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Interactive Comment

Interactive comment on "Atmospheric constraints on the methane emissions from the East Siberian Shelf" *by* A. Berchet et al.

Anonymous Referee #1

Received and published: 30 September 2015

It should be noted that this reviewer has an active project with one of the co-authors.

This is a professional analysis by a team of experts in inverse modelling and atmospheric trace gas measurement. It is an important evaluation of the atmospheric constraints on emissions claims in an important and dynamic region. It is a rare case of the top down modelling suggesting LOWER fluxes than bottom up data driven extrapolations (Kirschke et al. NatGeo 2013) e.g. McGuire et al. 2009 (p. 25478, ll. 2-3). Publicly available data are very few from the ESAS which lead to widely varying emissions scenarios that are highly uncertain.

The paper also contributes to the methodological groundwork for the use of inversion calculations in resolving such claims. It demonstrates the utility of using this approach

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to point out our gaps in our observational (e.g. too few in the Arctic) and accounting systems (e.g. uncertainty in wetland area resolution). This will become even more important as we will need to develop techniques with sufficient precision and resolution that will allow us to resolve changes in atmospheric burdens of climate forcing gases and, e.g. the veracity of national greenhouse gas reporting.

The paper is of significant importance and should be published.

However, before doing so, there are a number of clarifications that are required. The language needs to be tightened up (made more precise) and another look at the organization, specifically when and how figures are numbered and referenced in the text is needed. The figures that are used are appropriate and relevant to the discussion. More importantly the rationalizations for the data selection have to be made more explicit. There are very good and valid scientific rationales for this but the authors assume that these are obvious to the reader. The discussion of the use and interpretation of the isotope data in section 3.1 is particularly confusing. I have noted my opinion of my reading of this section in my comments below but the authors have to revisit this.

Particular issues:

p. 25478, ll. 10ff: Not certain of the meaning of: “Simulated mole fractions including a 8 TgCH4 y-1 source from ESAS are found largely overestimated compared to the observations in winter, whereas summer signals are more consistent with each other.” What are the mechanisms that force seasonality in coastal seas? Is it simply ice dynamics?

p. 25478, l. 18: “few”? is uncertain. Post the range of expected change.

p. 25479, ll. 4-5: a range is mentioned then a precise estimate is given.

p. 25479, l. 12: change “carbone” to “carbon”

p. 25479, ll. 13-14: The Shakhova 2010 paper does not offer hard evidence, data or references to data, only speculation for the existence of so much marine subsurface permafrost in this region. Check the Ruppel J. Chem. Eng. Data 2015 paper and

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references therein.

p. 25479, l. 19: change “that” to “which”

p. 25479, l. 20: change “at” to “to”

p. 25479, ll. 23-24: This is too modest a statement. The importance of ambient mole fractions and isotopes are underplayed here. These observations are critical and are required to constrain the emissions estimates. The gas has to go somewhere.

p. 25480, l. 3: change “a” to “an”

p. 25480, l. 4: change “emission” to “emissions” or use the specific article before “ESAS” in the previous line.

p. 25480, ll. 4-5: “. . .and the transport model are described as are the statistical analyses. . .” throughout: Use either “methane” or “CH4” (after its first use) consistently

p. 25480, l. 18: “Of the xx active observation sites. . .” Please be specific wherever possible.

p. 25480, l. 22: What is meant by “barely”? I suspect it might be closer to “infrequently influenced” or is there a better way to express this? The reasons must be made clear for the discrimination between and the ultimate choices of the specific sites that are used in the analysis. It looks like all the information is here but you need to be explicit about how you get from 22 stations to 4.

p. 25480, l. 25: I think “proposed” is a better word. Also the regions in the 2010 (which paper?) and 2014 papers are different and have different emissions mechanisms.

p. 25481, l. 3: Be careful of sequential figure numbering. These are entirely out of order and I can only find four figures in the Supplement to this paper. I imagine the “Figs S7, S8” actually refer to S2 and S3 (though they are numbered 2 and 3), again numbered or used out of order. Fig. 4 is referred to before Fig. 1.

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p. 25481, l. 3: “most of the time”? is very uncertain.

p. 25481, l. 7: There is no Figure S9. Does this refer to figure 4 in the Supplement?

p. 25481, l. 18-20: Here again is a filter that is not well explained. I think I understand it and it is certainly valid but it needs a word or two more to explain it.

p. 25481, ll. 21-22: This sentence needs to be clarified. What do you mean by “complete”? I suspect you are referring to those sampling periods at ZEP when there are both isotope and ambient measurements so the sampled air mass can be confidently traced back to the Siberian lowlands.

p. 25481, l. 25: Usually numbers ten or less are spelled out when they begin a sentence.

p. 25482, l. 1: Fig.3? Where is Fig. 2?

p. 25482, ll. 2-5: I do not entirely understand this statement. What are its implications?

p. 25482, ll. 17-19: Is the implication that tropospheric OH oxidation is included or expressed within the synoptic variation of the CH4?

p. 25482, l. 21: The use of “boundary” is unclear to me. Is this referring to the model constraints or to the boundary layer?

p. 25483, l. 6: Maybe specify the hatched area in Fig. 1. ESAS is not written on the map.

p. 25484, l. 8: “resp”?

p. 25484, l. 12: Does this constitute a “guess” or rather a calculated estimate based upon your modelling effort? I suggest not using the word. Why bother with the calculations if it is a mere conjecture?

p. 25484, l. 27: Does this refer to the uncertainties in the observed datasets used in the analysis?

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p. 25485, l. 2: change “wetland ones” to “wetlands”

p. 25485, l. 22: I am getting confused about the use of different years. Please be clear as to why you use 2010 sometimes and 2012 at other times.

p. 25486, ll. 2-6: Please break up this one long sentence into a readable paragraph.

p. 25486, l. 10: This implies that the isotopic analyses were done as part of this paper. As far as I can tell the isotope data from Fisher and Milkov are used to partition and constrain the specific source terms.

p. 25486, l. 10: Use “to” instead of “/”. The slash mark implies a ratio or fraction and its use here is ambiguous.

p. 25486, l. 14: change to “. . .and thawed permafrost . . .”. Thawing permafrost makes no sense as a source of methane. It can contain up to about 1 mM CH4 but rarely more. AFTER permafrost thaws then CH4 production can begin in earnest if the permafrost is waterlogged and then it will probably behave like the deeper layers of a wetland.

p. 25486, ll. 9ff: Section 3.1 is an important section and it is not clear. Is the point to CONFIRM or to TEST the assertion that thawing permafrost dominates the CH4 flux? First of all, this seems to depend a huge amount on the assumptions of what the source of the CH4 is and what the signature of that source might be. Is it thawing permafrost (mentioned at the beginning) or decomposing hydrates (mentioned at the end of the paragraph) that Shakhova et al. purport to be the source of the CH4? They seem to say decomposing shallow hydrates (but they offer no data to support that supposition) then the question becomes what is the source of the CH4 that forms those hydrates. Hydrates can have a broad range of 13CH4 isotopic compositions. Most researchers indicate the signatures are significantly heavier than biogenic sources.

The logical conclusion given the references in Fisher (Table 1) and in Milkov is that marine hydrates would provide a signature of around -55 to -50‰. Now the Fisher paper uses Keeling plots to determine the source signature of the late summer CH4 to suggest the source is biogenic then trajectory analyses to reasonably assign that source to Siberian wetlands.

Upon what are you basing your assumptions of the total flux (Kirshke et al maybe)? Are you saying that it is the assumption of the source (hydrate or wetland or biogenic) or the assumption of the magnitudes of those sources that is wrong? It APPEARS to me that you are assuming the very high magnitude of the Shakhova paper then making assumptions about what proportion of that total flux is from different sources with their different signatures then backing out the resulting signature of the total flux.

It is important to clarify this section. I THINK what you did was start with the Kirschke estimate (16 Tg/y) then take the 8 Tg/y magnitude from Shakhova (50%) and 6-7 Tg/y from wetland sources (35%, which you are assuming has a signature of -75 to -60‰ and then another 15% from somewhere (2Tg/y). What do YOU want say here?

p. 25486, l. 24: Again, this is a one sentence paragraph that is not clear. Significant CH4 emissions are only isotopically compatible with the values observed at ZEP if the CH4 source is biogenic. But if there was a doubling of the magnitude of the amount of methane then the molar ratios would shift adding an additional complication. And it is not clear what you mean with the last part that begins "…consistent with…".

The message is important to the evaluation. That the only way a purported source of the size posited by Shakhova et al. can be consistent with the observations is that those emissions have a biogenic signature. The signature is different for hydrate (or thermogenic) gas. And if an 8 Tg source has an expected hydrate signature (a hydrate source is conjectured) then the predicted signature would be inconsistent with what has been observed.

p. 25486, l. 19 and 20 and elsewhere: "resp." ?

p. 25487, l. 1: Probably should reference Fig. 4 in this first section.

p. 25487, l. 9: Explain what is meant by "…explained by boundary conditions…"

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p. 25487, l. 19: There are no Figs. S7 and S8.

p. 25488, l. 2: Please add the summer r values.

p. 25488, l. 8: "...not on the long run." is too idiomatic (as well as not quite correct) perhaps something like: "...but the calculations are inconsistent with a continuously sustained source."

p. 25488, l. 17: or from the directional quadrant of the ESAS region – especially given the discussion that follows from l. 20 ff.

p. 25488, l. 21: There is no Fig. S6.

p. 25488, l. 25: The coastal wetlands are very LIKELY to be poorly represented in at the 0.5o resolution of the LPJ model which is very likely to at least limit your ability to locate sources in space and time.

p. 25489, l. 5: Delete "...pieces of..."

p. 25489, ll. 5-6: and indicate that emissions that lead to an annual rate of 8 Tg cannot be sustained throughout the year nor identified in the atmosphere except possibly for the months of July and August.

p. 25489, l. 11: I would suggest moving the reference and discussion of Fig. 5 later. It is a product of the Monte Carlo analysis illustrated in Fig. 2 (which should be renumbered).

p. 25489, l. 13: Fig. 2 is referred to AFTER Figs. 1,3,4 and 5 (as well as the mystery Supplementary figures). Please arrange and renumber the Figures. Also Fig.2 needs a label on the y-axis.

p. 25489, l. 13: change "confirms" to "supports"

p. 25489, l. 16: change "uncorrelated" to "not correlated"

p. 25489, l. 18: change "proves" to "corroborates" or something else – very difficult to

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prove anything

p. 25489, l. 21: "confirms possible" is an interesting construction but ... Maybe just "indicates"

p. 25489, l. 23 and p. 25489, l. 25: change "...in a range of..." to "...to range from..."

p. 25489, l. 28: change "...from..." to "...to be..."

Note that this estimate is consistent with the aircraft observations of Kort et al. NatGeo, 2012

p. 25490, l. 7: A few words are needed to explain the meaning of "perturbed" here.

p. 25490, ll. 13-14: "...but adding such...would reduce..."

p. 25490, ll. 25 ff: It seems that the isotope discussion indicates that a large ESAS source is only consistent with that source being biogenic, i.e. light CH4.

p. 25491, l. 9: change "wetland" to "wetland area"

p. 25491, l. 15: I am probably biased but I do not think CRDS techniques are yet up to the precision we need for these analyses. I recommend generalizing this to "with laser spectrometry"

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 25477, 2015.