

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 #3 (3 June 2019):

I thank the reviewer for their comments, and I am glad they agree it is useful to explicitly consider shale gas in using data on the ^{13}C content of methane to evaluate methane emission sources over time. Below I respond to each comment.

“Overall Evaluation: Explicitly including information about the d^{13}C signature of shale gas into an isotope mass balance calculation to re-assess today’s global methane budget 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.”

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No previous work has explicitly considered the ^{13}C signal of shale gas when modeling changes in atmospheric sources of methane. In fact, Schwietzke et al. (2016) specifically excluded shale gas from their study. As for the flaw, I disagree that this is a fatal problem, although I do agree it should be addressed. See further discussion below after “specific comments: 1.”

“Specific Comments: 1. The approach of comparing methane source isotopic signatures directly to the mean global atmospheric $\text{d}^{13}\text{C}_{\text{CH}_4}$ value (graphically illustrated by Fig 3 in the manuscript and used in Equation 1) is conceptually flawed. The mean atmospheric $\text{d}^{13}\text{C}_{\text{CH}_4}$ is NOT the same as the emission-weighted sum of source d^{13}C signatures. The reason for this is that there is a relatively large isotopic fractionation associated with the atmospheric sink of CH_4 , with $^{12}\text{CH}_4$ being removed faster than $^{13}\text{CH}_4$. This results in a ~ 6 to 7 per mil enrichment of mean d^{13}C of atmospheric methane with respect to the mean d^{13}C of global methane emissions. To put it another way, for a steady-state mean atmospheric $\text{d}^{13}\text{C}_{\text{CH}_4}$ of -47.2 per mil (value during the pause in atmospheric CH_4 rise in early 2000s), the mean d^{13}C of global CH_4 emissions is ~ -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 d^{13}C , 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) d^{13}C value of -51.4 per mil proposed by the author for shale gas is still higher than mean d^{13}C of global emissions. Increases in emissions from shale gas would therefore still drive atmospheric $\text{d}^{13}\text{C}_{\text{CH}_4}$ up, not down. The author needs to re-do their analysis using a proper isotopic mass balance approach that incorporates the CH_4 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.”

I thank the reviewer for their very clear explanation of why I should have used a

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weighted mean value for the methane entering the atmosphere, and not the average value for the current atmospheric methane. In the revised manuscript, I have followed their guidance. In the revision, I now have written “The average $\delta^{13}\text{C}$ ratio for methane in the atmosphere in 2005 was - 47.15 o/oo (Schneising et al. 2016), which reflects a flux-weighted mean input of methane with a $\delta^{13}\text{C}$ ratio of - 53.5 o/oo. This flux-weighted mean value is approximately 6.3 o/oo more depleted in ^{13}C because of fractionation during the oxidation of methane in the atmosphere (Schneising et al. 2016; Sherwood et al. 2017). In our analysis, we use this flux-weighted mean value of - 53.5 o/oo.”

Note that the effect of this change on my calculations and conclusions is not large: the important part of the analysis is that the shale gas is more depleted in ^{13}C than are the values used for fossil fuels by Schaefer et al. (2016), Scwietzke et al. (2016), and Worden et al. (2017).

"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."

Reviewer #2 also expressed concern over my equation 1. In response, I have completely modified the approach and supporting language. The new text reads:

“To explore the contribution of methane emissions from shale gas, we build on the analysis of Worden et al. (2017). Figure 3-A shows the $\delta^{13}\text{C}$ values used by them as well as their mean estimates for changes in emissions since 2008 (as they estimated using the $\delta^{13}\text{C}$ data of Schwietzke et al. 2016). Figure 3-A represents a weighting for the change in emissions (y-axis) and the $\delta^{13}\text{C}$ values of those emissions (x-axis) by individual sources. Our addition is to separately consider shale gas emissions, recognizing that methane emissions from shale gas are more depleted in ^{13}C than for

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conventional natural gas or all other fossil fuels as considered by Worden et al. (2017). For this analysis, we accept that net total emissions increased by 24.7 Tg per year (\pm 14. Tg per year) since 2007, driven by an increase of \sim 28.4 Tg per year for the sum of biogenic emissions and emissions from fossil fuels and a decrease of \sim 3.7 Tg per year for emissions from biomass burning (Worden et al. 2017).

“We start with the Eq. (1) which explicitly considers methane emissions from shale gas:

$$(\text{BN} - \text{BW}) + (\text{FFN} - \text{FFW}) + \text{SG} = 0 \quad (1)$$

where BN is the estimate from Worden et al. (2017) for the increase in biogenic emissions of methane globally after 2007, BW is our new estimate for the increase in these biogenic fluxes, FFN is the estimate from Worden et al. (2017) for the increase in emissions of methane globally from fossil fuels after 2007, FFW is our new estimate for the increase in fossil fuel emissions after 2007 other than from shale gas, and SG is our estimate for emissions from shale gas after 2007. That is, the inclusion of an estimate for shale gas is matched by changes in the estimated fluxes from biogenic sources and other fossil fuels.

“Eq. (2) then reweights the information in Figure 3-A for the difference between most fossil fuels and shale gas, multiplying global mass fluxes for each source by the difference between the $\delta^{13}\text{C}$ ratio of each source and the flux-weighted mean for all sources:

$$(\text{BN} - \text{BW}) * \text{DB-A} = [(\text{FFN} - \text{FFW}) * \text{DA-FF}] + (\text{SG} * \text{DA-SG}) \quad (2)$$

where DB-A, DFF-A, and DSG-A are the differences in the $\delta^{13}\text{C}$ ratio of biogenic emissions, fossil fuels, and shale gas compared to the flux-weighted mean $\delta^{13}\text{C}$ ratio for all sources (A). The x-axis of Figure 3-B shows the $\delta^{13}\text{C}$ for each source; note that the y-axis is the estimate of the change in emissions for each of these sources that we derive below. Next, if we multiply both sides of equation 1 by DB-A and rearrange,

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$$(BN - BW) * (DB-A) = - [(FFN - FFW) * (DB-A)] - (SG * DB-A) \quad (3)$$

“Subtracting equation 3 from equation 2,

$$0 = [(FFN - FFW) * (DA-FF + DB-A)] + [SG * (DA-SG + DB-A)] \quad (4)$$

“Rearranging equation 4,

$$SG = - (FFN - FFW) * (DA-FF + DB-A) / (DA-SG + DB-A) \quad (5)$$

“Note that from Worden et al. (2017), FFN is 16.4 Tg per year.”

From here, the text closely follows that in the “discussion” manuscript, except using updated values in response to a comment from reviewer #1, and the change the use of the weighted mean value for methane entering the atmosphere (discussed under specific comment #1, above).

"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."

I have revised my choice of the d13C value for methane in response to reviewer #1, now using a value of – 46.9 o/oo, which is a weighted mean for three values from major shale-gas plays: the Bakken (North Dakota), Barnett (Texas), and Utica (Ohio). The value is more enriched than the - 51.4 o/oo value chose for my original submission; I now feel that value is too negative, and the organic-rich shales which were included there are probably not representative of most shale gas that has been developed over the past decade. Please see my detailed response to reviewer #1 (updated, 3 June 2019) for my logic.

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