

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

**Anonymous Referee #1**

Received and published: 3 May 2019

This manuscript takes up the widely discussed question of the origin of the renewed methane growth in recent years by analyzing the atmospheric  $\delta^{13}\text{C}$  methane isotopologue signature. As  $\delta^{13}\text{C}$  has decreased in recent years, several studies attributed the corresponding methane increase to biogenic sources, arguing that biogenic methane is more depleted in  $^{13}\text{C}$  than the average atmospheric value during the hiatus period at the beginning of the century, while methane from fossil fuels is not.

The author tries to revise previous estimates of the shares of biogenic sources and fossil fuels by explicitly distinguishing between shale gas and conventional gas claiming that the isotopologue signature from shale gas resembles biogenic gas more closely than conventional gas. Although the idea sounds promising at first glance, the representativity of the chosen shale gas plays for the entire sector is questionable and

C1

a revised and more comprehensive data set is needed to recommend publication in Biogeosciences and to support the far-reaching conclusions drawn by the author.

### **General Comments**

The main weakness of the analysis is that the  $\delta^{13}\text{C}$ -methane value of  $(-51.4 \pm 1.2)\text{‰}$  for shale gas is derived from a small data set from the review paper Golding et al.(2013) consisting of 61 samples in 3 studies, which is postulated here to be representative for the entire shale gas production sector.

In contrast to that, Tilley et al.(2013) present new and review former isotope data from several published papers including shale gas from the Barnett Shale, Fayetteville Shale, Marcellus Shale, Utica Shale and the Western Canada Sedimentary Basin. All of the various presented samples are less depleted than  $-51.4\text{‰}$  and almost all are even less depleted than  $-47.2\text{‰}$  which is the average  $\delta^{13}\text{C}$ -methane during the 2000-2008 period according to Figure 1B. Hence, these samples cannot explain the depletion of  $\delta^{13}\text{C}$  in the atmosphere since 2009 and do not point to a shale gas origin.

Furthermore, 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  $\delta^{13}\text{C}$ -methane values between  $-45\text{‰}$  and  $-35\text{‰}$  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.

### **Specific Comments**

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

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  $-47.3\text{‰}$  for shale gas, which is larger than the  $-51.4\text{‰}$  value used

C2

for the presented analysis and comparable to the average atmospheric  $\delta^{13}\text{C}$ -methane during the 2000-2008 period.

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?

Page 7, Lines 30-31: This sentence creates the impresssion 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).

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.

### Technical Corrections

Several instances: there is no space ahead of  $\delta^{13}\text{C}$ . Please check.

Page 4, Line 25: DA-CG should be  $D_{A-CG}$

Page 22, Line 7: Triangle indicates the average  $\delta^{13}\text{C}$ -methane...

### References

Tilley, B. and Muehlenbachs, K.: Isotope reversals and universal stages and trends of gas maturation in sealed, self-contained petroleum systems, *Chemical Geology*, 339, 194-204, <https://doi.org/10.1016/j.chemgeo.2012.08.002>, 2013.

Peischl, J., Eilerman, S. J., Neuman, J. A., Aikin, K. C., de Gouw, J., Gilman, J. B.,

C3

et al.: Quantifying methane and ethane emissions to the atmosphere from central and western U.S. oil and natural gas production regions. *Journal of Geophysical Research: Atmospheres*, 123, 7725–7740, <https://doi.org/10.1029/2018JD028622>, 2018.

Interactive comment on Biogeosciences Discuss., <https://doi.org/10.5194/bg-2019-131>, 2019.