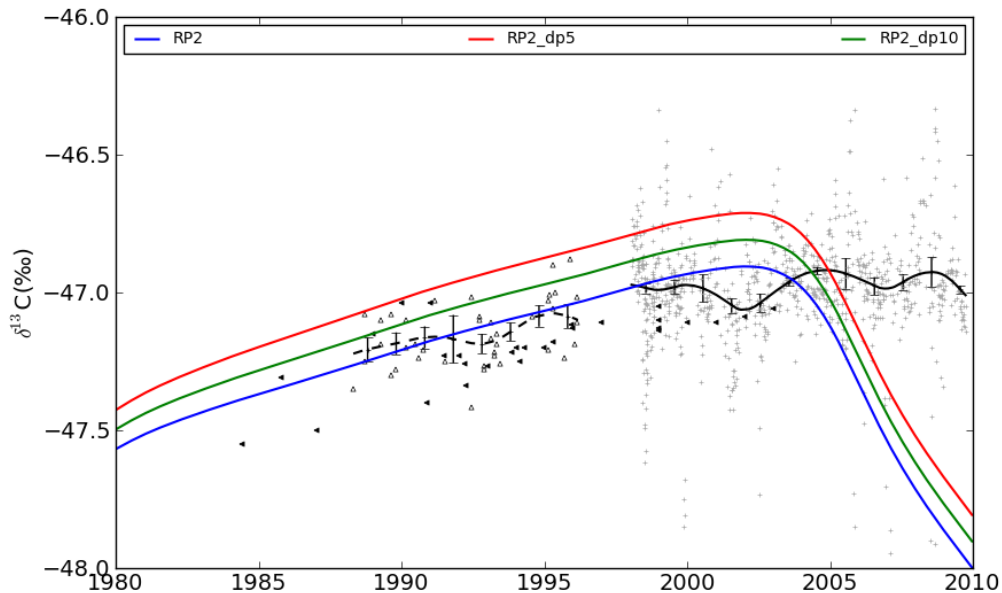
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**Fig. 1.** Methane isotopic ratio in scenario RP2, with different CH<sub>4</sub>+OH rate coefficients.

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