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

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Author response to anonymous review #1 (as of 9 May 2019):

I thank the reviewer for their comments and suggestions, which I will carefully consider as I revise my manuscript. Below are my preliminary responses.

Major Criticism – choice of value for δ13C content of methane from shale gas: The greatest criticism of the reviewer is my reliance on the studies reviewed by Golding et al. (2013) for the estimate of the δ13C content of methane from shale gas. My preliminary responses to this are:

1) the reviewer is correct that the 13C value used for the shale gas methane is critical to my analysis. However, I believe I have fairly and accurately reported representative

data in my discussion draft, albeit from an admittedly small set of studies. Nonetheless, as I revise the manuscript, I will consider further data as appropriate;

2) note that I submitted my manuscript under the "Ideas & Perspectives" category and not as a regular research paper. My intent was to draw attention to the failure of earlier papers that evaluated changes in atmospheric methane using the δ13C approach to explicitly consider that shale gas may differ from conventional natural gas, but I recognized the limited data behind my re-evaluation;

3) in that context, one of the major conclusions of my manuscript (lines 18-21, p. 8) was that ". . . we note an important caveat: our analysis of emissions with explicit consideration of the δ13C value for methane in shale gas is based on a small data set, only 61 samples in 3 studies. A clear priority should be to gather more data on the 13C content of shale-gas methane."

4) for the δ13C data I used, I chose the studies reviewed in Golding et al. (2013) because these were highlighted by Schwietzke et al. (2016) (one of the key earlier papers on changes in the global δ13C value of methane) as representative for shale gas. Note that Schwietzke et al. (2016) did not include these shale gas data (either explicitly as distinct from conventional natural gas or as part of their overall estimate for fossil fuels), stating in their supplemental materials: "Note that δ13CFF in this analysis excludes shale gas methane because the share of these sources to global total NG production increased from only 3% to 9% between 2007 and 201316." Reference #16 is to Golding et al. (2013). As an aside, I believe the important context for looking at changes in methane over the past decade is not the increase from 3% to 9% of total natural gas production, but rather the fact that "shale gas accounted for 63% of the global increase in all natural gas production between 2005 and 2015 (EIA 2016, IEA 2017)" as stated in my manuscript (lines 29-30, p. 4).

5) I welcome new data on the δ13C value for shale gas, and I thank the reviewer for pointing out the Tilley & Muehlenbachs paper. However, the Tilley & Muehlenbach

paper appears to report data both for actual shale gas (that is, the methane trapped in shale which is released by high-volume hydraulic fracturing) and for gas trapped in reservoirs that has migrated from the shale over geological time scales (which would be considered "conventional" gas in my manuscript). Before I revise my manuscript, I will consult with experts to ascertain which of the data in the Tilley & Muehlenbach paper might be relevant for my analysis (that is, those representing actual shale gas that would be developed from high-volume hydraulic fracturing).

Major Criticism – Tilley & Muehlenbach support no difference between shale and conventional gas: In the final paragraph of their General Comments, the reviewer states "... Tilley et al. identify three common maturation stages of shale gas systems and point out that the so called rollover zone may represent the peak of high productivity shale gas. The rollover zone roughly corresponds to δ 13C-methane values between -45‰ and -35‰ for the analyzed cases. This would support the implicit assumption of Worden et al. (2017) that the isotopologue signature of shale and conventional gas is similar." I disagree that the language in Tilley & Muehlenbach supports the implicit assumption of Worden et al. (2017) that shale gas and conventional gas are indistinct in their 13C content. Rather, I believe the preponderance of evidence indicates that shale gas is more depleted in 13C than is conventional gas. Specifically:

1) I find the reviewer's argument speculative, and note that Tilley & Muehlenbach (2013) state "..... the mechanism that created the reversed gases is still not well understood and is controversial."

2) as noted above, Tilley & Muehlenbach are reporting data both for actual shale gas (as defined in my paper) and for conventional gas that has migrated from shale gas (which I would expect to be more enriched in 13C; see point #4 below);

3) in contrast to the argument made by the reviewer, some of the sites with the most negative δ13C-methane values presented in the Tilley & Muehlenbach paper occur in the Marcellus shale; the Marcellus shale has been the largest shale-gas play in the

world over the past decade and has accounted for more than 25% of all shale gas production globally over this time period (EIA 2018). Further, these 13C-methane data for the Marcellus shale as reported in Tilley & Muehlenbach are apparently for methane that has migrated from the shale to a reservoir. If so, this methane is likely to be more enriched (heavier, less negative δ13C) than the actual shale gas (see point #4 below);

4) as I wrote in my discussion paper (line 30 on p. 3 through line 4 on p. 4, and Fig. 2-A), one should expect that methane that migrates out of shale and is trapped in reservoirs would be more enriched in 13C than the methane that remains in the shale: some of the methane is consumed by bacteria as it migrates, with fractionation by the bacteria causing this enrichment.

Specific Comments

"Page 1, Lines 11-12: The statement that shale gas is depleted in 13C relative to the atmospheric mean is not supported by sufficient evidence (see also general comments)."

Please see my detailed responses to the first "major criticism" above.

"Page 3, Lines 18-19: There are also several studies (Tilley et al., 2013 and references therein) which do not support this statement. Moreover, the cited paper of Bottner et al. assumes a value of -47.3‰ for shale gas, which is larger than the -51.4‰ value used for the presented analysis and comparable to the average atmospheric δ13C-methane during the 2000-2008 period."

There are two parts to this criticism. With regard to whether or not the Tilley & Muehlenbach paper supports my statement, please again refer to my detailed responses above to major criticism #1. Regarding Botner et al. (2018), that paper clearly states that the methane in shale gas is likely to be more depleted in 13C relative to conventional natural gas, as I stated in my discussion manuscript (line 18 to 19, p. 3). Yes, their shale gas estimate is heavier than the mean I used in my analysis (again, based on the papers reviewed in Golding et al. 2013) but is not statistically outside of the range I used.

Please note that before I submitted this manuscript to Biogeosciences, I consulted with a co-author of the Botner et al. (2018) paper (Amy Townsend-Small), sharing with her a draft of what I submitted.

“Page 6, Lines 16-19: Apart from the question of representativity of the presented analysis, is the derived difference to the Worden et al.(2017) estimates really significant? I find it hard to believe that the uncertainties of this study presented in Table 1 are that small although it is ultimately a reweight of the Worden et al. estimates. Are the prior uncertainties from Worden et al. considered correctly?”

The uncertainty for the Worden et al. estimates presented in my Table 1 come from that paper. For my new estimates in Table 1, the uncertainty is estimated as stated in the footnote: “Confidence bounds for estimates for shale gas, conventional natural gas, and biogenic sources are calculated using Eq. (9) and the upper and lower 95% confidence limits for the δ13C values shown in Figure 3-B.” I leave it to the reader to decide whether these new estimates are significantly different from those presented in Worden et al. (2017). There certainly is at least a trend, with my estimates strongly suggesting greater emissions from fossil fuels and smaller biogenic emissions.

One take home point for my discussion manuscript – which remember is in the “Ideas & Perspectives” category and is not a traditional research paper – is that small changes in assumptions can lead to major changes in the conclusions from these global analyses. Worden et al. (2017) showed that decreases in biomass burning over the past decade (Schaefer et al. 2016 and Schwietzke et al. 2016 had assumed a constant rate of biomass burning) qualitatively changed the conclusion on trends in fossil-fuel emissions: rather than fossil fuel emissions going down over the past decade by some 18 Tg/yr (Schwietzke et al. 2016, and my Table 1), they more likely increased by 15.5 Tg/yr (plus or minus 3.5 Tg/yr), according to Worden et al. (2017). My inclusion of a consideration that shale gas methane may be more depleted in 13C than the methane in conventional gas further increases the estimate for the change in emissions from fossil fuels, to a mean estimate of 20.4 Tg/yr greater emissions (my Table 1). There are

large uncertainties associated with any of these estimates, but the directional change in estimates from changes in assumptions is very clear.

“Page 7, Lines 30-31: This sentence creates the impression that the methane emissions in the Bakken shale are steadily increasing. However, it seems that after years of considerable increase (Schneising et al., 2014), emissions have been reduced again (Peischl et al., 2018).”

The time of reference for my analysis, following that of Worden et al. (2017), was 2005 to 2015. The Schneising et al. (2014) reference is quite pertinent to that time frame, as they compared 2006-2008 to 2009-2011. Nonetheless, as I revise, I will note that some evidence suggests emissions in Bakken shale may have decreased over the past few years (Peischl et al. 2018).

“Conclusions: The advice to move as quickly as possible away from natural gas based on this study does not appear sufficiently conclusive for the reasons mentioned above. A thorough analysis of the impact of shale gas and the adequacy of natural gas as a bridge fuel is highly desirable, but to draw such strong conclusions based on a small data set, which likely lacks representativity, is premature.”

I appreciate this comment, but again, my contribution is as an ““Ideas & Perspectives” piece. To date, previous analyses that used the change in the δ13C value for global methane over time have completely ignored the possible role of shale gas, even though shale gas was 63% of the total global increase in natural gas over the past decade. I do not view my manuscript as a final statement: it is a call to pay better attention to this potentially critical aspect of the story as to what is driving the increase in global methane emissions. And I note that my conclusion is qualified, as stated above: “an important caveat: our analysis of emissions with explicit consideration of the δ13C value for methane in shale gas is based on a small data set, only 61 samples in studies. A clear priority should be to gather more data on the 13C content of shale-gas methane” (lines 18-21, p. 8).

Both Schaefer et al. (2016) and Schwietzke et al. (2016) in very high profile papers (Science and Nature) concluded that methane emissions from fossil fuel sources had declined over the past decade, even though Schaefer et al. (2016) noted how surprising this conclusion was in the context of the massive global increase in unconventional (shale) oil and gas development. My analysis suggests a critical element these authors may have missed in their analyses. To point out this omission is overdue, in my opinion, not premature.

Technical Corrections I thank the reviewer for these technical corrections.

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