Author reply to Reviewers #1, #2, #3

We thank the three reviewers for their careful comments. The suggestions have greatly improved the manuscript! We have tried to address all of them in this document summarizing our replies and changes made to the document and the Supplement as a result.

R: Reviewer A: Author

Rev.#1

(…)

R: Inevitably there are shortcomings to this approach: â˘A ´c the number of source references (published papers, reports etc.) is vast: most, but not all, have been utilised. Table S4, for example, is not complete. â˘A ´c there is an enormous volume of data acquired by, for example, the petroleum industry, which has not been released to the public domain, so the data set cannot be comprehensive. â˘A ´c such data sets are never complete. As soon as a compilation is completed, more source references appear. For the above reasons it would perhaps be appropriate to recognise this a 'provisional' attempt to evaluate global geological methane emissions. However, I believe it be the only one available, and in its present form it is more than adequate to demonstrate the significance of geological sources of methane.

A: We agree with Reviewer#1, Table 4 (Submarine Seepage) may not be complete as it can only refer to published data (as indicated in Section 5.1); but we cannot include unpublished or confidential data from petroleum industry. We outline, however, that the data, for our purpose, must refer to methane emission into the atmosphere, not to the existence of submarine seeps or gas flux at seabed. We doubt that oil industry is interested in estimating the flux of methane at the sea surface. We can however better clarify the point by rephrasing the following sentence in Section 5.1.

A specific dataset….(…..).….was developed based exclusively on published literature (Table S4 in the Supplement).

R: Are there plans to maintain and update the data set?
A: Yes, the dataset can be updated when a significant number of new data will be available. This was already indicated at the end of the Conclusions.

Specific comments:

R: p3 l20: four major categories: is it worth noting here that other sources (e.g. deep water seeps) do occur, but are less likely to be responsible for direct methane emissions to the atmosphere?

A. In Section 5.1 we clarified that deep water seeps are not considered because of the limited or nil impact to the atmosphere. We have now clarified this point also in the Introduction, as suggested by the Reviewer.

R: P3 l23: Submarine (offshore) seeps: presumably this includes offshore mud volcanoes; if so this should be stated - if not they should have been included either here or in a separate category.
A: Yes, they include offshore mud volcanoes. This is now clarified in Section 5.1..

R: P3 l26: diffuse microseepage: presumably this category is exclusively onshore – this should be stated.
A: Yes, this is now clarified at the beginning of Section 6.1

R: P4 l 24 onshore cells without OS, GM or MS sources?
A: No, only, OS and GM; all onshore MS cells have a value

R: P16 l24 The double-sided Grubs test should either be explained or a reference should be provided.
A: OK reference is now provided

R: P16 l28 "as limit" – or "as the limit"?
A: "as the limit"

R: P25 l 14 (Table 5). The distribution of submarine seepages: "unknown % of global coverage (likely >80% ?)". What does this mean? If this means that more than 80% of the global distribution of submarine seeps is accounted for in the data set, I strongly dispute this. I suspect that many more than 20% of existing seeps and seep areas remain undiscovered (or are discovered but unreported).

A: We outline again that we are referring exclusively to shallow seeps (<300-400 m below sea level), with impact in the atmosphere. We considered conservatively 80% with a question mark; would 60 or 70% be more realistic?

R: The Supplement is first mentioned on p4 l8 –instructions on how to access it should be provided here and on p20 l20.
A: Following the editorial rule, the access to the Supplement must be indicated at the end of the manuscript. We did not know the doi during submission. I twill be added in the revised final version.

Technical comments:
General: â˘A ´c standardise "per": e.g. "Tg year-1" OR "Tg/y". â˘A ´c
p3 l11 "ad hoc" is correctly in italics. "et al." should also be in italics. â˘A ´c Several occurrences of "et al" should be corrected to "et al."
p2 l30: Bergamaschi et al. 2014: listed as 2014 in References
p3 l32: Etiope et al. (2007) not in References
p6 l 26-7: "OS emissions in the order of . . .. " Clumsy wording. Suggest: "There is a total of 76 OS with emissions in the order of 104 t CH4 year-1 ....."
p9 l1: California is in the USA!
p11 l10-11: Klusman et al. 2008 – not in References.
p11 l 28 "Sciarra": written "Sciarpa" in References. Which is correct?
p15 l24: Global Volcanism Program (2013): details of this should be included in the References
p15 l38 AND p16 l 6 Etiope et al (2007) not in References.
p16 l16 Procesi et al. – details should be added to References.
p17 l9 – there is a superfluous fullstop [".."] at the end of the line.
p19 l31 "JBC/PBL": should this be "JRC/PBL" as in References?
p22 l39 "breathdglobal" should be "breath global"
p22 l50 should Las Animas and Huerfano counties have capital leading letters?

The following appear in the References, but are not cited in the text: â˘A ´c Le Quéré et al., 2013 â˘A ´c Etiope, Baciu, Caracausi, Italiano & Cosma, 2004 â˘A ´c Etiope, Christodoulou et al. 2013 â˘A ´c Etiope Doezma & Pacheco, 2017 â˘A ´c Etiope, Feyzullaiev et al. 2004 â˘A ´c Etiope & Schoell, 2014 â˘A ´c Saunois et al. 2017 â˘A ´c USGS World Energy Assessment Team, 2000
I have not gone through the Supplementary References so I suggest that the authors re-check that it is correct and complete.

A: all minor technical comments have been addressed.
We are grateful to Reviewer#1 for the careful and positive review and comments.

Rev.#2

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: correct

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, as follows:

9 Note on temporal variability of geological methane emissions

The fluxes of natural gas seepage from the Earth's crust are not constant, either on short (hours, days, months, seasons) or long (years, centuries, millennia) time scales. Seepage variations can be induced by endogenous (geological) and exogenous (atmospheric) factors, including subsurface gas pressure variations (controlled mainly by gas migration and accumulation processes), changes of fracture permeability (tectonic stress, seismicity), hydrostatic aquifer variations, meteorological and climatic changes (atmospheric pressure, temperature, humidity and microbiological activity in the soil; Etiope, 2015). Mud volcano episodic eruptions (Mazzini and Etiope, 2017), seismicity-related degassing (e.g., Manga et al. 2009) and seasonal variability of microseepage (higher in winter due to lower methanotrophic consumption in the soil; Etiope and Klusman, 2010), are three, well studied, examples of geo-CH_4 emission variability. Anthropogenic activity, through modification of aquifer pressures (water pumping) and petroleum exploitation (with consequent decrease of reservoir pressures) can also induce seepage variability over time (e.g., Etiope, 2015). Therefore the global geo-CH_4 emission reported in this work, as well as in all other estimates available in the literature, must be interpreted as average, present-day degassing. Substantial decadal changes of seepage could occur as a result of decadal changes of hydrostatic aquifer pressure (e.g., Famiglietti, 2014) and decadal changes of seismicity (e.g., Mogi, 1979). Specific empirical studies are however missing, and with the present state of knowledge it is impossible to provide a temporal variability factor.

On longer, geological time scales, a series of proxies suggested that geo-CH_4 emissions could have been quite variable over the Quaternary period (Etiope et al. 2008b). Recent estimates on geo-CH_4 emission at the end of Pleistocene deserve a specific discussion. Based on radiocarbon (^{14}C) measurements in methane trapped in ice cores in Antarctica, Petrenko et al. (2017) estimated the absolute amount of ^{14}C-containing CH_4 in the atmosphere 11-12 k years ago, between the Younger Dryas and Preboreal intervals; this allowed to estimate that the maximum global natural, geological (^{14}C-free) CH_4 emission for that period was at most 15.4 Tg yr^{-1}. More recent analyses by the same authors confirmed this value (Dyonisius et al. 2018). These authors have then assumed that past geological methane emissions were no lower than today. They concluded, therefore, that present-day geological CH_4 emissions are much lower than present-day bottom-up estimates (54-60 Tg CH_4 yr^{-1}; Etiope 2015; Ciais et al., 2013). Without entering discussions on the accuracy and meaning of the ice core ^{14}C-based analyses and their temporal extrapolation to today, the following investigates whether the estimate by Petrenko et al. (2017) is compatible with:

(a) the estimates provided by authors other than Etiope (2015) and those reported in the present gridding work,

(b) the lowest bottom-up geo-CH_4 emission estimates available so far,

(c) present-day top-down geo-CH_4 emission estimates derived by different techniques, and
(d) pre-industrial geo-CH_4 emission estimates based on ice-core ethane measurements and observed geo-CH_4-to-ethane ratios.
Table 6 summarizes the data including individual literature references. In the bottom-up estimates table, the third column reports the lowest estimates proposed on the basis of more recent datasets and emission factors, which are updated in comparison with the earlier estimates (reported in the second column). The last column reports the overall lowest estimates, from old and new works, i.e. the minimum emission values derivable from different extrapolations. This comparison shows that the Petrenko et al. (2017) estimate is lower than any bottom-up estimate, regardless of authorship. The top-down estimates table reports geo-CH_4 emission derivable by three different procedures:
(a) assessing the portion of ^{14}C-free CH_4 in present day atmosphere (=30%; Lassey et al., 2007), then calculating the equivalent ^{14}C-free CH_4 emission (30% of total CH_4 emission, ~558 Tg yr^{-1} (Saunois et al. 2016) = 167 Tg yr^{-1}) and subtracting the anthropogenic ^{14}C-free component (fossil fuel fugitive emissions from inventories ~ 100-130 Tg yr^{-1}; EDGARv4.2; Saunois et al., 2016). The natural component (geo-CH_4 emission) would be 37-67 Tg yr^{-1}.
(b) Using methane concentration and isotopic data from ice-core records, based on box modelling by Schwietzke et al. (2016), suggest a geo-CH_4 emission of 30-70 (50) Tg yr^{-1}.
(c) With the same box model plus 3D forward modeling, but using current day atmospheric methane and isotopic data, Schwietzke et al. (2016) suggested a current day total fossil fuel (oil/gas/coal industries plus geological) CH_4 emission of 150–200 Tg yr^{-1}. Considering that oil/gas/coal emission inventories indicate 100-130 Tg yr^{-1}, geo-CH_4 emission could be 20–100 Tg yr^{-1}, consistent with approach (b) but with a wide uncertainty range.
(d) Using ethane concentration data from ice-core records, the 3 Tg yr^{-1} ethane top-down estimates by Dalsoren et al. (2018) confirm earlier bottom-up estimates of 2-4 Tg yr^{-1} ethane (Etiope and Ciccioli, 2009). Observed geo-CH_4-to-ethane emission ratios would then suggest 42-64 Tg CH_4 yr^{-1}.
Overall, geo-CH_4 emissions derived by top-down estimates range between 20 and 100 Tg yr^{-1}. These values are consistent with bottom-up estimates but substantially higher than Petrenko et al. (2017) estimate. The following options should then be considered:
(i) All current-day bottom-up and top-down geo-CH_4 emission estimates are biased high.
(ii) The Petrenko et al. (2017) estimate is biased low.
(iii) All estimates are reasonable, but the assumption that past Younger Dryas to Preboreal geo-CH_4 emissions were not lower than today does not hold.

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-CH_4 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 CH_4 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 $\delta^{13}C$ 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 ^{13}C-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

R: 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.

R: 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 16, 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

Rev.#3

R: 2/ You quote 20 self publications (Etiope or Etiope et al). It seems a bit too much regarding the total number of references and I recommend to keep only the main ones.

A: We really tried to use self-citations as little as possible, but the cited papers cannot be skipped as they refer to key reviews or are source of data used in the present work. Only two references could be deleted. References for specific seeps are reported in the inventories described in section 11.

R: Also, some recent relevant references are missing such as Petrenko 2017 (downward revision of geological source of methane), and Thornton 2017 (downward revision of ESAS methane emissions by a factor of about 8). I strongly suggest also to include in section 8.1 a short discussion about these recent papers and the implication for your work : you downward estimate of 37 Tg/yr is smaller than the previous 50 Tg/yr, but still well above the Petrenko suggested value of 15 Tg/yr.

A: We actually planned to add a discussion on Petrenko et al 2017 estimates in the revised version, as part of a chapter dealing with temporal variations of geological sources. This is now the new Section 9 (reported also above, as reply to Rev 2).
We have added Thornton et al 2016; we used Berchet et al 2016 as estimate of ESAS emission. It is not dissimilar from the estimate suggested by Thornton et al, but it seems to have considered a wider ESAS area than the one measured by Thornton et al.

R: 4/ You have to explain more clearly at the beginning that some part may be missing in the gridded map and that it means a possible underestimation of global emissions.

A: ok a sentence is now added at the end of Introduction

R: I am not convinced by the extrapolation made by the authors to complement the gridded estimate as it mostly rely on very rough estimates of the missing part (some additional areas emitting might be there and there, Arbitrary 50% flux, : : :).

A: The extrapolation only refers to the global estimate (for completeness of the paper and the obvious need to compare the gridded emissions with published global total estimates), and the limitations of the approach are discussed in the paper. The extrapolation has no impact on the gridded emission. For inverse modelling, the gridded emissions will be used.

R: In this sense column 3 of table 3 is a bit strange to me as roughly estimated whereas you spend a lot of time and energy to properly provide gridded estimates of column 2. This extrapolation has to be presented much more carefully and not put at the same level than the gridded estimate.

A: The extrapolation has the only scope to show that gridded emissions do not necessarily represent the actual global emission, because the datasets developed for the gridding may not be complete or may not contain the information necessary for improving the estimates (as happened for oil-gas seeps, SS and GM). This is now clarified at the end of Introduction and in Section 8.1.

R: In fine, I would just indicate in the conclusion that the gridded product will/may be revised regularly, upward or downward, when more data become available.

A: We agree, and this is stated at the end of Conclusions.

R: 5/ An uncertainty estimate has to be given for emissions of all categories (and reported in table 5), as for MS and isotopic signatures. This is critical for consistency of the paper and usage in atmospheric inversion. Although it might not be easy, the authors are the best choice we have to make such estimates, which else will be made by inverse modellers who probably know much less on the specific topic.

A: Yes, due to the nature of the data and derivation of the emission factors, uncertainty in final numbers is not of direct derivation and requires assumptions or arbitrary evaluations; this is why we preferred to report the factors controlling the uncertainty. However, we have now extended the discussion and provided approximate uncertainty values.

R: 6/ All along the text & tables : please harmonize the number of significant digits in the numbers provided. Considering the uncertainties I am not sure that 3.87 Tg/yr is relevant for instance for OS and I suggest to at least use 3.9 Tg/yr or possibly 4 Tg/yr. No more than 1 digit after the comma in any case.

A: Sure, one digit must be used.

Specific comments :
R: Abstract : “representativeness for many sources” suggested : and their isotopic signatures

A: OK

R: Abstract : "*This gap is particularly wide for geological CH4 seepage, i.e., the natural degassing of hydrocarbons from the Earth's crust. While geological seepage is widely considered the second most important natural CH4 source after wetlands, it has been mostly neglected in top-down CH4 budget studies, partly given the lack of detailed a priori gridded emission maps*".
This sentence is polemical and should be removed from the abstract which should reflect the work done.

A: These sentences explain the motivation for this work, and are therefore central to this study. They are not meant to devalue previous top-down studies, and we tried to reflect this in the language provided.

R: Considering the estimates of the CH4 emissions from geological seepage in the literature and in this paper, and the uncertain estimates from inland water systems, it is difficulet to say robustly that geological source is the 2nd. I would say a major source.

A: Geological emission is formally reported as 2nd CH4 source by the latest IPCC report (Ciais et al 2013). However, ok for using a more moderate "major source" expression.

R: And the lack of interest is true for past budgets but recent ones (e.g. Saunois et al., 2016) account for this source.

A: We do not mention a "lack of interest" but just the impossibility to use properly this source in top-down procedures because of the lack of priori gridded emission maps. We have rephrased as follows:

While geological seepage is widely considered a major source of atmospheric CH_4, it has been largely neglected in 3D inverse CH_4 budget studies given the lack of detailed a priori gridded emission maps.

R: P2 l5 : I suggest to update the ref to Saunois et al., 2016 and 558 MtCH4/yr
A: OK

R: P2 l7 : "emission inventories" and process-based models
A: OK

R: P2 l8-9 : TD and BU show strong disagreement only or natural sources, please precise.
A: OK

R: P2 l 9-12. The sentence has several problems. Schwietzke et al 2016 is not 3D inverse modelling but box modelling. The improvement brought by recent 3D modelling is arguable the recent study mentioned actually enlarge the range of emission estimates and needs to be further reproduced to pretend to get closer to the truth than other studies. I would rephrase to point that the usages of updated inventories of isotopic signatures has brought new constraints for the global methane budget. In any case, please rephrase.

A: Rephrased as follows: "Global box-modelling based on isotopic measurements (stable C isotope ratio, δ13C-CH4) of source signatures and the atmosphere combined with three-dimensional (3D) forward modelling using trends and spatial gradients recently improved the knowledge of major sources (fossil-fuel, agriculture and wetlands) and their spatio-temporal variation (e.g., Schwietzke et al 2016).

R: P2 l28 : geological degassing is today recognised as the second most important natural CH4 source after wetlands : see remark from the abstract. Also, the recent Petrenko paper should be quoted here (and commented later in the paper) as it proposes a downsizing of geological emissions to 15 Mt/yr at maximum.

A: OK for major source, but Petrenko et al refers to a late Pleistocene emission estimate, which can be used as reference for today's emission only assuming that geo-emissions are constant over time (which is not true). However, Petrenko et al is now discussed in a specific Section 9.

R: P2 l 29-30 : it is a bit unfair to quote specific papers when the highly visible synthesis from IPCC or GCP mention geological emissions in thei budget (e.g. Saunois et al., 2016). Please rephrase.

A: Ok, Saunois et al 2016 is added.

R: P3 l34 – figure 1 : References to other sources should be updated to the Saunois et al budget (GCP 2nd budget) instead of Kirschke et al. (GCP 1st budget). Please precise that figure 1 reflects literature and not the results of this paper.

A: OK, done

R: P5 l12-13 : what does it mean ? How can you know there is a seep of you cannot locate it ? Please precise and rephrase.

A: Because their existence, as number of seeps, is reported but their location is not provided.

R: P5 l 22 : Why not documented ? please provide a reason.

A: we suppose that many seeps in Africa and S.America are not documented because of the paucity of works (addressed to seeps) in these regions. It is clarified now.

R:Section 4.1 : How can you be sure that oil&gas seeps are not double counted in anthropogenic inventories as possibly located close to fossil fuel exploitation facilities? It is important to mention this somewhere in the paper and possibly discuss it as double counting is one clue to explain why bottom-up and top-down studies are not consistent for natural methane emissions.

A: But anthropogenic (fugitive emission) emissions are estimated by process-based modelling and specific emission factors. We do not see how there may be double counting with a natural phenomenon driven by different processes and with different emission factors. Double counting may happen with remote sensing (air, satellite) or micrometeorological (eddy-covariance) techniques, but these are not used for global emission estimates of fossil fuel. In fact, the gridded maps of this paper may help avoid double-counting in top-down studies by providing for the first time a geological seep grid that can be overlaid with a fossil fuel industry grid. Atmospheric measurements can then better constrain the sum of both while both grids help attribute sources.

R: P5 l 30 : "few tens" : can't you be more precise ? it is important to have a more precise idea of the fraction compared to the total number.

A: We estimated a total of about 100-200 seeps; it is not possible to be more precise as some papers (e.g., Walter Anthony et al. 2012) do not specify the number of seeps where the flux was measured. We have however referred to a summary table reported in Etiope (2015).

R: P5 l30-39 : the methodology should be a bit more detailed here (the supplementary does not bring much

more on this). How did you use the direct measurements to calibrate ? How did you attribute a measurement to a type of seeps ? how many types did you use ?

A: 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. We do not think it is appropriate to include in this paper all technical (gas-geochemical) details (how a seep flux is calibrated, etc.), as it is outside the scope of the work.

R: P5 l 38 : how do you account for miniseepages ? please provide ref or explanation.

A: A reference was given.

R: Section 4.2.3 big emitters. What fraction of these big emitters has been directly obvserved? It would be important to mention as they are not so numerous and a strategy to refine the estimate would be to measure them all (if not done yet). Please precise here.

A: Emission from the big emitters (almost all mud volcanoes) is estimated using the emission factor and area approach described in Section 4.2.2. This is now indicated.

R: Section 4.5.1 :
l15-16, if you do not do this work to update or improve estimates, why doing it so ? I am pushing a bit what you write but please rephrase.

A: Gridding has the scope to provide a priori maps for inverse modelling, as indicated in the Introduction. The datasets developed for the gridding may not contain the information necessary for improving the estimates. See also our response above: The extrapolation only refers to the global estimate (for completeness of the paper and the obvious need to compare the gridded emissions with published global total estimates), and the limitations of the approach are discussed in the paper. The extrapolation has no impact on the gridded emission. For inverse modelling, the gridded emissions will be used.

R: L28-29 : where does the 30% and 50% come from ? The 50% looks like a bit arbitrary ?

A: As stated in the text, 30% comes from knowing the total number of seeps on Earth (as discussed in Section 4.1). Yes, 50% is arbitrary, and we use hypothetical terms “may contribute at least 50% of the global emission..”, “…could…”.

R: is this 100% error reflected in column 3 of table3, moving from 3.8 to 8.1 Tg/yr for OS ?

A: Only partially. About 3.1 Tg/y are due to mud volcano eruptions, not accountable in the gridding.
All is explained in Table 3 footnote.

R: It is not clear to me why producing a gridded map if it cannot be used directly for global scale and needs re-assessment of emissions. Please clarify this section and the meaning of column 3 of table 3.

A: The grid can be used for global modeling, but the represented global flux therein may be incomplete. We have explained above that the datasets developed for the gridding may not contain be complete and/or may not have the information necessary for improving the estimates. Gridding has the scope of providing a priori maps for inverse modelling. Note that the same may be true for other published and widely used natural and

anthropogenic CH_4 flux grids. For example, different wetland flux grid products vary widely in their spatial distribution due to different data sources used (which may not be complete either). The geological seep maps developed here are unique in the sense that they are the first comprehensive product of this source, and thus no ensemble inverse runs are possible (like it is for wetlands). Anthropogenic grids are similar in the sense that it is very unlikely that every landfill in the world is included.

R: Section 4.6 : It is strange to me that you do not provide an uncertainty attached to emissions and signature in this section as in 6.6 for MS. "Order of magnitude" means a factor of 10 uncertainty. Does it means that OS emissions range from 0 to 38 Tg/yr ?

A: This section refers to emissions (and uncertainties) of individual seeps, not of their global emission (which is now determined and discussed).

R: Please be more precise in this section of possible or explain whay you cannot provide a range or a sigma for uncertainties.

A: This is now better explained in the text.

R: P9 l 27: there is no section 5.5.1.
A: Corrected

R: Section 5.5 : The total of 20Tg/yr has been highly controversial in the past years and recent papers related to ESAS largely reduced emission estimates (Berchet 2016, Thornton 2017). I would not present this number as a target to reach in the text.

A: We agree and we do not mean to use 20 Tg/y (Kvenvolden et al) as a target, but only as reference. This is now clarified in the text. Anyway, there are no updated global emission estimates for submarine seepage after that value. Berchet and Thornton papers refer only to ESAS and their finding do not imply that Kvenvolden et al estimate was wrong.

R: Lines 15 to 20 are highly arbitrary and should be identified as so. Why 5 to 10 Tg/yr ? These extrapolations should be taken with caution to me and mentioned as so.

A: Of course, these are just hypothetical, potential numbers. We rephrased however as follows:
"…it is plausible that global SS emission exceeds 5 Tg yr^{-1}".

R: Again do these estimate refer to column 3 of table 3 (5-12 Tg/yr, where text mentions 7-12) ?

A: Yes, but now rather than a range, a lower value is indicated (>5 Tg/y, i.e., >5 + 2 including Berchet's upper limit)

R: Section 8.1: This section has to be enriched to reflect a more complete spectrum of estimates than the ones provided by the co-author of this paper. At least the estimate from the recent Petrenko 2017 paper is important because it lowers to at maximum 15 MT/yr the total global value of geological emissions. Also, the 14C constraint on total 14C free methane from Lassey 2007 could be quoted. These elements should be quoted and discussed briefly in this section.

A: We actually planned to add a discussion on Petrenko et al 2017 estimates in the revised version, as part of a chapter dealing with temporal variations of geological sources (now Section 9). As noted above, however, Petrenko et al refers to a late Pleistocene emission estimate, which can be used as reference for today's emission only assuming that geo-emissions are constant over time (which is not true).

R: Section 5.6 : same remark as for OS : can you provide an uncertainty number for emissions (sigma or range) as in 6.6 for MS ?

A: The uncertainty of Berchet's ESAS (2 Tg/y as average) is 2 Tg/y. The uncertainty of other SS (1 Tg/y), from literature, cannot be assessed, because that literature does not provide uncertainties (as clearly indicated in the text). We may use arbitrarily 10% of uncertainty for the 1 Tg/y, so overall uncertainty would be 2.1 Tg/y. If a number is wanted, this can be given.

R: Section 6 : Even more critical than with OS emissions, the possible double counting with anthropogenic emissions should be addressed. How can we be sure that this diffuse source is not part of the oil&gas estimates of inventories ? OS are precisely located so the risk may be smaller than for diffuse MS. But for diffuse sources in the middle of oil&gas fields it seems more tricky. Please at least mention/discuss this in the text as a cause of uncertainty in section 6.6

A: Similar to our previous comment concerning double counting with OS: anthropogenic (fugitive emission) emissions are estimated by process-based modelling and specific emission factors. We do not see how there may be double counting with a natural phenomenon. Double counting may happen with remote sensing (air, satellite) or micrometeorological (eddy-covariance) techniques, but these are not used for global emission estimates of fossil fuel.

R: P15 l29 : what is the impact of the 4 km choice on the emission estimate ?

A: The 4 km choice does not influence the overall emission estimate, it is only a parameter guiding the gridding, as it served to convert the point data into more realistic areal data (polygons). This is now clarified in the text (section 7.1).

R: P15 l 37 : "few cases" : please provide a more precise number if possible.

A: OK, (<100 sites) is now indicated

R: P16 l 7 Again this sentence is unclear to me. Please rephrase

A: See our comments above. We rephrased as follows: " Although the GM emission grid developed here is expected to improve global CH_4 inverse modeling (as it includes previously neglected GM sources), the total GM emission estimate suggested by the gridding, because of the uncertainty of the theoretical emissions, is not meant to update or refine the previous global GM emission estimate (derived by process-based modelling; Etiope, 2015).

R: P16 l25 : "It is known : : :" : any reference to justify this ? Any explanation ? please provide a reference or explanation

A: Ok reference + brief explanation added.

R: Section 7.6 : as for other categories please provide a number (sigma/range) for the uncertainty on GM emissions as in 6.6

A: This is now provided.

R: P17 l34 : again please clarify this sentence.

A: Same comment above (why gridding could improve emission estimates only for some types of emission)

R: Table 5 : As already mentioned, please provide an uncertainty estimate for emissions from OS, SS and GM and fill it in table 5, column 3.

A: Done for all

R: P18 l27 : is there a risk that some SH emissions are forgotten because of les knowledge of the terrain ?

A: Yes.

R: P19 l4-6 : The 20 Tg/y value previously widely used for SS has been revised downward by several studies at least because of ESAS region (Berchet 2016, Thornton 2017).

A: See responses above. This is not correct. There is no downward revision of the global SS emission. Berchet and Thornton papers refer only to ESAS and their finding do not imply that Kvenvolden et al global estimate was too high.

R: As already noticed, one should stop giving the idea that this value is kind of a target to reach, as suggested here and in the corresponding paragraph of the text (see previous comment). The reference given here (Kvenvolden et al. 2001) seems a bit old regarding the past years activity on these emissions. Can the author provide a more recent reference and rephrase according to this remark ?

A: No, there is no updated global estimate for SS emission. We have however clarified that Kvenvolden et al number is not a target to reach, but just a theoretical reference for the gridded estimate.

R: P19 l31-33 : if no description of geological is given in an inverse modelling exercise, all the flux is spread on other distribution, possibly for onshore emissions, but with no guaranty, on the anthropogenic fossil emissions. So the term low bias should be rephrased (while the high bias is possibly correct for anthropogenic fossil). Please rephrase.

A: We re-phrased as follows: In the absence of a comprehensive gridded geological CH_4 seepage product, global or regional inverse model studies would erroneously attribute a low-bias to CH_4 emissions from geological seepage. This is because of a de-facto zero geological a priori estimate. At the same time, the inverse studies would erroneously attribute a high-bias to CH_4 emissions from fossil fuel industry activity (and potentially other sources) while correctly reporting total emissions of all sources

MARKED CHANGES

Gridded maps of geological methane emissions and their isotopic signature

Giuseppe Etiope [1,2], Giancarlo Ciotoli [3,1], Stefan Schwietzke [4], Martin Schoell [5]

[1] Istituto Nazionale di Geofisica e Vulcanologia, Roma Italy
[2] Faculty of Environmental Science and Engineering, Babes Bolyai University, Cluj-Napoca, Romania
[3] Istituto di Geologia Ambientale e Geoingegneria, CNR-IGAG, Roma, Italy
[4] Cooperative Institute for Research in Environmental Sciences, University of Colorado, Boulder, Colorado, USA, and NOAA Earth System Research Laboratory, Global Monitoring Division, Boulder, Colorado, USA.
[5] Gas-Consult Int., Pleasanton, California, USA

Correspondence to: Giuseppe Etiope (giuseppe.etiope@ingv.it)

Abstract

Methane (CH_4) is a powerful greenhouse gas, whose natural and anthropogenic emissions contribute ~20% to global radiative forcing. Its atmospheric budget (sources and sinks), however, has large uncertainties. Inverse modelling, using atmospheric CH_4 trends, spatial gradients and isotopic source signatures, has recently improved the major source estimates and their spatial-temporal variation. Nevertheless, isotopic data lack CH_4 source representativeness for many sources, and their isotopic signatures are affected by incomplete knowledge of the spatial distribution of some sources, especially those related to fossil (radiocarbon-free) and microbial gas. This gap is particularly wide for geological CH_4 seepage, i.e., the natural degassing of hydrocarbons from the Earth's crust. While geological seepage is widely considered a major source of atmospheric CH_4, it has been largely neglected in 3D inverse CH_4 budget studies given the lack of detailed a priori gridded emission maps. Here, we report for the first time global gridded maps of geological CH_4 sources, including emission and isotopic data. The 1°x1° maps include the four main categories of natural geo-CH_4 emission: (a) onshore hydrocarbon macro-seeps, including mud volcanoes, (b) submarine (offshore) seepage, (c) diffuse microseepage and (d) geothermal manifestations. An inventory of point sources and area sources was developed for each category, defining areal distribution (activity), CH_4 fluxes (emission factors) and its stable C isotope composition ($\delta^{13}C$-CH_4). These parameters were determined considering geological factors that control methane origin and seepage (e.g., petroleum fields, sedimentary basins, high heat flow regions, faults, seismicity). The global geo-source map reveals that the regions with the highest CH_4 emissions are all located in the northern hemisphere, in North America, the Caspian region, Europe, and in the East Siberian Arctic Shelf. The globally gridded CH_4 emission estimate (37 Tg yr^{-1} exclusively based on data and modelling specifically targeted for gridding, and 43-50 Tg yr^{-1} when extrapolated to also account for onshore and submarine seeps with no location specific measurements available) is compatible with published ranges derived by top-down and bottom-up procedures. Improved activity and emission factor data allowed to refine previously published mud volcanoes and microseepage emission estimates. The emission-weighted global mean $\delta^{13}C$-CH_4 source signature of all geo-CH_4 source categories is -48.5‰ to -49.4‰. These values are significantly lower than those attributed so far in inverse studies to fossil fuel sources (-44‰) and geological seepage

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Stefan Schwietzke 13/11/y 12:21
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Giuseppe Etiope 25/10/y 14:14
Eliminato: the second most important natural CH_4 source after wetlands

Giuseppe Etiope 25/10/y 14:18
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(-38‰). It is expected that using these updated more ^{13}C-depleted, isotopic signatures in atmospheric modelling will increase the top-down estimate of the geological CH_4 source. The geo-CH_4 emission grid maps can now be used to improve atmospheric CH_4 modelling, thereby improving the accuracy of the fossil fuel and microbial components. Grid csv files are available at https://doi.org/10.25925/4j3f-he27.

1. Introduction

Methane (CH_4) is a powerful greenhouse gas, whose concentrations in the atmosphere increased about 2.5 times since the pre-industrial era (1750), approaching 1.9 ppm in 2018. With a global emission of about 558 Tg CH_4 yr^{-1} (Saunois et al., 2016), CH_4 contributes ~20% to global radiative forcing (Ciais et al. 2013). The CH_4 budget, i.e. natural and anthropogenic sources and sinks, estimated by either bottom-up (emission inventories and process-based models) or top-down (inverse modelling) approaches (e.g., Saunois et al., 2016 and Refs. therein), is subject to considerable uncertainties, however. With respect to natural sources, top-down estimates show strong disagreement with bottom-up estimates, both globally and regionally. Global box-modelling based on isotopic measurements (stable C isotope ratio, δ^{13}C-CH_4) of source signatures and the atmosphere combined with three-dimensional (3D) forward modelling using trends and spatial gradients recently improved the knowledge of major sources (fossil-fuel, agriculture and wetlands) and their spatio-temporal variation (e.g., Schwietzke et al 2016). Nevertheless, isotopic data lack representativeness of CH_4 source signatures for many sources, and source attributions are limited by incomplete knowledge of the spatial distribution of some major sources, especially fossil fuel and microbial. In this respect, geological CH_4 emissions, i.e. the natural component of fossil fuel (^{14}C-free) emission, play a critical role. Geological CH_4 sources are the natural degassing of hydrocarbons from the Earth's crust (e.g., Etiope and Klusman, 2002; Kvenvolden and Rogers, 2005; Etiope, 2015). Geo-CH_4 originated in deep rocks by biotic (i.e. microbial and thermogenic) processes related to petroleum fields in sedimentary basins, as described in a wide petroleum geology literature (see for example Etiope, 2017 for a recent overview). Relatively minor amounts of CH_4 can also be produced by abiotic processes, which do not involve organic matter in rocks (e.g., magma degassing, high temperature post-magmatic process, CO_2 hydrogenation or Sabatier reaction, in geothermal/volcanic systems and ultramafic igneous rocks; e.g., Etiope and Sherwood Lollar, 2013). Surface emissions of geological CH_4 occur through the process known as "gas seepage", which includes point sources (gas-oil seeps, mud volcanoes, springs, geothermal manifestations) and area sources (diffuse "microseepage"). Once considered a minor natural CH_4 source globally (e.g., Lelieveld et al., 1998; Prather et al., 2001), geological degassing is today recognised as a major contributor to atmospheric CH_4, as indicated by the agreement between bottom-up and top-down estimates converging to 40-60 Tg yr^{-1} (Etiope et al. 2008; Ciais et al. 2013; Etiope, 2015; Saunois et al. 2016; Schwietzke et al. 2016). Nevertheless, geological seepage has mostly been neglected in global top-down CH_4 budget studies (e.g., Bousquet et al. 2006; Bergamaschi et al. 2013). In addition, geological CH_4 has erroneously been considered to be typically ^{13}C-enriched, thus with relatively high δ^{13}C-CH_4 values compared to biological sources such as wetlands (a global average of -38‰ was assumed for seepage by Sapart et al. 2012). In petroleum geochemistry it is well known, in fact, that in addition to the common thermogenic gas produced by moderate to high maturity source rocks, typically with δ^{13}C-CH_4 from -30‰ to about -50‰, vast amounts

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Eliminato: Three-dimensional (3D) inversion modelling, using trends, spatial gradients and isotopic measurements (stable C isotope ratio, δ^{13}C-CH_4) of source signatures and the atmosphere, recently improved the knowledge of major sources (fossil-fuel, agriculture and wetlands) and their spatio-temporal variation (e.g., Schwietzke et al 2016).

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of methane in sedimentary basins are microbial (thus with $\delta^{13}C\text{-}CH_4$ ranging from -55 to about -90‰) and thermogenic from low maturity source rocks, with $\delta^{13}C\text{-}CH_4$ from -50‰ to about -70‰ (Etiope, 2017; Milkov and Etiope, 2018). Degassing (seepage) to the atmosphere of ^{13}C-depleted geo-CH_4 sources is also widely documented (e.g., Etiope et al. 2009 and references therein). In addition to using unrepresentatively heavy $\delta^{13}C\text{-}CH_4$ geo-CH_4 values in previous studies, detailed a priori gridded maps of geo-CH_4 emissions and its isotopic signatures, which are essential for 3D inverse modelling and to discriminate between natural and anthropogenic microbial emissions, are currently lacking.

Here, we report the first global grid maps of geological CH_4 sources, including emissions and isotopic source signatures. The maps, elaborated by ArcGIS at 1°x1° resolution, include the four main categories of natural geological CH_4 sources: (a) onshore hydrocarbon macro-seeps (including mud volcanoes), (b) submarine (offshore) seeps, (c) diffuse microseepage and (d) geothermal manifestations. For each category we have developed an inventory of point sources and area sources, including coordinates (areal distribution, i.e. activity), estimated CH_4 fluxes (emission factors) and $\delta^{13}C\text{-}CH_4$ values. These parameters have been determined considering several geological factors that control CH_4 origin and seepage (petroleum fields, sedimentary basins, faults, earthquakes, geothermal/volcanic systems), based on published and originally *ad-hoc* developed datasets, as described in Sections 4, 5, 6 and 7. Integrated (total geo-CH_4) maps and associated text files (csv, comma- separated-values) have been generated to facilitate atmospheric CH_4 modelling to improve the accuracy of fossil fuel and microbial components. Gridded geo-CH_4 emissions were compared with published global estimates, derived by different approaches (e.g., Etiope et al. 2008; Etiope, 2012; 2015; Schwietzke et al. 2016). Gridded emissions do not necessarily represent the actual global geo-CH_4 emission or improve previous estimates, because the datasets developed for the gridding may not be complete or may not contain all the information necessary for improving previous estimates. A refinement of bottom-up estimates has only been possible for mud volcanoes and microseepage, because their gridding implied a careful assessment of the spatial distribution and emission factors.

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Giuseppe Etiope 26/10/y 09:45
Eliminato: for mud volcanoes and microseepage, thus allowing for new bottom-up estimates of their global emissions.

2. Classification of the geological CH_4 sources

Geological CH_4 sources can be classified into four major categories:

(a) Onshore hydrocarbon seeps (or macro-seeps) in sedimentary (petroliferous) basins including CH_4-rich gas-oil seeps, mud volcanoes (MV) and gas-bearing springs. Hereafter referred as **OS**.

(b) Submarine (offshore) seeps, where CH_4 released from shallow seafloor (coastal areas or shelves, generally up to 300-400 m below sea level) can cross the water column and enter the atmosphere. Hereafter referred as **SS.** Deep-sea seeps that are unlikely responsible for methane emission into the atmosphere are not considered.

(c) Diffuse microseepage in sedimentary (petroliferous) basins, the widespread, invisible exhalation of CH_4 typically detected in correspondence with gas-oil fields. Hereafter referred as **MS.**

(d) Geothermal and volcanic manifestations, where CH_4 is a minor component (subordinated to CO_2) but with potentially significant fluxes to the atmosphere. Hereafter referred as **GM.**

These “geo-methane” sources are extensively described and discussed in a wide body of literature; for

details and definitions the reader may refer to Etiope and Klusman (2002); Judd (2004); Kvenvolden and Rogers (2005); McGinnis et al (2006); Etiope et al. (2007); Judd and Hovland (2007); Etiope et al. (2008); Etiope and Klusman (2010); Etiope (2015), Mazzini and Etiope (2017). Their global bottom-up and top-down emissions, compared with other natural CH_4 sources, are summarized in Fig.1.

3. Methodology

Methods for creating CH_4 emission and $\delta^{13}C$-CH_4 grids vary by geo-CH_4 category, based on the data availability and specific seepage characteristics. Methods are therefore described in detail for each category in Sections 4 (OS), 5 (SS), 6 (MS) and 7 (GM). First, a brief overview of the different types of input data and gridding procedure is given below.

3.1 Data sources

Table 1 summarizes how the four categories of geo-CH_4 sources were elaborated, showing data sources, the parameters used to define the "activity" (spatial distribution), the "emission factors" (fluxes), and the attribution of the isotopic CH_4 values. The list and web links of the sources of databases are reported in the Supplement (S6).

3.2. Gridding procedure

The gridding procedure is the same for each geo-CH_4 source category. Geo-CH_4 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 CH_4 values, plus geographical information, such as country and region). The grid was then joined with single OS, SS, MS, GM shapefiles. The final csv files include data fields that define the coordinates of each cell centroid, the variable name and its unit of measurement (tonnes year^{-1} per cell for CH_4 emission and ‰ for $\delta^{13}C$-CH_4, according to VPDB, Vienna Pee Dee Belemnite, standard). For the grid cell values, the number zero (0) is used to indicate an actual or best emission estimate of zero (no seepage), whereas -9999 indicates lack of knowledge, of the emission. Specifically:

In the CH_4 output files:

- zero (0) value is used for:
  - all offshore cells of the onshore seepage shape files (OS, MS and GM)
  - all onshore cells of the offshore seepage (SS).
  - all onshore cells outside the potential MS area
  - onshore cells without OS or GM sources
  - offshore cells outside the SS areas
- the number -9999 is used for:
  - cells within SS areas where emissions are unknown.

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The categories OS, MS and GM, due to the method of emission derivation (see related sections below) have always an emission value.

In the isotope files:

- an isotopic value is reported in each cell that has a flux value;
- where specific values are not available (as occurred in OS and SS), the global weighted average $\delta^{13}C$ for the relative emission category is reported;
- four decimals are used for global weighted average isotope values; this can help to trace back which cells are based on cell-specific data (with one decimal), and which contain weighted averages (four decimals);
- the value -9999 is used only for cells with no emissions in the corresponding CH_4 output files.

The application of such rules is described in the specific chapters of the four emission categories. Once individual OS, SS, MS, GM maps/files were produced, they were merged into a unified, total geo-CH_4 gridding: emissions per cell were summed and $\delta^{13}C$ values were averaged.

4. Onshore seeps (OS)

4.1 Global seep count and distribution

The spatial distribution (activity) of onshore seeps is derived from geographic coordinates of 2827 seeps, from 89 countries, reported in a global onshore seep dataset, which includes 1119 oil seeps, 846 gas seeps, 741 mud volcanoes and 121 gas-bearing springs. This dataset is an updated version of a previous inventory (named GLOGOS, reporting 2100 seeps) available from CGG (2015) and described in Etiope (2015). The global distribution of OS is reported in Fig. 2.

The seeps listed in the dataset generally refer to individual focused vents (single macro-seep manifestations) but in several cases they refer to groups or clusters, or even wide zones of multiple seep points. 612 seeps (569 gas-oil seeps and 43 mud volcanoes) could not be geographically located with precision and they are listed without coordinates (in addition to the 2827 seeps). The dataset, therefore, actually mentions a total of 3439 seeps or seepage sites, including 3396 gas-oil seeps and 784 mud volcanoes. The total number of 3439 OS represents about 30% of total seeps assumed to exist on Earth (≈10,000 was proposed by Clarke and Cleverly, 1991), but the present dataset includes the largest and more active seeps (especially for MV) because they are more easily documented and have attracted attention for scientific research, petroleum exploration, and natural heritage protection. Small or inactive seeps tend to be less observed and reported. In particular, the MV inventory is almost complete, probably missing smaller MVs in Asia. The gas-oil seeps in the dataset likely contribute more than 50% of the previously estimated total gas-oil seep emission. Africa and South America likely host a larger number of gas-oil seeps and springs not documented in the dataset, because of the paucity of specific investigations, especially in remote areas.

4.2 Attribution of CH_4 emissions to individual seeps

The attribution of CH_4 emission magnitudes to individual seep locations follows two different approaches for (a) gas-oil seeps or springs and (b) mud volcanoes (MV).

4.2.1 Emission of gas-oil seeps and springs

Direct measurements of CH_4 flux are available for about 100-200 gas-oil seeps in Europe, Asia and North America (see Table 6.1 in Etiope, 2015). In general, therefore, potential or theoretical flux values have been attributed to the inventoried seeps. Theoretical emission values can be reasonably provided only in terms of order of magnitude (i.e. 10^0, 10^1, 10^2, 10^3, 10^4 tonnes $year^{-1}$). For gridding purposes, however, theoretical values (approximate working values) were used taking into account basic characteristics of the several seeps, i.e. the type of seep (for example, gas seeps generally release more methane than oil seeps), the activity and size of the seep (according to specific literature, reports, web images), and taking into account, as "calibration", experimental data, i.e., flux values measured in the field from seeps covering a wide range of activity and size (data are taken from the wide literature considered in Table 6.1 of Etiope (2015)). The theoretical values also take into account the gas emission from the invisible miniseepage, the diffuse degassing from the ground surrounding the macro-seep craters and vents (see definitions in Etiope, 2015), and which adds an amount of gas that may be three times higher than that released from the macro-seep (Etiope, 2015). This resulted in the attribution of the values reported in Table S1 in the Supplement. These values should be considered as first-order estimates and care should be taken when using individual seep flux estimates from this product to derive global emission estimates, as discussed in section 4.5.

4.2.2 Emission of mud volcanoes (MV)

For MV, emission values refer to the continuous quiescent degassing, i.e. they do not include emissions during episodic eruptions, as these are practically impossible to estimate for each MV. Eruptions were considered separately for the global emission estimate as discussed below. The quiescent emissions were attributed to each MV following the activity (area) and emission factor approach as follows.

A precise evaluation of the MV areas was performed by accurate image (Google Earth) analysis. For each MV visible on Google Earth images, the area of the entire MV structure, including central craters and flanks, was estimated by drawing a polygon encompassing the mud cover and mound flanks. For smaller MV, not visible on low resolution Google Earth images or covered by vegetation, photos or information from published literature or web sources were considered to define the order of magnitude of the MV size. From two repeated image analyses the global MV area resulted to be about 680±40 km^2.

The MV emissions were then assessed using an updated dataset of fluxes measured from 16 MV in Azerbaijan, Romania, Italy, Taiwan, China and Japan (Table S2 in Supplement), distinguishing between the macro-seepage (the focused emission from craters and vents) and miniseepage. Regression analysis between MV area, miniseepage and macro-seep flux of these measured MV was used to derive miniseepage and macro-seep flux (and thus the total CH_4 emission) for each MV of the inventory, whose area was determined as previously indicated. The procedure is described in detail in the Supplement (Section S1.1)

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4.2.3 The "big emitters"

There is a total of 76 OS with emissions in the order of 10^4 tonnes CH_4 yr^{-1} (i.e. that may emit at least 10,000 tonnes CH_4 yr^{-1} individually), and they can be considered "big emitters". They typically refer to large, very active and frequently erupting MVs, so their emission is estimated based on emission factor and area approach described in the previous section. The 76 big emitters likely dominate the spatial distribution of CH_4 emissions (they represent 63% of the total OS emission) and the weighted global mean isotopic value. As shown in Fig. 3, it is clear that, on a global scale, the Caspian and Mid-East regions represent the main OS emission areas.

4.3 Attribution of the δ^{13}C-CH_4 value

For each seep the δ^{13}C-CH_4 value is:

(a) measured, as indicated in the literature (available in the OS inventory; CGG, 2015), or (b) estimated on the basis of isotopic values using 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.

The OS emission-weighted value (Section 4.5.2) was used for gridding where the isotopic value could not be assessed. The global distribution of three classes of δ^{13}C-CH_4 value is shown in Fig. S4 in the Supplement.

4.4 OS gridding

The OS shapefile generated in ArcGIS was spatially joined to the 1°x1° vector square grid. OS occur in 616 cells, for a total emission of 3.9 Tg yr^{-1} (Fig. 4). This is about 0.1 Tg yr^{-1} higher than the actual sum of the seep emission in the inventory because of multiple counting of 57 seeps that occur exactly on the boundary of a cell.

4.5 Evaluation of global OS emission and δ^{13}C-CH_4

4.5.1 Re-assessing global OS emission

Because the OS inventory is not complete and the uncertainty of the theoretical flux values considered for individual oil-gas seeps is large (see Section 4.6), the OS flux grid is not meant to update or refine the previous global OS CH_4 emission estimate (Etiope et al. 2008). A comparison with the published bottom-up estimates can establish whether the OS inventory data used in the OS flux grids are plausible. However, the procedure developed to attribute CH_4 emissions to MVs can represent a refinement of the global MV emission estimate.

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Published bottom-up estimates of CH_4 emission from onshore macro-seeps are reported in Table 2. Some estimates included, without a clear distinction, shallow submarine MV (e.g., Dimitrov, 2003; Milkov et al. 2003), which must be considered within the category SS in this work. Therefore, those estimates are indicated in the table as upper limit. Because the data of the OS inventory, as explained in Section 4.2, refer only to quiescent degassing, the table distinguishes emissions that exclude MV eruptions (quiescent degassing) and those that include MV eruptions.

Concerning gas-oil seeps and springs, the use of the theoretical values, as described in Section 4.2, results in global CH_4 emission of about 1 Tg yr^{-1} (Table 2). As indicated in Section 4.1, the OS dataset, although representing only 30% of all seeps existing on Earth, includes the largest and more active seeps, which may contribute at least 50% of the global emission; accordingly, the total gas-oil seep emission could be likely around 2 Tg yr^{-1}. Any further or more detailed extrapolation to a global seep emission estimate would be inappropriate.

The global MV emission from quiescent degassing, i.e. the sum of the MV emission values reported in the OS dataset, amounts to ~2.8 Tg yr^{-1}. The total CH_4 emissions from the 2827 OS seeps is, therefore, about 3.8 Tg yr^{-1} (1 + 2.8 Tg yr^{-1}). The OS-MV dataset likely represents about 90% of total MVs assumed to exist on Earth (≈900; Dimitrov, 2002; Etiope and Milkov, 2004); extrapolating to the total MV number would result in a global MV emission of approximately 3 Tg yr^{-1}. This is within the range suggested by Etiope and Milkov (2004). Compared to previous emission estimates of Etiope and Milkov (2004) and Etiope et al (2011), the present MV estimate used a lower activity, i.e. lower global area, 680 km^2 instead of 2800 km^2 (which was suggested by data provided by Azerbaijan Geological Institute) but relatively higher emission factors. Concerning the MV eruptions, we can only use, again, the rough estimations indicated in Dimitrov (2003), Milkov et al. (2003) and Aliyev et al. (2012) (i.e., average gas flux during eruptions of MVs in Azerbaijan $2.5x10^8$ m^3, the proportions of eruptive MVs: 27%, and the frequency of eruption: 1.35 eruptions/year), which translate into a total eruptive emission of 3.1 Tg yr^{-1} (Milkov et al. 2003). Therefore, the global OS emission, including MV eruptions and assuming the theoretical values for the gas-oil seeps and springs, would be ~ 8.1 Tg yr^{-1}, which is within 10% of the lower range proposed by Etiope et al. (2008).

4.5.2 The average emission-weighted $\delta^{13}C$-CH_4

The total mean value of $\delta^{13}C$-CH_4 from all OS is -47.8‰, and that from the 76 big emitters is -46.7‰. The global OS emission-weighted mean value of $\delta^{13}C$-CH_4 is -46.6‰.

4.6 OS uncertainties

Spatial distribution uncertainty: In the 1°x1° grid, the uncertainty of the geographic distribution of the OS is practically zero, as all identified seeps have geographic coordinates within an error <1°.

Emission uncertainty: The uncertainty of the modeled oil-gas seep emission (based on the method of value attribution described in Section 4.2.1) is maximum 90% (1 ±0.9 Tg yr^{-1}). The uncertainty of global MV emission (48%) was estimated by summing (a) the uncertainty of the estimated MV areas (6%, see Section 4.2.2) and (b) the uncertainty of the modelled MV emission factor (42%; see Supplementary S1.1). Because

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oil-gas seeps and MVs account for 26% and 74% of total OS emission, respectively, the overall gridded OS emission uncertainty is about 58% (3.8 ±2.2 Tg yr^{-1}).

$\delta^{13}C$-CH_4 uncertainty: The uncertainty of measured $\delta^{13}C$ values (from literature) practically corresponds to laboratory analytical uncertainty (typically <0.1‰). The maximum uncertainty of the estimated $\delta^{13}C$ values (based on criteria described in Section 4.3) is approximately within 15‰, i.e. half of the range of $\delta^{13}C$ values for typical microbial (-80 to -60‰) and thermogenic (-50 to -20‰) gas. The uncertainty of the emission-weighted mean (-46.6‰) is mainly induced by the 76 big emitters, for which the $\delta^{13}C$ values are available or estimated with good approximation (<±5‰), leading to a mean value of -46.7‰. The difference between global emission-weighted and 76 big emitters average $\delta^{13}C$ values is 0.1‰. The average order of magnitude of ±1‰ can be considered for the uncertainty of global emission-weighted $\delta^{13}C$ value.

5. Submarine seepage (SS)

5.1 Assessment of global SS area

A specific dataset of offshore seepage areas, in coastal regions and shallow seas (typically <500 m deep, which is generally the maximum depth of seeps that may affect the atmosphere; e.g., Solomon et al. 2009), was developed based exclusively on published literature (Table S4 in the Supplement). The dataset includes:

a) Submarine seeps (including mud volcanoes) where gas was observed to reach the sea surface via bubble plumes and the emission to the atmosphere was estimated: flux emission estimates are available from 15 zones (from focused, point-source, manifestations to wide regional areas) in the seas of Australia, Bulgaria, Brunei, Canada, Chile, China, Denmark, Georgia, Greece, Norway, Spain, Romania, Russia, Turkey, Ukraine, United Kingdom, USA.

b) Submarine seeps in shallow seas where gas was actually observed (also through hydro-acoustic images) to reach the surface but the output to the atmosphere was not provided, or where, due to the shallow seabed (<400-500 m), the methane is expected to enter the atmosphere. These areas (16 zones) are in the offshore of USA, Canada, Mexico, The Netherlands, Denmark, France, Italy, Greece, Russia, Azerbaijan, Turkmenistan and Pakistan.

The dataset does not include deep-sea seeps or areas with gas-charged sediments (e.g. as those inventoried by Fleischer et al. 2001) that may release methane into the water column, but for which the possibility of injection into the atmosphere is scarce or unknown. The area and methane flux estimates reported in the several papers were used here without critical evaluation. Geo-referenced polygons were created for each area (Fig. 5).

5.2 Attribution of seepage levels

CH_4 fluxes from the original publications (Table S4) are used in the gridded emission dataset.

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5.3 Attribution of the $\delta^{13}C$ value

The $\delta^{13}C$-CH_4 values of SS are attributed on the basis of available literature or considering the geological setting (type of petroleum system, origin of the gas) of the seepage areas (*italic* values in Table S4) following the same criteria adopted for OS. For four areas in Table S4 (China-Brunei offshore, Laurentian Channel and Grand Banks Downing Basin in Canada, and East Kamtchatka shelf in Russia) it was not possible to attribute any theoretical $\delta^{13}C$ value because the gas may actually derive from either microbial or thermogenic sources, covering a wide range of isotopic values. In these cases, the global emission-weighted $\delta^{13}C$ value of SS (see Section 5.5) was used for these regions in the $\delta^{13}C$ grids. The global map of $\delta^{13}C$ for SS is shown in Fig. S5 in the Supplement.

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5.4 SS gridding

The SS grid dataset was generated digitizing polygons of the SS areas from literature maps (see references in Table S4). The final shapefile contains 31 polygons characterized by the following variables: country, longitude and latitude of the polygon centroid, CH_4 output flux, area, and average $\delta^{13}C$ value of the emissions in each polygon. The value -9999 is reported for the missing emissions at 16 sites (sites 16 to 31 in Table S4). The SS layer was joined with the 1°x1° vector grid and the resulting map is shown in Fig. 6.

5.5 Evaluation of global SS emission and $\delta^{13}C$-CH_4

The sum of CH_4 emissions from the 15 SS areas in Table S4 (which refer to published estimates) is ~3.9 Tg yr^{-1}. This represents about 20% of the theoretical estimate of global SS emission to the atmosphere (~20 Tg yr^{-1}), derived by process-based models, proposed by Kvenvolden et al (2001). SS emissions also occur in the other 16 areas reported in Table S4 and likely in many other sites not investigated yet. Among the areas with missing emission values, the Gulf of Mexico and the Caspian Sea are very likely major methane emitters, followed by the North US Atlantic margin. It is difficult to evaluate whether adding these missing SS emissions, the total sum would approach the Kvenvolden et al. estimate of 20 Tg yr^{-1} (however, we consider this global value as a theoretical reference for our SS gridded emission, not a target or actual value to reach). Evaluation of the SS emission factor (based on the reported area and total fluxes in Table S4) is also difficult because the areas indicated in the several works (see References in Table S4) often refer to the surveyed area and not to the actual area of seepage; in these cases, using the surveyed area would result in a strongly underestimated emission factor. However, using the relationship observed for the 15 "investigated" sites between area (actual seepage or surveyed) and emission factor, the other 16 sites would yield total emissions of about 1 Tg yr^{-1}. This would bring the total CH_4 emission from the 15+16 sites of Table S4 to about 5 Tg yr^{-1}. Assuming that (a) SS generally do not take into account the release of dissolved methane (i.e., only methane bubbles are accounted for) and (b) today unknown SS areas (not listed in Table

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S4) may have a seepage extent not exceeding that of the investigated areas, it is plausible that global SS emission exceeds 5 Tg yr^{-1}. If the upper estimate for the East Siberian Arctic Shelf, 4 Tg yr^{-1} (Berchet et al., 2016; i.e., twice the mean used in Table S4), is considered, then the global SS emission would exceed 9 Tg yr^{-1}. The SS emission-weighted mean value of $\delta^{13}C$-CH_4 is -59‰. The non-weighted mean value is -51.2‰.

5.6 SS uncertainties

Spatial distribution uncertainty: In the 1°x1° grid, the uncertainty of the geographic distribution of the SS is practically zero, as all seepage zones have geographic coordinates within an error <1°.

Emission uncertainty: The main uncertainty and control on the global gridded 3.9 Tg yr^{-1} value is associated with the estimate of CH_4 emissions from the East Siberian Arctic Shelf, for which we used the central value (2 Tg yr^{-1}) of the range indicated by Berchet et al (2016), i.e. 0-4 Tg yr^{-1} (not very dissimilar from the estimate of 2.9 Tg yr^{-1} suggested by Thornton et al. 2016). The other 15 SS areas, totaling ~1 Tg yr^{-1}, have variable uncertainty, often not defined in the individual publications. With a ±2 Tg yr^{-1} uncertainty for the Siberian Arctic Shelf and assuming arbitrarily 10% uncertainty for the other estimates, the overall SS gridded emission uncertainty would result ±2.1 Tg yr^{-1} (54%).

$\delta^{13}C$-CH_4 uncertainty: The maximum uncertainty of the estimated $\delta^{13}C$ values (based on criteria described in Section 5.3) is approximately within ±15‰, i.e. half of the range of $\delta^{13}C$ values for typical microbial (-80 to -60‰) and thermogenic (-50 to -20‰) gas. The uncertainty of the emission-weighted mean (-59‰) is mainly controlled by emissions from Eastern Siberian Arctic Shelf, North Sea and Black Sea, whose $\delta^{13}C$ values are available or estimated, ranging from -50 to -63‰. The overall uncertainty of the global emission-weighted mean is thus reasonably < ±7‰.

6 Microseepage (MS)

6.1 Assessment of global MS area

The diffuse exhalation of CH_4, called microseepage (MS), is widespread throughout onshore petroleum fields all over the world. It is systematically observed in correspondence with anticlines and marginal (faulted) areas of gas-oil fields (Etiope and Klusman, 2010; Tang et al. 2017). The existence of macro-seeps (OS) in a given region also implies a high probability that MS exists in that region, even if that region falls outside a known petroleum field. Therefore, as a proxy of the activity (spatial distribution) of MS, we considered the global area of petroleum fields and a global area including OS defined as described below. This criterion is conservative as MS may also occur in sedimentary basins without known petroleum fields and OS (Klusman et al. 2000; Etiope and Klusman, 2010). The assessment of the global petroleum field area (PFA) and global OS area (OSA) is discussed in the Supplement (Sections S3.1 and S3.2). The total potential MS area (PMA) resulted to be PFA + OSA = 13,033,000 + 85,900 = 13,118,900 km^2.

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6.2 Attribution of MS levels

The level of MS CH_4 emissions was established on the basis of a statistical analysis of a MS flux data-set (see Section 6.2.1) and considering the theory of seepage migration mechanisms, for which the gas flux greatly depends on the permeability of the rocks, especially when induced by faults and fracture networks (Etiope and Klusman, 2010; Etiope, 2015; Tang et al. 2017). Accordingly, the attribution of the flux within the PMA (PFA+OSA) was done considering the presence/absence, in each cell, of three major geological factors, which are proxies of methane seepage and gas permeability, i.e. OS, faults and seismicity, as explained in Section 6.2.2.

6.2.1 Statistics of MS data

A dataset of 1509 MS CH_4 flux measurements was compiled based on available literature and unpublished works. The data are from 19 petroleum areas: 8 in the USA (Klusman et al 2000; Klusman, 2003; Klusman, 2005; Klusman, unpublished; LTE, 2007), 6 in Italy (Etiope and Klusman, 2010; Sciarra et al. 2013; Etiope, 2005; Etiope, unpublished), 1 in Romania (Etiope, 2005), 1 in Greece (Etiope et al. 2006; Etiope, unpublished), and 3 in China (Tang et al. 2007; 2010; 2017). The resulting descriptive statistics are reported in Table S5 in the Supplement.

The data are divided into two groups: (a) negative and near-zero values (<0.01 mg m^{-2} d^{-1}, considering minimum analytical error), which represent the normal CH_4 flux in dry (not flooded) soils, and (b) positive values, >0.01 mg m^{-2} d^{-1} (i.e. microseepage). The similar order of magnitude between the median and the geometric mean flux indicates a log-normal behavior of the positive CH_4 flux distribution. The positive values represent about 57% of total measurements. This implies that MS does not occur throughout the entire PFA. This is well known, as CH_4 flux from the ground, in addition to underground rock permeability and fluid pressures, depends also on soil conditions (humidity, porosity, temperature) and methanotrophic activity. Accordingly, and taking into account that the MS measured sites are geographically dispersed with a relatively homogeneous spatial distribution (and the measurements were taken in different seasons), we reduced the PFA by removing 43% of the area as described in Section 6.4. A new MS area was therefore defined as “Effective Microseepage Area”, EMA, which is OSA + 57% of PFA. The derivation of the EMA area is described in Section 6.4. Frequency histogram and Normal Probability Plot (NPP) of MS data (logarithmic values of positive values) confirm that flux values have a log-normal distribution (Fig. S7). Values exceeding 1000 mg m^{-2} d^{-1} (up to 7078) were excluded as they represent special and rare cases of MS (often not distinguishable from miniseepage, which is the halo surrounding macro-seeps).

The combined analysis of NPP and frequency histogram (Fig. S7) resulted in the identification of 4 main groups of positive flux data, i.e. 4 levels of MS:

Level 1 0.01-12 (median: 1.3) mg m^{-2} d^{-1}

Level 2 12-60 (median: 31.1)

Level 3 60-300 (median: 110)

Level 4 300-1000 (median: 493.5)

Level 0 implies absence of MS. The median of each level was assigned to the 0.05°x0.05° grid cells

included in the area with expected MS defined in Section 6.2.2 and according to the presence of the factors influencing MS. The median was chosen because it is not affected by outliers within each level, providing conservative flux values.

6.2.2 Factors influencing MS level: presence of macro-seeps, faults and seismicity.

The 4 MS levels (1, 2, 3 and 4) are associated with 4 different combinations of the three factors influencing the gas flux, following MS theory and experimental data. The three factors are:

(a) faults;

(b) seismic activity;

(c) presence of macro-seeps (OS), which are themselves expression of regional seepage activity;

(a) Fault data were taken from 17 different datasets (see Sources of databases in the Supplement): the main one is the Global Faults layer of ArcAtlas (Finko, 2014). It includes two types of faults: (1) faults created by the dislocation of rocks that define the geological structures of the continents (tectonic contacts and thrust-faults) and (2) faults created by the morphology of the present-day relief and morphostructure (steps and rifts). The first type of faults refers to ancient structures, while those revealed by relief are comparatively young structures that appeared during the neotectonic stage of the Earth's evolution (mostly in the Neogene and Quaternary periods). The other 16 fault datasets are national or regional datasets from Afghanistan, Australia, Bangladesh, Caribbean region, Central Asia, Europe including Turkey, Georgia, Greece, Ireland, Italy, New Zealand, South America, Southern Mediterranean area, Spain, Switzerland, United Kingdom (see Supplement). The final merged fault dataset includes 156,095 tectonic elements (Fig. S8); obviously it does not include all actual existing faults on Earth. The dataset must be interpreted as a global distribution of the main regional fault systems and fractured zones.

(b) The epicenters of earthquakes are proxies of fault location and activity (permeability), so they also represent the presence of active faults, which may not be reported in the fault dataset. It is also known that gas migration and escape to the surface may increase with seismic activity. We used the seismicity dataset of USGS Earthquake Lists, Maps and Statistics (see Sources of databases in the Supplement). We extracted only onshore seismic events with magnitude M>4.5 recorded from 2005 to 2017. This resulted in a dataset of 18,157 onshore epicenters covering 177 countries (Fig. S9).

(c) Presence of macro-seeps (OS). The OS area is described in the Supplement (Section S3.2).

The three factors, faults, seismicity and presence of seeps, were applied on the gridded EMA as described in Section 6.4.

6.3 Attribution of the $\delta^{13}C$ value

Measured and published data of $\delta^{13}C\text{-}CH_4$ in gas MS are scarce and available only for a few petroleum fields. However, during the seepage process (migration driven by pressure gradients, i.e. advection), the CH_4 isotopic composition does not change significantly, so that surface CH_4 flux has basically the same $\delta^{13}C$ value of the original gas in the reservoir (e.g., Etiope et al. 2009). Therefore, for each field or basin, the MS $\delta^{13}C$ value was taken from published data related to subsurface reservoirs. A limitation of this strategy is that in a given basin 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.

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Accordingly we adopted the following procedure:

- when one or more seeps (OS) occur in a petroleum field (in the Petrodata list), the average $\delta^{13}C\text{-}CH_4$ of those seeps was used for MS;
- in absence of seeps, reservoir $\delta^{13}C\text{-}CH_4$ data were used; they were taken from the inventory described by Sherwood et al. (2017) or published literature. For the fields (in the Petrodata list) whose $\delta^{13}C\text{-}CH_4$ value is not reported either in Sherwood et al. (2017) or literature, a theoretical $\delta^{13}C\text{-}CH_4$ value was estimated on the basis of the type of gas (microbial or thermogenic) and maturity of the petroleum system.

The file contains 349 $\delta^{13}C\text{-}CH_4$ data points (from 891 petroleum fields). It was not possible to estimate a specific $\delta^{13}C$ value for the remaining 542 petroleum fields. In these cases, the global emission-weighted isotopic value was used in the resulting empty cells, as described in Section 6.4.

6.4 MS gridding

PFA and OSA (described in the Supplement) were intersected with a high-resolution (0.05°x0.05°) global grid. The 0.05°x0.05° cell dimension corresponds to the maximum resolution that can be obtained using ArcGIS software (the software cannot handle shapefiles > 2 Gbyte). The high-resolution gridding was used to match, as much as possible, the PFA: gridded PFA is in fact 14,791,897 km^2, while the original PFA was 13,033,750 km^2. The high-resolution gridding also served to reduce the boundary effect, and thus the overestimation of the areas with MS enhancing factors, i.e. faults, earthquake and seeps (the larger the cells, the higher the probability that the cells include MS enhancing factors).

As discussed in Section 6.2.1, only 57% of PFA cells were considered to host MS. It was then necessary to delete 43% of PFA cells. The cells were randomly deleted only among those that do not host MS enhancing factors (faults, earthquakes and seeps), i.e. empty cells (which are 93% of total PFA). The overall PFA reduction of 43% was obtained by deleting 54% of the empty cells (resulting in a PFA of 8,408,360 km^2). Combining PFA and OSA results in EMA (Table S6). The sequence of MS modelling is summarized in the block diagram of Fig. S10. The MS levels were then assigned to the 0.05°x0.05° gridded EMA according to the presence of the factors influencing MS: a) presence of faults; b) presence of seismic activity; c) presence of macro-seeps (OS), as follows:

Level 1 was applied to cells without any geological factor.

Level 2 was applied to cells with faults or earthquakes

Level 3 was applied to cells with faults plus earthquakes or oil-seeps or gas-bearing springs
Level 4 is applied to cells with gas-seeps or mud volcanoes.
The resulting global MS CH_4 emissions are about 24 Tg yr^{-1}. The emissions per cell range from 14.7 tonnes $year^{-1}$ (cells of about 30 km^2) to 29,446 tonnes $year^{-1}$ (cells of about 169 km^2). The grid was then converted into 1°x1° resolution for atmospheric modelling applications (Fig. 7). MS emissions occur in 3,039 cells, ranging from 15 to 471,000 tonnes yr^{-1}. The cell with the highest emission is located in the Caspian region (Azerbaijan). The sensitivity of the MS modelling is discussed in the Supplement (Section S3.3).

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6.5 Evaluation of global MS emission and $\delta^{13}C$-CH_4

The global MS emission derivable by summing the emission from the cells of the 4 MS classes, about 24 Tg yr^{-1} (Table S6), is within the range, 10-25 Tg yr^{-1}, previously suggested by Etiope and Klusman (2010). The emission-weighted $\delta^{13}C$-CH_4 resulting from gridded MS is -51.4‰ (non-weighted average is -46.4‰). This value is mostly influenced by areas with elevated MS of microbial gas, such as the Po Basin (Italy), the Transylvania Basin (Romania) and the Powder River Basin (USA). The global emission-weighted value was applied to cells without isotopic value.

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6.6 MS uncertainties

Spatial distribution uncertainty: The uncertainty of the spatial distribution of MS depends on the assumption, supported by field measurements, that MS occurs significantly only within petroleum fields (PFA) and areas with seeps (OSA). The uncertainty of PFA depends on the "Petrodata" dataset of Päivi et al. (2007), discussed in section 5.1, and it cannot be quantified. The uncertainty of OSA depends on the buffer applied to individual seeps, which was however defined by geospatial analysis (see Supplement, S3.2).

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Emission uncertainty: The uncertainty of the MS emission depends on the activity (EMA) and on the process-based model of attribution of the seepage levels (emission factors), and their statistical elaboration, discussed in section 6.2 (see also Fig. S10). Changing activity by ± 20% and emission factor by the 95% confidence interval of the median, with different combinations, resulted in a total MS output ranging from 15 to 32.7 Tg yr^{-1}, with a mean of 23 Tg yr^{-1}, matching the first estimate (see Supplement). We can therefore set, approximately, a maximum uncertainty in the total MS output of ±9 Tg yr^{-1} (about ±38%).

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The model was then tested comparing its output values with measured values. This comparison was possible for 9 areas where the coordinates of the measurement points were identified. In all cases, measured and modeled values have the same order of magnitude, and in many cases the range of the MS level attributed by the model includes the mean value measured.

$\delta^{13}C$ uncertainty: The uncertainty of individual MS $\delta^{13}C$ values depends on the assumptions discussed in Section 6.3. The uncertainty of the emission-weighted mean (-51.4‰) is mainly controlled by the cells with larger MS emissions where $\delta^{13}C$ values are estimated. When the cells with emission-weighted mean are excluded, the remaining 536 cells (at 0.05°x0.05°, over a total of 192,166) have emission ranging from 5623 to 8296 tonnes $year^{-1}$ and $\delta^{13}C$ values from -65 to -35‰ (mean -53.4‰). The difference of this value with the

emission-weighted mean, i.e., 2‰, may be considered as approximate expression of the uncertainty of the global emission-weighted mean.

7. Geothermal manifestations (GM)

7.1 Global GM distribution

The global distribution of CH_4-emitting geothermal/volcanic sites (GM) generated here is based on an inventory of volcanoes and geothermal sites developed by Global Volcanism Program (2013) (see Sources of databases in the Supplement). This inventory reports all major volcanic-geothermal systems on Earth (2,378 sites; Fig. S12). They include both Holocene systems (1,307 sites distributed in 128 countries), and older, Pleistocene volcanic systems (1,071 sites distributed in 119 countries), which represent geothermal areas. In order to convert the point data into more realistic areal data (polygons), an arbitrary buffer area of 4 km of radius was created for each GM point (the buffer area does not influence the overall emission estimate, being only a parameter guiding the gridding). It is important to outline that this inventory reports the "zones" of volcanic/geothermal sites, and does not list individual manifestations: for example, the numerous geothermal manifestations in Central Italy are cumulatively included in a few lines, e.g., "Vulsini complex", "Sabatini complex" and "Vulture". Therefore each emission value, attributed as explained in Section 7.2, represents a "regional" GM emission.

7.2 Attribution of CH_4 emission levels

Methane flux measurements and regional total estimates in GM are available only in a few cases (<100 sites), mostly in Europe (as reviewed by Etiope et al. 2007). The GM inventory refers to geothermal-volcanic areas where GMs are expected to occur, but their actual surface area is unknown. Therefore, even assuming an emission factor (from the limited flux dataset) it cannot be translated into emission for each GM site. In this work, theoretical numbers were adopted considering 3 classes of "regional" emissions: 500, 5000, 10,000 tonnes yr^{-1}, as central values of the ranges 100-1000, 1000-10,000 and 5000-15,000 tonnes yr^{-1}, respectively. These ranges were derived from emission factors ranging from 1 to 150 tonnes km^{-2} yr^{-1} (Etiope et al. 2007) applied on a area of 100 km^2, as average order of magnitude of the extension of geothermal/volcanic zones (derived from Global Volcanism Program, 2013). Although the GM emission grid developed here is expected to improve global CH_4 inverse modeling (as it includes previously neglected GM sources), the total GM emission estimate suggested by the gridding, because of the uncertainty of the theoretical emissions, is not meant to update or refine the previous global GM emission estimate (derived by process-based modelling; Etiope, 2015).

The emission level was attributed based on:

(a) the location of the geothermal site, which may be within or outside a sedimentary basin (Fig. S13),

(b) the concentration of CH_4 measured in the geothermal fluids, within and outside a sedimentary basin.

The amount of methane in a geothermal-volcano area depends, in fact, on the presence of sediments rich in

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organic matter, which may be source of thermogenic gas in addition to the geothermal abiotic gas. The CO_2/CH_4 ratio of emissions to the atmosphere is in the order of 1000-10,000 in volcanic sites, with limited sedimentary contribution, and it ranges from 1 to 100 in geothermal systems characterized by important sedimentary covers. Sediment-Hosted Geothermal Systems (SHGS) in sedimentary basins (e.g., Etiope, 2015; Procesi et al, submitted) show the highest CH_4 concentrations (lowest CO_2/CH_4 ratio). In addition, sedimentary basins hosting petroleum fields reasonably contain larger amounts of methane. The three classes of methane emissions are reported in the Supplementary Table S9.

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7.3 Attribution of the $\delta^{13}C$ value

A specific dataset was compiled listing 98 published $\delta^{13}C\text{-}CH_4$ values of various, geographically dispersed, geothermal/volcanic systems in the world. The isotopic $\delta^{13}C\text{-}CH_4$ values range from -43.2 to -6.4‰, with an average of -26.7‰. The double-sided Grubbs test (Grubbs, 1969) identified 4 outliers; the mean $\delta^{13}C\text{-}CH_4$ value of the 94 values excluding the outliers is -26.5‰. It is known that geothermal methane in sedimentary basins, due to the presence of organic matter and related thermogenic gas, has a lower $\delta^{13}C\text{-}CH_4$ value compared to magmatic, sediment-free, systems (e.g., Welhan, 1988). The NPP of the $\delta^{13}C\text{-}CH_4$ data shows a sharp deviation at about -29‰ (Fig. S14). This value is actually consistent with the isotopic boundary of dominantly thermogenic gas; we used therefore this value as the limit between GM falling outside sedimentary basins and GM within sedimentary basins. The mean values of the two classes (excluding the outliers) are summarized in Table S8.

7.4 GM gridding

The GM shapefile generated in ArcGIS environment was spatially joined to the 1°x1° vector square grid. The result is reported in Table S9 and mapped in Fig. 8.

7.5 Evaluation of global GM emission and $\delta^{13}C\text{-}CH_4$

The 2,378 GM sites yield a total methane emission of about 5.7 Tg yr^{-1}, which is within the range of the latest global GS emission estimate (2.2-7.3 Tg yr^{-1}; Etiope, 2015). The emission-weighted mean value of $\delta^{13}C\text{-}CH_4$ for the GM emission is -30.6‰ (non-weighted mean is -27.5‰).

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Eliminato: are theoretical estimates, consistent however with the emission factor dataset and with the global GM emission, published in the literature, derived from process-based models

7.6 GM uncertainties

Spatial distribution uncertainty: The uncertainty of the spatial distribution of GM has the same uncertainty as the global distribution of geothermal-volcanic areas, derived from Global Volcanism Program (2013).

Emission uncertainty: The gridded GM emission, equivalent to the sum of individual regional values attributed as described in Section 7.2, has an uncertainty of about 75% (5.7±4.3 Tg yr^{-1}).

$\delta^{13}C$ *uncertainty:* The uncertainty of emission-weighted GM $\delta^{13}C$ may refer to the average of the two values corresponding to the 95% confidence interval of the means of the two groups of isotopic data (outside and within sedimentary basins) discussed in section 6.3, i.e. ±2.5‰.

8. Merging OS, SS, MS and GM: total geo-CH_4 emission gridding

8.1 Global geo-CH_4 emission

The global geo-CH_4 emission distribution, obtained merging OS, SS, MS and GM grids, is shown in Fig. 9. The total gridded CH_4 emission is 37.4 Tg yr^{-1} (Table 3, second column). The extrapolated gridded emission estimate including the factors not considered in the gridding procedure (i.e. mud volcano eruptions, existence of onshore and offshore seeps not included in the OS-SS inventories) is between about 43 and 50 Tg yr^{-1} (Table 3, third column). These values are within the published global bottom-up estimates (Table 3, fourth column). The global extrapolated geo-CH_4 emission is then compatible with recent top-down estimates (about 50 Tg yr^{-1} by Schwietzke et al. 2016; see also Section 9 for a wider discussion addressing the temporal variability of geological methane emissions). The scope of columns 3 and 4 in Table 3 (extrapolated and published emission estimates) is only to show that gridded emissions do not necessarily represent the actual global geo-CH_4 emission, because the datasets developed for the gridding may not be complete or may not contain the information necessary for improving previous estimates. Considering the four geo-CH_4 source categories individually, the gridded MS and GM emission totals are, however, within published ranges. The differences between gridded and published OS and SS are largely due to:

- incomplete OS dataset (it represents only 30% of global number of seeps assumed to exist on Earth)
- lower estimate of the global MV area (680 km^2 instead of 2800 km^2 assumed in previous works)
- incomplete SS flux dataset (flux data missing from at least 16 areas with known gas emissions).

The gridded emissions may represent an updated assessment of the global emissions only for MS and MVs (part of OS), because the gridding implied a careful assessment of the spatial distribution and emission factors for these types of geo-sources.

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8.2 Global geo-CH_4 $\delta^{13}C$

Based on the emission-weighted $\delta^{13}C$ value for each category of emission (using the respective emissions from Table 3), the global geo-CH_4 emission-weighted average $\delta^{13}C$ is -49.4‰, considering global emission estimates and -48.5‰ for gridded emissions (Table 4). The global distribution of the isotopic signature is shown in Fig. 10.

8.3 Uncertainties of gridded geo-CH_4 distribution, emission and isotopic value

The overall uncertainties of the spatial distribution of the geo-CH_4 sources, CH_4 emissions and emission-weighted average values of $\delta^{13}C$, depend on individual uncertainties of the four categories of seepage, as discussed in the respective Sections 4.6, 5.6, 6.6 and 7.6. These are summarized in Table 5.

9 Note on temporal variability of geological methane emissions

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The fluxes of natural gas seepage from the Earth's crust are not constant, either on short (hours, days, months, seasons) or long (years, centuries, millennia) time scales. Seepage variations can be induced by endogenous (geological) and exogenous (atmospheric) factors, including subsurface gas pressure variations (controlled mainly by gas migration and accumulation processes), changes of fracture permeability (tectonic stress, seismicity), hydrostatic aquifer variations, meteorological and climatic changes (atmospheric pressure, temperature, humidity and microbiological activity in the soil; Etiope, 2015). Mud volcano episodic eruptions (Mazzini and Etiope, 2017), seismicity-related degassing (e.g., Manga et al. 2009) and seasonal variability of microseepage (higher in winter due to lower methanotrophic consumption in the soil; Etiope and Klusman, 2010), are three, well studied, examples of geo-CH_4 emission variability. Anthropogenic activity, through modification of aquifer pressures (water pumping) and petroleum exploitation (with consequent decrease of reservoir pressures) can also induce seepage variability over time (e.g., Etiope, 2015). Therefore the global geo-CH_4 emission reported in this work, as well as in all other estimates available in the literature, must be interpreted as average, present-day degassing. Substantial decadal changes of seepage could occur as a result of decadal changes of hydrostatic aquifer pressure (e.g., Famiglietti, 2014) and decadal changes of seismicity (e.g., Mogi, 1979). Specific empirical studies are however missing, and with the present state of knowledge it is impossible to provide a temporal variability factor.

On longer, geological time scales, a series of proxies suggested that geo-CH_4 emissions could have been quite variable over the Quaternary period (Etiope et al. 2008b). Recent estimates on geo-CH_4 emission at the end of Pleistocene deserve a specific discussion. Based on radiocarbon (^{14}C) measurements in methane trapped in ice cores in Antarctica, Petrenko et al. (2017) estimated the absolute amount of ^{14}C-containing CH_4 in the atmosphere 11-12 k years ago, between the Younger Dryas and Preboreal intervals; this allowed to estimate that the maximum global natural, geological (^{14}C-free) CH_4 emission for that period was at most 15.4 Tg yr^{-1}. More recent analyses by the same authors confirmed this value (Dyonisius et al. 2018). These authors have then assumed that past geological methane emissions were no lower than today. They concluded, therefore, that present-day geological CH_4 emissions are much lower than present-day bottom-up estimates (54-60 Tg CH_4 yr^{-1}; Etiope 2015; Ciais et al., 2013). Without entering discussions on the accuracy and meaning of the ice core ^{14}C-based analyses and their temporal extrapolation to today, the following investigates whether the estimate by Petrenko et al. (2017) is compatible with:

(a) the estimates provided by authors other than Etiope (2015) and those reported in the present gridding work,

(b) the lowest bottom-up geo-CH_4 emission estimates available so far,

(c) present-day top-down geo-CH_4 emission estimates derived by different techniques, and

(d) pre-industrial geo-CH_4 emission estimates based on ice-core ethane measurements and observed geo-CH_4-to-ethane ratios.

Table 6 summarizes the data including individual literature references. In the bottom-up estimates table, the third column reports the lowest estimates proposed on the basis of more recent datasets and emission factors, which are updated in comparison with the earlier estimates (reported in the second column). The last column reports the overall lowest estimates, from old and new works, i.e. the minimum emission values derivable from different extrapolations. This comparison shows that the Petrenko et al. (2017) estimate is lower than any bottom-up estimate, regardless of authorship. The top-down estimates table reports geo-CH_4 emission derivable by three different procedures:

(a) assessing the portion of ^{14}C-free CH_4 in present day atmosphere (=30%; Lassey et al., 2007), then calculating the equivalent ^{14}C-free CH_4 emission (30% of total CH_4 emission, ~558 Tg yr^{-1} (Saunois et al. 2016) = 167 Tg yr^{-1}) and subtracting the anthropogenic ^{14}C-free component (fossil fuel fugitive emissions from inventories ~ 100-130 Tg yr^{-1}; EDGARv4.2; Saunois et al., 2016). The natural component (geo-CH_4 emission) would be 37-67 Tg yr^{-1}.

(b) Using methane concentration and isotopic data from ice-core records, based on box modelling by Schwietzke et al. (2016), suggest a geo-CH_4 emission of 30-70 (50) Tg yr^{-1}.

(c) With the same box model plus 3D forward modeling, but using current day atmospheric methane and isotopic data, Schwietzke et al. (2016) suggested a current day total fossil fuel (oil/gas/coal industries plus geological) CH_4 emission of 150–200 Tg yr^{-1}. Considering that oil/gas/coal emission inventories indicate 100-130 Tg yr^{-1}, geo-CH_4 emission could be 20–100 Tg yr^{-1}, consistent with approach (b) but with a wide uncertainty range.

(d) Using ethane concentration data from ice-core records, the 3 Tg yr^{-1} ethane top-down estimates by Dalsoren et al. (2018) confirm earlier bottom-up estimates of 2-4 Tg yr^{-1} ethane (Etiope and Ciccioli, 2009). Observed geo-CH_4-to-ethane emission ratios would then suggest 42-64 Tg CH_4 yr^{-1}.

Overall, geo-CH_4 emissions derived by top-down estimates range between 20 and 100 Tg yr^{-1}. These values are consistent with bottom-up estimates but substantially higher than Petrenko et al. (2017) estimate. The following options should then be considered:

(i) All current-day bottom-up and top-down geo-CH_4 emission estimates are biased high.

(ii) The Petrenko et al. (2017) estimate is biased low.

(iii) All estimates are reasonable, but the assumption that past Younger Dryas to Preboreal geo-CH_4 emissions were not lower than today does not hold.

10. Summary and Conclusions

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Gridded maps of global geological CH_4 emissions at 1°x1° resolution have been developed comprehensively for the first time for atmospheric modelling and evaluation of global CH_4 sources. The maps, elaborated by ArcGIS and provided as csv files, include the four main categories of natural geological CH_4 emissions: onshore hydrocarbon seeps (OS), submarine (offshore) seepage (SS), diffuse microseepage (MS), and geothermal manifestations (GM). A combination of published and originally *ad-hoc* developed datasets was used to determine the emission factors and the areal distribution and extent (activity) of the several geo-CH_4 sources and their stable carbon isotope signature ($\delta^{13}C$). Due to the limited number of direct CH_4 flux measurements, globally and regionally representative CH_4 emission factors for OS, MS and GM were estimated based on experimental emission factors (measurements) and statistical approaches. Methane emission estimates for SS were adopted directly from published regional emission estimates. The results of this work can be summarized as follows:

(a) The global geo-CH_4 source map reveals that the regions with the highest CH_4 emissions are all located in the northern hemisphere, in North America, in the Caspian region, Europe, and in the East Siberian Arctic Shelf.

(b) The globally gridded CH_4 emission estimate (37.4 ± 17.6 Tg yr^{-1} exclusively based on data and modelling specifically targeted for gridding, and 43-50 Tg yr^{-1} when extrapolated to also account for onshore and submarine seeps with no location specific measurements available) is compatible with published ranges derived by top-down and bottom-up procedures.

(c) The procedures adopted to attribute CH_4 fluxes to mud volcanoes (MV, a OS sub-class) and microseepage (MS) are based on a detailed assessment of the activity (areas) and emission factors, and the resulting gridded total output can be considered a refinement of previously published emission estimates. Specifically, the global MV emission estimate (2.8 Tg yr^{-1}, excluding eruptions) is compatible with early estimates by Dimitrov (2002), Milkov et al (2003), Etiope and Milkov (2004) and Etiope et al (2008). Global MS emissions (previously estimated between 10 and 25 Tg yr^{-1}; Etiope and Klusman, 2010) are now estimated to be ~24 (±9) Tg yr^{-1}.

(d) Regional emissions of SS are available from the literature for only a limited number of cases. The regions with missing emission data in the literature are not included in the gridded dataset developed here. As a result, the gridded CH_4 emission estimate (3.9 Tg yr^{-1}) is substantially smaller than a previously published global total estimate (20 Tg yr^{-1}, which would include extrapolated values to regions without region-specific estimates (Kvenvolden et al. 2001). However, the published SS estimate has large uncertainties (at least 10 Tg CH_4 yr^{-1} since two separate estimates of 10 and 30 Tg CH_4 yr^{-1} were actually provided without indication of their uncertainties) and it was purely based on process-based modelling (Kvenvolden et al. 2001). This work verified that SS emissions also occur in other regions where emission values are missing (among these, the Gulf of Mexico, Caspian Sea and the North US Atlantic margin). Given an estimated SS emission factor, we propose that global SS CH_4 emissions may range between 5–12 Tg yr^{-1}, with a best guess (central value) of 8.5 Tg yr^{-1}.

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(e) The emission-weighted global mean of $\delta^{13}C$-CH_4 is -48.5‰ for the gridded emissions, and -49.4‰ when gridded OS and SS emissions are extrapolated to include all global regions. The second value is therefore more realistic. Both values are significantly lower (about 4-5‰ lighter) than typical values attributed to fossil fuel industry sources (-44‰ by Schwietzke et al, 2016) and much lower (10-11‰ lighter) than seepage values considered in inverse studies (-38‰ by Sapart et al. 2012). Clearly, natural geological sources are more ^{13}C-depleted than generally assumed (and this mostly occurs as microseepage and submarine seepage). Low maturity thermogenic gas and microbial gas are, in fact, a neglected, but considerable, fraction of the global fossil CH_4 budget (Sherwood et al. 2017). It is expected that using the updated, more ^{13}C-depleted, isotopic signatures in atmospheric modelling studies will increase the top-down estimate of the global geological CH_4 sources (all else equal).

The maps developed here represent important inputs for future atmospheric modelling of the global CH_4 cycle. Fossil fuel industry "upstream" activities (exploration, production, and some processing of fossil fuels) and associated CH_4 emissions occur largely on land surface above sedimentary basins that are also the habitat for geological CH_4 seepage. Thus, there is substantial spatial overlap in CH_4 emissions from the fossil fuel industry and geological seepage. Nevertheless, there is substantial spatial variability in CH_4 emission intensity for both the fossil fuel industry (Maasakkers et al. 2016; JRC/PBL, 2017) and geological seepage (this work). In the absence of a comprehensive gridded geological CH_4 seepage product, global or regional inverse model studies would erroneously attribute a low-bias to CH_4 emissions from geological seepage. This is because of a de-facto zero geological a priori estimate. At the same time, the inverse studies would erroneously attribute a high-bias to CH_4 emissions from fossil fuel industry activity (and potentially other sources) while correctly reporting total emissions of all sources. The geological seepage data and maps developed here can be used to refine fossil fuel industry and microbial CH_4 emission budgets at the regional and global level. Finally, methane/ethane and methane/propane source composition ratios are available for the four categories of geo-sources (preliminary data were used in Etiope and Ciccioli, 2009) for use beyond the scope of this work. Combining the gridded geo-CH_4 emissions and the available source composition data, gridded ethane and propane maps could be developed in the future. The gridded geo-CH_4 maps shall be updated when additional, statistically significant gas flux data for the several seepage categories become available. Geo-CH_4 emission from a fifth, recently discovered, geological category, the seepage from serpentinized peridotites (e.g., Etiope and Schoell, 2014; Etiope et al. 2017 and references therein) shall also be gridded when sufficient flux data become available.

11 Data availability

The free availability of the data does not constitute permission for their publication. If the data are essential to new modelling, or to develop results and conclusions of a paper, co-authorship may need to be considered. Grid csv files (emission and isotopic composition for each geological source and integrated grid files) and microseepage and geothermal manifestations inventories are available at

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https://doi.org/10.25925/4j3f-he27 (Etiope et al. 2018), including full contact details and information on how to cite the data. The SS inventory is provided in the Supplement (Table S4). Due to CGG (2015) license restrictions, the OS inventory can be requested at *www.cgg.com/en/What-We-Do/Multi-Client-Data/Geological/Robertson-Geochemistry*.

The datasets of petroleum fields, faults, volcanic-geothermal sites, earthquakes, sedimentary basins are available on the web as described in the Supplement.

The Supplement related to this article is available online at ……

Competing interests. The authors declare that they have no conflict of interest.

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Tables

Table 1. Parameters and data sources used to generate grid maps of geological CH_4 sources. Complete references and links to data sources are provided in the Supplement.

	Onshore seeps (OS)	Submarine seeps (SS)	Microseepage (MS)	Geothermal manifestations (GM)
Activity data	Global seep distribution (georeferenced points)	Global distribution of marine seepage zones (georeferenced areas)	Global distribution of petroleum fields (georeferenced area)	Global distribution of volcanoes and geothermal sites (georeferenced points)
Data source	Updated GLOGOS dataset (after CGG, 2015; Etiope, 2015)	Originally developed data-set	"Petrodata" from Päivi et al. (2007)	Global Volcanism Program (2013)
Emission factors	Measurements and estimates based on size and activity	Measurements and estimates based on size, activity and depth	- Statistical evaluation of flux data - presence of faults - seismicity	- Measurements and estimates based on size and activity - presence of sediments
Data source	Literature, web sources	Literature	Merged global and regional databases USGS Earthquake Lists, Maps and Statistics	Literature Sedimentary basins world map (CGG data services)
δ^{13}C-CH_4	Measured or estimated value for each seep	Mean value for each seepage zone	Mean value for each basin or sub-basin	Global mean value based on statistical analysis
Data source	Updated GLOGOS (CGG, 2015), reservoir data (Sherwood et al. 2017) and petroleum system data (literature)	Published data or estimates based on local petroleum system	Petroleum reservoir data (Sherwood et al. 2017 and literature), seeps (OS data-set) and estimates based on the type of petroleum system	Literature data and estimates based on the type of system

Table 2. Estimates of global CH_4 emission from OS (onshore MV and other seeps)

	MV quiescent degassing	MV quiescent + eruption	Gas-oil seeps	Total quiescent	Total (incl. MV eruptions)
Dimitrov (2002)	0.3 – 2.6	10 - 12	nd	nd	nd
Dimitrov (2003) [a]	< 2.3	< 5	nd	nd	nd
Milkov et al (2003)[a]	< 2.9	< 6	nd	nd	nd
Etiope and Milkov (2004)	2.8 - 4	5.6 - 8	nd	nd	nd
Etiope et al (2008) [a]	< 3 - 4.5	< 6 - 9	3-4	6-8.5	9-13
Etiope et al (2011)	9	< 10-20	3-4	12-13	13-24
This work – 2827 seeps	2.83	nd	1	**3.8**	nd
This work – total extrapol.	~3	6.1	~ 2	~ 5	~ 8.1

nd: not determined

[a] Values include shallow submarine MV, therefore they can be considered as upper limits for onshore emission.

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Table 3. Global gridded, global extrapolated and global published geo-CH_4 emissions

Emission category	CH_4 gridded emission ($Tg\ yr^{-1}$)	CH_4 extrapolated * emission ($Tg\ yr^{-1}$)	Published ranges (best guess) ($Tg\ yr^{-1}$)
OS - Onshore Seeps	3.8 [a, b]	8.1	9 – 24 [d]
SS - Submarine Seeps	3.9 [c]	>7	10 - 30 (20) [e]
MS - Microseepage	24	24	10 – 25 [f]
GM - Geothermal Manifestations	5.7	5.7	2.2 - 7.3 [g]
Total	37.4 [a, b, c]	42.8 – 49.8	41- 76 (58)

* Including estimates from notes a, b, and c. See also text below. [a] Not including MV eruptions
[b] Partial (estimated <50%) gas-oil seeps emissions. [c] Excluding unidentified or not-investigated offshore seepage sites
[d] Etiope et al. 2008; Etiope et al. 2011 (see also Table 3). [e] Kvenvolden et al (2001)
[f] Etiope and Klusman (2010). [g] Etiope (2015)

Table 4. Global emission-weighted $\delta^{13}C$ values (‰)

Emission category	Emission-weighted $\delta^{13}C$
OS - Onshore Seeps	-46.6
SS - Submarine Seeps	-59
MS - Microseepage	-51.4
GM - Geothermal Manifestations	-30.6
Global weighted average (based on gridded emissions, 2nd column in Table 3)	**-48.5**
Global weighted average (based on globally extrapolated gridded emissions, 3rd column in Table 3)	**-49.4**
Global weighted average (based on published emissions, 4th column in Table 3)	**-49.8**

Table 5. Summary of uncertainty factors for the four types of gridded geological emissions

Emission category /Uncertainty	**Spatial distribution**	**Emission**	**$\delta^{13}C$***
OS - Onshore Seeps	Zero uncertainty on global scale Coverage 30% gas-oil seeps (but all biggest seeps reported) Almost complete MV coverage	Gas-oil seeps uncertainty: max. 90% (order of magnitude theoretically assessed) MV uncertainty: 48% (statistically assessed values) Overall OS uncertainty 58% (±2.2 Tg yr^{-1})	±1‰
SS - Submarine Seepage	Zero uncertainty for central values of gridded area Area extent from published papers Unknown % of global coverage (likely >80% ?)	From published data (central value used) Uncertainty 54% (±2.1 Tg yr^{-1})	±7‰
MS - Microseepage	Theoretically predicted (measurements and process-based model) Possibility that microseepage occurs outside petroleum fields (unknown gas pools) is accounted for	Process-based modelling Uncertainty max. 38% (< ±9 Tg yr^{-1})	±2 ‰
GM - Geothermal Manifestations	Zero uncertainty		±2.5‰

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		Process-based modelling (regional emissions) Uncertainty 75% (±4.3 Tg yr^{-1})	

* Uncertainty of the global emission-weighted $\delta^{13}C$ values of Table 4.

Table 6. Combinations of bottom-up and top-down estimates of geological methane emissions (Tg yr^{-1})

Bottom-up

	Lowest estimates from other authors	Lowest updated estimates	Lowest overall estimates
Onshore macro-seeps (includ. mud volcanoes)	5[a]	3.8[e]	3.8[e]
Global submarine emissions	10[b]	5[e]	5[e]
Global microseepage	7[c]	10[f]	7[c]
Geothermal	5.5[d]	2.2[g]	2.2[g]
Total	28	21	18

Top-down

Atmoshere ^{14}C-based (Etiope et al. 2008; Lassey et al., 2007)	37-67
Ice-core ^{13}C-CH_4 based (Schwietzke et al., 2016)	30-70
Current day emission data (Schwietzke et al., 2016)	20-100
Ice-core ethane (Dalsoren et al., 2018) and observed geo-CH_4-to-ethane ratios (Etiope et al., 20009)	42-64

[a] Dimitrov (2003); [b] Kvenvolden et al. (2001); [c] Klusman (1998); [d] Lacroix (1993); [e] this work; [f] Etiope and Klusman (2010); [g] Etiope (2015).

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