Using global isotopic data to constrain the role of shale gas production in recent increases in atmospheric methane

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The accelerated increase in global methane (CH₄) in the atmosphere, accompanied by a decrease in its δ¹³C/δ¹²C isotopic ratio (δ¹³C(CH₄)) from −47.1‰ to −47.3‰ observed since 2008, has been attributed to increased emissions from wetlands and cattle, as well as from shale gas and shale oil developments. To date both explanations have relied on poorly constrained δ¹³C(CH₄) source signatures. We use a dataset of δ¹³C(CH₄) from >1600 produced shale gas samples from regions that account for >97% of global shale gas production to constrain the contribution of shale gas emissions to observed atmospheric increases in the global methane burden. We find that US shale gas extracted since 2008 has volume-weighted-average δ¹³C(CH₄) of −39.6‰. The average δ¹³C(CH₄) weighted by US basin-level measured emissions in 2015 was −41.8‰. Therefore, emission increases from shale gas would contribute to an opposite atmospheric δ¹³C(CH₄) signal in the observed decrease since 2008 (while noting that the global isotopic trend is the net of all dynamic source and sink processes). This observation strongly suggests that changing emissions of other (isotopically-lighter) CH₄ source terms is dominating the increase in global CH₄ emissions. Although production of shale gas has increased rapidly since 2008, and CH₄ emissions associated with this increased production are expected to have increased overall in that timeframe, the simultaneously-observed increase in global atmospheric CH₄ is not dominated by emissions from shale gas and shale oil developments.

Methane (CH₄) is the third-most important greenhouse gas (after water vapor and carbon dioxide) and a significant contributor to global climate change1–8. Its globally averaged marine surface annual mean mole fraction in the atmosphere steadily increased from ~1600 parts per billion (ppb) to ~1775 ppb in the 1980–1990s, stabilized around ~1775 ppb during the period 1999–2006, and then returned to the earlier pattern of increases leading to ~1860 ppb in 2018.⁴,⁹,₁₀ There are anthropogenic (e.g., agriculture, wastes, fossil fuels, biomass burning) and natural (e.g., wetlands, freshwaters, geological seepage, wild fires) sources of CH₄ to the atmosphere. The carbon-isotopic composition of CH₄ (the ratio of stable isotopes ¹²C and ¹³C expressed as δ¹³C (‰) relative to the Vienna Pee Dee Belemnite standard), together with estimates of flux from each source-type, can be used to infer the relative contributions of various CH₄ emitters to the global budget by matching co-constrained global observations of CH₄ and its δ¹³C.⁴ Furthermore, temporal variations in δ¹³C of atmospheric CH₄ are a highly useful indicator of changes in the trends of various emitters and/or CH₄ sinks. The global mean atmospheric δ¹³C(CH₄) trended upward between 1980 (approximately −47.7‰) and 1997 (approximately −47.1‰), and remained relatively stable until 2008, before decreasing towards the most recent values of approximately −47.3‰.⁷,⁸

This recent increase in the atmospheric CH₄ burden, coincident with the depletion of ¹³C, has been attributed to the increasing contribution of biogenic CH₄ from wetlands and from agricultural activities such as cattle husbandry that produce CH₄ with δ¹³C usually more negative than −55‰.⁴,⁸ While atmospheric CH₄ sinks are less extensively studied, changes in the sink strength may at least partially explain some of the long-term observed trends in δ¹³C(CH₄).¹¹ Fossil fuel production also contributes to increasing atmospheric CH₄.¹²,¹³ However, CH₄ production...
in fossil fuels is, on average, enriched in $^{13}$C ($\delta^{13}$C $= -44.9‰$) relative to globally-averaged atmospheric CH$_4$. Decreasing $\delta^{13}$C of atmospheric CH$_4$ since 2008 implies that emissions from biogenic sources are therefore increasing at a greater rate relative to emissions from fossil fuels. However, recent studies have suggested that emissions from conventional petroleum developments and from shale gas/oil developments in particular (see Text S1 in Supplementary Information) have been the greatest single cause of the recent global increase of atmospheric CH$_4$. Here, we use a large global dataset of $\delta^{13}$C$_{CH4}$ from produced shale formations, which leads us to conclude that emissions from shale gas and oil production have not played a dominant role in the increase in atmospheric CH$_4$ since 2008.

Materials and Methods

Global isotopic dataset. We analyzed $\delta^{13}$C$_{CH4}$ data for 1619 samples of produced natural gas from 38 shale formations around the world originally presented in 73 studies (Table S1). This shale gas dataset is a subset of a larger global inventory of gas samples from conventional reservoirs, shales, coals, seeps and other geological settings originally published by Sherwood et al. and further expanded and discussed by Milkov and Etiope and Milkov et al. Although most gas samples come from formations dominated by true shale lithology (e.g., the Marcellus Formation, USA), we also include samples collected from unconventional low-permeability (tight) reservoirs dominated by very fine-grained sandstone or siltstone (e.g., the Montney Formation, Canada) or mixed clastic/carbonate lithologies (e.g., the Niobrara Formation, USA) developed through hydraulic fracturing and commonly included in the inventories of produced shale gas$^{17}$. The produced gas may be free gas associated with relatively little condensate liquids (e.g., in the Haynesville Formation) or oil-dissolved gas (e.g., in the Eagle Ford Formation). Most samples come from the USA (n=1238), followed by China (n=252), Canada (n=124), United Kingdom (n=2), Sweden (n=2) and Australia (n=1).

Calculation of weighted $\delta^{13}$C$_{CH4}$ values. Values of production volume-weighted $\delta^{13}$C$_{CH4}$ for shale gases were derived by first calculating the proportion of gas production from each shale formation in the total production, then multiplying that value by the average $\delta^{13}$C$_{CH4}$ for the corresponding shale formation, and then summing up the results. Emission volume-weighted $\delta^{13}$C$_{CH4}$ values were derived by first calculating the proportion of CH$_4$ emissions from each shale formation in total emissions, multiplying that value by average $\delta^{13}$C$_{CH4}$ for corresponding shale formation, and then summing up the results.

Results

The arithmetic mean $\delta^{13}$C$_{CH4}$ for all shale gas samples is $-41.3 \pm 0.2‰$ (n=1619, range from $-70.0‰$ to $-23.3‰$, median $-41.4‰$) (Fig. 1). The mean value is slightly more positive than $-42.5 \pm 0.3‰$ reported by Sherwood et al. based on a smaller dataset of 647 samples. Methane from produced shales is, on average, more enriched in $^{13}$C than CH$_4$ produced from conventional oil and gas reservoirs (mean $\delta^{13}$C$_{CH4} = -44.0 \pm 1.1‰$, n=6079 in the study of Sherwood et al.); mean $\delta^{13}$C$_{CH4} = -42.8 \pm 0.1‰$, n=12,697 in the study of Milkov et al.) and significantly more enriched in $^{13}$C than the modern atmospheric $\delta^{13}$C$_{CH4}$ ($-47.3‰$). We note that shale gas is even more enriched in $^{13}$C relative to the global average $\delta^{13}$C$_{CH4}$ (about $-54‰$) of all atmospheric sources prior to isotopic fractionation of atmospheric CH$_4$ by all sinks resulting in the modern atmospheric value above.

Global shale gas production increased from about 31 billion cubic meters (bcm) in 2005 to about 434 bcm in 2015$^{18}$. In the USA, the cumulative production of shale gas from 2000 to mid-2019 reached approximately 4.5 trillion cubic meters (tcm), including about 4.1 tcm produced since 2008 (Fig. 2, based on dry gas production). Half of the cumulative shale gas was produced from the Marcellus, Barnett and Haynesville formations. Figure 3 summarizes $\delta^{13}$C$_{CH4}$ data on gases produced from these and other principal shale formations in the USA.

In this study, we use the global $\delta^{13}$C$_{CH4}$ dataset to derive $\delta^{13}$C$_{CH4}$ representative of both produced gas (volume-weighted average) and the $\delta^{13}$C$_{CH4}$ signature when weighted for measured emissions across plays (emission-weighted average). Table 1 presents average $\delta^{13}$C$_{CH4}$ for the main producing shale plays in the USA. The 1002 available gas samples with $\delta^{13}$C$_{CH4}$ data are from plays that account for 94% of cumulative US shale gas production. The average $\delta^{13}$C$_{CH4}$ when weighted by the amount of cumulative production from each shale play during 2008–2019, is $-39.6‰$. A large proportion (28%) of cumulative shale gas production comes from the Marcellus Formation where CH$_4$ is significantly enriched in $^{13}$C (mean $\delta^{13}$C$_{CH4} = -32.0‰$, n=98). This latter source significantly influences the average volume-weighted isotope signature of CH$_4$ produced from shales in the USA.

The average shale $\delta^{13}$C$_{CH4}$ weighted by the amount of emissions measured in 2015 from the main USA shale plays is $-41.8‰$ (Table 2). Sensitivity analysis suggests that this value changes little when emission measurements from other years are considered (see Table S2, Text S3). We also calculated how the average $\delta^{13}$C$_{CH4}$ signature of shale-emitted gas changed over time. When weighted by production or emissions, the US average signature becomes heavier (thus, opposite to the direction of the atmospheric trend) by about $-4.7‰$ from 2000 to mid-2019 (Fig. 4). This is because the relative contribution of shales with relatively more positive $\delta^{13}$C$_{CH4}$ (e.g., Marcellus and Haynesville formations) to both production and emissions increased in that period.

Gas samples from three other countries currently producing shale gas commercially (Canada, China, Argentina) indicate somewhat more positive $\delta^{13}$C$_{CH4}$ values than the USA (Fig. 3), resulting in a global volume-weighted $\delta^{13}$C$_{CH4}$ signature of $-38.8‰$ (Table S3, Text S4). The volume- and emission-weighted $\delta^{13}$C$_{CH4}$ values calculated here do not account for shale plays for which production has become negligible after the 1990s (see Text S4).

The US shale $\delta^{13}$C$_{CH4}$ weighted by the amount of cumulative production from 2000 to mid-2019 for each shale play is $-40.0‰$ (Table S4). The mean $\delta^{13}$C$_{CH4}$ of produced shale gas in the USA since 2008 is $-39.6‰$ (Table 1). The slight (by 0.4‰) enrichment in $^{13}$C during 2008–2019, relative to 2000–2019, is due to a relatively larger contribution of production from the Marcellus and Haynesville formations in 2008–2019, and a smaller...
contribution from the Barnett Formation during that period. The mean δ¹³C.CH₄ of globally produced shale gas in 2018 is −38.9‰ (Table S3). We also estimated an emission-weighted δ¹³C.CH₄ of −41.8‰ for the principal US shale plays in 2015 (Table 2). These values are appropriate for utilization in models that constrain CH₄ emissions from shale developments to the atmosphere based on matching modelled global δ¹³C to observed δ¹³C. Methane from produced shales is, on average, significantly enriched in ¹³C relative to atmospheric CH₄ (δ¹³C.CH₄ ~ −47‰).

Discussion

Recently, atmospheric CH₄ became more abundant but also depleted in ¹³C, as δ¹³C decreased from about −47.1‰ in 2007 to −47.3‰ in 2017. If shale gas (with δ¹³C.CH₄ around −40‰ as documented in this study) and conventional oil and gas (with δ¹³C.CH₄ around −43‰) were conceived to collectively dominate recent emissions of CH₄ to the atmosphere, then atmospheric CH₄ would very simply become more enriched in ¹³C relative to the current global mean δ¹³C, which is not consistent with global observations. While we agree that

Figure 1. δ¹³C.CH₄ values from a global dataset of 1619 samples of produced shale gases from around the world. The data are displayed using a box plot, which shows distribution of values as histogram, average (mean) value (−41.3‰) as black star, median value (−41.4‰) as dotted line, first quartile (Q1), third quartile (Q3), lower adjacent value, upper adjacent value, and outliers. The first quartile (Q1) is the median of the lower half of the data set. This means that about 25% of the values in the data set lie below Q1 and about 75% lie above Q1. The third quartile (Q3) is the median of the upper half of the data set. This means that about 75% of the values in the data set lie below Q3 and about 25% lie above Q3. The lower adjacent value is the smallest observation that is greater than or equal to the lower inner fence, which is the first quartile minus 1.5 × IQR, where IQR stands for the interquartile range. The upper adjacent value is the largest observation that is less than or equal to the upper inner fence, which is the third quartile plus 1.5 × IQR. Outliers are all values that fall outside of either of the fences. Original data are in Table S1.

Figure 2. Cumulative production (in billion cubic meters or bcm) of dry gas from shale plays in the USA. Data are from Energy Information Administration. 

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shale developments (and fossil fuel in general) represent an important CH4 source, and that emissions from those sources have been likely increasing due to growing production, we conclude that the increases in global atmospheric CH4 concentrations since 2008 are not as strongly attributable to shale gas and conventional oil and gas emissions as some studies claim2,5, based on our global observations of isotopic fractionation.

Additionally, we must emphasize that the measured atmospheric δ13CCH4 signal is the sum-total of all CH4 source and sink terms. For example, a decrease in biomass burning emissions (significantly enriched in 13C (δ13CCH4 = −22.3 ± 1.9‰4), and an increase in fossil fuel emissions (including shale gas), could in principle result in the same global average atmospheric δ13CCH4 signal over time as if both sources had no trend1,3. The biomass burning category includes fires and solid biofuels (e.g., for use in cook stoves). Data on global CH4 emissions from fires is not entirely conclusive. Remote sensing data of CH4 and CO (and assuming (i) biomass burning CH4/CO emission ratios and (ii) a partitioning of CO emissions across sectors) suggests decreased fire CH4 emissions of

Table 1. Data used to calculate the average δ13CCH4 in shale gases produced in the USA since 2008. Production data are from Energy Information Administration17. See Text S2 in Supplementary Information for specific details on Permian, Utica and Niobrara-Codell samples. na – not available.

Figure 3. Main statistics on δ13CCH4 for gases produced from main shale formations in the USA, Canada and China. The data are displayed using box plots (see Fig. 1 for legend). Original data are in Table S1.
~3.7 Tg/yr from the 2001–2007 to the 2008–2014 periods. In contrast, remote sensing of burned fire area suggests no such trend (no trend over this period apart from inter-annual variation; Fig. S1). Furthermore, CH$_4$ emissions from solid biofuels are reported to have increased from 12.2 to 13.6 Tg/yr from 2000–2012 (latest time series available). While this data does not indicate an immediately apparent decrease in global biomass burning CH$_4$ emissions, more research is needed. Potential trends in the various CH$_4$ sink processes such as the soil sink and the tropospheric OH sink can further complicate the diagnosis of source trends. As a result, it is important to account for these processes, as well as other existing evidence such as latitudinal and seasonal CH$_4$ trends, when attributing the global signal.

From the above, it follows that attributing ~1/3 of the global CH$_4$ increase to North American shale gas production and another ~1/3 to conventional gas and oil with a simple mass balance approach is not supported by observations because of unconstrained uncertainties. Based on long-term airborne CH$_4$ measurements over the US, previous analysis concludes that oil and gas industry CH$_4$ emissions (shale and conventional) over the past decade have increased at about the same rate as natural gas production volume. The existence of unaccounted and poorly characterized emission sources within the oil and gas industry has also been demonstrated through intensive field studies in the USA, and additional international studies paint a similar picture, although little independent measurement data exist for many world regions including the Middle East, the Former Soviet Union, and Africa. Further research targeted for these areas, in addition to changing biogenic sources and sinks, will serve to further constrain the conclusions made in this work.

Based on existing knowledge of CH$_4$ source and sink terms and isotopic signatures, additional CH$_4$ emissions associated with increased shale gas development in the USA cannot account for a large fraction of the recent increase in atmospheric CH$_4$. Yet, oil and gas industry expansion remains a significant factor in the complex patterns of global atmospheric CH$_4$ emissions and concentrations. And, of equal importance, fossil fuel CH$_4$ sources may be mitigated with policy and best (or better) industrial practice that can effectively reduce emissions. We suggest that the rise in global CH$_4$ concentrations is most effectively seen not through a lens of what is the most important or dominant source of emissions, but rather understanding all sources and how they can collectively explain the observed patterns of atmospheric increases. Indeed, a reduction in emissions from any major source (such as fossil fuels or cattle husbandry) would be expected to lead to a reduction in the global CH$_4$.

### Table 2

Data used to calculate the emission-weighted average $\delta^{13}$C of CH$_4$ emitted from, mostly, shale gas production in selected plays and areas in the USA in 2015. Gas production, percentage of emitted gas, and CH$_4$ emissions are from Peischl et al. and references therein. These results account for ~60% of total USA shale gas production in 2015.

| Shale formation and basin/area | Gas production (m$^3$/d) | Emitted gas (% of production) | CH$_4$ emissions (tones/hr) | Average $\delta^{13}$C (%o) of emitted CH$_4$ |
|-------------------------------|--------------------------|------------------------------|---------------------------|----------------------------------|
| Eagle Ford                    | 1.54E+08                 | 2.5                          | 83                        | −42.9                            |
| Haynesville                   | 1.50E+08                 | 1                            | 42                        | −39.5                            |
| Marcellus (N.E. PA)           | 1.40E+08                 | 6.4                          | 18                        | −32.0                            |
| Barnett                       | 1.20E+08                 | 1.5                          | 46                        | −42.7                            |
| Fayetteville                  | 6.80E+07                 | 1.5                          | 31                        | −38.2                            |
| Niobrara-Codell (Denver)      | 3.80E+07                 | 2.1                          | 18                        | −47.6                            |
| Bakken                        | 3.70E+07                 | 5.4                          | 29                        | −47.3                            |
| **Total**                     | **7.07E+08**             | **1.6**                      | **267**                   | **−41.8**                       |

### Figure 4

Increasing $\delta^{13}$C$_{CH_4}$ signature of produced and emitted gas from shale developments in the USA from 2000 to mid-2019. Only formations with sufficient isotopic data (Marcellus, Barnett, Haynesville, Permian, Eagle Ford, Fayetteville, Woodford, Niobrara-Codell and Bakken) and emission data (Marcellus, Barnett, Haynesville, Eagle Ford, Fayetteville, Niobrara-Codell and Bakken) are used to construct this figure.
Conclusions

CH$_4$ recently increased in the atmosphere and simultaneously became more depleted in $^{13}$C. In this study, we compiled a large global dataset of isotopic composition of CH$_4$ produced from shale formations that account for most global shale gas production. Developments of shale gas and oil on average emit CH$_4$ significantly more enriched in $^{13}$C than the atmospheric CH$_4$ signal. Given current knowledge of global isotopic data and processes, the increase in US shale oil and gas apparently does not dominate the recent increased emissions of global CH$_4$ to the atmosphere. It is important to understand all sources of CH$_4$ that collectively contribute to recent atmospheric increases, and isotopic data provide key constraints for this.

Data availability

The dataset used in this study is available as Supplementary information.

Received: 6 December 2019; Accepted: 19 February 2020;
Published: 6 March 2020

References

1. Nisbet, E. G. et al. Very strong atmospheric methane growth in the 4 years 2014–2017: Implications for the Paris Agreement. Global Biogeochem. Cy. 33, 318–342, https://doi.org/10.1029/2018GB006009 (2019).
2. Howarth, R. W. Ideas and perspectives: Is shale gas a major driver of recent increase in global atmospheric methane? Biogeoosciences 16, 3033–3046, https://doi.org/10.5194/bg-16-3033-2019 (2019).
3. Schaefer, H. et al. A 21st century shift from fossil-fuel to biogenic methane emissions indicated by $^{13}$CH$_4$, Science 352, 80–84, https://doi.org/10.1126/science.aad2705 (2016).
4. Schwietzke, S. et al. Upward revision of global fossil fuel methane emissions based on isotope database. Nature 538, 88–91, https://doi.org/10.1038/nature19797 (2016).
5. Worden, J. R. et al. Reduced biomass burning emissions reconcile conflicting estimates of the post-2006 atmospheric methane budget. Nat. Commun. 8, 2227, https://doi.org/10.1038/s41467-017-02246-0 (2017).
6. White, J., Vaughn, B. & Michel, S. E. Stable isotopic composition of atmospheric methane (13C) from the NOAA ESRL Carbon Cycle Cooperative Global Air Sampling Network, 1998–2018. University of Colorado, Institute of Arctic and Alpine Research (INSTAAR), ftp://afdp.cmdl.noaa.gov/data/traces/gch4c13-flask/ (2019).
7. Lan, X. et al. Long-term measurements show little evidence for large increases in total U.S. methane emissions over the past decade. Geophys. Res. Lett. 46, 4991–4999, https://doi.org/10.1029/2018GL081731 (2019).
8. Intergovernmental Panel on Climate Change. 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, chap. 8, 659–740 (2013).
9. Nisbet, E. G. et al. Rising atmospheric methane: 2007–2014 growth and isotopic shift. Global Biogeochem. Cy. 30, 1356–1370, https://doi.org/10.1002/2016GB005406 (2016).
10. Dlugokencky, E. Global CH$_4$ monthly means, NOAA/ESRL. Available at: www.esrl.noaa.gov/gmd/ccgg/trends_ch4/ (Accessed: 11th October 2019).
11. Rigby, M. et al. Role of atmospheric oxidation in recent methane growth. Proc. Natl. Acad. Sci. USA 114, 5373–5377, https://doi.org/10.1073/pnas.1616426114 (2017).
12. Rice, A. L. et al. Atmospheric methane isotopic record favors fossil sources flat in 1980s and 1990s with recent increase. Proc. Nat. Acad. Sci. 113, 10791–10796, https://doi.org/10.1073/pnas.1522923113 (2016).
13. Saunois, M. et al. The global methane budget 2000–2012. Earth Syst. Sci. Data 8, 697–751 (2016).
14. Sherwood, O. A., Schwietzke, S., Arling, V. A. & Etiope, G. Global inventory of gas geochemistry data from fossil fuel, microbial and biomass burning sources, version 2017. Earth Syst. Sci. Data 9, 659–666 (2017).
15. Milkov, A. V. & Etiope, G. Revised genetic diagrams for natural gases based on a global dataset of >20,000 samples. Org. Geochem. 125, 109–120, https://doi.org/10.1016/j.orggeochem.2018.09.002 (2018).
16. Milkov, A. V., Faiz, M. & Etiope, G. Geochemistry of shale gases from around the world: Composition, origins, isotope reversals and rollovers, and implications for the exploration of shale plays. Org. Geochem. in press https://doi.org/10.1016/j.orggeochem.2020.103997 (2020).
17. Environmental Information Administration. Available at: https://www.eia.gov/naturalgas/weekly/ (Accessed: 22nd August, 2019).
18. Environmental Information Administration. Shale gas production drives world natural gas production growth. Available at: https://www.eia.gov/todayinenergy/detail.php?id=27512 (Accessed: 3rd October, 2019).
19. Peischl, J. et al. Quantifying methane and ethane emissions to the atmosphere from central and western U.S. oil and natural gas production regions. J. Geophys. Res. Atmos. 123, 7725-7740, https://doi.org/10.1029/2018JD028622 (2018).
20. Global Fire Emissions Database. Available at: https://www.gevo.vu.nl/~gwerf/GFED/GFED4/tables/GFED4.1s_CH4.txt (Accessed: 15th February 2020)
21. EDGARv4.3. European Commission, Joint Research Centre (JRC)/Netherlands Environmental Assessment Agency (PBL). Emission Database for Global Atmospheric Research (EDGAR), release version 4.3. Available at: https://edgar.jrc.ec.europa.eu/overview.php?v=432 (Accessed: 16th February 2020).
22. Ni, X. & Groffman, P. M. Declines in methane uptake in forest soils. Proc. Natl. Acad. Sci. USA 115, 8587–8590, https://doi.org/10.1073/pnas.1807377115 (2018).
23. Alvarez, R. A. et al. Assessment of methane emissions from the US oil and gas supply chain. Science 361, 186–188, https://doi.org/10.1126/science.aar7204 (2018).
24. Johnson, M. R., Tyner, D. R., Comley, S., Schwietzke, S. & Zavala-Araiza, D. Comparisons of airborne measurements and inventory estimates of methane emissions in the Alberta upstream oil and gas sector. Environ. Sci. Technol. 51, 21, https://doi.org/10.1021/acs.est.7b03525 (2017).
25. Yacovitch, T. I. et al. Methane emissions in the Netherlands: The Groningen Field. Elem. Sci. Anth. 6, 57, https://doi.org/10.1525/elementa.308 (2018).

Acknowledgements

This work was supported by Gates Foundation Environmental Development Fund at Colorado School of Mines.
Author contributions
A.V.M. conceived the study, compiled the shale gas dataset and calculated reported statistical values. S.S. compiled and evaluated the shale emissions dataset. A.V.M., S.S., G.A., O.A.S. and G.E. discussed the data, interpreted the results and wrote the paper with input from all authors.

Competing interests
A.V.M. worked in petroleum industry (BP, Sasol and Murphy Oil) for 13 years as geoscientists and manager. He is Director of Potential Gas Agency (PGA) at Colorado School of Mines, and PGA receives financial support from oil&gas companies, gas pipeline companies and distributors, and trade associations. He also regularly teaches technical courses at oil&gas companies and consults them on the issues of petroleum geochemistry and petroleum exploration. The other authors do not have any competing interest.

Additional information
Supplementary information is available for this paper at https://doi.org/10.1038/s41598-020-61035-w.

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