

Interactive comment on “Interpreting methane variations in the past two decades using measurements of CH₄ mixing ratio and isotopic composition” by G. Monteil et al.

Anonymous Referee #2

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Monteil et al. have done a suite of model simulations of atmospheric CH₄ and 13C/12C isotopic ratios in methane to evaluate different scenarios against the observed atmospheric growth rate of these species over recent decades. This manuscript makes use of excellent atmospheric datasets to explore a topic of great interest to the readership of Atmospheric Chemistry and Physics. However, I would appreciate it if the authors would please address the questions and concerns raised below prior to publication.

In particular, I am curious about the authors' use of the atmospheric methane and methane isotope data. In section 2.2.4, they discuss observations from three

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NOAA/INSTAAR stations and two NIWA stations. The methane isotope measurements are then extended using observations from Quay et al. over the 1990's and ice core data from Ferretti et al.

The authors note briefly in the methods section that there may be internal inconsistencies between these data sets, but they do not go into any details about how large offsets between these datasets might be and how much they might influence trends in the data that they are trying to match with the model scenarios. Does the smoothed curve at Cape Grim, which has a three-year gap between two different data sources, look similar to the smoothed curve at Baring Head and Arrival Heights, which don't have a gap over this period? It is difficult for me to determine this from the plots shown. Given the author list of this paper, I have a high degree of confidence that these issues have been given careful consideration during the analysis. However, I would like to see some discussion of them in the manuscript.

Also, it seems odd that the authors focus on Cape Grim for figures 3 and 4 when the longest running stations (Baring Head and Arrival Heights, which run from 1991-present) have records that are long enough to provide a reasonable test for the model simulations without combining datasets that may contain offsets. Similarly, five stations are discussed in the text, but only four are shown in the figures.

There is little discussion of uncertainty in the isotopic signatures of the sources or isotopic fractionation of the sinks. How big is the range in the reported values of these quantities? Would it be appropriate to include scenarios evaluating the impact of changing isotopic signatures?

The authors do not include a scenario to test for a Cl sink in the marine boundary layer, which has already been mentioned by Keith Lassey in his short comment in response to this paper. This is particularly relevant because even a relatively modest Cl sink would have a substantial impact on the 13C/12C ratio in methane due to the strong isotopic signature, and because the Cl sink may have shifted considerably over the

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period being considered by Monteil et al (Allan et al., 2007). Monteil et al., conclude that two possible scenarios may lead to an accurate representation of the methane and methane isotope growth rate: an increase in methane oxidation by OH or a decrease in wetland emissions. I'm curious about whether the authors have looked at how well these two scenarios are able to reproduce the seasonal cycles of these species at the observing stations? I suspect this may help to narrow it down to one scenario.

Minor issues:

Quite a few people who are interested in the methane growth rate over recent decades are much less familiar with the $^{13}\text{C}/^{12}\text{C}$ isotopic signatures of the methane sources and the fractionation of the sinks. It would be nice to include a paragraph generally outlining the roles of the source and sink processes in enriching or depleting the atmosphere in ^{13}C in the introduction.

P. 6773, second paragraph. While everything said here is accurate, I'm concerned that switching between discussing the growth rate and concentration could lead to confusion. For example, while the methane growth rate over the 1990's was decreasing, the atmospheric mixing ratio was still increasing. However, since the authors have just contrasted slowing growth rate in the 1990's with the "period of continuous increase of concentrations during the 20th century", it could give the impression that concentrations were decreasing over the 1990's.

P.6773, first paragraph. In the stratosphere, methane is also destroyed by Cl and O(1D), as discussed later in this manuscript.

P. 6773, third paragraph. Bousquet et al. also made some use of the ^{13}C isotope data, which would be appropriate to mention here.

P. 6774, second paragraph. The Bousquet et al. (2006) paper cited elsewhere in this paper covers over 20 years. Also, Fiore et al. (2004) looks at trends over more than a decade.

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P. 6776, first paragraph. It would be nice to give a one-sentence overview of the Velders (1995) model here.

P. 6777, last paragraph. What about the Walter et al. (2001) wetland model, which suggests substantial natural wetland emission variability in response to changes in temperature and precipitation?

P. 6781, first paragraph. It would be helpful to include a table here showing the positions of the sites used and some details about what data sets are available for each of the five sites and what time periods these data span.

P. 6783, first paragraph. Dlugokencky et al., used the fine grid resolution of TM3 while Montiel et al. use the coarse grid version. These two model resolutions can behave quite differently from one another (see for example the back of the TM3 User's Manual by Heimann and Korner).

Table 1. Did the authors consider spatial variability in the isotopic signature of biomass burning due to whether C3 or C4 plants were being burned? Also, it would be useful to include some form of uncertainty or acceptable range in both the methane fluxes and isotopic signatures/sink fractionation.

Figure 1. It seems odd that the more recent atmospheric measurements are not shown here, where these datasets are combined in subsequent plots.

Figures 2 and 3. Missing a 'ts' at the end of 'Arrival Heights'

Unfortunately, I noticed quite a few grammatical errors and typos in this paper, which the authors should take care to correct prior to resubmission.

References:

Fiore, A. M., L. W. Horowitz, E. J. Dlugokencky, and J. J. West (2006), Impact of meteorology and emissions on methane trends, 1990–2004, *Geophys. Res. Lett.*, 33, L12809, doi:10.1029/2006GL026199.

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Walter, B. P., M. Heimann, and E. Matthews (2001), Modeling modern methane emissions from natural wetlands 2. Interannual variations 1982–1993, *J. Geophys. Res.*, 106, 34,207–34,220

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