

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

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Author reply to Reviewer #3

R: Reviewer A: Author

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.

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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. 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

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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

C3

A: OK

R: Abstract : “This gap is particularly wide for geological CH<sub>4</sub> seepage, i.e., the natural degassing of hydrocarbons from the Earth’s crust. While geological seepage is widely considered the second most important natural CH<sub>4</sub> source after wetlands, it has been mostly neglected in top-down CH<sub>4</sub> 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 CH<sub>4</sub> emissions from geological seepage in the literature and in this paper, and the uncertain estimates from inland water systems, it is difficult to say robustly that geological source is the 2nd. I would say a major source.

A: Geological emission is formally reported as 2nd CH<sub>4</sub> 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. Saunio 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 a priori gridded emission maps. We have rephrased as follows:

While geological seepage is widely considered a major source of atmospheric CH<sub>4</sub>, it has been largely neglected in 3D inverse CH<sub>4</sub> budget studies given the lack of detailed a priori gridded emission maps.

R: P2 I5 : I suggest to update the ref to Saunio et al., 2016 and 558 MtCH<sub>4</sub>/yr A: OK

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R: P2 I7 : “emission inventories” and process-based models A: OK

R: P2 I8-9 : TD and BU show strong disagreement only or natural sources, please precise. A: OK

R: P2 I9-12. 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,  $\delta^{13}\text{C-CH}_4$ ) 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).

R: P2 I28 : geological degassing is today recognised as the second most important natural CH<sub>4</sub> 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 I29-30 : it is a bit unfair to quote specific papers when the highly visible synthesis from IPCC or GCP mention geological emissions in their budget (e.g. Sauniois et al., 2016). Please rephrase.

## C5

A: Ok, Sauniois et al 2016 is added.

R: P3 I34 – figure 1 : References to other sources should be updated to the Sauniois 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 I12-13 : what does it mean ? How can you know there is a seep if 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 I22 : 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.

## C6

R: P5 I 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 I30-39 : 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 I 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 observed? 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 : I15-16, 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.

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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<sub>4</sub> flux grids. For example, different wetland flux

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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 mean 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 why you cannot provide a range or a sigma for uncertainties.

A: This is now better explained in the text.

R: P9 | 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

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as follows: "...it is plausible that global SS emission exceeds 5 Tg yr<sup>-1</sup>".

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

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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 I29 : 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 I 37 : "few cases" : please provide a more precise number if possible.

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

R: P16 I 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 CH4 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 I25 : "It is known : : ." : any reference to justify this ? Any explanation ? please provide a reference or explanation

A: Ok reference + brief explanation added.

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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 I34 : 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 I27 : is there a risk that some SH emissions are forgotten because of less knowledge of the terrain ? A: Yes.

R: P19 I4-6 : 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 I31-33 : if no description of geological is given in an inverse modelling exercise,

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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<sub>4</sub> seepage product, global or regional inverse model studies would erroneously attribute a low-bias to CH<sub>4</sub> 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<sub>4</sub> emissions from fossil fuel industry activity (and potentially other sources) while correctly reporting total emissions of all sources.

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

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