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# *Interactive comment on* "Global Inventory of Gas Geochemistry Data from Fossil Fuel, Microbial and Biomass Burning Sources, Version 2017" *by* Owen A. Sherwood et al.

**Owen A. Sherwood et al.**

owen.sherwood@colorado.edu

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Reviewer 1: Abstract: the database DOI could be given more prominence by putting it on a separate line, and then mentioning it at the start of the main text. -We follow ESSD conventions on placement of the database DOI.

L54 – Bruhwiler et al. could be mentioned here. -Reference added

L65 – Reference Rigby et al. 2017, Turner et al. 2017, Nisbet et al. 2016. -References added

L72 – 'increases' – in the box model – not necessarily in real nature. -Wording



changed: "… a 5 ‰ downward adjustment in the global weighted average fossil fuel $\delta$13CCH4 source signature increases modeled estimates of anthropogenic fossil fuel emissions…"

L86 – the sharp criticism of Dlugokencky et al. 2011 for being without reference to primary data is a bit unfair: : :look for example at their Fig. 3. -Wording softened; Dlugokencky et al. omitted from citations.

L87 – no fossil-fuel CH4 measurements? – what about Zazzeri et al on coal? And there is lots of industry information, as L91 makes clear. -We apologize for the clumsy wording. Sentence omitted completely.

L133 – "isotopic signatures are unaffected by gas processing" – this is rather sweeping. It depends what is done to the gas. -Changed to: "In comparison with the intentional changes of the molecular composition of natural gas, isotopic signatures are thought to be relatively unaffected by gas processing"

L220 – no quality assessment – this is a key problem. Inverse modellers seem happy with accuracy to 1 or 2 per mil, but some numbers are likely worse than that, as the paper points out in L224. In particular for Siberia's gas, a few numbers from the academic studies of the gas industry are probably more valuable than a large database of inaccurate data. -We thank the reviewer for their insights on data quality. While it is true that "a few numbers from high quality studies are probably more valuable than a large database of inaccurate data", there were no objective criteria by which to cull low quality data consistently across the entire database. As a case in point: the AAPG Bulletin is a rich source of gas geochemistry data; but, as a journal intended for "practicing geologists", AAPG Bulletin papers provide few details on analytical methods and therefore no way to evaluate data quality. Instead, we compiled a large database of uneven quality, in which "By sheer number of samples…, systematic errors inherent to any single dataset average out over the whole database…". We also warn that "We make no attempts to correct for these systematic errors; rather we caution users of this database to evaluate and use the data appropriately".

Maybe check the old Meth-MonitEUr report from 2005. -To the best of our knowledge, the Meth-MonitEUr program was concerned with measurements of background atmosphere – they did not directly sample point source emissions.

But note also that some work – such as Galimov and Rabbani 2001 – is very valuable indeed (for example for the giant Iranian Pars field). -Data from Galimov and Rabbani (2001) are included in the database.

Probably should also cite the old classic by Grace J.D., and Hart G. F. 1986. Giant gas fields of northern west Siberia. American Association of Petroleum Geologists Bulletin, 70: 830-852. -Grace and Hart (1986) did not explicitly address data quality. The Mikov (2010) paper that we cite provides a more recent and comprehensive description of Soviet era data problems.

L320 – This line may need a bit of rewriting: hard to understand what exactly is meant by samples in the abiotic field not overlapping with thermogenic. Note that coal mines can give an enormous range within the same mine. -Sentence reworded

L328 – isotopic signatures of biomass burning emissions vary significantly depending on the C3 to C4 mix. Thus African savanna grasses can give very heavy methane, while bushes nearly can emit methane that is significantly more negative in d13C. -This is addressed in section 3.5

L390 – C3/C4 proportion is very important here – because the seasonal African C4 grass fires are so major in the MODIS monitoring, and have such a large leverage on global isotopic balance. -Agreed. "Biomass burning falls furthest from the hinge point (mean ïĄď13CCH4 = -26.2 ± 4.8 ‰ unweighted by proportion of C3 and C4 vegetation), therefore it has the most leverage on the isotopic mass balance".

L404 – this is the unweighted mean for sources, not for emissions. -Agreed. We provide the "unweighted mean $\delta$13CCH4 for conventional natural gas", not the emissions



from this source.

L410 – for coal the jury is probably still out. The problem is that there is not enough information from the Chinese coal industry, which has grown extremely rapidly (see BP Energy reports), and where the older less-modernised mines probably dominate methane emissions (but we don't know!). The 2007 World Energy Council data, presumably complied some years earlier, must surely be very out of date now. Note also that Australia and South Africa are very large-scale open cast producers nowadays. -Agreed. However, we are limited to coal production data that can be accessed at present.

L422 – these are important comments. -Thank you

L427 emissions (: : :) are– not emissions (: : :.) 'is'. -Corrected

L437-438 – is this a fair and supported comment? How about the alternative hypothesis that the frackers realised after Karion et al and Pétron et al that they were leaking a lot of money to the sky, and then – given the competitive impacts of Saudi oil price cutting – the frackers did a lot to cut their emissions, to cut costs and boost profits in a tough market? -Not clear what the reviewer objects to here. Our argument does not compare "fracking" with conventional oil and gas production; instead it compares dry gas fields with wet gas fields, which exist for both "fracking" and conventional oil and gas production. We wrote: "A basin with mature dry gas and no associated oil production (and thus relatively heavy $\delta$13CCH4) typically employs less gas processing infrastructure (e.g. gas separators, combustors, storage tanks) than a basin with associated gas production (and thus relatively light $\delta$13CCH4). The former is therefore likely to emit less CH4 per unit of gas production than the latter." The text in italics has been added to clarify the meaning of "gas processing infrastructure". References are provided later in the paragraph to support the assertion. As for their alternative hypothesis, market forces could indeed accelerate the implementation of emissions controls, but this goes far beyond the scope of our ESSD database paper.

L443 Accent missing on Pétron. -Fixed throughout manuscript

L472 – shale gas. Very valuable. Most likely, each fracking play differs from the next, but they're probably mostly pretty heavy. -Agreed.

Reviewer 2: Overall, I thought the paper emphasized the fossil fuel sources and their isotopic composition too much – which, I understand that fossil fuel CH4 is the authors' "thing" and that fossil fuels are the hot topic right now. But despite the inclusion of all of the alkane ratios and even the isotopic ratios of some of the alkanes in the database, these are not discussed in the paper. -We focus on a description of the isotopic ratios because of their prominence in the CH4 inversion literature. For the fossil fuel part of the database, alkane ratios and isotopic ratios of the alkanes are included as a "bonus" set of parameters that may prove useful to the wider community.

Also, the data are heavily weighted toward fossil fuel sources: the amount of biogenic data in the database is so much less, and so much older! Where are all of the isotopic measurements of Arctic methane sources in your biogenic data, just as one example? People have been making measurements of methane emitted from lakes and tundra there for decades. Biasing your database toward fossil fuel sources doesn't really help your argument that models have relied on "outdated source signature values". Much more recent data are available in this area (see below for some suggestions). -Agreed that our database is more heavily weighted to fossil fuel sources as compared to microbial sources. We will correct this deficiency and the low number of Arctic data in future versions of the database. However, even the microbial data (n = 1972 individual datapoints from 41 references and 19 countries; note that we corrected the country count from 15 to 19 in the conclusions) far exceeds in number and geographic coverage any other compilation of microbial source signature data in the literature. We thank the reviewer for the list of additional data sources. We will incorporate these data into the next version of the database. To avoid version confusion and to maintain consistency with the database used in Schwietzke et al. (2016), we opt not to make any further additions to the current (2017) version of the database.



Specific comments: Lines 27-28: "Most global models: : :": this is a strong statement – some of us use recent measurements of sources, sometimes even measurements of sources we make ourselves! -Table 1 lists a large number of global scale inversion studies (up to 2016) and the source signature values used therein. It can be seen from Table 1 that virtually all of these studies reference older papers or do not specify the origin of the source signature values used.

Plus, as mentioned above, your biogenic data are not "globally representative" if you don't include Arctic wetlands and more recent sources. -Agreed that our database is weak on Arctic wetlands data. Please see response to reviewer's second comment above.

32: Permanent? -Changed to "Gas molecular compositions. . ."

34: What does mean (unweighted) mean? -Added "unweighted for production intensity" in parentheses.

35: be specific about the d13C values you calculate here versus what has been used previously -We purposely do not provide the actual numbers in the abstract, so that readers are not tempted to use those values without appreciating the difference between raw and weighted values, as discussed in the last paragraph of section 3.5.

35-36: "thus highlighting potential underestimation of fossil fuel CH4 emissions in previous CH4 budget models" – it is not clear how this conclusion was reached. -This conclusion is discussed in section 3.5 of the manuscript

47: today? This reference is from 1992. Please update. -Reference changed to Saunois et al. 2016

53-59: other papers have indicated that changes in CH4 consumption by the OH radical may be the cause -Sentence with Rigby et al. (2017) reference added to paragraph.

61: I know you wrote "in part", but you didn't define the counterpoint to top-down: the bottom-up inventory. -We do not feel that it is necessary to define bottom-up inventories in this paper.

75-86: I take issue with this somewhat. Many of these papers you cite were written by people who pioneered the measurements of methane isotopes and made the source signature measurements, so you should try not to disparage these older studies. The measurements were really, really hard to make back then (they are still not a picnic, and it's not fun when modelers think they are). -We thank the reviewer for these insights. Wording has been softened, as per comments from Reviewer #1. However, our message remains the same: the vast majority of global scale methane inversions have been using "canonical" and out of date source signature values, as shown in Table 1.

Finally, there have been efforts at least at the regional level to match source isotope/alkane measurements to top-down measurements in the same area. Townsend-Small et al., 2012; 2015; 2016 would be good examples. -Agreed, there has been progress at the regional scale. But global inversions are still using canonical values. Please see Table 1.

87-88: What do you mean, there have been no original measurements of fossil fuel isotopic signatures since 1994? Are you being serious? This is a review paper of fossil fuel isotopic measurements that you discuss in your next paragraph. -We apologize for the clumsy wording. Sentence omitted completely.

182-185: you cite global top-down studies that utilize C2:C1 in your introduction, and several of these studies that you cite do not use alkanes anyway. -Changed to "less commonly used". The inclusion of Townsend-Small (2016) was a mistake; reference omitted. The other four references all consider non-methane hydrocarbons.

189-200: The issue of instrument calibration is a huge problem here – each lab uses different calibration standards. I realize you can't really screen for that but you should note it somewhere in your paper -New paragraph on isotopic calibration added under section 2.5.



234-241: The database that NOAA sent me has 8734 rows for fossil fuel sources, as you note here, but only 107 for non-fossil sources. You wrote there are 1,972 non-fossil data records. -In section 2.3 we state: "…the literature on non-fossil fuel sources of CH4 more commonly reports statistical summaries (e.g., multiple measurements at a given location/time) as opposed to discrete sample data; because of this, the non-fossil data comprise n = 1973 measurements represented in 107 rows of data". Text in italics has been added for clarity.

380-389: I don't understand the "hinge point". Needs more explaining. -Reworded: "The $\delta$13C of the atmosphere before fractionation represents the "hinge point" upon which CH4 emissions fluxes are estimated by isotopic mass balance (e.g. Whiticar and Schaefer, 2007)". Note the reference to Whiticar and Schaefer: our figures 7 and 8 and the "hinge point" concept are modeled on their figures 5 and 6.

394: the +/-5‰ in d2H of CH4 is the measurement variability, not the variability of CH4 in the atmosphere. That's about as good as the measurement can get. -Changed "variability" to "uncertainty". For reference, in Quay 1999: "The global mean delta D of atmospheric CH4 between 1989 and 1995 was -86+3 per mil (n=95)."

395-398: still not understanding the "hinge point" aspect of this figure. -Please see comments above

417: Do you mean shale gas versus conventional gas? Or dry gas versus wet gas? Shales can have oil too. -Changed to: "Shale gas also exhibits lower mean $\delta$13CCH4 (-42.5 ‰ and $\delta$2HCH4 (-167 ‰ than indicated in the CH4 budget literature".

438-445: I'm not sure where you are going here – neither the Peischl, Karion, nor Petron papers used d13C for source apportionment in their papers, so I am failing to see how this is relevant. -The references to Peischl, Karion and Petron support the statement that dry gas plays have lower emissions than wet gas plays. "For example, the dry gas basins Marcellus Shale and Fayetteville are estimated to emit on average 0.3% and 1.9%, respectively, per unit of gas produced (Peischl et al., 2015), whereas the wet gas Denver and Uinta Basins emit on average 4.1% and 8.9%, respectively, per unit of gas produced (Karion et al., 2013; Petron et al., 2014)." It does not matter that they did not use $\delta$13C for source apportionment in this particular context.

451: you have not discussed the patterns in C2:C1 in the data at all in this paper (nor any of the other alkanes). -C2:C1 statistics have been omitted from the conclusions.

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