Manuscript prepared for Atmos. Chem. Phys.
with version 2015/11/06 7.99 Copernicus papers of the LaTeX class copernicus.cls.
Date: 17 January 2016

Detailed reply to comments from the two anonymous referees of "Atmospheric constraints on the methane emissions from the East Siberian Shelf"

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1 Introduction

We thank the reviewers for their detailed, precise, and very useful comments and suggestions on our manuscript. We acknowledge the fact that the text has to be improved in precision and clarity in the specified zones. The numerous comments and suggestions of the reviewer will be of great help to do so. In the following, we begin with answering the main points risen by the referees; then we treat point by point all their remarks and comments.

Comments by referee #1 are reported in red; those of referee #2 in blue. Our answers are in black font. Attached to our replies (at the end of the present document), one can find an updated manuscript with highlighted in red the revised sentences and paragraph.

2 General comments

2.1 On the choice of atmospheric data

– **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.**

– **When employing top down approaches to constrain emissions, the density of sites are important. This is particularly true for inverse modelling methods, but also the forward modelling as used in this work. There are few sites with quality assured CH4 measurements with high time resolution in the Arctic. Hence, it is surprising that the authors do not include the CH4 observations from the Zeppelin observatory in the study. In particular since Pallas is not located in the core of the domain influenced by ESAS emissions (Figure 1), and the Zeppelin site experiences influence from ESAS (see, Figure 1), and isotopic data from Zeppelin are used to confirm CH4 dominating Arctic sources at this sites. Harmonised observations of CH4 with high time resolution (1 h) is available from this site over the full year investigated (2012). Figure 1 indicates that Zeppelin will experience an influence of ESAS of ca 30-50 ppb (hard to see accurately from the figure), and the site will contribute to constraining the model simulations and fluxes, hence these data should be addressed to have a stronger fundament for the constrain of the flux estimate.**

– **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.**

We acknowledge that some clarifications are required about our choice of observations.

We agree that atmospheric measurements are scarce in the Arctic and one should take full advantage of all the available data. We need continuous measurements as we are inquiring into the synoptic signal. This means that the number of available site is in principle 22. To date, the year 2012 is the year with the highest number of available continuous observations around the Arctic. For more recent years, not all the observation sites have made their data available when we submitted the paper. For instance, at this time, we had no access to ZEP continuous data. This was mainly due to a change of instrument during 2012 and to some dysfunction in the instrumentation during the first three months of 2012. We have now obtained hourly measurements for ZEP from April to December 2012 and we have included them in the revised version of the manuscript (see Section 3.1 and figure 4). The addition of ZEP data only slightly modifies our results. We observe very marginal changes in the seasonal cycle of estimated ESAS emissions and the final annual range of emission is 0-4.5 $TgCH_4.y^{-1}$ instead of 0-4.3 $TgCH_4.y^{-1}$ in the previous version.

In order to complement our analysis on methane concentrations, we also use isotopic measurements to inquire into the origin of ESAS emissions. Unfortunately, we have no access to isotopic data in 2012. This is why we analyse measurements at ZEP in end summer 2008 and 2009 (this was mis-reported in the manuscript and we make it clearer in the revised version).

2.2 Analysis of isotopic measurements

- **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.**

- **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.

We thank the reviewer for his detailed, precise, and very useful comments and suggestions on our manuscript. We acknowledge the fact that the text has to be improved in precision and clarity in isotopic section.

We agree that the text of section 3.1 presenting the use of ^{13}C observation to constrain the source of air sampled at ZEP and coming from ESAS and Ob regions needs some clarification. Our main point is that only a dominant biogenic source is consistent with observed ^{13}C when air is coming from these regions. This can be due 1) to the microbial decomposition of recently thawed C-rich sub-sea permafrost, producing methane which eventually reach the atmosphere, 2) to the destabilization of marine hydrates trapped below a thawing sub-sea permafrost, and 3) to a biogenic continental source (wetlands, freshwaters, or microbial production after continental permafrost melting). Quantifying the fraction of air coming from the different source regions (ocean, continent) with our transport model, it is possible to show that emissions coming from ESAS should be very depleted in ^{13}C (-70 to -75‰) and that hydrates cannot explain the observed signals, but we cannot make a partition between oceanic or continental biogenic source. We revised the specific text of this paragraph and propose the following (hopefully clearer) text:

The isotopic composition in ^{13}C of Arctic air brings insights on the origin of the regional methane sources. Indeed, Arctic surface emissions mixed into the atmosphere own very different isotopic signatures (Fisher et al., 2011; Milkov, 2005): typically of -40 to -55 ‰ for gas leaks (thermogenic origin), -52 to -68 ‰ for marine hydrates (thermogenic and biogenic origin; range for methane in surface waters) and -60 to -75 ‰ for wetlands and biological degradation of thawing permafrost (biogenic origin). We use here δ^{13}C measured at ZEP observatory in combination with methane concentration measurements in September 2008 and September-October 2009 and compare them to CHIMERE simulations of atmospheric transport for the same period (Fisher et al., 2011). We assume that the estimation of ESAS fluxes for late summer 2012 can be transpose to the periods of isotope measurements in 2008 and 2009. Emissions are expected to vary from a year to the other, but this variability unlikely impacts on the qualitative isotopic calculations.

During the observation campaigns, episodes with identified air origin from River Ob and Eastern Siberia exhibited a mean signature of -65 ± 3‰ in September 2008 (Fisher et al., 2011) and of -62 ± 3‰ in 2009 (see Fig. 6). These values point toward a dominant biogenic origin of emitted methane. More precisely, in these air masses, the contribution of the different methane sources can be estimated as they are run separately in the CHIMERE model. ESAS emissions are found to contribute 35–45% to the observed signals (with ESAS emission strengths of 6 TgCH$_4$.y^{-1} as computed in Sect. 4.2), continental wetlands contributing to 35–40% and fossil fuels to 20–25%. Using these relative weights, together with the range of

associated source isotopic signatures, it is possible to calculate the integrated isotopic signature of sources at ZEP during the above-mentioned episodes. With a scenario of 6 $TgCH_4.y^{-1}$ for ESAS emissions in August–September (as deduced from Sect. 4.2), and depending on the range of the isotopic signature of other sources, it is found that only isotopic signatures in the range of -60 to -75 ‰ for ESAS source are compatible with the observations. This points at a purely biogenic origin when the sampling was performed. Conversely, if ESAS emissions were made entirely by degassing of hydrates trapped under the sub-sea permafrost, the simulated $\delta^{13}C$ signature at ZEP would be in the range of -52 to -61 ‰ thus only marginally compatible with the $\delta^{13}C$ observations.

Our simple methodology does not allow proposing a partition of this biogenic contribution between degrading thawing marine permafrost, degassing of marine hydrates and continental biogenic emissions, which are mostly related to wetlands and freshwaters, but it is possible to eliminate a dominant thermogenic and pyrogenic contribution. To go further, a full atmospheric inversion assimilating both ^{13}C and ^{12}C observations in addition to methane concentrations in the transport model would be necessary, which is beyond the scope and objectives of the present paper.

3 Point-by-point answers

- **Page 25478 - line 8: summer 2010 was this the year with isotopic data? According to section "3.1 Summer isotopic observations in the Arctic" this does not seem to be the case. The selection and description of years used throughout the manuscript are confusing, please clarify and check this thoroughly.**

  Indeed, we misreported the dates in the manuscript. Isotope measurements have been collected in September 2008 and September-October 2009. Other continuous measurements are done in 2012. This is now more clearly written in the text.

- **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?**

  According to Shakhova et al. (2010), ESAS emissions are stronger in summer than in winter, mostly related to ice cover and dynamics as suggested by the reviewer. Our prior emission scenario includes this variability. After analysis, it appears that summer emissions are somewhat realistic in Shakhova et al studies, but winter emissions are largely overestimated. The sentence is clarified in the new manuscript.

- **p. 25478, l. 18: "few"? is uncertain. Post the range of expected change.**

The range is highly uncertain for Arctic: 2–8°C depending on IPCC scenarios. This new range is explicitly reported in the manuscript.

- **Page 25478 - line 25: I propose to rephrase the sentence. The expression "... the regional methane budget still has significant uncertainties, depending on the method used for its assessment" can be misinterpreted, indicating that it is the uncertainty that depends on the choice of method, not the estimates. At this stage, I believe that no methods are proven better than others and the uncertainty is rising more from the lack of agreement between the approaches, I assume that was the meaning as well.**

  Indeed, the different methods suffer from high uncertainties due to too few available information on the region to constrain them at regional scale, and they do not agree with each other, making a consensus hard to reach in the region. We propose to rephrase : "'The regional methane budget still has significant uncertainties, revealed by the large spread of the emissions given by the different methods used for its assessment"'

- **Page 25478 – line 26: remove "the" before "one side"**

  OK.

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

  We report only the range in the amended version of the manuscript

- **p. 25479, l. 12: change "carbone" to "carbon"**

  OK.

- **Page 25479 – line 11: Carbone -> carbon**

  OK.

- **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 references therein.**

  Thank you for suggesting this enlightening reference, we adapt our statement to it and cite the paper.

- **p. 25479, l. 19: change "that" to "which"**

  OK.

- **p. 25479, l. 20: change "at" to "to"**

  OK.

– **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.**

We add two sentences to stress the key role of atmospheric measurements (methane and its isotopes) in inquiring into atmospheric emissions.

– **p. 25480, l. 3: change "a" to "an"**

OK.

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

We changed the sentence to: "to propose atmospheric insights on the magnitude of ESAS methane emissions"

– **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**

We revised this sentence in the new manuscript. We agree that "methane" and "CH4" are not used consistently. Now we use only "methane" in the text. We still use "CH4" in the units.

– **p. 25480, l. 18: "Of the xx active observation sites..." Please be specific wherever possible.**

We try to be as specific as possible now. To our knowledge, 12 continuous sites were active North of 55°N. Our concern was to have sites detecting ESAS signals, which correspond more to coastal sites around the Arctic ocean considering the atmospheric transport. Sites in Siberia are too influenced by regional fossil fuel and wetland emissions, making it difficult to detect ESAS signal. Some other sites are too remote (e.g. Greenland) or inland (e.g. in Canada) and hardly get ESAS signals.

– **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.**

OK.

– **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.**

We did not compile well the referencing in the submitted manuscript. We now pay attention to that problem to avoid any mis-numbering.

– **p. 25481, l. 3: "most of the time"? is very uncertain.**

We acknowledge the lack of precision of this statement. What we meant is that when a significant signal from ESAS is detected at these sites, the air masses have been directly transported across the Arctic ocean, with no other influence along the way. We revise this statement in the new manuscript.

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

Again a bug in latex. Corrected in the new manuscript.

– **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.**

We filter out night time observations as atmospheric models generally cannot reproduce hourly variability during the night, under shallow planetary boundary layer conditions. We then keep only afternoon observations. This is clarified in the revised manuscript.

– **Page 25385 - line 14: The rationale behind using afternoon averaged mixing ratios is not clear, bearing in mind the lifetime of methane, and also the long distance to local sources with high diurnal variation. Furthermore, is the OH reaction included? This is not clear from the description, please add. In any case, using afternoon mixing rations is probably not influencing the results, but the set up do not seem tuned to Arctic studies, and this might be addressed in later potential studies.**

OH reactions are not included in our system. We give more detail on this point in the model section.

The filtering of night-time observations is further explained now in Sect. 2.1. It is true that for most stations, no significant local emissions would influence the observations and the simulations during the night. However, even very little emissions could lead to large night-time biases in the simulations when the surface vertical mixing is really weak. This could be the case in the Arctic, as we experienced it in Siberia (Berchet et al., 2014). In any case, the influence of this filtering could not be important as most of ESAS illuminating episodes (in time series of Fig. 4) last a least a few days.

– **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.**

We use isotopic observations to get further information on the physical processes underlying ESAS emissions. The sentence is clarified now.

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

Thank you for this remark. We make it consistent in the updated manuscript.

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

We amended our use of figure reference to make it consistent.

– Page 25482- line 3: Please double check the use of years; it is confusing a few places. E.g. What is the relevance of mentioning 2010? The isotopic data is from 2008-2009, the other period is 2012, as it seems. Or?

We acknowledge the fact that we misreported the years in the submitted manuscript. We correct that in the new version of the manuscript.

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

This statement was here to justify the fact that we did not use ZEP data for the statistical analysis. But actually, we now realise that observations are available and usable for 9 months of 2012. ZEP is included in the new version of the manuscript, with very limited impact on the final numbers.

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

We do not take chemistry into account. We do so as air masses remain only a few days or weeks in the domain of CHIMERE. During this time, the influence of OH chemistry is really low on the concentrations. Moreover, OH-oxidation is expected to act smoothly compared to the synoptic variability, only having a limited influence on our statistical analysis.

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

We clarify this point in the new manuscript. We are referring to the lateral boundary conditions, i.e. to the concentration fields on the side of the limited domain of CHIMERE

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

Thank you for this remark. Updated in the new manuscript.

– Page 25483-line 20/21 ->: The use of the emission inventories in section 2.3 is adequate and appropriate, and this is essential studying the influence of an additional source. However, if I understand this correctly, emission for 2010 from EDGAR is used, and not 2012. The reason for this (which is obvious, but nevertheless) should be clearly stated, and also the potential impact of this approximation. Furthermore, it is not clear which year the LPJ modelled wetland emissions are used, and also not the GFED (fire emissions). This should also be clearly stated. My interpretation is that all emissions are for 2010, and the

meteorological conditions and input for 2012. Is this correct? I suggest to add a figure showing the monthly variations of the emissions throughout the year + total emissions to make this clearer, also with information of years. This can potentially be in the supplementary. Figure 5 is nice and useful, and this additional Figure suggested can be referred to in the context of this.

This needs to be clarified. All the available emissions at the time we wrote the paper treat years no later than 2010 and not 2012. Thus, we take anthropogenic emissions from 2010. The change in anthropogenic emissions reported in the inventory is annual and a potential update to 2012 numbers will not affect the synoptic variations a lot at atmospheric sites and only marginally the mean values. For wetland and fires, we compute a climatology over the ten previous years. This is a reason why we take large uncertainties in the Monte Carlo sampling. As requested by the reviewer, we add a figure in Sect. 2.3 showing the seasonal cycle of emissions, to highlight the monthly variations of emissions and their relative magnitude.

- **p. 25484, l. 8: "resp"?**

  Inappropriate abbreviation of respectively. This is corrected in the udpated manuscipt.

- **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?**

  Thank you for the remark. We directly use "The magnitude of ESAS emissions" now.

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

  The observational uncertainties are included into the 60 ppb of random noise applied to the simulations. We are referring to uncertainties in the observations, the transport model and the emissions. This is now clarified.

- **p. 25485, l. 2: change "wetland ones" to "wetlands"**

  OK.

- **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.**

  We acknowledge some errors in the use of years in the submitted manuscript. Continuous measurements are used only in 2012. The statistical estimates of ESAS emissions are thus based on 2012 data.

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

  Clarified in the updated manuscript.

– **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.**

The sentence is indeed unclear. We only use the measurements done by Fisher et al. (2011) for our isotope section. Milkov and Fisher papers were used to assess the isotopic signatures. This is clearer in the updated version.

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

Thank you for the suggestion. This is updated in the new manuscript.

– **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.**

Thank you for the clarification. This is corrected in the new manuscript.

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

Again a misuse of abbreviation. We now write fully "respectively"

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

The reference to figure 4 comes a little bit late indeed. We refer to it directly in the first paragraph.

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

We mean here the lateral boundary conditions. As PAL is close to the side of our domain, the atmospheric signal is mostly explained by air masses coming to the domain, and not by emissions within the CHIMERE domain.

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

This is now corrected in the manuscript.

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

We recall now summer and winter r values to clarify our statement here.

– **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."**

Thank you for this suggestion, that makes the text clearer.

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

The ESAS emissions with a 8 $TgCH_4.y^{-1}$ scenario flood all TIK surroundings with a 20 ppb background, no matter the wind direction. This makes it difficult to analyse ESAS signal in term of directional quadrant. This is why we cannot really go beyond hypotheses about the surrounding wetlands.

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

The new manuscript provides proper figure numbering.

– **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.**

This is indeed what we expect. This is a reason why we take rather high uncertainties in the Monte Carlo computation of uncertainties, so that we do not underestimate the uncertainties in our estimates.

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

Corrected.

– **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.**

Thank you for clarifying this sentence.

– **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).**

We have reorganized the beginning of this section. Our point was to make it clear that PAL does not influence the analysis much and is used to demonstrate that the CHIMERE model is able to reproduce Arctic methane concentrations at a site not influenced by ESAS emissions. Thus, only TIK, ZEP and ALT play a significant role in estimating ESAS emissions.

– **Page 25489 – line 11: "Figure 5 shows..." Should it be Fig. 5 or Figure 5? This is not consistent throughout the text.**

In the updated manuscript, we have made the references to figures consistent.

– **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.**

Again, we acknowledge the issues in the figure numbering. Figure 2 was not properly defined in method Sect. 2.4. We add some comments on this figure in the method section. Figure 2 is based on a polar projection, so that the x- and y-axis are labelled the same way. We add a sentence in the caption to clarify this point and also add some dash quadrants to link x and y axes.

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

  OK.

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

  OK.

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

  OK.

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

  We modify this construction to "points at"

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

  OK.

- **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**

  Thank you for pointing at this interesting paper with similar fluxes in north Canada. We cite it out of comparison with our current estimates.

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

  We are referring to the "perturbation" applied to monthly emissions for generating the Monte Carlo ensemble as described in the method section. This is not clear in the submitted manuscript. We slightly modify this sentence to make it clearer.

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

  OK.

- **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.**

  We agree with that. The conclusions on the isotopic section were not well reported. We move this sentence down in the conclusion and make it more explicit in the section dedicated to isotopes.

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

OK.

– **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"**

– **Page 25491 – Line 15: I would not recommend the use of commercial cavity ring down spectrometry for isotopic measurements at Arctic sites yet, as the precision is still, unfortunately, too low. I suggest to modify this statement. The sites are normally far from many of the sources and the dilution is high resulting in low variability over the year. Hence, very high precision is necessary**

We thank the referees for correcting a probably too optimistic statement. We use the general expression "laser spectrometry" in the amended manuscript.

– **Figure 4 (Page 25500): It would be interesting to see the difference between the simulations and observations more clearly. I propose an additional figure or table, potentially in supplementary. I suggest to add a figure (with bars?) showing the seasonal or monthly mean of observations, together with model simulations with and without ESAS. Preferably, also with standard deviations included to visualise the simulations and the observations better (or in a table)**

The monthly to seasonal biases between the observations and the simulations are dominated by the lateral boundary conditions forced by global re-analyses. Thus, no clear signal can be inferred from the seasonal cycle of the bias. Though, looking at the biases at ZEP and TIK clearly indicates that the scenario with 8 $TgCH_4.y^{-1}$ from ESAS is not unrealistic in summer. We give some details on the biases in Section 4.1

– **Related to this: Thank you for Table 1 (Page 25496). I miss more comments to this table and the correlation coefficients, and particularly the low correlation with Tiksi in the reference simulation during summer. Also a comment to the simulation after adding ESAS emissions would be very interesting**

We indeed missed any comments on low correlation in summer. We amend this point in the new manuscript. The low correlation clearly indicates that something is missing in the basic scenario, i.e. ESAS emissions and/or local wetland emissions have a significant influence on TIK.

– **I suggest to use either "methane" or "CH4", but consistently**

We now use it consistently. "Methane" is used all over the text and CH_4 only in units

- **Numbering of Figures in "Supplementary" is wrong. It is confusing referring to figure S7, S8, S9 etc. in the manuscript, as they are nowhere.**

  We experienced some bugs when compiling our manuscript with latex. This issue will be paid attention before resubmission.

- **I think it might be relevant to refer to Shakhova et al, 2015: http://rsta.royalsocietypublishing.org/content/373/2052/20140451**

  Thank you for pointing to this very recent paper with a clear acknowledgement of the very high variability of ESAS emissions, and thus the difficulty to quantify them with in situ observations. We cite it in the revised manuscript.

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Manuscript prepared for Atmos. Chem. Phys.
with version 2015/11/06 7.99 Copernicus papers of the LaTeX class copernicus.cls.
Date: 17 January 2016

Atmospheric constraints on the methane emissions from the East Siberian Shelf

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Abstract. Sub-sea permafrost and hydrates in the East Siberian Arctic Ocean Continental Shelf (ESAS) constitute a substantial carbon pool, and a potentially large source of methane to the atmosphere. Previous studies based on interpolated oceanographic campaigns estimated atmospheric emissions from this area at 8–17 $TgCH_4.y^{-1}$. Here, we propose insights based on atmospheric observations to evaluate these estimates. The comparison of high-resolution simulations of atmospheric methane mole fractions to continuous methane observations during the entire year 2012 confirms the high variability and heterogeneity of the methane releases from ESAS. **A reference scenario with ESAS emissions of 8 $TgCH_4.y^{-1}$, in the lower part of previously estimated emissions, is found to largely overestimate atmospheric observations in winter, likely related to overestimated methane leakage through sea-ice. In contrast, in summer, simulations are more consistent with observations.** Based on a comprehensive statistical analysis of the observations and of the simulations, annual methane emissions from ESAS are estimated to range from 0.0 to 4.5 $TgCH_4.y^{-1}$. Isotopic observations suggest a biogenic origin (either terrestrial or marine) of the methane in air masses originating from ESAS **during late summer 2008 and 2009**.

1 Introduction

Most long-range global climate projections forecast a warming in the Arctic of 2–8°C over the next decades (Collins et al., 2013). Warmer Arctic temperatures could induce the thawing of continental

and submarine permafrost and the destabilization of marine hydrates, causing massive methane emissions into the atmosphere, and hence, generating positive feedbacks to the regional and global warming. Monitoring methane emissions at high latitudes in the North Hemisphere is therefore of critical importance to anticipate and to interpret future climate changes. The different potential sources emitting methane in the Arctic are identified, **but no consensus has been reached concerning their magnitudes. The regional methane budget still has significant uncertainties, revealed by the large spread of the emissions given by the different methods used for its assessment.** For example, on one side, emissions of methane from the Arctic tundra estimated by flux observations and process-based models (i.e. bottom-up approaches) for the 2000s have been synthesized respectively at 20 [11 to 51] $TgCH_4.y^{-1}$ and 28 [18 to 37] $TgCH_4.y^{-1}$ (McGuire et al., 2009). On the other side, top-down atmospheric inversions, based on methane atmospheric observations, show a range for total natural Arctic methane emissions north of 60°N of 12 to 28 $TgCH_4.y^{-1}$ (Kirschke et al., 2013), i.e. smaller and slightly narrower than the bottom-up range, but still statistically consistent with bottom-up estimates. In addition, anthropogenic emissions are estimated at 9 [7 to 11] $TgCH_4.y^{-1}$ above 60°N by top-down inversions (Kirschke et al., 2013).

Methane emissions from the Arctic Ocean are lower than land emissions, but more uncertain relatively, as synthesized by McGuire et al. (2009), with a range of 1–12 $TgCH_4.y^{-1}$. The East Siberian Arctic Shelf (ESAS), which covers 2×10^6 km^2 or 14% of the Arctic Ocean, constitutes a large pool of carbon for potential Arctic methane emissions as a large part of Arctic marine permafrost (up to 40%; Ruppel, 2015) is located in this region after the flooding of Siberian tundra during the Holocene transgression (7–15 ky ago). When thawing, microbial activity can produce significant to large amount of methane but the fraction reaching the atmosphere remain largely disputed. Marine hydrates are a large pool of sub-sea methane, with very uncertain global emissions (2–9 $TgCH_4.y^{-1}$; Kirschke et al., 2013). Based on oceanographic measurements performed over almost a decade, Shakhova et al. (2010) suggested that ESAS emits 8 $TgCH_4.y^{-1}$ into the Arctic atmosphere, **which** is 2/3 of the 1–12 $TgCH_4.y^{-1}$ range by McGuire et al. (2009). Shakhova et al. (2014) revised ESAS emissions upwards **to** 17 $TgCH_4.y^{-1}$, accounting for methane emissions above several oceanic hotspots due to bubbling in the water-column and methane degassing to the atmosphere during storms. **However, due to very high spatial and temporal variability in methane fluxes, estimates of ESAS emissions are still uncertain (e.g, Shakhova et al., 2015).**

Observations of atmospheric methane mole fractions and of methane isotopes in the Arctic can improve our understanding of ESAS emissions (Dlugokencky et al., 2011; Fisher et al., 2011). **Arctic regional emissions of methane drive the variability of the atmospheric signal at distant locations through transport and mixing. This makes it possible to inversely constrain emissions with atmospheric observations and simulations of transport and mixing.** North of 55°N, 22 atmospheric stations measure methane mole fractions, among which 12 sites provide continuous observations and 3 sites sample the isotopic composition of air on weekly basis or during intensive

campaigns. Although sparse, these stations are well illuminated by ocean and land methane emissions because of fast horizontal transport of air masses around the North Pole (e.g., Bousquet et al., 2011, and supplementary material).

In this paper, atmospheric methane observations and high-resolution simulations of atmospheric transport in the Arctic are combined to evaluate the potential of an $8\,\mathrm{TgCH_4.y^{-1}}$ source from ESAS and to propose atmospheric insights on **the magnitude of ESAS methane emissions**. In Sect. 2, the observations and the set-up of the transport model are described as is the statistical analysis used to compare simulations to measurements. In Sect. 3.1, simulations from a $8\,\mathrm{TgCH_4.y^{-1}}$ reference scenario are compared to observed time series of methane concentrations to assess the likelihood of such a reference scenario. In Sect. 3.2, a comprehensive statistical analysis based on Monte Carlo experiments (described in Sect. 2.4) is carried out to propose a range of ESAS emission magnitudes compatible with circumpolar atmospheric observations. In Sect. 3.3, Arctic isotopic methane measurements are analysed to confirm the geophysical origin of ESAS methane emissions.

2 Materials and Methods

2.1 Observation sites

This study is based on the statistical analysis of one year of synoptic atmospheric signal (days to weeks) reaching atmospheric observation sites. Therefore, continuous observations are needed as weekly or biweekly sampling does not allow to capture synoptic changes. As the focus here is on emissions from ESAS, continuous observations that are sensitive to these emissions are needed. Year 2012 was chosen as the year with the largest number of available observations at the time the paper was written. The double constraint of data availability and of data sensitivity to ESAS emissions leaves 4 sites relevant for our analysis (see detailed characteristics in Tab. 1), out of the **12** observation sites carrying out continuous measurements of atmospheric methane mole fractions around the Arctic Ocean in 2012: one nearby site (TIK) and three sites remotely but regularly illuminated by ESAS emissions (ALT, BRW, ZEP). On top of these four sites, we selected one background site (PAL) poorly influenced by ESAS emissions (see Fig. 1) to evaluate the ability of the model to represent Arctic atmospheric methane. The remaining sites are either barely influenced by ESAS (e.g., the Ivituut site in Greenland, Bonne et al., 2014; or Canadian sites from Environment Canada, Worthy et al., 2003), or local and regional influences are dominant (e.g., fossil fuel and wetland emissions in the Siberian lowlands for observation sites of JR-STATION network; Sasakawa et al., 2010). Tiksi (TIK) is located closest to the shores of the Laptev sea, a few tens of kilometres only away from the emitting region proposed by Shakhova et al. (2010, 2014). Barrow (BRW) and Alert (ALT) are located at the northern edge of North America, in north Alaska and north Canada respectively, about 2000–2500 km away from the ESAS but still influenced by this region (see typical footprints in Fig. 2 and 3 in supplementary material). **Zeppelin observatory (ZEP) is operated on a summit**

of Svalbard island, about 2400 km away from ESAS (see Fig. 5 in supplementary material), but also illuminated by it. When these three remote sites are illuminated by ESAS emissions, methane-enriched air masses are transported to them directly across the Arctic ocean in 2–3 days. Therefore, usually no major continental emission areas lie along the air mass paths from ESAS to BRW, ZEP or ALT stations. Pallas (PAL), in northern Scandinavia, is taken as a distant site, with dominant influence from Europe (see Fig. 4 in supplementary material) and very limited influence by Laptev Sea emissions (typical contributions < 2 ppb; maximum 20 ppb in a few plumes; Fig. 1). Here, PAL is used for evaluating the capacity of our transport model CHIMERE (see Sect. 2.2) to reproduce the methane mole fraction variability at high latitudes and at synoptic scales in a basic scenario (see Sect. 2.3).

The methane mole fractions at the observation sites are analysed with instruments maintained by Environment Canada (EC; ALT), NOAA/Earth System Research Laboratory (NOAA/ESRL; BRW), the Norwegian Institute for Air Research (NILU; ZEP) and the Finnish Meteorological Institute (FMI; PAL and TIK). They are calibrated with standards traceable to the WMO X2004 CH_4 mole fraction scale (Dlugokencky et al., 2005). The combined standard uncertainty on individual measurement remains below the ± 3.7 ppb requested by the World Meteorological Organization (WMO/GAW, 2009).

The continuous observations are hereafter compared to simulated mixing ratios. Atmospheric transport models have known bias at night-time when the vertical mixing close to the surface is very small (e.g., Berchet et al., 2013). This bias deteriorates the model performance in reproducing the influence of local and regional sources to the observation sites during the night. To minimize this documented issue, only afternoon averages of observed mole fractions are compared to simulated equivalents in our analysis.

For enhancing atmospheric insights on ESAS emissions, **especially about the underlying physical processes causing emissions, we also analyse isotope measurements from** ZEP with clear identified origin from East Siberia (Fisher et al., 2011). Isotopes measurements of $\delta^{13}C_{CH_4}$ at ZEP are carried out by the Royal Holloway University of London (RHUL). Five litre tedlar bags are collected and analysed with modified gas chromatography isotope ratio mass spectrometry (GC-IRMS) at RHUL (Fisher et al., 2011). Methane emissions from the Arctic ocean are expected to dominantly come from microbial activity in the ESAS seabed and thawing carbon-rich permafrost as suggested by Shakhova et al. (2010), and less from hydrate methane destabilization. Isotopic compositions measured at ZEP during September 2008 and September–October 2009 are compared to CHIMERE simulations in Sect. 3.3 for assessing methane emission processes in ESAS.

2.2 Polar CHIMERE transport model

Atmospheric transport is simulated with the Eulerian meso-scale non-hydrostatic chemistry transport model CHIMERE (Vautard et al., 2001; Menut et al., 2013) over a limited-area domain. The

Table 1. Observation site characteristics. The site location is displayed in Fig. 1.

Station	ID	Location			Network / Institute (Data server)	Reference correlations r	
		Lon (° E)	Lat (° N)	Alt (m a.s.l)		Winter	Summer
Alert	ALT	-62.5	82.5	200	EC (WDCCGG[1])	0.79	0.56
Barrow	BRW	-156.6	71.3	11	NOAA/ESRL (ESRL[2])	0.76	-
Tiksi	TIK	128.9	71.6	29	FMI (ESRL[3])	0.56	-0.04
Pallas	PAL	24.12	68.0	560	FMI (WDCGG[1])	0.89	0.63
Zeppelin	ZEP	11.9	79.9	475	NILU (Pedersen et al., 2005)	0.87	0.70
					RHUL (Fisher et al., 2011)		

[1] World Data Center for Greenhouse Gases ($http://ds.data.jma.go.jp/gmd/wdcgg/$)

[2] Dlugokencky et al., 1995, 2014. $ftp://aftp.cmdl.noaa.gov/data/trace_gases/ch4/$

[3] $ftp://ftp.etl.noaa.gov/psd3/arctic/tiksi/greenhouse_gas/ghg_concentration/raw/$

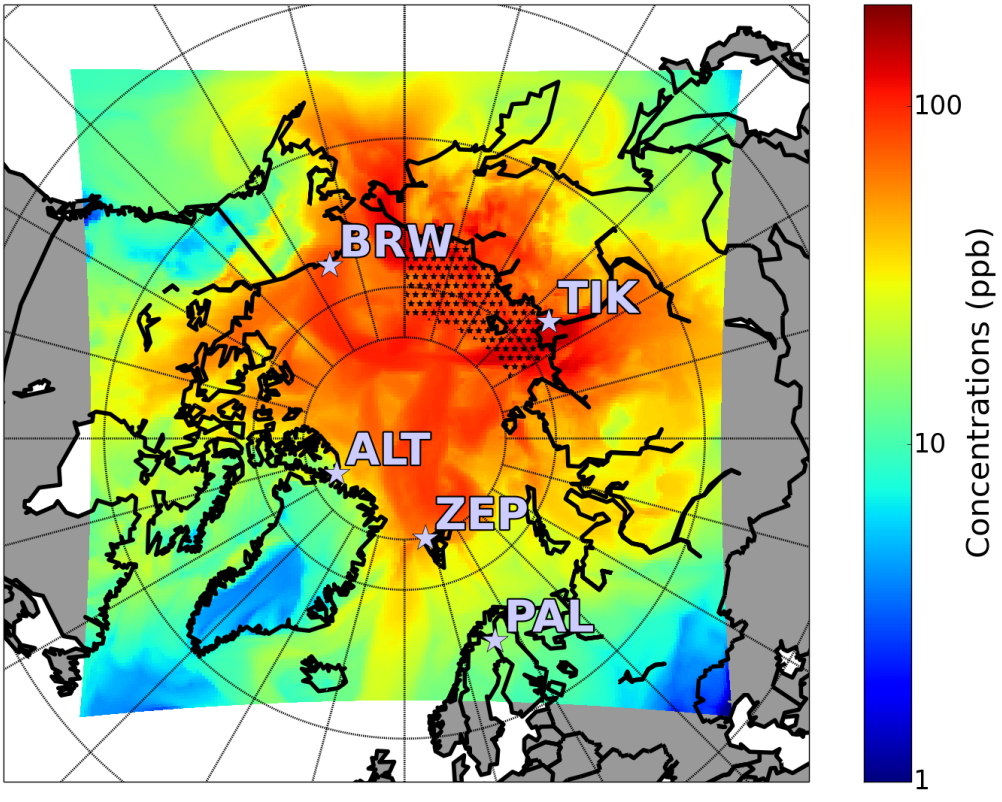

Figure 1. Map of the domain of CHIMERE simulations (see Sect. 2.2) with the emission zone from ESAS (black stars; see Sect. 2.3) and the stations used in the analysis. Shaded colours show the maximum over the whole year 2012 of near-surface simulated influence (in ppb) of the ESAS methane emissions after transport.

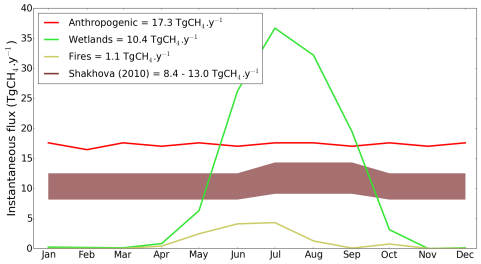

Figure 2. Seasonal cycle of prior emissions as used in the model CHIMERE.

model is constrained by meteorological fields interpolated at a spatial resolution of $0.5° \times 0.5°$ every 3 hours from re-analyses of the European Centre for Medium-range Weather Forecasts (ECMWF, ERA–INTERIM; Uppala et al., 2005). The original model has been modified to simulate atmospheric transport over polar regions with a regular kilometric resolution of 35×35 km^2 covering all latitudes from 50°N up to the North Pole (as illustrated by Fig. 1). Such a kilometric resolution avoids the numerical issues in grid cells becoming very small close to the pole, as it is the case for longitude/latitude regular grids. The transport simulations represent the troposphere in the region from the surface to 300 hPa ($\sim$ 9000 m) with geometrically spaced vertical layers of 10 m close to the surface and 300 m in the upper troposphere.

Methane has a lifetime of 8–9 years regarding oxidation by the OH radicals (e.g., Voulgarakis et al., 2013). As the focus is put here on synoptic variations within days or weeks of atmospheric methane mole fractions at the surface, methane chemistry is not accounted for in our set-up of the model CHIMERE.

2.3 Transport inputs and emission scenarios

The regional transport model CHIMERE requires boundary conditions to its limited-area domain: i) surface emissions within the domain and ii) lateral and top 3-d concentration fields accounting for transport and emission outside the domain to force the open sides of the domain (lateral sides and top side). Lateral boundary 3-d fields of mole fractions are interpolated from global analyses obtained by assimilating surface mole fraction measurements in the global circulation model LMDz (Locatelli et al., 2014). The 3-hourly global analyses at $3.75° \times 1.875°$ of resolution are interpolated at the **lateral and top** sides of CHIMERE domain for the required dates.

Surface emissions for the CHIMERE domain are deduced from state-of-the-art models and inventories: (1) EDGAR v4.2 FT2010 inventory for anthropogenic emissions (http://edgar.jrc.ec.europa.eu), (2) LPJ model for wetland emissions (Spahni et al., 2011), (3) GFED v3 model at a daily scale for fire emissions (Giglio et al., 2009; van der Werf et al., 2010), and (4) emissions from ESAS (see below and hatched area in Fig. 1). The EDGAR inventory uses up-to-date economic activity maps by sector, convolved with emission factors estimated in laboratories or with statistical studies (Olivier

et al., 2005). LPJ model includes a dynamical simulation of inundated wetland areas (Stocker et al., 2014), dynamic nitrogen cycle (Stocker et al., 2013), and dynamic evolution of peatlands (Spahni et al., 2013; Stocker et al., 2014). The model uses CRU TS 3.21 input data (temperature, precipitation rate, cloud cover, wet days), observed atmospheric CO_2 and prescribed nitrogen deposition (Lamarque et al., 2011) for each year for the simulation of dynamic forest and peatland vegetation growth. The GFED v3 database is built from the 500 m Collection 5.1 MODIS DB burned-area mapping algorithm (Giglio et al., 2009). Methane emissions at monthly and daily scales are deduced from the burnt areas using Carnegie-Ames-Stanford-Approach (CASA model; Potter et al., 1993) and emission factors (van der Werf et al., 2010).

EDGAR v4.2 FT2010 reports emissions for the year 2010, and not 2012. Anthropogenic emissions are reported on an annual basis in this inventory and have been found to only change slightly for the Arctic in the fast track recent release for 2012 (http://edgar.jrc.ec.europa.eu/). Moreover, as we analyse here synoptic signals, our results are not very sensitive to annual small changes. We thus directly transpose 2010 anthropogenic emissions to the year 2012. GFED v3 database and LPJ model also do not provide emission fluxes for years later than 2010. We thus take a climatology of biomass burning and wetland emissions computed over the years 2000 to 2010 to represent fire and wetland emissions in 2012. In the absence of the actual year, this is a conservative approach in order to represent all potential emitting zones for these two sources.

The first three types of emissions are projected from their original grids of $0.1° \times 0.1°$ (EDGAR) and $0.5° \times 0.5°$ (LPJ, GFED) to CHIMERE grid. ESAS emissions are directly built on CHIMERE grid from Shakhova et al. (2010) as they provide a detailed description of the emission zones and emission strengths per period (winter/summer). As it is suggested in Shakhova et al. (2010), hot spots are separated from background emissions and summer fluxes (mid-June to mid-September) from winter ones (the rest of the year). We prescribe uniform and constant emissions by emission type (hot spots and background) and period (summer and winter). Doing so, we underestimate the variability of methane emissions from ESAS which likely vary on shorter time scales, especially in winter in relation with sea ice breaks and ice displacements after periods of accumulation under the ice. This means that simulated mole fractions are less contrasted with smaller peaks and higher background values than with a more realistic (but unknown) flux map. We scale ESAS emissions, so that annual emissions are 8 $TgCH_4.y^{-1}$, **in the lower range of the previous estimates**.

Fig. 2 presents the seasonal cycle of prior emissions used as CHIMERE inputs. Anthropogenic emissions are constant over the year, the small variations on the monthly emissions simply coming from the different numbers of days in each month. Wetland and fire emissions have a smooth cycle with high emissions in summer and almost no emissions in winter. Considering the magnitudes of each type of emission, ESAS emissions are expected to be noticeable in the atmospheric signals, especially in winter.

The four types of emissions are run as separate passive tracers in polar CHIMERE for 2012, which allows analysing the contribution of each source separately at observation sites. The combination of the contributions from the four types of emissions and from the transported **lateral** boundary mole fractions provides the modelled methane mole fractions including the ESAS contribution. The emission scenario not including (respectively including) ESAS emissions is hereafter called the basic (respectively reference) scenario. The basic and reference scenarios are compared to observed time series in Sect. 3.1.

2.4 Statistical assessment of ESAS emissions

The magnitude of ESAS emissions can be derived by summing scaled ESAS emissions to the basic scenario (see Sect. 2.3), so that simulated time series fit the best to observed time series (see Eq. 1 below for agreement score definition). However, the emission databases used in the transport model, as much as the lateral boundary conditions and the transport representation itself, suffer from uncertainties. A tolerance interval for ESAS emission magnitude as seen by atmospheric sites is computed through Monte Carlo experiments to account for these uncertainties.

The Monte Carlo ensemble (20000 samples hereafter) is generated by randomly scaling for every month of the year the anthropogenic emissions, the wetland emissions and the lateral boundary conditions. The distributions used for these perturbations are Gaussian distributions of respectively 50%, 75% and 50% of errors. In addition to the emission scaling, we also add a random Gaussian noise on the simulated mixing ratios of 60 ppb of standard deviation. This noise on the simulations is expected to represent the errors on the transport and from the imperfect distribution of the emissions. **The scaling factors applied on emissions and the random noise in the Monte Carlo sampling** have been chosen in the upper range of known uncertainties in the used data sets (e.g., Kirschke et al., 2013 for emissions and Patra et al., 2011 for transport), so that the final uncertainties on ESAS emissions are not under-estimated.

For every Monte Carlo sample i (i.e. a specific perturbed set of anthropogenic emissions, wetlands, and lateral boundary conditions, with added transport noise), model-observations agreement scores S are computed for ESAS emissions from 0 to 20 $\mathrm{TgCH_4.y^{-1}}$; other emission rates (wetlands and anthropogenic) are not changed. The model-observations agreement score S is the sum of the local scores s at ALT, TIK, ZEP and PAL (BRW is not used in the computation of the score S as no observations are available between June and December 2012). Local scores s are defined by the centered RMS distance (i.e. the distance to the reference observation point in a Taylor diagram; Taylor, 2001):

$$s^2 = 1 + \left(\frac{\sigma_s}{\sigma_o}\right) - 2\frac{\sigma_s}{\sigma_o} r \tag{1}$$

where σ_s (respectively σ_o) is the simulated (respectively observed) standard deviation and r the correlation coefficient between the observations and the simulations at the selected site.

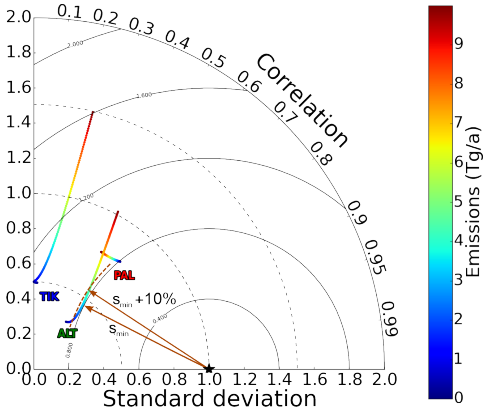

Figure 3. Taylor diagram representation of the statistical analysis of ESAS emissions (see Sect. 2.4). For each observation site, ESAS emission scenarios from 0 to 10 $TgCH_4.y^{-1}$ are located on the Taylor diagram, depending on the compatibility (correlation and standard deviation) between observations and simulations, thus generating a compatibility trajectory. **The Taylor plot is based on a polar projection with the standard deviation as the radius and the correlation coefficient as the polar angle.** Point colours depict the ESAS emission magnitude for each scenarios. To compare the different sites, each trajectory has been normalized by the site standard deviation.

With this definition of the scores, varying ESAS emissions results in trajectories in the Taylor diagram, as illustrated in Fig. 3. For all samples i of the Monte Carlo ensemble, we define a minimum agreement score S_{min}, which corresponds to the points of the emission trajectories closest to the reference point (perfect correlation and no bias; black star in Fig. 3). Tolerance intervals TI_i for ESAS emissions are deduced for all samples of the Monte Carlo ensemble, so that all ESAS emissions with associated scores within $[S_{\text{min}}, S_{\text{min}} + 10\%]$ are considered compatible with the atmospheric signal. These tolerance intervals are computed for every month of the year 2012. In the end, for every month of the year 2012, we compute aggregated tolerance intervals for ESAS methane emissions such that 95% (equivalent to 2σ interval for Gaussian distributions) of the Monte Carlo ensemble is within the interval.

This statistical analysis is not performed on the whole available dataset, but on afternoon averaged mixing ratios. This processing protocol is widely used in atmospheric quantitative studies and reduces the impact of local emissions not-well mixed in the meso-scale transport model (see Sect. 2.1).

3 Results

In the following, simulated mole fractions for the four source contributions described above are compared with methane continuous observations. Then, the Monte Carlo statistical analysis is applied to estimate the methane emissions from the ESAS which best fit the atmospheric

methane observations. Finally, isotopic remote observations are used to confirm the origin of ESAS methane emissions.

3.1 Model–observation comparisons at four Arctic sites

At PAL, BRW, ZEP and ALT, **as shown in Fig. 4**, the continuous methane observations exhibit similar seasonal variations with a minimum during summer (June–July) and a maximum during winter (December–January). At PAL, ZEP, and less evidently at ALT, the synoptic variations appear larger in winter than in summer. At TIK, the seasonal maximum is observed in August, associated with large synoptic variations and a less pronounced seasonal cycle, suggesting an influence of local to regional emissions during summer months.

At PAL, a site scarcely influenced by ESAS emissions, most of the atmospheric signal is explained by **the lateral boundary conditions (i.e. by air masses coming from outside the CHIMERE domain)**, especially the large synoptic variations during winter months. Polar–CHIMERE computed with our basic emission scenario demonstrates a very good (respectively good) skill in winter (respectively summer) in representing the atmospheric methane mole fraction variability at high latitude sites. As shown in Fig. 4A, the variability of the daily averages observed methane mole fractions is indeed well captured by CHIMERE (annual temporal correlation of $r = 0.87$; winter $r = 0.89$; summer $r = 0.63$; Tab. 1). Discrepancies between the observed signal and the simulated one at other sites can then be reasonably interpreted in terms of mis-specified regional emissions.

At ZEP, ALT and BRW (Fig 4B, C and D), three sites remote from ESAS but influenced by long-range transport from ESAS across the Arctic ocean (see Fig. 2 and 3 in supplementary material), non-summer mole fractions (i.e. all the year but June–September) are well reproduced by the basic scenario ($r = 0.87$, $r = 0.79$ and $r = 0.76$, respectively). In the reference scenario (see Sect. 2.3), the contribution of ESAS is much too large at ALT, ZEP and BRW for non-summer months as shown by the large blue spikes of Fig. 4 between January and April (ALT, BRW), between March and June (ZEP), and between October to December (ALT only, no data available for BRW during summer 2012). Moreover, as discussed in Sect. 2.3, the actual time distribution of ESAS emissions is not represented. A realistic time distribution would have led to enhanced simulated spikes, reinforcing the inconsistency of winter ESAS fluxes.

In summer, at ALT and ZEP, the fit of the reference scenario to the observations is less favourable than in winter **($r = 0.56$, respectively $r = 0.70$ in summer against $r = 0.79$, respectively $r = 0.87$ in winter for ALT and ZEP).** Adding ESAS emissions may fill some gaps in July–August, less in June and September, though some spikes at ALT are too high and phases are not always in agreement with observations. Some summer peaks from the ESAS are very well reproduced by the model (Fig. 4) at ALT and ZEP in July/August. This would suggest that sudden bursts of methane may be released on short periods (typically days) during July and August, with instantaneous rates correspond-

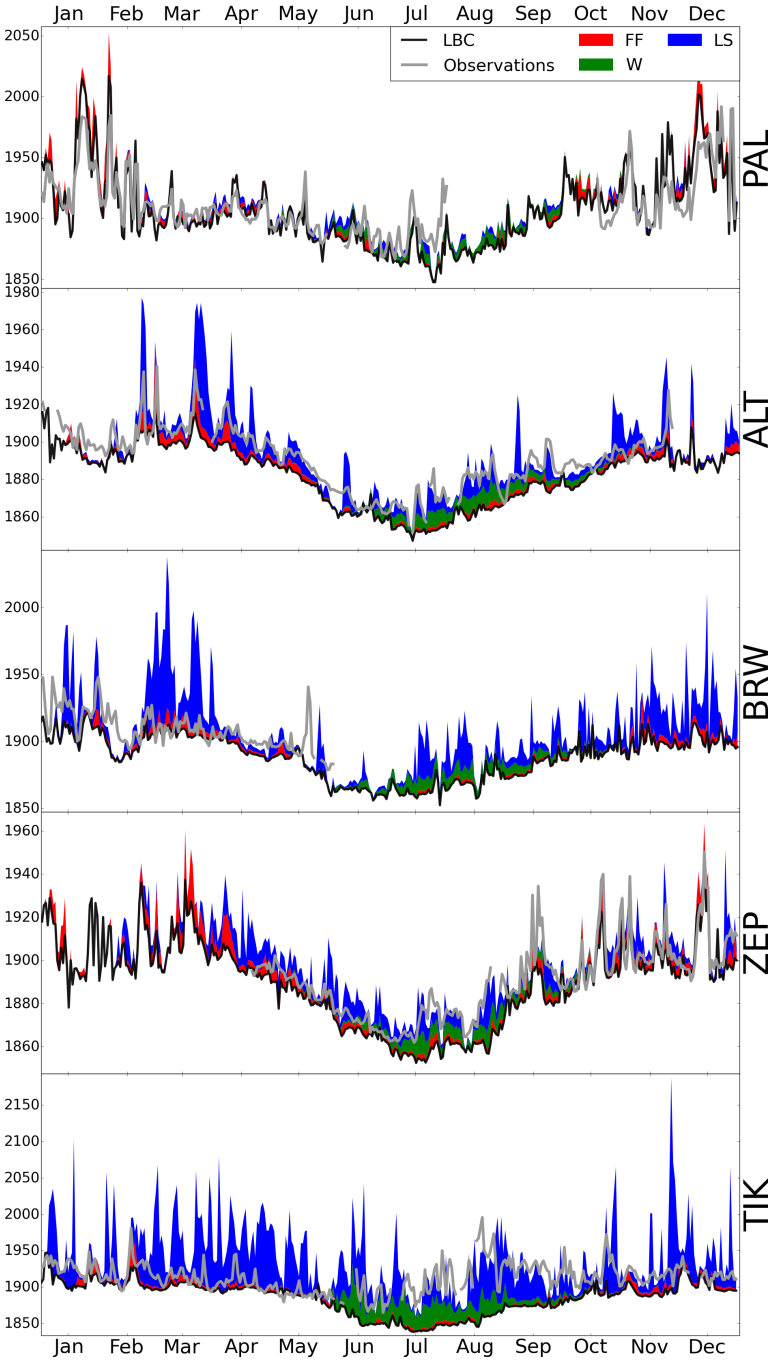

Figure 4. Time series of observed and simulated methane mole fractions at **five** Arctic sites in 2012. The filled areas depict the daily afternoon contributions from wetlands (W, green), fossil fuels and other anthropogenic emissions (FF, red) and from the ESAS (LS, blue; 8 $TgCH_4.y^{-1}$ scenario; Sect. 2.3). The LBC line (black) represents the contribution of the lateral boundary conditions transported into the domain. Grey lines are observations (daily averages of continuous measurements). Fire emissions are not represented in this figure due to very low influence on the studied sites.

ing to 8 $TgCH_4.y^{-1}$, **but a sustained source from ESAS is inconsistent with the observation-simulation comparison**.

Getting closer to the ESAS, TIK methane observations compared to simulations confirm that the simulated contribution of ESAS emissions from January to April and from October to December is over-estimated (Fig. 4E). Indeed, the baseline of observations is well reproduced by the basic scenario, despite some unexplained spikes in winter (Fig. 4E), which slightly decorrelate the fit of the basic scenario to observations ($r = 0.56$ in winter; Tab. 1). These spikes can be attributed either to small and short-term releases of methane from ESAS or to other emissions not properly represented or transported to TIK. In June, the contribution from the ESAS is still too large compared to observations. However, from July to September, the observed mole fractions are higher and more variable than the basic scenario. **Additionally, from July to September, the simulations decorrelate from the observations at TIK ($r = -0.04$ in summer), and the average simulations–observations differences are -36 ppb at TIK in the basic scenario, while they average at -1 ppb in the reference scenario with ESAS. The same applies to ZEP, where the bias is reduced from -6 ppb to 0 ppb when adding the 8 $TgCH_4.y^{-1}$ scenario from ESAS.** This suggest emissions from ESAS that are compatible with the 8 $TgCH_4.y^{-1}$ scenario, or even higher, for these 3 months.

However, as confirmed by the footprint analysis at TIK (supplementary material; Fig. 1), observations from July to September are mostly influenced by regional emissions (closer than 200 km), including ESAS. Within this radius of influence, wetland emissions from north Yakutia (mainly along Laptev sea shores between Lena and Indigirka rivers) could also significantly contribute to observed methane mole fractions at TIK. If such wetlands are ill-represented in LPJ model at $0.5°$ of resolution (either in magnitude or timing), this could dampen the compatibility of the 8 $TgCH_4.y^{-1}$ scenario with TIK observations for summer months. **Nevertheless, methane emissions from surrounding wetlands only have a significant influence on TIK site, and not on remote sites, as their emission magnitude is low compared to ESAS emissions. The improved compatibility of the reference scenario with ESAS compared with the basic scenario at ZEP from July to September indicates plausible high summer methane emissions from ESAS.**

In summary, the emission scenario from Shakhova et al. (2010) shows a large over-estimation of methane mole fractions at Arctic stations during all months but from July to September. Definite conclusion on the exact magnitude of ESAS methane releases cannot be obtained from TIK alone, due to possibly mis-accounted regional influences from natural wetlands, but also to the simplified spatial and temporal scenario used here for ESAS emissions. The distant observation sites (ZEP, BRW and ALT) are more likely to provide integrated information about the methane fluxes from ESAS. **These three sites indicate that emissions that lead to an annual rate of 8 $TgCH_4.y^{-1}$ cannot be sustained throughout the year nor identified in the atmosphere except for the months of July to September.** In the following, we estimate the ESAS emissions that are compatible with

atmospheric observations using a comprehensive statistical approach (see Sect. 2.4), accounting for the uncertainties of our atmospheric approach.

3.2 Estimation of methane emissions from the ESAS

As seen in Sect. 3.1, PAL is not affected by a change in simulated ESAS emissions, resulting in a very short trajectory in Fig. 3. This supports its status of background site regarding ESAS emissions. Thus, the Monte Carlo statistical analysis detailed in Sect. 2.4 is mostly influenced by sites well illuminated by ESAS emissions all over the year: ALT, ZEP and TIK. BRW is not used as 6 months of data are missing in 2012.

Fig. 5 shows the monthly methane emissions deduced from the statistical analysis. Despite the large uncertainties prescribed in the Monte Carlo experiment, the posterior uncertainties on ESAS emissions are low (1-$\sigma < 1.5\,\mathrm{TgCH_4.y^{-1}}$). The signal emitted by ESAS is not correlated with other signals of atmospheric methane in the Arctic, which makes it easier to analyse from the atmospheric point of view. This corroborates that the chosen observation sites are relevant for constraining the ESAS emissions and are robust to errors in the quantification method.

The score analysis points at high methane emissions (up to $1.6\,\mathrm{TgCH_4}$ in July) in summer from July to September, as suggested by the time series in Fig. 4. Mean summer flux rates are estimated **to range from** 4 to $11.5\,\mathrm{TgCH_4.y^{-1}}$, consistent with Shakhova et al. (2010) estimates from intensive summer campaigns. However, for other time periods, which are less documented by in-situ campaigns, low emissions are found **to range from** 0.3 to $1.9\,\mathrm{TgCH_4.y^{-1}}$ on average. This is roughly 4 times lower than previous winter estimates. Overall, on a yearly basis, our statistical analysis suggests methane emissions **to be** 0.0 to $4.5\,\mathrm{TgCH_4.y^{-1}}$ from the ESAS, **somewhat similar to estimates of methane fluxes from the Arctic ocean North of Canada as deduced from aircraft measurements (Kort et al., 2012)**.

The estimate computed here is to be considered as an upper bound for ESAS emissions for the two following reasons. First, the monthly flat temporal emission profile from ESAS in our emission scenario underestimates the impact of ESAS region on synoptic methane variations at observation sites. In the real world, concentration peaks due to shorter and more intense methane release from ESAS would be larger, thus reducing further the estimated emissions in order to match atmospheric observations. **Second, the local and regional influence of wetland emissions may be systematically under-estimated in the global LPJ model at high latitudes (e.g. around TIK station; as suggested by intercomparison of wetland emission models in Siberia; Bohn et al., 2015). We do not fully account for this potential bias in our Monte Carlo analysis as wetland emissions have been monthly rescaled with a centered Gaussian distribution in the Monte Carlo ensemble.** Indeed, the most extensive wetland area (a 200–300 km wide coastal lowland) in the vicinity of TIK is located to the East. Wetland emissions from this area may be either missing or partly displaced in a global model such as LPJ. More work is needed to provide a more realistic regional wetland

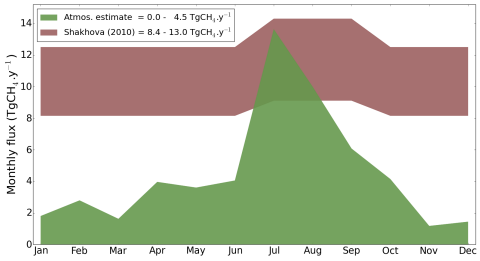

Figure 5. Monthly fluxes in $TgCH_4.y^{-1}$ as deduced from agreement scores (green; see Sect. 2.4) computed for every month of the year 2012, compared to Shakhova et al. (2010) fluxes (red).

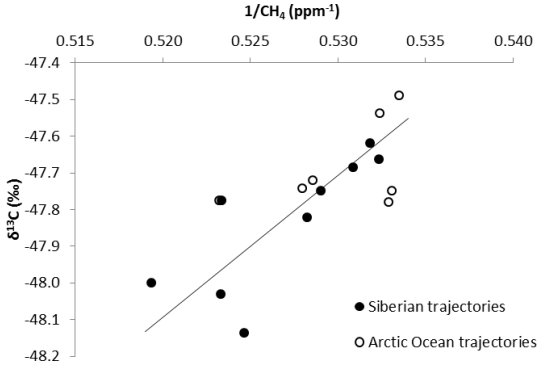

Figure 6. Keeling plot for observations carried out at ZEP observatory in Sept.–Oct. 2009. Only the observations with a dominant origin from the ESAS and Siberia or from the Arctic Ocean are kept here. The y axis intercept of the Keeling plot is $-62 \pm 5‰$.

scenario, **but adding such unaccounted for or underestimated wetland emissions would reduce** our ESAS emission estimates (in order to match the observed concentration at TIK).

3.3 Summer isotopic observations in the Arctic

We assume that the estimation of ESAS fluxes for late summer 2012 as computed in Sect. 3.2 can be transposed to the periods of isotope measurements in 2008 and 2009. Emissions are expected to vary from a year to the other, but this variability unlikely impacts on the qualitative isotopic calculations.

The isotopic composition in ^{13}C of Arctic air brings insights on the origin of the regional methane sources. Indeed, Arctic surface emissions mixed into the atmosphere own very different isotopic signatures (Fisher et al., 2011; Milkov, 2005): typically of -40 to -55 ‰ for gas leaks (thermogenic origin), -52 to -68 ‰ for marine hydrates (thermogenic and biogenic origin; range for methane in surface waters) and -60 to -75 ‰ for wetlands and biological degradation of thawing permafrost (biogenic origin). We use here $\delta^{13}C$ measured at ZEP observatory in combination with methane concentration measurements in September 2008 (Fisher et al., 2011) and September-October 2009

and compare them to CHIMERE simulations of atmospheric transport for the same period. **We assume that the estimation of ESAS fluxes for late summer 2012 as computed in Sect. 3.2 can be transposed to the periods of isotope measurements in 2008 and 2009. Emissions are expected to vary from a year to the other, but this variability unlikely impacts on the qualitative isotopic calculations.**

During the observation campaigns, episodes with identified air origin from River Ob and Eastern Siberia exhibited a mean signature of $-65 \pm 3‰$ in September 2008 (Fisher et al., 2011) and of $-62 \pm 3‰$ in 2009 (see Fig. 6). These values point toward a dominant biogenic origin of emitted methane. More precisely, in these air masses, the contribution of the different methane sources can be estimated as they are run separately in the CHIMERE model. ESAS emissions are found to contribute 35–45% to the observed signals (with ESAS emission strengths of 6 $TgCH_4.y^{-1}$ as computed in Sect. 3.2), continental wetlands contributing to 35–40% and fossil fuels to 20–25%. Using these relative weights, together with the range of associated source isotopic signatures, it is possible to calculate the integrated isotopic signature of sources at ZEP during the above-mentioned episodes. With a scenario of 6 $TgCH_4.y^{-1}$ for ESAS emissions in August–September (as deduced from Sect. 3.2), and depending on the range of the isotopic signature of other sources, it is found that only isotopic signatures in the range of -60 to -75 ‰ for ESAS source are compatible with the observations. This points at a purely biogenic origin when the sampling was performed. Conversely, if ESAS emissions were made entirely by degassing of hydrates trapped under the sub-sea permafrost, the simulated $\delta^{13}C$ signature at ZEP would be in the range of -52 to -61 ‰ thus only marginally compatible with the $\delta^{13}C$ observations.

Our simple methodology does not allow proposing a partition of this biogenic contribution between degrading thawing marine permafrost, degassing of marine hydrates and continental biogenic emissions, which are mostly related to wetlands and freshwaters, but it is possible to eliminate a dominant thermogenic and pyrogenic contribution. To go further, a full atmospheric inversion assimilating both ^{13}C and ^{12}C observations in addition to methane concentrations in the transport model would be necessary, which is beyond the scope and objectives of the present paper.

4 Conclusions

We suggest some insights on methane emissions from the Eastern Siberian Arctic Shelf using atmospheric methane observations, to complement the intensive in-situ oceanographic measurement campaigns carried out mostly in summer in the region. We test the consistency of a methane emission scenario including a 8 $TgCH_4.y^{-1}$ source from the ESAS. This scenario is run in a high-resolution model representing Arctic atmospheric transport and confronted to continuous methane concentrations performed at remote and nearby continuous atmospheric stations. The analysis of the modelled and observed time series suggests a large overestimation of ESAS emissions for all months but sum-

mer months, but still a high contribution of ESAS emissions from July and August, also consistent with isotopic observations. Over 2012, a statistical analysis based on model/observations comparison is performed to estimate ESAS emissions and address the uncertainties of our approach. Our method suggests methane emissions from ESAS of 0.0 to 4.5 $TgCH_4.y^{-1}$. Although significant at the regional scale, especially in summer, these revised emissions are about 2 to 5 times smaller than previous estimates from Shakhova et al. (2010) and 6 to 10 times smaller than the most recent estimates (Shakhova et al., 2014). The time series from the different sites also confirm a very likely heterogeneous temporal variability and spatial distribution, with very short and local methane releases from ESAS. **Finally, remote $\delta^{13}C_{CH_4}$ observations are also used to identify the processes emitting methane in ESAS, pointing at dominant biogenic processes, excluding any thermogenic and pyrogenic processes.**

A multi-year analysis with more observation sites and an improved representation of the regional wetland **area** should be carried out in order to reduce the uncertainties in ESAS emission estimates and to properly identify the sensitivity of the emissions to the ice cover or to other meteorological conditions and the distribution and short-scale variability of the fluxes. The use of another transport model would also be important to address biases in the representation of transport, not addressed by our statistical analysis based on centered perturbations. The development of continuous $^{13}CH_4$ observations at Arctic observation sites, now possible through laser spectrometry, would provide additional constraints for partitioning emissions between marine hydrates, gas leaks, thawing permafrost and continental wetlands. Finally, the observatories operated around the Arctic Ocean could also provide more quantitative estimates of Arctic emissions from ESAS using direct and inverse modelling of both methane and $^{13}CH_4$ observations.

Acknowledgements. We thank the PI from the observation sites we used for maintaining methane measurements at high latitudes and sharing their data. We are grateful to the anonymous referees for their valuable comments which led to a substantial improvement in the presentation of the paper. We also thank Patrick Crill (Department of Geological Sciences, Stockholm University) for his kind advices and remarks on the manuscript and F. Marabelle (LSCE) and the LSCE IT team for the computer resources. This study was supported by the CEA, ANR-CLIMSLIP project and iZomet Franco-Swedish project.

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