Interactive comment on "Is Shale Gas a Major Driver of Recent Increase in Global Atmospheric Methane?" *by* Robert W. Howarth et al.

Anonymous Referee #3

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Manuscript Summary This manuscript attempts to re-examine the role of fossil emissions in the recent rise of atmospheric methane. This is an important topic that has been the subject of vigorous scientific debate. The author argues that emissions from shale gas are considerably more depleted in 13C than conventional gas, which should change the estimates of relative contributions from different methane sources in an isotopic mass balance calculation. Explicitly including shale gas in the isotopic analysis and making some assumptions about the shale: conventional gas ratio, the author arrives at revised estimates for contributions from fossil (higher than prior estimates) and biogenic (lower than prior estimates) sources to the recent methane increase.

Overall Evaluation: Explicitly including information about the d13C signature of shale gas into an isotope mass balance calculation to re-assess today's global methane bud-

get seems like a useful contribution, if it has not been done already. However, unfortunately this study contains a major flaw in the isotope mass balance analysis that invalidates its results.

Specific Comments: 1. The approach of comparing methane source isotopic signatures directly to the mean global atmospheric d13CH4 value (graphically illustrated by Fig 3 in the manuscript and used in Equation 1) is conceptually flawed. The mean atmospheric d13CH4 is NOT the same as the emission-weighted sum of source d13C signatures. The reason for this is that there is a relatively large isotopic fractionation associated with the atmospheric sink of CH4, with 12CH4 being removed faster than 13CH4. This results in a $\sim$ 6 to 7 per mil enrichment of mean d13C of atmospheric methane with respect to the mean d13C of global methane emissions. To put it another way, for a steady-state mean atmospheric d13CH4 of -47.2 per mil (value during the pause in atmospheric CH4 rise in early 2000s), the mean d13C of global CH4 emissions is $\sim$ -53.7 per mil. This is an effect that has been known for a very long time and is incorporated into all recent atmospheric methane budget analyses that use d13C, including the Schwietzke et al. (2016, Nature) and Schaefer et al. (2016, Science) papers the author cites. When this atmospheric fractionation effect is taken into account, even the much more negative (compared to conventional gas) d13C value of -51.4 per mil proposed by the author for shale gas is still higher than mean d13C of global emissions. Increases in emissions from shale gas would therefore still drive atmospheric d13CH4 up, not down. The author needs to re-do their analysis using a proper isotopic mass balance approach that incorporates the CH4 sink fractionation and also accounts for the fact that methane in the atmosphere today is not in steady-state. Examples of such calculations are described in detail in the supplements to the Schwietzke and Schaefer papers mentioned above, among others.

2. Equation 1 is a strange approach to isotopic mass balance, and it is difficult to judge whether or not it is correct (the issue above aside). The author should either provide a detailed derivation of their form of this equation to illustrate why it's valid or use a

more conventional isotopic mass balance approach – again see the Schwietzke and Schaefer papers for examples.

3. Page 3, line 25. How representative is this d13C value (which seems to be based on a limited number of measurements and sites) of the cumulative shale gas emissions? Is this a simple arithmetic mean? Is it possible to estimate an emissions-weighted mean (which would be more appropriate for an isotope mass balance calculation)? The 95% confidence limit stated seems very narrow to me.