Earth Syst. Sci. Data Discuss.,
https://doi.org/10.5194/essd-2018-108-AC2, 2018

Earth System Science Data Discussions

Interactive comment on "Gridded maps of geological methane emissions and their isotopic signature" *by* Giuseppe Etiope et al.

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Received and published: 16 November 2018

Author comments to Anonymous Referee #2

R: Reviewer A: Author

GENERAL COMMENTS R: According to the abstract the global mean isotopic signature of geologic emissions is -48.5 to -49.4 per mil. In addition to the global average, a global map of fractionation factors is provided. The question is how those numbers were derived.

A: This is explained below (last comment in this page).

R: The section on on-shore seeps mentions (section 4.3) that it is a combination of

measurements and estimates. The measurements are 'as indicated in the literature' without references.

A: It was implicit that the "literature" reporting isotopic data is in the onshore seeps inventory, which is described in section 4.1. This is now clarified in the ms.

R: The estimates follow 3 rules that are listed without specifying which rule applies when.

A: The 3 rules are applied depending on the availability of the data. For clarity, we have rephrased as follows: ....or (b) estimated on the basis of isotopic values following one of the following three procedures, in priority order: - of similar seeps occurring in the same basin (when these data are available) - of reservoir gas in the same petroleum field, from Sherwood et al. (2017) dataset or literature - suggested by local petroleum geology (existence of microbial gas, thermogenic gas, oil), when the previous procedures cannot be applied.

R: A global mean is used in regions for which no data exist. It is unclear, however, where this applies.

A: Since onshore seeps are 2827, it is not possible to list in the text the seeps that have no δ13C value, also because the OS inventory cannot be provided due to license restrictions, as indicated in the Data Availability chapter. However, the regions with emission-weighted mean are shown in Figure 10.

R: The descriptions of the other subsections on isotopic information are similarly general, and unspecific.

A: For SS, the isotopic information is given in Table S4, as indicated in the text. For MS, we have described in detail the procedure (section 6.3), and some data are linked to OS inventory (which cannot be released for license restrictions). For GM, the isotopic data are shown in the GM dataset (available at https://doi.org/10.25925/4j3f-he27) and the derivation of the mean values is described in detail (section 7.3).

R: It is unclear how the range between -48.5 and -49.4 should be interpreted. Is this supposed to be an uncertainty range?

A. As indicated in Section 8.2, the value -48.5 is emission-weighted using gridded emissions, the value -49.4 uses global emission estimates. Since the gridded emissions are not complete (as explained in section 8.1) the second value is likely more realistic, as now clarified in the Conclusions (point (e)).

R: Some of the paragraphs dealing with uncertainty in d13C mention 15 per mil as a range of reported numbers. Then how is it possible to arrive at an uncertainty on the global fractionation within 1 per mil?

A: The global uncertainties of Table 5 refer to the uncertainty of global emission-weighted values, not to individual data. This is now clarified with a note at the bottom of the table. The 15 per mil uncertainty was mentioned (in Section 4.6) only for OS, referring to maximum uncertainty of individual estimated data. In Table 5, for OS we reported 1 per mil referring to an approximate expression of the uncertainty of the global emission-weighted mean, which is dominated by 76 big emitters, whose specific uncertainty is 0.1 per mil (see last line of Section 4.6). We adopted then 1 per mil (Table 5) as average order of magnitude of the uncertainty. This is now clarified in Section 4.6.

R: Important for the application of the geologic emission map to inverse modelling is the information on uncertainty that is provided. However, the method that is used to quantify uncertainties is questionable. For example, for some sources (e.g. OS and SS) it is stated that the uncertainty in the geographical distribution is practically zero. While this may apply to specific sources that have been located, there should be uncertainty from sources that have not yet been found. The question then becomes to what fraction of the emissions this may apply.

A: Uncertainties are now better described and assessed for all geo-sources (see additions in sections 4.6, 5.6 and 7.6). Of course, we refer only to the uncertainty of the

data provided. If we do not know whether and where other sources exist, how can we define an uncertainty?.

R: Section 4.6 on onshore seeps describes contributions to uncertainty, but does not provide a single number. For the uncertainty in the isotopic fractionation factor a maximum difference between estimates is mentioned, but it is unclear how representative that maximum is. Table 5 mentions ± 1 per mil, which seems quite accurate, but it is unclear where this number comes from.

A: OS uncertainties are now better described and assessed (see additions in section 4.6).

R: In the description of GM uncertainties, the emission estimates are discussed without providing a clue of what the uncertainties may be. To summarize: The treatment of uncertainties requires several clarifications.

A: GM uncertainties are now better described and assessed (see additions in section 7.6).

R: If I understand correctly the gridded emission maps do not account for the global extrapolation, i.e. they account for 37 Tg/yr of methane emissions.

A: right.

R: For the remainder no specific information is available, which raises the question what the extrapolation of almost a quarter of the emissions is based on. The assumptions underlying the extrapolation are not spelled out, which is critical not only for trying to understand how they were derived, but also to guide users of the emission dataset as to where missing emissions are to be expected (see my earlier remark about the uncertainty in the spatial distribution of the emissions). Besides a clearer description of the assumptions underlying the extrapolation, some discussion is needed of what are reasonable assumptions (in terms of geographical regions) to put the remaining sources or to account for their uncertainty.

A: The extrapolated emissions (and related assumptions) are explained in Table 3 note. Extrapolated emissions include factors that cannot be accounted in the gridding, such as MV eruptions, the OS foreseen but not included in the gridded inventory, unidentified or not-investigated offshore seepage. Geographical regions of these extra emissions (difference between gridded and extrapolated emission) can be understood reading the specific Sections describing the distribution of OS, SS, MS and GM.

R: Some discussion is needed of how to distribute the emissions not only spatially but also temporally. Without this information, modelers will probably assume that all emissions are constant over the year. Besides eruptions, for which you would obviously need to know the timing, continuous emissions may vary with environmental conditions (temperature, soil water content?). A few sentences of discussion would be useful to provide information to guide the choice of temporal distribution, and the uncertainty assigned to it (for atmospheric modelers it is relevant to know within what bounds continuous emission may vary over the year)

A: This is a good point. We were actually planning to add a short section discussing temporal geo-emission variations, also with reference to recent works reporting estimates of geo-CH4 emissions in the geological past (Younger Dryas and Preboreal intervals). The new section is added in the final revised version of the manuscript.

SPECIFIC COMMENTS R: Page 3, line 10: What are 'originally ad hoc developed datasets'? Which parts of the inventory are based on such data?

A: Everything is described in Section 4, 5, 6 and 7. This is now clarified in Introduction.

R: Page 4, line 8: '. . . reported in the supplement'. Add '(S6)'. OK

Page 4, line 14: How were the 'single OS, SS, MS, and GS shapefiles' derived? Aren't these just lists of coordinates of reported seeps? If so, this needs to be made clear to avoid the impression that unspecified information went into these shape files.

A: The shapefiles were in point or polygon format depending on the type of the source.

We have now added the following text for clarity:

Geo-CH4 emission and isotope datasets were imported in ArcGIS environment and saved in either point (OS and GM) or polygon (SS and MS) shapefile format, including coordinates and attributes (i.e., type of emission, area, emission factor, isotopic CH4 values, plus geographical information, such as country and region).

R: Page 5, line 13: '. . . listed without coordinates' Does this mean that no geographical information is provided at all?

A: Only the country or rough indication of the basin (region) is reported.

R: Page 5, line 15: 'The total number of 3439 OS represents about 30% . . .' Does this mean that the remainder is part of the 'global extrapolation'? This question applies as well to the 50% mentioned later in this paragraph.

A: yes, and this is indicated in Table 3.

R: Page 5, line 33: 'theoretical values were used'. The explanation that follows makes clear what these values account for, but it is unclear what the values are and based on which criteria they are assigned.

A: The values are reported in Table S1, as indicated in the text. The value assignment is based on experimental data (measured fluxes) and flux modelling (mainly depending on seep size) reported in various papers, listed in Etiope (2015). This is now clarified in the text.

Page 5, line 38: What is the difference between 'micro' and 'mini' seepage?

Microseepage is the diffuse seepage, independent of macro-seeps. Miniseepage is the diffuse seepage surrounding a macro-seep. Definitions are given in Etiope (2015) and this is now indicated in the text. In Section 2 we outlined that, for details on seepage processes and terminology, the reader may refer to a series of fundamental works.

R: Page 6, line 38: As discussed on Etiope et al, 2009, the isotopic signature of the

reservoir may be a poor indicator of the isotopic signature of the emissions due to fractionation due to advective segregation. In light of this, what is justifying the use of the reservoir signature here?

A: The reviewer has misinterpreted Etiope et al 2009. Advective segregation produces molecular fractionation but not isotopic fractionation. Etiope et al 2009 shows, in fact, that generally there is no substantial difference between reservoir and seep δ13C values.

R: Page7, line 8: '. . . because of multiple counting of 57 seeps . . .' but I thought the point of using ArcGIS was to deal with this kind of issues. Why not assign the emission proportional to area or something like that? There must have been an easy way to avoid double counting.

A: To our knowledge there is no way to assign the value of the point, located on the boundary of two or four cells, to one specific cell. The only way would be to shift the point, changing its coordinates. We preferred to avoid this procedure and keep the original coordinates, as the double counting is actually negligible compared to the uncertainty of the total emission.

R: Page 7, line 16: '. . . OS grids are not meant to update or refine . . .' but the reference is to a paper that was published 10 years ago. Table 1 lists data sources for category OS that are of more recent dates. What does this statement mean for those updates?

A: Those updates basically refer to the inventory (existence and location) of seeps, not necessarily to flux values. Almost all OS flux values for the grid are theoretically derived (for the reasons described in Section 4.2.1), so OS gridding cannot be considered an advance of previous OS emission estimates procedures.

R: Page 7, line 22: '. . .those estimates are indicated in the Table as upper limits . . .' Are marine MV larger emitters as onshore MV's? Otherwise I don't understand

this statement. It seems not obvious to me, since a fraction of the emissions from submarine MV's will be oxidized in the water column.

A: Shallow marine MV can be important emitters, but regardless, formally, for the comparison with gridded emissions, they should be considered as part of SS. The problem is that Dimitrov, 2003 and Milkov et al. 2003 did not distinguish clearly the emission from submarine MV and that from onshore MV. This is now clarified in section 4.5.1.

R: Page 8, line 10: The total mean value is just the average of all reported d13C values? A: YES

R: Since emission weighting seems such an obvious improvement of this estimate why are both estimates mentioned?

A: This is done to show how big emitters control the weighted-emission mean, leading to a final value of -46.6 per mil.

R: Page 8, line 35: '< 500 m deep . . . McGinnis et al, 2006' According to McGinnis it is unlikely that seeps from deeper than 100m can contribute significant amounts of methane to the atmosphere. Therefore, the threshold should be 100m instead of 500m.

A: We actually used a misleading reference. McGinnis et a. (2006) is just one of the several studies reporting variable depths on submarine seeps reaching the atmosphere. The 100 m threshold indicated by McGinnis et al (2006) is a minimum value, valid for some observations in the Black Sea, and it neglects bubble plume induced upwelling and excludes plume dynamics of large gas releases, as outlined in successive modeling (e.g., Schmale et al. 2009, doi: 10.1016/j.jmarsys.2009.10.003; Yamamoto et al 2009, doi:10.1016/j.epsl.2009.05.026) and direct observations (Solomon et al. 2009; doi: 10.1038/NGEO574). Greinert et al (2006, doi:10.1016/j.epsl.2006.02.011) and Solomon et al (2009) reported methane bubble columns ascending for >1000 m and >500m respectively. We used therefore an approximate maximum threshold of 500 m (<500 m). We now refer to Solomon et al (2009) in the revised manuscript.

R: Page 11, line 34: 'The similar order of magnitude . . . log normal behavior' If the mean and median are the same than this suggests rather a normal distribution. I don't see why the mean and median in the same order would point to a log normal distribution.

A: We wrote "geometric mean" not "mean".

R: Page 13, line 23: Implicit here is that the shallower reservoirs have a heavier isotopic signature than a deep reservoir. Is this generally true? If so, then why?

A: NO, the opposite. Shallower reservoirs have a lighter isotopic signature. For better understanding, the sentence has been rephrased as follows: . . ..the MS gas may actually come from shallower reservoirs, not necessarily or not dominantly from the deep productive reservoirs, which are more frequently the literature source of the isotopic value. Therefore, in some cells the real isotopic value could be lighter than that used in the grid maps.

R: Page 14, line 2: What explains the _10% difference between gridded and reported area in this case?

A: The difference depends on the cell size and it increases when cell size increases. The perfect match would be only when the grid cells are infinitely small.

Page 14, line 37: 'buffer applied to individual seeps' I don't understand what this means. A: This is explained in Supplement S3.2 (as now indicated in the text).

R:The reference should probably be to section 6.2.1. A: No, S3.2 (now corrected).

R: Page 15, line 16: The result of 2 different ways of averaging does not sound as a reliable estimator of uncertainty. This seems confirmed by 2 per mil being very small given the range of the fractionation values.

A: We considered it as "approximate expression of the uncertainty"; it is not easy to assess more precisely the uncertainty on those type of data. See also responses to

Reviewer #1 above.

R: Page 16, line 6: 'Accordingly, the total emission estimate . . . (Etiope et al, 2008)' Why is this? Because the accounting approach that is adopted here is not considered meaningful?

A: Simply because accounting approach adopted is not better than previous estimates. Our gridding operation had a specific objective, which is not necessarily the one of improving global emission estimates.

R: Page 17, line 4: What explains the exceptionally heavy isotopic signature of GM emissions?

A: Geothermal methane includes both over-mature thermogenic and abiotic gas, and both are typically 13C-enriched.

R: Page 18, line 2-5: If I understand correctly, the emission maps are referred to here as gridded emissions. It means that they only account for 37 Tg/yr.

A: YES

R: Page 18, line 10: The combined d13C uncertainty depends on the individual uncertainties. The more important question of how they are combined should also be answered.

A: Their combination is explained in the gridding procedure, Section 8.1

Page 19, line 23-24: 'It is expected that using the updated . . . (all else equal)' I don't understand why this would be the case. At -49 per mil atmospheric 13C is really insensitive to geological emissions as it is so close to the mean atmospheric composition.

A: The global source attribution (top-down either via simple box models or via 3D inverse models) includes all methane sources, whether their isotopic source signatures are "close" or "not close" to the atmospheric signal. In fact, these models quantitatively

estimate the magnitude of each source based on how close (on a continuum) each source is from the atmospheric signal.

Page 19, line 32-34: It is unclear to me why this would be the case (see my previous point).

A: Previous top-down studies have lumped fossil fuel industry and natural geologic seepage together for two reasons: (i) Source signatures were assumed to be the same, and (ii) source locations largely overlap. Hence, it has been difficult for any top-down model to distinguish the two in the absence of an a priori natural geologic seepage map. Additionally, global methane emissions from natural geologic seepage have been assumed to be minor in many previous top-down studies. As a result, almost the entire sum of both sources was previously attributed to fossil fuel industry.

R: Suppl. Page 9: '. . . considered for the text file' Which text file?

A: the csv MS grid file (now clarified).

R: Suppl. Fig S3: To what extend could the difference in slope between the two regression lines be explained by the use of the erroneous syringe method for the larger MV's (increasing the micro seepage would bring the lines closer together)

A: Difficult to say because both small and large MV data are underestimated

R: Suppl. Page 8: 'tested' or 'evaluated' i.o. 'checked'. The latter suggests that the validity of the sensitivity was verified using some external information, which is not the case.

A: OK, tested

TECHNICAL CORRECTIONS Page 6, line 31: 'emission' i.o. 'output' Page 8, line 37: 'emission' i.o. 'output' Page 6, line 24: 'Grubbs' i.o. 'Grubs' Table S3, caption: 'Azerbaijan' i.o. 'Azerbaiajn'

OK, all technical corrections have been done.

We thank the anonymous reviewer for the valuable and careful revision of the work

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