

# Cornell University

Department of Ecology and Evolutionary Biology

---

July 1, 2019

Prof. Jack Middleburg  
Associate Editor  
*Biogeosciences*

Dear Jack:

I have extensively revised my submission #bg-2019-131 entitled “Is shale gas a major driver of recent increase in global atmospheric methane” in response to the comments of three reviewers. I found the comments of all of the reviewers to be constructive, and I feel my submission is substantially improved. I have previously written responses to each of the reviews (two responses, in the case of reviewer #1), and these have been posted on the web page for my discussion article since the 3<sup>rd</sup> of June. Below, I indicate in detail how I have revised my manuscript in light of these reviewer comments, but I do not repeat the detailed responses I provided earlier. I am submitting two versions of the revision, one showing full edit mode with all of the changes clearly indicated, and another where all of the changes have been accepted, making it easier to read the final version. The take-home message of my paper remains largely unchanged from that of my original submission.

I look forward to hearing your response.

Best regards,

Robert W. Howarth, Ph.D.  
*David R. Atkinson Professor of Ecology  
and Environmental Biology*

---

## List of revisions made:

Page 1, abstract: In response to reviewer #1, I am now using a different value for the d13C of methane for shale gas; this is not as different from the value for conventional natural gas, and I have changed the language here to reflect that. I have also somewhat toned down the conclusions.

Page 2, lines 4-6: Reviewer #2 had suggested I include mention to Nisbet et al. (2016); I have done so, but also found a more recent relevant paper, Nisbet et al. (2019); that paper makes an important point about the urgency of controlling methane emissions, and I have added reference to it here.

Page 2, line 14: I have added the Nisbet et al. (2016) reference here.

Page 2, line 29: I have added Sherwood et al. (2017) as a reference here, to strengthen the importance of getting biomass burning right in the 13C analysis.

Page 3, line 26, through page 14, line 14: In response to the comment by reviewer #1, I have changed the d13C value used for shale gas, which has resulted in extensive changes in the language here (as well as later, see new paragraph on page 6, lines 2-19). Note that I have added several new references that support that methane is partially oxidized and therefore fractionated as it migrates from shales into conventional reservoirs.

Page 4, line 28, through page 5, line 32; page 6, line 27 through page 7, line 15; and page 7, line 32, through page 9, line 5: In response to comments by both reviewer #2 and reviewer #3, I have substantially altered the equations used to estimate shale gas emissions.

Page 6, line 2-19: as noted above, I have added a new paragraph here, presenting a new value for the d13C of shale gas. I believe my logic is fully explained in this paragraph, as well as in the updated June 3 reply to reviewer #1.

Pge 6, lines 21-25: reviewer #3 argued that my analysis should be based not on the d13C value for methane currently in the atmosphere, but rather on the mass-weighted mean value for the methane emitted to the atmosphere that would lead to that value, after fractionation due to the oxidation sink in the atmosphere. I have taken this advice, and presented the new approach in this paragraph, with supporting references.

Page 6, line 30: I added a new header, to break up this rather long section.

Page 7, line 23: as suggested by reviewer #2, I have changed “measured” to “estimated base don.”

Page 9, lines 9-17: I have made some minor adjustments in language to reflect the change in results from the the new conceptual approach and the new d13C for methane from shale gas.

Page 9, lines 19-34: at the suggestion of reviewer #2, I have added a new paragraph to discuss how my results inform the discussion over the changes in methane emission geographically since 2007. As I discuss in my response to reviewer #2, I do not include reference here to Rice et al., as he had suggested, as their analysis ends in 2009, just as the shale gas revolution was accelerating.

Page 10, lines 2-21: I have made a few small changes here, mostly reflecting the new results. I deleted language on line 17 in response to a comment I received orally during the question section of a talk I gave on this paper in Amsterdam on June 13 at the 8th International Symposium on Non-CO2 Greenhouse Gases.

Page 11, line 1-6: I have made small changes in response to the new results.

Page 11, line 15: I have modified this language in response to the comment by the reviewer suggesting that emissions from the Bakken shale have decreased in the past few years. The Schneising et al. study only evaluates emissions through 2011.

Page 11, lines 17-28: I have made small changes in response to the new results.

Page 12, lines 2-4: I deleted some text to reflect the use of a new value for the mean d13C content of shale gas.

Page 12, lines 19-25: I have deleted some text in response to comments by reviewer #2.

Pge 12, ines 29-31: I have made some small changes to reflect the new results.

Page 13, line 15, through page 14, line 9: these calculations are changed to reflect the new conceptual approach used.

Page 15, line 16, through page 16, line 27: these calculations are change to reflect the new conceptual approach used.

References, page 17 through page 22: I added 14 new references to support the choice of d13C for shale gas methane and in response to suggestions made by the reviewers.

Tables 1 and 2, pages 24 and 25: numbers are updated to reflect the new results.

Figure 1-A, page 26: 1880 was changed to 1800 on the y-axis, correcting an error caught by reviewer #2.

Figures 3-A and 3-B, pages 30 and 31: these figures are updated to reflect the new results and to use the mean mass-weighted d13C for all emission sources, as suggested by reviewer #3.

Note that other figures remain unchanged, but I needed to re-enter them into the draft due to some issue with Microsoft Word.

# Is Shale Gas a Major Driver of Recent Increase in Global Atmospheric Methane?

Robert W. Howarth<sup>1</sup>

5 <sup>1</sup>Department of Ecology & Evolutionary Biology, Cornell University, Ithaca, NY 14853 USA

*Correspondence to:* Robert W. Howarth (howarth@cornell.edu)

**Abstract.** Methane has been rising rapidly in the atmosphere over the past decade, contributing to global climate change. Unlike the late 20<sup>th</sup> Century when the rise in atmospheric methane was accompanied by an enrichment in the heavier carbon stable isotope (<sup>13</sup>C) of methane, methane in recent years has become more depleted in <sup>13</sup>C. This depletion has been widely

10 interpreted to indicate a primarily biogenic source for the increased methane. Here we show that ~~the part of the~~ change may instead be associated with emissions from shale gas and shale oil development. ~~While methane in conventional natural gas is enriched in <sup>13</sup>C relative to the atmospheric mean Previous studies have not explicitly considered shale gas, even though most of the increase in natural gas production globally over the past decade is from shale gas. The methane in shale gas is somewhat depleted in <sup>13</sup>C relative to conventional natural gas at this atmospheric level.~~ Correcting 15 ~~earlier analyses~~ for this difference, we conclude that ~~emissions from~~ shale gas production in North America over the past decade may ~~have contributed more than half of all of the increased emissions from fossil fuels globally and approximately one third of the total increased emissions from all sources globally over the past decade. well be the leading cause of the increased flux of methane to the atmosphere. Increased fluxes from biogenic sources such as animal agriculture and wetlands are far less important than indicated by some other recent papers using <sup>13</sup>C data.~~

20

## 1. Introduction

Methane is the second most important greenhouse gas behind carbon dioxide causing global climate change, contributing approximately 1-watt m<sup>-2</sup> to warming when indirect effects are included, compared to 1.66 watt m<sup>-2</sup> for carbon dioxide (IPCC 2013). Unlike carbon dioxide, the climate system responds quickly to changes in methane emissions, and 25 reducing methane emissions could provide an opportunity to immediately slow the rate of global warming (Shindell et al. 2012) and perhaps meet the ~~UNFCCC~~ COP21 target of keeping the planet well below 2° C above the pre-industrial baseline (IPCC 2018). Methane also contributes to the formation of ground-level ozone, with large adverse consequences for human health and agriculture. Considering these effects as well as climate change, Shindell (2015) estimated the social cost of methane is 40 to 100 times greater than that for carbon dioxide: \$2,700 per ton for methane compared to \$27 per ton for carbon

dioxide when calculated with a 5% discount rate, and \$6,000 per ton for methane compared to \$150 per ton for carbon dioxide when calculated with a 1.4% discount rate.

Atmospheric methane levels rose steadily during the last few decades of the 20<sup>th</sup> Century before leveling off for the first decade of the 21<sup>st</sup> Century. Since 2008, however, methane concentrations have again been rising rapidly (Fig. 1-A). [This](#)

5 [increase, if it continues in coming decades, will significantly increase global warming and undercut efforts to reach the COP21 target \(Nisbet et al. 2019\).](#) The total atmospheric flux of methane for the period 2008-2014 was ~24.7 Tg per year greater than for the 2000-2007 period (Worden et al. 2017), an increase of 7% in global human-caused methane emissions. The change in the stable carbon  $\delta^{13}\text{C}$  ratio of methane in the atmosphere over the past 35 years is striking and seems clearly related to the change in the methane concentration (Fig. 1-B). For the final 20 years of the 20<sup>th</sup> Century, as atmospheric methane

10 concentrations rose, the isotopic composition became more enriched in the heavier stable isotope of carbon,  $^{13}\text{C}$ , relative to the lighter and more abundant isotope  $^{12}\text{C}$ , resulting in a less negative  $\delta^{13}\text{C}$  signal. The isotopic composition remained constant from 1998 to 2008 when the atmospheric concentration was constant. And the isotopic composition has become lighter (depleted in  $^{13}\text{C}$ , more negative  $\delta^{13}\text{C}$ ) since 2009 as atmospheric methane concentrations have been rising again (Schaefer et al. 2016; [Nisbet et al. 2016](#)). Since biogenic sources of methane [are tend to be](#) lighter than the methane released from fossil-

15 fuel emissions, Schaefer et al. (2016) concluded that the increase in atmospheric methane in the late 20<sup>th</sup> Century was due to increasing emissions from fossil fuels, but that the increase in methane since 2006 [was](#) due to biogenic sources, most likely tropical wetlands, rice culture, or animal agriculture. Their model results indicated that fossil fuel sources have remained flat or decreased globally since 2006, playing no major role in the recent atmospheric rise of methane. Schaefer et al. (2016) noted that their conclusion [contradicted](#) many reports of increased emissions from fossil fuel sources over this time, and stated that

20 20 their conclusion [was](#) “[is](#)-unexpected, given the recent boom in unconventional gas production and reported resurgence in coal mining and the Asian economy.” Six months after the Schaefer et al. (2016) study was published in *Science*, Schwietzke et al. (2016) presented a similar analysis in *Nature* that used a larger and more comprehensive data set for the  $\delta^{13}\text{C}$  values of methane emissions sources. They too concluded that fossil fuel emissions have likely decreased during this century, and that biogenic emissions are the probable cause of any recent increase in global methane emissions.

## 25 2. Sensitivity of emission models based on $\delta^{13}\text{C}$ in methane to biomass burning

Model analyses that use  $\delta^{13}\text{C}$  methane data to infer emission sources are highly sensitive to changes in the rate of biomass burning: although biomass burning is a relatively small contributor to global methane emissions, those emissions are quite enriched in  $^{13}\text{C}$  relative to the atmospheric methane signal (Rice et al. 2016; [Sherwood et al. 2017](#)). Both Schaefer et al. 30 (2016) and Schwietzke et al. (2016) assumed that biomass burning had been constant in recent years. However, Worden et al. (2017) estimated that [reductions in](#) biomass burning globally [went down](#) for the period 2007-2014 compared to 2001-2006, resulting in decreased methane emissions of 3.7 Tg per year ( $\pm 1.4$  Tg per year) [and contributing to a lower  \$\delta^{13}\text{C}\$  for](#)

atmospheric methane. Using the data set of Schwietzke et al (2016) for  $\delta^{13}\text{C}$  values of methane emission sources, but including changes in biomass burning over time, Worden et al. (2017) concluded that the recent increase in methane emissions was likely driven more by fossil fuels than by biogenic sources, with an increase of 165.45 Tg per year from fossil fuels ( $\pm 3.65$  Tg per year) compared to an increase of 12 Tg per year from biogenic sources ( $\pm 2.5$  Tg per year) when comparing 2007-2014

5 vs 2001-2006.

Clearly global models for partitioning methane sources based on the  $\delta^{13}\text{C}$  approach are sensitive to assumptions about seemingly small terms such as decreases in biomass burning. In this paper, we explore for the first time another assumption: that the global increase in shale gas development may have caused some of the depletion of  $^{13}\text{C}$  in the global average methane 10 observed over the past decade. Shale gas emissions were not explicitly considered in the models presented by Schaefer et al. (2016) and Worden et al. (2017) and were explicitly excluded in the analysis of Schwietzke et al. (2016).

### 3. What is shale gas?

15 Shale gas is a form of unconventional natural gas (mostly methane) held tightly in shale-rock formations. Conventional natural gas, the dominant form of natural gas produced during the 20<sup>th</sup> Century, is composed largely of methane that migrated upward from the underlying sources such as shale rock over geological time, becoming trapped under a geological seal (Fig. 2-A). Until this century, shale gas was not commercially developable. The use of a new combination of technologies in the 21<sup>st</sup> century – high precision directional drilling, high-volume hydraulic fracturing, and clustered multi-20 well drilling pads -- has changed this. In recent years, global shale gas production has exploded 14-fold, from 31 billion m<sup>3</sup> per year in 2005 to 435 billion m<sup>3</sup> per year in 2015 (Fig. 2-B), with 89% of this production in the United States and 10% in western Canada (EIA 2016). Shale gas accounted for 63% of the total increase in natural gas production globally over this time period past decade (EIA 2016, IEA 2017). The US Department of Energy predicts rapid further growth in shale gas production globally, reaching 1,500 billion m<sup>3</sup> per year by 2040 (EIA 2016; Fig. 2-B).

25

Several studies have suggested shown that the  $\delta^{13}\text{C}$  signal of methane from shale gas can is often be lighter (more depleted in  $^{13}\text{C}$ ) than that from conventional natural gas (Golding et al. 2013; Hao and Zou 2013; Turner et al. 2017; Botner et al. 2018). Here, we use the data from Figure 1 in the review by Golding et al. (2013) that were explicitly identified as shale gas. The samples are from the New Albany shale (Martini et al. 1998), the Antrim shale (McIntosh et al. 2002), and an organic-rich shale in the northern Appalachian basin (Osborn and McIntosh 2010). Note that these studies appear to be the only ones included in the  $\delta^{13}\text{C}$  methane data repository published by Sherwood et al. (2017), which is the data set underlying the analysis by Schwietzke et al. (2016). Out of 61 data points for shale gas in the Golding et al. (2013) figure, only 5 had  $\delta^{13}\text{C}$  values similar to those for conventional natural gas, while many samples more closely resembled the signal for biogenic gas. From the 61 values, we calculate a mean value  $\delta^{13}\text{C}$  for shale gas of  $-51.4\text{ ‰}$ , with a 95% confidence limit of  $\pm 1.2\text{ ‰}$ . Thus,

emissions of methane from shale gas are on average depleted in  $\delta^{13}\text{C}$  relative to atmospheric methane, while methane from conventional natural gas is more  $\delta^{13}\text{C}$  enriched than atmospheric methane. This should

It should perhaps not be surprising that the  $\delta^{13}\text{C}$  of methane from shale gas tends to be lighter than for conventional

5 natural gas. In the case of conventional gas, the methane has migrated over geological time frames from the shale and other source rocks through permeable ~~strata~~ rocks until trapped below a seal (Fig. 2-A). During this migration, some of the methane  
is likely can be oxidized both by bacteria, perhaps using iron (III) or sulfate as the source of the oxidizing power, and by  
thermochemical sulfate reduction (Whelan et al. 1986; Burruss and Laughrey 2010; Rooze et al. 2016). This pPartial  
10 oxidation and consumption of methane by bacteria would fractionate the methane by preferentially consuming the lighter  $^{12}\text{C}$   
isotope and so, gradually enriching the remaining methane in  $^{13}\text{C}$  (Hao and Zou 2013; Baldassare et al. 2014), resulting in a  
 $\delta^{13}\text{C}$  signal that is less negative. The methane in shales, on the other hand, is tightly held in the highly reducing rock formation  
and therefore very unless likely to have been subject to bacterial oxidation and the resulting fractionation. The expectation,  
therefore, is that methane in conventional natural gas should be heavier and less depleted in  $^{13}\text{C}$  than is the methane in shale  
gas.

15

#### 4. Calculating the effect of $^{13}\text{C}$ signal of shale gas on emission sources: Conceptual framework

To explore the contribution of methane emissions from shale gas, we build on the analysis of Worden et al. (2017).

20 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  $^{13}\text{C}$  than for 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  
25 Tg per year ( $\pm 14$  Tg per year) since 2007~~78~~, 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:

30 
$$(B_N - B_W) + (FF_N - FF_W) + SG = 0 \quad (1)$$

where  $B_N$  is the estimate from Worden et al. (2017) for the increase in biogenic emissions of methane globally after 2007,  $B_W$   
is our new estimate for the increase in these biogenic fluxes,  $FF_N$  is the estimate from Worden et al. (2017) for the increase in  
emissions of methane globally from fossil fuels after 2007,  $FF_W$  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.

which reweights the information in Figure 3-A for the difference between most fossil fuels and shale gas:

$$12 * D_{B-A} = (B * D_{B-A}) + (SG * D_{SG-CG}) - (CG * D_{A-CG}) \quad (1)$$

Formatted: Indent: First line: 0.5"

where 12 Tg per year is the mean estimate from Worden et al. (2017) for the increase in biogenic emissions,  $D_{B-A}$  is the difference in the  $\delta^{13}\text{C}$  value for biogenic emission sources and atmospheric methane in 2005, B is the estimate for the increase in biogenic emissions, SG is the estimate for the increase in methane emissions from shale gas,  $D_{SG-CG}$  is the difference in the  $\delta^{13}\text{C}$  value for shale gas and conventional natural gas, and  $D_{A-CG}$  is the difference in the  $\delta^{13}\text{C}$  value for atmospheric methane in 2005 and for emissions from conventional natural gas. 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.

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:

$$(B_N - B_W) * D_{B-A} = [(FF_N - FF_W) * D_{A-FF}] + (SG * D_{A-SG}) \quad (2)$$

Formatted: Subscript

where  $D_{B-A}$ ,  $D_{A-FF}$ , and  $D_{A-SG}$  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  $D_{B-A}$  and rearrange,

$$(B_N - B_W) * (D_{B-A}) = -[(FF_N - FF_W) * (D_{B-A})] - (SG * D_{B-A}) \quad (3)$$

Subtracting equation 3 from equation 2,

$$0 = [(FF_N - FF_W) * (D_{A-FF} + D_{B-A})] + [SG * (D_{A-SG} + D_{B-A})] \quad (4)$$

Rearranging equation 4,

$$SG = - (FF_N - FF_W) * (D_{A-FF} + D_{B-A}) / (D_{A-SG} + D_{B-A}) \quad (5)$$

Formatted: Indent: First line: 0.5"

Note that from Worden et al. (2017),  $FF_N$  is 16.4 Tg per year.

Although our expectation is that the methane in shale gas is depleted in  $^{13}\text{C}$  relative to conventional natural gas, the  $\delta^{13}\text{C}$  ratios for the methane in both conventional gas reservoirs and in shale gas vary substantially, changing with the maturity of the gas and several other factors (Golding et al. 2013; Hao and Zou 2013; Tilley and Muehlenbachs 2013). The large data set of Sherwood et al. (2017) suggests no systematic difference between the average ratio for shale gas and the average for conventional gas. However, some of the data listed as shale gas in that data set are actually for methane that has migrated from shale to reservoirs (Tilley et al. 2011) and therefore may have been partially oxidized and fractionated (Hao and Zou 2013). In other cases, the data appear to come both from conventional vertical wells and shale-gas horizontal wells in the same region, making interpretation ambiguous (Rodriguez and Philp 2010; Zumberge et al. 2012). Note that in the Barnett shale region, Texas, the  $\delta^{13}\text{C}$  ratio for methane emitted to the atmosphere (- 46.5 ‰; Townsend-Small et al. 2015) is more depleted than the average for wells reported in the Sherwood et al. (2017) data set: - 44.8 ‰ for “group 2A and 2B” wells and - 38.5 ‰ for “group 1” wells (Rodriguez and Philp 2010) and - 41.1 ‰ (Zumberge et al. 2012). For our analysis, we use the mean of the  $\delta^{13}\text{C}$  ratio (- 46.9 ‰) from three studies where the methane clearly came from horizontal, high-volume fractured shale wells: - 47.0 ‰ for Bakken shale, North Dakota (Schoell et al. 2011), - 46.5 ‰ for Barnett shale, Texas (Townsend-Small et al. 2015), and - 47.3 ‰ for Utica shale, Ohio (Botner et al. 2018). Note that several studies have reported mean  $\delta^{13}\text{C}$  ratios for methane from organic-rich shales that are more depleted in  $^{13}\text{C}$  (more negative) than this: - 50.7 (Martini et al. 1998) for Antrim shale, Michigan; - 53.3 (McIntosh et al. 2002) and - 51.1 (Schlegel et al. 2011) for New Albany shale, Illinois; and - 49.3 (Osborn and McIntosh 2010) for a Devonian shale in Ohio. However, these shales are not typical of the major shale plays supporting the huge increase in gas production over the past decade.

20

The average  $\delta^{13}\text{C}$  ratio for methane in the atmosphere in 2005 was - 47.15 ‰ (Schneising et al. 2016), which reflects a flux-weighted mean input of methane with a  $\delta^{13}\text{C}$  ratio of - 53.5 ‰. This flux-weighted mean value is approximately 6.3 ‰ more depleted in  $^{13}\text{C}$  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 ‰. Therefore, the mean value for  $D_{\text{A-FF}}$  is 9.5 ‰, for  $D_{\text{B-A}}$  is 9.0 ‰, and for  $D_{\text{A-SG}}$  is 6.6 ‰ (Fig. 3-B). Substituting these values into Eq. (5), we see that:

25

$$\text{SG} = -1.19 \text{FF}_{\text{N}} + 19.4 \quad (6)$$

### 30 5. Estimating increased methane fluxes for coal, oil, and natural gas

As noted above, shale gas accounted for 63% of the global increase in all natural gas production between 2005 and 2015 (EIA 2016, IEA 2017). If we make the simplifying assumption that for both shale gas and conventional natural gas, emissions are equal as a percentage of the gas produced, then

$$SG = 0.63 * TG \quad (2)$$

and

$$5 \quad CG = 0.37 * TG \quad (3)$$

where TG is total increase in emissions from all natural gas. Note that we test this assumption later in our sensitivity analyses, since some research indicates emissions from shale gas are higher than for conventional gas as a percentage of gas production. Rearranging Eq. (2) for TG and substituting into Eq. (3),

$$10 \quad CG = 0.37 * (SG / 0.63), \text{ or } CG = 0.59 * SG \quad (4)$$

Substituting CG from Eq. (4) into Eq. (1),

$$15 \quad 12 * D_{B,A} = (B * D_{B,A}) + (SG * D_{SG,CG}) - (0.59 * SG * D_{A,CG}) \quad (5)$$

Next, we estimate the likely contributions from coal and oil to the increased methane emissions over the past decade. We estimate the increase in methane emissions from coal between 2006 and 2016 as 1.3 Tg per year, based on the rise in global coal production of 27%, with almost all of this due to surface-mined coal in China (IEA 2008, 2017), and using a well-  
20 accepted emission factor of 870 g methane per ton of surface-mined coal (Howarth et al. 2011). Methane emissions from surface-mined coal tend to be low, as much of the whatever methane that was once associated with the coal has degassed over geological time. This estimate is very close to the 1.1 Tg per year increase from coal emissions in China between 2009 and 2015 as measured-estimated based on from satellite observationsdata (Miller et al. 2019). For oil, global production increased by 9.6% (IEA 2008, 2017), thereby increasing methane emissions by approximately 1.6 Tg per year (using emission factors  
25 from NETL 2008, as detailed in Howarth et al. 2011). Therefore, of the increase in 28.4 Tg per year from fossil fuels plus biogenic sources since 2005 (see discussion above), we estimate 2.9 Tg per year from increased emissions from coal and oil, leaving an increase of approximately 25.5 Tg per year from natural gas (including shale gas) plus biogenic sources.

As noted above, shale gas accounted for 63% of the global increase in all natural gas production between 2005 and 30 2015 (EIA 2016, IEA 2017). If we make the simplifying assumption that for both shale gas and conventional natural gas, emissions are equal as a percentage of the gas produced, then

$$SG = 0.63 * TG \quad (7)$$

and

$$CG = 0.37 * TG \quad (8)$$

5 where TG is total increase in emissions from all natural gas. Note that we test this assumption later in our sensitivity analyses, since some research indicates emissions from shale gas are higher than for conventional gas as a percentage of gas production. Rearranging Eq. (7) for TG and substituting into Eq. (8),

$$CG = 0.37 * (SG / 0.63), \text{ or} \quad CG = 0.59 * SG \quad (9)$$

10

$FF_N$  is the sum of CG ( $0.59 * SG$ ) plus the emissions from oil and coal (2.9 Tg per year), or

$$FF_N = (0.59 * SG) + 2.9 \quad (10)$$

15 Substituting Eq. (10) into Eq. (6) and solving for SG, we estimate that the increase in shale gas emissions between 2005 and 2015 was 9.4 Tg per year (Table 1; Fig. 3-B). That is,

Formatted: Indent: First line: 0"

Formatted: English (United Kingdom)

$$SG + CG + B = 25.5 \quad (6)$$

20 Substituting CG from Eq. (4) into Eq. (6) and rearranging yields

$$B = 25.5 - (1.59 * SG) \quad (7)$$

Substituting B from Eq. (7) into Eq. (5),

25

$$12 * D_{B,A} = [(25.5 - (1.59 * SG)) * D_{B,A}] + (SG * D_{SG,CG}) - (0.59 * SG * D_{A,CG}) \quad (8)$$

And rearranging to solve for SG,

30  $SG = (13.5 * D_{B,A}) / [(1.59 * D_{B,A}) - (D_{SG,CG}) + 0.59 * D_{A,CG}] \quad (9)$

Formatted: Indent: First line: 0"

If we use mean values for the differences in the  $\delta^{13}\text{C}$  terms in Eq. (9) (ie,  $D_{B,A} = 15.35\text{‰}$ ,  $D_{SG,CG} = 7.4\text{‰}$ , and  $D_{A,CG} = 3.15\text{‰}$ ), then we estimate the increase in methane emissions from shale gas between 2005 and 2015 (SG) as 11 Tg per year (Table 1). From Eq. (9), increased emissions from conventional natural gas are then estimated as 5.56.5 Tg per year, and

Formatted: Indent: First line: 0"

from all natural gas (shale plus conventional) as 14.97.5 Tg per year, and from all fossil fuels (including coal and oil) as 17.8 Tg per year. From Eq. (17), increased emissions from biogenic sources are estimated as 10.68.0 Tg per year. While the biogenic sources are important, the increase in fossil fuel emissions has been greater, and shale gas makes up more than half of these increased fossil fuel emissions. The confidence bounds on these estimates, calculated using Eq. (9) and the upper and lower 95% confidence limits for the  $\delta^{13}\text{C}$  ratio terms (Fig. 3-B), are relatively small (Table 1).

5

## 65. Comparison with prior estimates

Our best estimate for the increase in methane emissions from all fossil fuels since 2008 (shale gas, conventional natural gas, coal, and oil) is 17.820.4 Tg per year is (Table 1), 9% substantially larger than the mean estimate of Worden et al. (2017) of 16.45.5 Tg per year (Table 1). Our estimate for the increased emissions from biogenic sources, 10.68 Tg per year, is substantially 12% lower than the Worden et al. (2017) estimate of 12 Tg per year (Table 1). Thus, our estimates are not greatly different from those of Worden et al. (2017), although our estimate for fossil fuels is larger and our estimate for biogenic fluxes lower than their estimates. On the other hand, comparing emissions for the 2003-2013 period with those from the late 20<sup>th</sup> Century, Schwietzke et al. (2016) concluded that biogenic emissions had risen by ~ 27 Tg per year while fossil fuel emissions had decreased by ~18 Tg per year. And Schaefer et al. (2016) concluded that increased methane emissions since 2006 are “predominantly biogenic” and that fossil fuel emissions likely have fallen.

10 We estimate that shale gas contributed 33% of the global increase in all methane emissions in recent years (Table 1).  
20 Since virtually all shale gas development globally through 2015 occurred in North America (mostly the United States but also western Canada), we conclude that at least 33% of the increase in methane fluxes came from North America. This is consistent with the work of Turner et al. (2016) who used satellite data to conclude that 30% to 60% of the global increase in methane emissions between 2002 and 2014 came from the United States. Our finding is also consistent with Schneising et al. (2014), who analyzed data from another satellite and concluded that most of the global increase in methane emissions between 2006-25 2008 and 2009-2011 came from mid latitudes in the northern hemisphere, with shale gas and oil operations in the United States likely playing a role. On the other hand, Nisbet et al. (2016, 2019) used monitoring data to infer spatial changes in methane emissions over time and emphasized that much of the increase in recent years originated in the tropics and southern hemisphere, although they noted that the north temperate latitude played a major role in the large increase in emissions in 2014 (Nisbet et al. 2019), a time of major increase in shale gas development (EIA 2016). Overall, our results tend to support the conclusions 30 of Turner et al. (2016) and Schneising et al. (2014) on the spatial source of increased methane emissions. While our estimate for increased emissions from fossil fuels is only marginally greater than that of the Worden et al. (2017) paper upon which we build our analysis, we demonstrate the importance of shale gas as a major part of these increased fossil fuel emissions and thereby explicitly link the increased emissions to North America.

Formatted: Indent: First line: 0.5"

Our estimate of increased emissions of [9.411 Tg](#) per year from shale gas development is quite reasonable in light of the growing body of evidence from measurements made at local to regional scales. Between 2005 and 2015, global shale gas production rose by 404 billion m<sup>3</sup> per year (Fig. 2-B) (EIA 2016). Assuming that 93% of natural gas is composed of methane

5 (Schneising et al. 2014), our estimate of the increase in methane emissions from shale gas ([11 Tg per year](#)) represents [3.54.1](#) % of the [shaleincreased](#) gas production ([an increase of](#) 270 Tg per year of methane produced from shale-gas operations [on average in 2015](#)). This estimate of [3.54.1](#) % (based on global change in the <sup>13</sup>C content of methane) represents full life-cycle emissions, including those from the gas well site, transportation, processing, storage systems, and final distribution to customers. Our estimate is well within the range reported in several recent studies for shale gas, and in fact is at the low end

10 for many (but not all) of these studies (Howarth et al. 2011; Pétron et al. 2014; Karion et al. 2013; Caulton et al. 2014; Schneising et al. 2014; Howarth 2014). Alvarez et al. (2018) recently presented a summary estimate for natural gas emissions in the United States (both conventional and shale gas) of 2.3% using bottom-up, facility-based data. However, they noted that top-down estimates from approaches such as airplane flyovers give higher values than the bottom-up estimates they emphasized. In fact, a careful comparison of bottom-up and top-down approaches for one shale-gas field showed 45% higher

15 emissions from the top-down approach, due to under sampling of some emission events by the bottom-up, facility-based approach (Vaughn et al. 2018). Further, Alvarez et al. (2018) used a very low value for the methane emissions from local distribution pipelines, only 0.08 % [based on non-peer-reviewed data from the US Environmental Protection Agency from 1996](#) (see discussion in Howarth et al. 2011). Many studies suggest distribution emissions in Boston, Los Angeles, Indianapolis, and Texas cities may be as high as 2.5% or more, not 0.08 % (Howarth et al. 2011; McKain et al. 2015; Lamb et al. 2016;

20 Wunch et al. 2016), so that a full life-cycle of [3.54.1](#) % emissions from shale gas over the past decade is quite plausible, [and perhaps even low.](#)

25 **[26. Sensitivity analyses](#)**

Our analysis contain two major assumptions: 1) that methane emissions as a percentage of gas produced are the same for shale gas and conventional natural gas (Eq. [\(72\)](#) and Eq. [\(83\)](#)); and 2) that emissions from oil have remained proportional to the global rate of oil production. Here we explore the sensitivity of our analysis to these assumptions. With regard to the

30 first assumption, some evidence suggests that percent emissions may be higher from shale gas than from conventional natural gas, perhaps due to venting at the time of flow-back following high-volume hydraulic fracturing of shale-gas wells (Howarth et al. 2011) and also due to release of methane from trapped pockets when drilling down through a very long legacy (often a century or more) of prior fossil fuel operations ([coal, oil, and gas](#)) to reach the deeper shale formations (Caulton et al. 2014; Howarth 2014). For this first sensitivity analysis, we modify equations Eq. [\(72\)](#) through Eq. [\(109\)](#) with new equations Eq.

(A1) through Eq. (A47) to reflect a 50% higher emission factor for shale gas than for conventional gas, as proposed in Howarth et al. (2011). ~~See appendix A~~. With this change in assumptions, estimated shale gas emissions increase by 1225% (10.83.9 instead of 9.411 Tg per year), ~~which corresponds to a life-cycle emission factor of 4.0% rather than 3.5%.~~ Biogenic emissions ~~decrease by 23% remain virtually unchanged~~ (10.46.2 instead of 10.68 Tg per year), ~~as do while~~ total fossil fuel emissions ~~increase~~ (1822.2 instead of 17.820.4 Tg per year). ~~The fossil fuel emission estimate is now 3.6-fold larger than the biogenic emission estimate~~ (Table 2).

Our second major assumption in the base analysis is that methane emission factors for oil production have remained constant over time as a function of production. This may not be true, since 60% of the increase in global oil production between 10 2005 and 2015 was due to tight oil production from shales using the same technologies that allowed shale gas development, high-precision directional drilling and high-volume hydraulic fracturing (calculated from data in EIA 2015 and EIA 2018). Large quantities of methane are often co-produced with this tight shale oil, and because oil is a much more valuable product than natural gas, for shale-oil fields removed from easy access to natural gas markets, much of the methane may be vented or flared rather than delivered to market. This may be part of the reason for the large increase in methane emissions ~~in~~   
15 ~~recent between 2008 and 2011 years~~ in the Bakken shale fields of North Dakota (Schneising et al. 2014).

For this sensitivity scenario #2, we modify equations Eq. (74) through Eq. (109) with new equations Eq. (B1) through Eq. (B48) to allow for higher emissions associated with shale oil than from conventional oil production. ~~S~~ ~~(see appendix B)~~. For this, we follow the approach of Schneising et al. (2014) in combining shale gas and shale oil, scaling the increase in 20 production since 2005 by the energy value of the two products. As in our baseline analysis developed in equations Eq. (1) through Eq. (109), we assume that conventional natural gas and shale gas have the same percentage methane emission per unit of produced gas. Here we further assume that shale oil has the same emission rate as well, scaled to the energy content of oil compared to natural gas. This sensitivity analysis ~~again has very little influence on either increases~~ total emissions from fossil fuels ~~by 18%~~ (18.222.8 instead of 17.820.4 Tg per year) ~~or, while~~ biogenic emissions ~~fall~~ (10.25.6 instead of 10.68.0 Tg per   
25 year) (Table 2). The contribution from shale gas falls somewhat (from 9.411 to 7.89.9 Tg per year), as does that from conventional natural gas (from 5.5 to 4.26.5 to 5.4 Tg per year), while shale oil becomes an important emission source (4.25.5 Tg per year). Overall in this scenario, increased emissions from fossil fuels extracted from shales (gas plus oil) are 1215.4 Tg per year, two-thirds of the total increase due to fossil fuels.

### 30 87. Conclusions

We conclude that increased ~~methane~~ emissions from fossil fuels ~~are far more~~ likely ~~exceed those from~~ ~~than~~ biogenic ~~sources~~ ~~emissions to have driven the observed global increase in methane~~ over the past decade (since 20078). The increase in emissions from shale gas (perhaps in combination with those from shale oil) makes up more than half of the total increased

fossil fuel emissions. That is, the commercialization of shale gas and oil in the 21<sup>st</sup> Century has dramatically increased global methane emissions. ~~However, we note an important caveat: our analysis of emissions with explicit consideration of the  $\delta^{13}\text{C}$  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  $\delta^{13}\text{C}$  content of shale gas methane.~~

5

Note that while methane emissions are often referred to as “leaks,” ~~some of the~~ emissions include purposeful venting, including the release of gas during the flowback period immediately following hydraulic fracturing, the rapid release of gas from blowdowns during emergencies but also for routine maintenance on pipelines and compressor stations (Fig. 4-A), and the steadier but more subtle release of gas from storage tanks (Fig. 4-B) and compressor stations to safely maintain pressures (Howarth et al. 2011). This suggests large opportunities for reducing emissions, but at what cost? Do large capital investments for rebuilding natural gas infrastructure make economic sense, or would it be better to move to phase natural gas out as ~~fuel an energy source~~ and instead invest in a 21<sup>st</sup> Century energy infrastructure that embraces renewable energy and much more efficient heat and transportation through electrification (Jacobson et al. 2013)?

15 In October 2018, the Intergovernmental Panel on Climate Change issued a special report, responding to the call of the United Nations COP21 negotiations to keep the planet well below 2° C from the preindustrial baseline (IPCC 2018). They noted the need to reduce both carbon dioxide and methane emissions, and they recognized that the climate system responds more quickly to methane: reducing methane emissions offers one of the best routes to immediately slowing the rate of global warming (Shindell et al. 2012). ~~Nonetheless, the model scenarios presented in the IPCC report emphasize reducing carbon dioxide emissions first, and these scenarios begin to reduce methane emissions only after 2030. This may reflect the belief of the IPCC authors that methane emissions are dominated by biogenic sources, which are difficult to reduce. Given our conclusion finding that the oil and gas industry is natural gas (both shale gas and conventional gas) are more likely responsible for much of the recent increases in methane emissions, we suggest that the best strategy is to move as quickly as possible away from natural gas, reducing both carbon dioxide and methane emissions. Natural gas is not a bridge fuel (Howarth 2014). Doing so will in fact make it easier to reach the COP21 target than predicted by the IPCC (2018).~~

Finally, in addition to contributing to climate change, methane emissions lead to increased ground-level ozone levels, with significant damage to public health and agriculture. Based on the social cost of methane emissions of \$2,700 to \$6,000 per ton (Shindell 2015), our baseline estimate for increased emissions from shale gas of ~~9.411~~ Tg per year ~~corresponds to has resulted in~~ damage to public health, agriculture, and the climate of ~~\$2530 to \$565~~ billion USD per year for each of the past several years. This ~~is comparable to exceeds~~ the wholesale value for this shale gas over these years.

**Appendix A.** Sensitivity case #1: emissions per unit of gas produced assumed to be 50% greater for shale gas than for conventional gas.

First we modify Eq. (72) and Eq. (83) as follows to reflect that methane emissions per unit of gas produced are 50% greater for shale gas than for conventional natural gas:

$$SG = 1.2 * (0.63 * TG), \text{ or} \quad SG = 0.76 * TG \quad (A1)$$

and

$$CG = 0.8 * (0.37 * TG), \text{ or} \quad CG = 0.30 * TG \quad (A2)$$

10

Rearranging Eq. (A1) for TG and substituting into Eq. (A2),

$$CG = 0.30 * (SG / 0.76), \text{ or} \quad CG = 0.40 * SG \quad (A3)$$

15 Since  $FF_N$  is the sum of CG and the 2.9 Tg per year emissions for oil and coal,

$$FF_N = 0.40 SG + 2.9 \quad (A4)$$

Substituting Eq. (A4) into Eq. (6) and solving for SG, we estimate that the increase in shale gas emissions between 2005 and 20 2015 was 10.8 Tg per year (Table 2).

Substituting CG from Eq. (A3) into Eq. (1),

$$12 * D_{B,A} = (B * D_{B,A}) + (SG * D_{SG,CG}) - (0.40 * SG * D_{A,CG}) \quad (A4)$$

25 Substituting CG from Eq. (A3) into Eq. (6),

$$SG + (0.40 * SG) + B = 25.5, \text{ or with rearrangement,}$$

$$B = 25.5 - (1.40 * SG) \quad (A5)$$

30

Substituting B from Eq. (A5) into Eq. (A4),

$$12 * D_{B,A} = \{[25.5 - (1.40 * SG)] * D_{B,A}\} + (SG * D_{SG,CG}) - (0.30 * SG * D_{A,CG}) \quad (A6)$$

Formatted: Indent: First line: 0"

Formatted: Subscript

Formatted: Subscript

Formatted: Indent: First line: 0"

And rearranging to solve for SG,

$$SG = (13.5 * D_{B,A}) / [(1.40 * D_{B,A}) - (D_{SG,CG}) + 0.30 * D_{A,CG}] \quad (A7)$$

5 If we use mean values for the differences in the  $\delta^{13}\text{C}$  terms in Eq. (A7) (as we did previously for Eq. (9)), for shale gas,  $SG = 13.8 \text{ Tg per year}$ . From Eq. (A3), for conventional natural gas,  $CG = 4.355 \text{ Tg per year}$ . From Eq. (A5), for biogenic emissions,  $B = 6.2 \text{ Tg per year}$ . The increase in total fossil fuel emissions are estimated as the contributions from coal (1.3 Tg per year) and oil (1.6 Tg per year) plus SG plus CG, or  $1822.2 \text{ Tg per year}$ . From Eq. (1), biogenic emission are estimated to have increased by  $10.4 \text{ Tg per year}$ . These values are reported in Table 2.

10

#### Appendix B. Sensitivity case #2: explicit consideration of shale oil (tight oil).

15 For the base analysis presented in the main text using equations Eq. (1) through Eq. (109), we assumed that increased emissions from the additional oil development over the past decade were proportional to the increase in that rate of development. That is, the oil produced in recent years had the same emission factor as for oil produced a decade or more ago. However, 60% of the increase in oil production globally between 2005 and 2015 was for tight oil from shale formations (calculated from data in EIA 2015 and EIA 2018), and methane emissions from this shale oil may be greater than for 20 conventional oil. In this sensitivity case #2, we consider increased emissions from conventional oil and from tight shale oil separately. For conventional oil, the increase in emissions is 40% of the total oil emissions from the base analysis (40% of 1.6 Tg per year, or 0.65 Tg per year, rounded to 0.7 in Table 2), reflecting that conventional oil contributed 40% to the growth in oil production between 2005 and 2015.

25 For the tight shale oil, we follow the approach used by Schneising et al. (2014): the increase in methane emissions from shale gas and shale oil are considered together, normalized to the energy content of the two fuels. Shale gas production increased by 405 billion m<sup>3</sup> per year between 2005 and 2015 (EIA 2016). With an energy content of 37 MJ per m<sup>3</sup>, this reflects an increase in 15.9 trillion MJ per year. For shale oil, production increased by 230 liters per year between 2005 and 2015 (EIA 2015, 2018). With an energy content of 38 MJ per liter, this reflects an increase in 8.9 trillion MJ per year. Conventional 30 natural gas production increased by 238 billion m<sup>3</sup> per year between 2005 and 2015 (EIA 2016). With an energy content of 37 MJ per m<sup>3</sup>, this reflects an increase in 8.8 trillion MJ per year. Therefore, the sum of the increase in production for shale gas, shale oil, and conventional natural gas is 33.6 trillion MJ per year. Shale gas represents 48% of this, shale oil 26%, and conventional natural gas represents 26%. The sum of shale gas and shale oil represents 74% of the total.

For this sensitivity analysis, we further assume that shale gas and conventional natural gas have the same percentage emissions, as in our base case analysis in the main text, and that the  $^{13}\text{C}$  content of methane from shale oil is the same as for shale gas. Using these assumptions, we modify Eq. (72) and Eq. (83) as follows,

5  $\text{SG\&O} = 0.74 * \text{TG\&SO}$  (B1)

and

10  $\text{CG} = 0.26 * \text{TG\&SO}$  (B2)

where SG&O is shale gas plus shale oil and TG&SO is total natural gas plus shale oil. Rearranging Eq. (B1) for TG&SO and substituting into Eq. (B2),

15  $\text{CG} = 0.26 * (\text{SG\&O} / 0.74), \text{ or} \quad \text{CG} = 0.35 * \text{SG\&O}$  (B3)

Since  $\text{FF}_N$  is the sum of CG and the 2.0 Tg per year emissions for coal and conventional oil,

20  $\text{FF}_N = 0.35 \text{ SG} + 2.0$  (B4)

Substituting Eq. (B4) into Eq. (6) and solving for SG, we estimate that the increase in methane emissions from shale oil plus shale gas between 2005 and 2015 was 12 Tg per year. From Eq. (B3), increased emissions from conventional natural gas are 4.2 Tg per year (Table 2).

← Formatted: Indent: First line: 0"

25 Substituting CG from Eq. (B3) into Eq. (1) modified by substituting SG&O for SG,

~~$12 * D_{\text{B,A}} = (B * D_{\text{B,A}}) + (\text{SG\&O} * D_{\text{SG,CG}}) - (0.35 * \text{SG\&O} * D_{\text{A,CG}})$~~  (B4)

30 Modifying Eq. (6) for SG&O rather than SG, and estimating 1.95 Tg per year from increased emissions from coal and conventional oil, leaves an increase of approximately 26.45 Tg per year from natural gas (including shale gas) plus tight shale oil plus biogenic sources, so

~~$\text{SG\&O} + \text{CG} + B = 26.45$~~  (B5)

Substituting CG from Eq. (B3) into Eq. (N5),

$$SG\&O + (0.35 * SG\&O) + B = 26.45, \text{ or with rearrangement,}$$

5  $B = 26.45 - (1.35 * SG\&O) \quad (B6)$

Substituting B from Eq. (B6) into Eq. (B4),

$$12 * D_{B,A} = [(26.45 - 1.35 * SG\&O) * D_{B,A}] + (SG\&O * D_{SG,CG}) - (0.35 * SG\&O * D_{A,CG}) \quad (B7)$$

10 Rearranging to solve for SG&O,

$$SG\&O = (14.45 * D_{B,A}) / [(1.35 * D_{B,A}) - (D_{SG,CG}) + 0.35 * D_{A,CG}] \quad (B8)$$

15 If we use mean values for the differences in the  $\delta^{13}C$  terms in Eq. (B8) (as we did previously for Eq. (9) and Eq. (A7)), for shale gas plus shale oil emissions,  $SG\&O = 15.4$  Tg per year. From Eq. (B3), for conventional natural gas,  $CG = 5.4$  Tg per year. From Eq. (B6), for biogenic emissions,  $B = 5.6$  Tg per year.

20 The total increase in fossil fuel emissions are estimated as the contributions from coal (1.3 Tg per year), conventional oil (0.765 Tg per year), conventional natural gas (4.254 Tg per year), plus the sum for shale gas plus shale oil (125.4 Tg per year), or 18.222.8 Tg per year. We can separately estimate shale gas and shale oil, estimating the proportion of the sum of the two made up by shale gas as follows,

$$15.9 \text{ trillion MJ per yr} / (15.9 \text{ trillion MJ per year} + 8.9 \text{ MJ per year}) = 0.64 \quad (B59)$$

25

Therefore, of the increased in emissions of 1245.4 Tg per year for SG&O, the increase for shale gas emissions are 7.89.9 Tg per year and those for shale oil emissions are 4.25.5 Tg per year. These values are reported in Table 2.

30

## 8. Author contributions and competing interests

Robert Howarth is the sole author, responsible for all aspects of this work. The author declares that he has no conflicts of  
5 interest.

## 9. Acknowledgements

Financial support was provided by the Park Foundation and an endowment given by David R. Atkinson to support the  
10 professorship at Cornell University held by RWH. We thank Tony Ingraffea, Amy Townsend-Small, [Alexander Turner](#), Euan  
Nisbet, Martin Manning, Dennis Swaney, [Rand Roxanne Marino](#), [and three anonymous reviewers](#) for comments on earlier  
versions of this manuscript. We particularly thank Dennis Swaney for help [withful discussion and review of](#) the analyses we  
report. We thank Gretchen Halpert for the art work in Figures 1 and 2; Sharon Wilson for the photographs in Figure 4-A; and  
Jack Ossont for the photograph in Figure 4-B. Tony Ingraffea helped interpret these photographs.

15

## 10. References

Alvarez, R.A., Zavalao-Araiza, D., Lyon, D.R., Allen, D.T., Barkley, Z.R., Brandt, A.R., Davis, K.J., Herndon, S.C., Jacob,  
D.J., Karion, A., Korts, E.A., Lamb, B.K., Lauvaux, T., Maasakkers, J.D., Marchese, A. J., Omara, M., Pacala, J.W., Peiachl,  
20 J., Robinson, A.J., Shepson, P.B., Sweeney, C. Townsend-Small, A., Wofsy, S. C., and Hamburg, S. P., Assessment of  
methane emissions from the U.S. oil and gas supply chain, *Science* 361, 186-188, doi:10.1126/science.aar7204, 2018.

Baldassare, F.J., McCaffrey, M.A., and Harper, J.A., A geochemical context for stray gas investigations in the northern  
Appalachian Basin: Implications of analyses of natural gases from Neogene-through Devonian-age strata, *AAPG Bulletin*, 98,  
25 341–372, doi:10.1306/06111312178, 2014.

Botner, E.C., Townsend-Small, A., Nash, D.B., Xu, X., Schimmelmann, A., and Miller, J.H., Monitoring concentration and  
isotopic composition of methane in groundwater in the Utica Shale hydraulic fracturing region of Ohio, *Environ. Monit.  
Assess.*, 190, 322, doi:10.1007/s10661-018-6696-1, 2018.

30

[Burruss, R.C., and Laughrey, C.D., Carbon and hydrogen isotopic reversals in deep basin gas: evidence for limits to the stability  
of hydrocarbons. \*Organic Geochemistry\* 41, 1285–1296, doi:10.1016/j.orggeochem.2010.09.008, 2010.](#)

Caulton, D.R., Shepson, P.D., Santoro, R.L., Sparks, J.P., Howarth, R.W., Ingraffea, A., Camaliza, M.O., Sweeney, C., Karion, A., Davis, K.J., Stirm, B.H., Montzka, S.A., and B. Miller, B., Toward a better understanding and quantification of methane emissions from shale gas development, *P. Natl. Acad. Sci. USA*, 111, 6237-6242, doi:10.1073/pnas.1316546111, 2014.

5 EIA, Shale gas and tight oil and commercially produced in just four countries, Energy Information Administration, U.S. Department of Energy, <https://www.eia.gov/todayinenergy/detail.php?id=19991>, 2015.

EIA, [Shale gas production drives world natural gas production growth](#), Energy Information Administration, U.S. Department of Energy, <https://www.eia.gov/todayinenergy/detail.php?id=27512>, 2016.

10

EIA, Table 1.2, World crude oil production 1960-2017, Monthly energy review, June 2018, Energy Information Administration, U.S. Department of Energy, 2018.

Golding, S.D., Boreham, C.J., and Esterle, J.S., Stable isotope geochemistry of coal bed and shale gas and related production waters: A review, *Int. J. Coal Geol.*, 120, 24-40, doi:10.1016/j.coal.2013.09.001, 2013.

**Field Code Changed**

Hao, F., and Zou, H., [Cause of shale gas geochemical anomalies and mechanisms for gas enrichment and depletion in high-maturity shales](#), *Mar. Petrol. Geol.*, 44, 1-12, doi:10.1016/j.marpetgeo.2013.03.005, 2013.

**Formatted:** Font: 10 pt

**Formatted:** Font: (Default) +Body (Times New Roman), English (United Kingdom)

20 Howarth, R.W., Santoro, R., and Ingraffea, A., Methane and the greenhouse gas footprint of natural gas from shale formations, *Climatic Change Letters*, 106, 679–690, doi:10.1007/s10584-011-0061-5, 2011.

Howarth, R.W., A bridge to nowhere: Methane emissions and the greenhouse gas footprint of natural gas, *Energy Science & Engineering*, 2, 47-60, doi: 10.1002/ese3.35, 2014.

25

IEA, World Energy Outlook, International Energy Agency, 2008.

IEA, Key World Energy Statistics, International Energy Agency, 2017.

30 IPCC, Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change, Intergovernmental Panel on Climate Change, 2013.

IPCC, Summary for Policymakers. In: Global warming of 1.5°C. An IPCC Special Report on the impacts of global warming of 1.5°C above pre-industrial levels and related global greenhouse gas emission pathways, in the context of strengthening the

global response to the threat of climate change, sustainable development, and efforts to eradicate poverty, Intergovernmental Panel on Climate Change, <http://www.ipcc.ch/report/sr15/>, 2018.

Jacobson, M.Z., Howarth, R.W., Delucchi, M.A., Scobies, S.R., Barth, J.M., Dvorak, M.J., Klevze, M., Katkhuda, H., Miranda, B., Chowdhury, N.A., Jones, R., Plano, L., and A.R. Ingraffea, A.R., Examining the feasibility of converting New York State's all-purpose energy infrastructure to one using wind, water, and sunlight, *Energy Policy*, 57, 585-601, doi:10.1016/j.enpol.2013.02.036, 2013.

Karion, A., Sweeney, C., Pétron, G., Frost, G., Hardesty, R.M., Kofler, J., Miller, B.R., Newberger, T., Wolter, S., Banta, R., and Brewer, A., Methane emissions estimate from airborne measurements over a western United States natural gas field, *Geophys. Res. Lett.*, 40, 4393–4397, doi:10.1002/grl.50811, 2013.

Lamb, B.K., Cambaliza, M.O.L., Davis, K.J., Edburg, S.L., Ferrara, T.W., Floerchinger, C., Heimburger, A.M.F., Herndon, S., Lauvaux, T., Lavoie, T., Lyon, D.R., Miles, N., Prasad, K.R., Richardson, S., Roscioli, J.R., Salmon, O.E., Shepson, P.B., Stirm, B.H., and Whetstone, J., Direct and indirect measurements and modeling of methane emissions in Indianapolis, Indiana, *Environ. Sci. Technol.*, 50, 8910-8917, doi:10.1021/acs.est.6b01198, 2016.

Martini, A.M., Walter, L.M., Budai, J.M., Ku, T.C.W., Kaiser, C.J., and Schoell, M., Genetic and temporal relations between formation waters and biogenic methane: Upper Devonian Antrim Shale, Michigan Basin, USA, *Geochim. Cosmochim. Acta*, 62, 1699–1720, doi:016-7037(98)00090-8, 1998.

McIntosh, J.C., Walter, L.M., and Martini, A.M., Pleistocene recharge to mid-continent basins: effects on salinity structure and microbial gas generation, *Geochim. Cosmochim. Acta*, 66, 1681–1700, doi:10.1016/S0016-7037(01)00885-7, 2002.

McKain, K., Down, A., Raciti, S.M., Budney, J., Hutyra, L.R., Floerchinger, C., Herndon, S.C., Nehrkorn, T., Zahniser, M.S., Jackson, R.B., Phillips, N., and Wofsy, S.C., Methane emissions from natural gas infrastructure and use in the urban region of Boston, Massachusetts, *Proc. Natl. Acad. Sci. USA*, 112, 1941-1946, doi:10.1073/pnas.1416261112, 2015.

Miller, S.M., Michalak, A.M., Detmers, R.R., Hasekamp, O.P., Bruhwiler, L.M.P., and Schwietzke, S., China's coal mine methane regulations have not curbed growing emissions, *Nat. Communic.*, 10, 303, doi:10.1038/s41558-019-0432-x, 2019.

Nisbet, E.G., Dlugokencky, E.J., Manning, M.R., Lowry, D., Fisher, R.E., France, J. L., Michel, S.E., Miller, J.B., White, J.W.C., Vaughn, B., Bousquet, P., Pyle, J.A., Warwick, N.J., Cain, M., Brownlow, R., Zazzeri, G., Lanoiselle, M., Manning,

A.C. Gloor, E., Worthy, D.E.J., Brunke, E.G., Labuschagne, C., Wolff, E.W., and Ganesan, A.L., Rising atmospheric methane: 2007–2014 growth and isotopic shift. Global Biogeochemical Cycles, 30, 1356–1370, doi: 10.1002/2016GB005406, 2016.

Nisbet, E.G., Manning, M.R., Dlugokencky, E.J., Fisher, R.E., Lowry, D., Michel, S.E., Myhre, C.L., Platt, S.M., G. Allen, G., Bousquet, Brownlow, P.R., Cain, M., France, J.L., Hermansen, O., Hossaini, R., Jones, A.E., Levin, I., Manning, A.C., Myhre, G., Pyle, J.A., Vaughn, B.H., Warwich, N.J., and White, J.W.C. Very strong atmospheric methane growth in the 4 years 2014–2017: Implications for the Paris Agreement. Global Biogeochemical Cycles, 33, 318–342, doi: 10.1029/2018GB006009, 2019.

10 Osborn, S.G., and McIntosh, J.C., 2010. Chemical and isotopic tracers of the contribution of microbial gas in Devonian organic-rich shales and reservoir sandstones, northern Appalachian Basin, Appl. Geochem., 25, 456–471, doi:10.1016/j.apgeochem.2010.01.001, 2010.

15 Pétron, G., Karion, A., Sweeney, C., Miller, B., Montzka, S.A., Frost, G.J., Trainer, M., Tans, P., Andrews, A., Kofler, J., Helming, D., Guenther, D., Dlugokencky, E., Lang, P., Newberger, T., Wolter, S., Hall, B., Novelli, P., Brewer, R., Conley, S., Hardesty, M., Banta, R., White, A., Noone, D., Wolfe, D., Schnell, R., A new look at methane and nonmethane hydrocarbon emissions from oil and natural gas operations in the Colorado Denver-Julesburg Basin, J. Geophys. Res. Atmos., 119, 6836–6852, doi:10.1002/2013JD021272, 2014.

20 Rice AL, Butenhoff CL, Tema DG, *et al.* 2016. Atmospheric methane isotopic record favors fossil sources flat in 1980s and 1990s with recent increase. P. Natl. Acad. Sci. USA, 13, 10791–10796, doi:10.1073/pnas.1522923113, 2016.

25 Rodriguez, N.D., and Philip, R.P., Geochemical characterization of gases from the Mississippian Barnett shale, Fort Worth Basin, Texas, AAPG Bull., 94, 1641–56, doi:10.1306/04061009119, 2010.

Rooze, J., Egger, M., Tsandev, I., and Slomp, C.P., Iron-dependent anaerobic oxidation of methane in coastal surface sediments: Potential controls and impact, Limnol. Oceanogr., 61, S267–S282, doi:10.1002/lno.10275, 2016.

30 Schaefer, H., Mikaloff-Fletcher, S.E., Veidt, C., Lassey, K.R., Brailsford, G.W., Bromley, T.M., Dlugokencky, E.J., Michel, S.E., Miller, J.B., Levin, I., Lowe, D.C., Martin, R.J., Vaughn, B.H., and White, J.W.C., A 21st century shift from fossil-fuel to biogenic methane emissions indicated by  $^{13}\text{CH}_4$ , Science, 352, 80–84, doi:10.1126/science.aad2705, 2016.

**Formatted:** Font color: Text 1

**Formatted:** Font color: Text 1

**Formatted:** Font: 10 pt

**Formatted:** Line spacing: 1.5 lines

**Formatted:** Font: +Body (Times New Roman)

**Formatted:** Font: 10 pt

**Formatted:** Font: 10 pt

**Formatted:** Font: +Body (Times New Roman)

**Formatted:** Font: +Body (Times New Roman)

**Formatted:** Font: 10 pt

**Formatted:** Font: +Body (Times New Roman)

**Formatted:** Font: 10 pt

**Formatted:** Font: +Body (Times New Roman)

**Formatted:** Font: 10 pt

**Formatted:** Font: +Body (Times New Roman)

**Formatted:** Font: 10 pt

**Formatted:** Font: +Body (Times New Roman)

**Formatted:** Font: 10 pt

**Formatted:** Font: +Body (Times New Roman)

**Formatted:** Font: 10 pt

**Formatted:** Font: +Body (Times New Roman)

**Formatted:** Font: 10 pt

**Formatted:** Font: +Body (Times New Roman)

**Formatted:** Font: 10 pt

**Formatted:** Font: +Body (Times New Roman)

**Formatted:** Font: 10 pt

**Formatted:** Font: +Body (Times New Roman)

**Formatted:** Font: (Default) +Body (Times New Roman)

**Formatted:** Font: (Default) +Body (Times New Roman)

Schlegel, M.F., McIntosh, J.C., Bates, B.L., Kirk, M.F., and Martini, A.M., Comparison of fluid geochemistry and microbiology of multiple organic-rich reservoirs in the Illinois Basin, USA: Evidence for controls on methanogenesis and microbial transport, *Geochim. Cosmochim. Acta.*, 75, 1903-1919, doi: 10.1016/j.gca.2011.01.016, 2011.

Formatted: Font: 10 pt

Formatted: Line spacing: 1.5 lines

Formatted

Field Code Changed

Formatted: Font: 10 pt

5 Schneising, O., Burrows, J.P., Dickerson, R.R., Buchwitz, M., Reuter, M., and Bovensmann, H., Remote sensing of fugitive emissions from oil and gas production in North American tight geological formations, *Earth's Future*, 2, 548-558, doi:10.1002/2014EF000265, 2014.

10 Schoell, M., Lefever, J.A., and Dow, W., Use of maturity-related changes in gas isotopes in production and exploration of Bakken shale plays, *AAPG Search and Discovery Article #90122©2011, AAPG Hedberg Conference, December 5-10, 2010, Austin, Texas,* [http://www.searchanddiscovery.com/abstracts/pdf/2011/hedberg-beijing/abstracts/ndx\\_schoell.pdf](http://www.searchanddiscovery.com/abstracts/pdf/2011/hedberg-beijing/abstracts/ndx_schoell.pdf), 2011.

Schneising, O., Burrows, J.P., Dickerson, R.R., Buchwitz, M., Reuter, M., and Bovensmann, H., Remote sensing of fugitive emissions from oil and gas production in North American tight geological formations, *Earth's Future*, 2, 548-558, doi:10.1002/2014EF000265, 2014.

Formatted

Formatted: Font: 10 pt

15 Schwietzke, S., Sherwood, O.A., Bruhwiler, L.M.P., Miller, J.B., Etiope, G., Dlugokencky, E.J., Michel, S.E., Arling, V.A., Vaughn, B.H., J.W.C White, and Tans, P.P., Upward revision of global fossil fuel methane emissions based on isotope database, *Nature*, 538, 88-91, doi:10.1038/nature19797, 2016.

20 Sherwood, O.A., Schwietzke, S., Arling, V.A., and Etiope, G.: Global inventory of gas geochemistry data from fossil fuel, microbial and burning sources, version 2017. *Earth Syst. Sci. Data*, 9, 639-656, doi:10.5194/essd-9-639-2017, 2017.

Shindell, D., The social cost of atmospheric release, *Climatic Change*, 130, 313-326, doi:10.1007/s10584-015-1343-0, 2015.

25 Shindell, D., Kuylenstierna, J.C., Vignati, E., van Dingenen, R., Amann, M., Klimont, Z., Anenberg, S.C., Muller, N., Janssens-Maenhout, G., Raes, R., Schwartz, J., Falvegi, G., Pozzoli, L., Kupiainent, K., Höglund-Isaksson, L., Emberson, L., Streets, D., Ramanathan, V., Kicks, K., Oanh, N.T., Milly., G., Williams, M., Demkine, V., and Fowler, D., Simultaneously mitigating near-term climate change and improving human health and food security, *Science*, 335, 183-189, doi:10.1126/science.1210026, 2012.

30 Tilley, B., and Muehlenbachs, K., Isotope reversals and universal stages and trends of gas maturation in sealed, self-contained petroleum systems, *Chem. Geol.*, 339, 194-204, doi:10.1016/j.chemgeo.2012.08.002, 2013.

Formatted: Font: (Default) +Body (Times New Roman)

Formatted

Formatted: Font: 10 pt

Tilley, B., McLellan, S., Hiebert, S., Quartero, B., Veilleux, B., and Muehlenbachs, K., Gas isotope reversals in fractured gas reservoirs of the western Canadian Foothills: Mature shale gases in disguise, *AAPG Bull.*, 95, 1399–1422, doi:10.1306/01031110103, 2011.

5 Townsend-Small, A., Marrero, J.E., Lyon, D.R., Simpson, I.J., Meinhardt, S., and Blake, D.R., Integrating source apportionment tracers into a bottom-up inventory of methane emissions in the Barnett shale hydraulic fracturing region, *Env. Sci. Tech.*, 49, 8175–8182, doi:10.1021/acs.est.5b00057, 2015.

10 Turner, A.J., Jacob, D.J., Benmergui, J., Wofsy, S.C., Maasakker, J.D., Butz, A., Haekamp, O., and Biraud, S.C., A large increase in U.S. methane emissions over the past decade inferred from satellite data and surface observations, *Geophysical Res. Letters*, 43, 2218–2224, doi:10.1002/2016GL067987, 2016.

15 Turner, A.J., Frankenberg, C., Wennber, P.O., and Jacob, D.J., Ambiguity in the causes for decadal trends in atmospheric methane and hydroxyl, *P. Natl. Acad. Sci. USA*, 114, 5367–5372, doi:10.1073/pnas.1616020114 PNAS, 2017.

20 Vaughn, T.L., Bella, C.S., Picering, C.K., Schwietzke, S., Heath, G.A., Pétron, G., Zimmerle, D.J., Schnell, R.C., and Nummedal, D., Temporal variability largely explains top-down/bottom-up difference in methane emission estimates from a natural gas production region, *P. Natl. Acad. Sci. USA*, 115, 11712–11717, doi:10.1073/pnas.1805687115, 2018.

25 Whelan, J.K., Oremland, R., Tarata, M., Smith, R. Howarth, R., and C. Lee, C., Evidence for sulfate reducing and methane producing microorganisms in sediments from sites 618, 619, and 622, *Reports of the Deep-Sea Drilling Project*, 47, 767–775, doi:10.2973/dsdp.proc.96.147.1986, 1986.

Worden, J.R., Bloom, A.A., Pandey, S., Jiang, Z., Worden, H.M., Walter, T.W., Houweling, S., and Röckmann, T., Reduced biomass burning emissions reconcile conflicting estimates of the post-2006 atmospheric methane budget, *Nat. Communic.*, 8, 2227, doi:10.1038/s41467-017-02246-0, 2017.

30 Wunch, D., Toon, G.C., Hedin, J.K., Vizenor, N., Roehl, C.M., Saad, K.M., Blavier, J.L., Blake, D.R., and Wennberg, P.O., Quantifying the loss of processed natural gas within California's South Coast Air Basin using long-term measurements of the ethane and methane, *Atmos. Chem. Phys.*, 16, 14091–14105, doi:10.5194/acp-16-14091-2016, 2016.

Zumberge, J., Ferworn, K., and Brown, S., Isotopic reversal (“rollover”) in shale gases produced from the Mississippian Barnett and Fayetteville formations, *Mar. Petrol. Geol.*, 31, 43–52, doi:10.1016/j.marpetgeo.2011.06.009, 2012.

**Formatted**  
Formatted: Line spacing: 1.5 lines  
Formatted: Font: +Body (Times New Roman)  
Formatted: Font: +Body (Times New Roman)  
Formatted  
Field Code Changed  
Formatted  
Formatted: Font: 10 pt  
Formatted: Font: 10 pt  
Formatted: Line spacing: 1.5 lines  
Formatted: Font: (Default) +Body (Times New Roman)



← Formatted: Left, Line spacing: single

Table 1. Estimates for sources of increased or decreased methane emissions to the atmosphere in recent years (Tg per year).  
All values are positive, except as specified.

5

	This study <sup>a</sup>	Worden et al. <sup>b</sup> (2017)	Schwietzke et al. <sup>c</sup> (2016)	
10	<b>All fossil fuels</b>	<b>20.17.84</b>	<b>16.45.5</b>	negative ~ 18
15	-- shale gas	<u>9.411.0</u> ( $\pm 0.7$ )		
20	-- conventional gas	<u>6.5.55</u> ( $\pm 0.4$ )		
25	-- coal	1.3		
	-- oil	1.6		
	<b>Biogenic sources</b>	<b>8.010.6</b> ( $\pm 1.0$ )	<b>12.0</b> ( $\pm 2.5$ )	~ 27

<sup>a</sup> Time period is 2008-2014 compared to 2000-2007.

<sup>b</sup> 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  $\delta^{13}\text{C}$  values shown in Figure 3-B.

<sup>c</sup> Time period is 2008-2014 compared to 2000-2007, with values from their Figure 4, using with the Schwietzke et al. (2016) data set and assuming a decrease in biomass burning of 3.7 Tg per year. Uncertainty is as shown in original publication.

<sup>c</sup> Time period is for 2003-2013 compared to 1985-2002, with values from their Figure 2B. Uncertainties are large, and only mean differences shown here.

35

Table 2. Exploration of sensitivity to assumptions for estimates of increase in global methane emissions in recent years (Tg per year). **Only mean values presented.**

	Base analysis <sup>a</sup>	Increased emission factor for shale gas (sensitivity test #1) <sup>b</sup>	Explicit consideration of shale oil (sensitivity test #2) <sup>c</sup>
5			
10	All fossil fuels <u>18.2</u> <u>22.8</u>	<u>20.4</u> <u>17.8</u>	— <u>218.0</u> <u>2.2</u>
15	-- all natural gas <u>5.3</u>	<u>47.5</u> <u>14.9</u>	— <u>15.1</u> <u>9.3</u> —
20	-- shale gas <u>9.7</u> <u>7.8</u> <u>9</u>	— <u>11.0</u> <u>9.4</u>	— <u>410.8</u> <u>3.8</u> —
25	-- conventional gas <u>5.4</u>	— <u>6.5</u> <u>5.5</u>	— <u>4.3</u> <u>5.5</u> —
30	-- all oil <u>5.5</u>	1.6	— <u>1.6</u> — <u>6.2</u> <u>4.9</u>
35	-- shale oil <u>0.7</u>	—	— <u>4.2</u> —
	-- conventional oil <u>1.3</u>	—	— <u>0.7</u> —
	-- coal <u>1.3</u>	—	— <u>1.3</u> —
	<b>Biogenic sources</b> <u>5.6</u>	<b><u>-10.6</u></b> <b><u>8.0</u></b>	<b>— <u>10.4</u> <u>6.2</u> —</b> <b><u>-</u></b> <b><u>10.2</u></b>

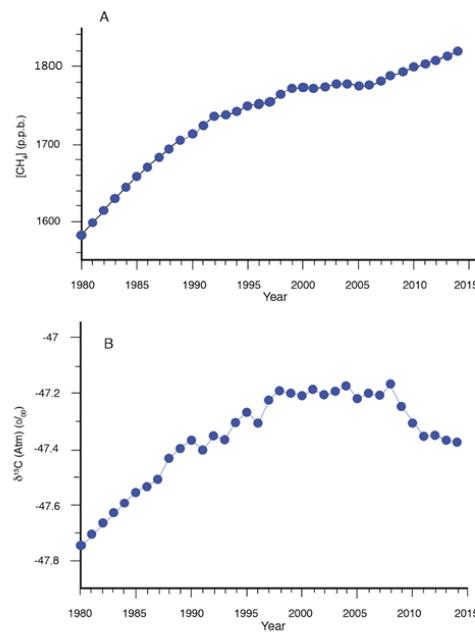
<sup>a</sup> Base analysis is from equations Eq. (1) through Eq. (109) and is also presented in Table 1. Assumptions include equivalent percentage emissions as a function of production for shale gas and conventional natural gas, and no contribution of <sup>13</sup>C-depleted

40 methane from tight shale oil production.

<sup>b</sup> Same assumptions as for the base analysis, except shale gas emissions are assumed to be 50% greater than those from conventional natural gas, expressed as a percentage of production.

<sup>c</sup> Same assumptions as for the base analysis, except emission of <sup>13</sup>C-depleted methane from shale oil is explicitly considered.

5

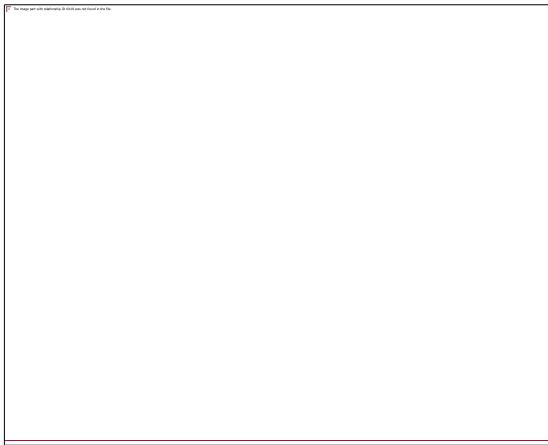


**F**

The image you uploaded is blank and contains no data.

Figure 1. A, top: global increase in atmospheric methane between 1980 and 2015. B, bottom: change in  $\delta^{13}\text{C}$  value of atmospheric methane globally between 1980 and 2015. Both adapted from Schaefer et al. (2015).

← Formatted: Left, Line spacing: single



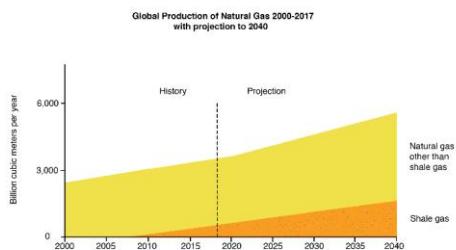
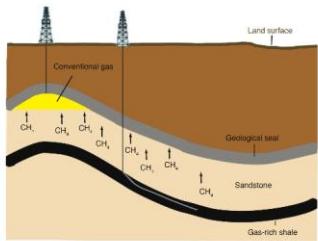
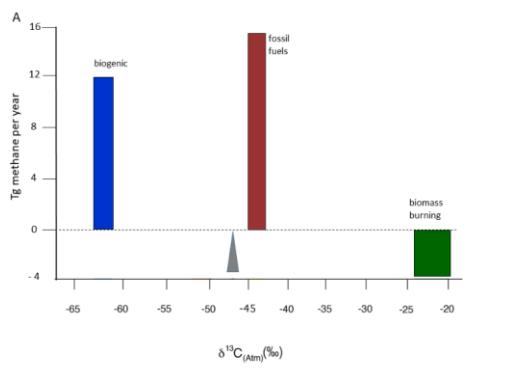
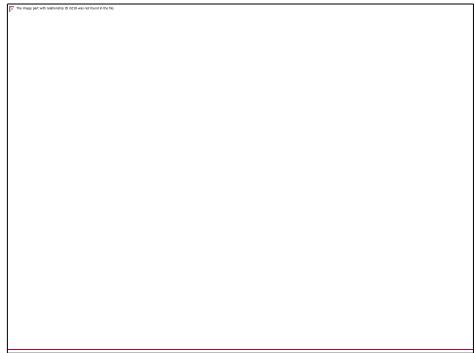


Figure 2. A, top: schematic comparing shale gas and conventional natural gas. For conventional natural gas, methane migrates from the shale through semi-permeable formations over geological time, becoming trapped under a geological seal. Shale gas

5 is methane that remained in the shale formation and is released through the combined technologies of high-precision directional drilling and high-volume hydraulic fracturing.

B, bottom: global production of shale gas and other forms of natural gas from 2000 through 2017, with projections into the future from EIA (2016). Redrawn from EIA (2016) with data from IEA (2017).



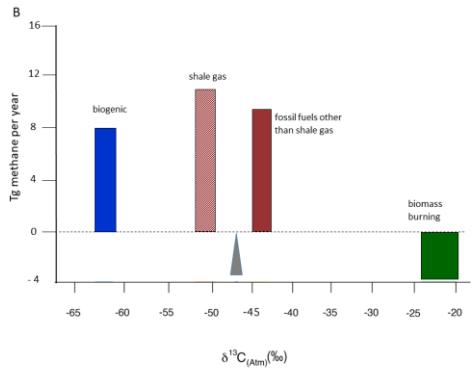
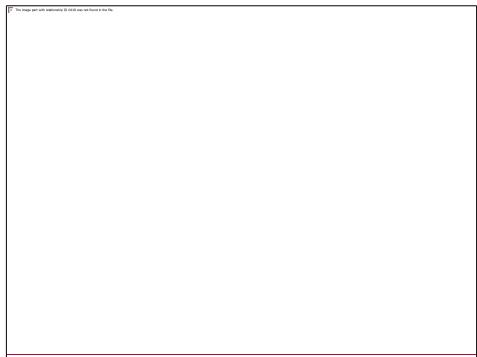


Figure 3. A, top: on the x-axis,  $\delta^{13}\text{C}$  values for methane from biogenic sources, fossil fuel, and biomass burning as presented in Worden et al. (2017) for values from Schwietzke et al. (2016); width of horizontal bars represent the 95% confidence limits

5 for these values. Triangle indicates the [average methane flux-weighted mean input of methane to the atmosphere in the atmosphere during the 2000 to 2008 period](#). The y-axis shows mean estimates from Worden et al. (2017) for the increase and decrease in methane emissions from particular sources since 200<sup>78</sup> as calculated using the  $\delta^{13}\text{C}$  values of Schwietzke et al. (2016).

B, bottom: on the x-axis,  $\delta^{13}\text{C}$  values as in Figure 3-A except the value for fossil fuels does not include shale gas, and a 10 separate estimate for shale gas value is included [based on the mean and 95% confidence level for 61 observations from three different shales and studies summarized by Golding et al. \(2013\): mean value is -51.4 ‰, with a 95% confidence limit of ± 4.2 ‰ \(see text\)](#). The y-axis indicates estimates developed in this paper for the increase or decrease in methane emissions since 2008.





Figure 4. A, top: gas blowdown for maintenance on a pipeline in Yates County, NY. While methane is invisible, the cooling caused by the blowdown condenses water vapor, leading to the obvious cloud. Photo courtesy of Jack Ossont.

5 B, bottom: Gas storage tanks receiving natural gas from feeder pipelines before compression for transport in high-pressure pipelines, Haynseville shale formation, Texas. Photo on left was taken with a normal camera. Photo on the right was taken with a FLIR camera tuned to the infra-red spectrum of methane, allowing visualization of methane, which is invisible in the normal camera view and to the naked eye. Photo courtesy of Sharon Wilson.