The original comments are in bold text and the answers are in normal text.

Answers to:

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

by N. Shakhova
nshakhov@iarc.uaf.edu

Received and published: 2 November 2015

We acknowledge the time spent by Dr. Shakhova to read and comment our paper. Considering the size of her (short) comment (10 pages of very dense comments and questions, organized around three main topics), we propose answers at two levels (see below): a synthesis of the main points of our answers and a detailed point-by-point answer.

NS: Thank you for acknowledging my time spent to read and comment on your paper – I very much appreciate this and promise to spend more time commenting on every assumption used in your model and in the "mother" model, in an article specifically devoted to this topic that I am currently preparing. Commenting on your paper is the only opportunity for me to learn from you first-hand; I am grateful to you for this opportunity.

1/ Synthesis of our detailed answers

A) The number, accuracy, and spatial-temporal coverage of observations constraining the inversion;

Atmospheric transport integrates the heterogeneity of surface emissions and, before suppressing gradients by mixing, it produces peaks of concentrations at sampling stations located downwind the emissions zones. This is physics and it is used every day by all atmospheric scientists working on greenhouse gases, air pollution, aerosol pollution, and even radio-element pollution, at various spatial scales from local to global. With few atmospheric sites, and continuous observations, it is therefore possible to test emission scenarios using a transport model to link emissions to concentrations. This is what we do here to test ESAS methane emissions reported in Dr Shakhova's papers. To do so, we do not perform a full formal Bayesian inversion but a simple optimization based on a model/observations comparison. Doing so, we find that ESAS emissions as reported by Dr Shakhova's papers are too large on a yearly basis. We understand that atmospheric transport is not the main research topic of Dr Shakhova. This is visible in many questions about atmospheric transport and modeling, concentrations peaks, background/initial conditions, boundary conditions… Anyway, we provide detailed answers below to all the questions.

NS: Yes, you are absolutely right, atmospheric transport is not my main research topic, but I do not claim that it is; I just hope that common sense and knowledge of very basic physics serves my purposes. In addition, I am constantly learning by communicating with professionals like you. In this regard, I would appreciate it very much if you would provide me with clear and direct responses to the following comments.

You say that atmospheric transport is able to produce methane concentration peaks downwind from the emission zones (this is how you track down the atmospheric signal from the ESAS). From the content of your paper, it follows that such peaks reflect increases in downwind concentrations as compared to upwind concentrations. You also state that such increases could have been clearly registered at downwind stations due to specific features of air transport in the Arctic. I have doubts about this. First, if it were true, we would have observed such peaks reflecting downwind increases in atmospheric

concentrations of CH_4 while conducting a survey along the Siberian coast, all the way from the western to the eastern reaches of the Siberian shelf (>4000 km along the Northeast Passage) in 2005 (see figure 3A in Shakhova et al., 2010, Science 327, 1246). These peaks, according to your explanation, should have been determined either by the CH_4 emissions from the west Siberian wetlands (the largest source in the Northern Hemisphere), or by the CH_4 emissions from the ESAS. Contrary to what we should have been observing according to your explanation, while crossing the Kara Sea (west Siberia) we observed atmospheric concentrations of CH_4 to be within the LSMM (around 1.85 ppm) and then we observed significantly increased atmospheric concentrations of CH_4 when the survey crossed into the Laptev and the East Siberian seas. This observation runs contrary to your modeling assumptions/results.

Second, if CH_4 emissions in the ESAS that range from 3-13 mg m^2 d^{-1} (reported in Shakhova et al., 2010) could have produced peaks in downwind atmospheric concentrations of CH_4 by up to 100 ppb, should one expect that in the tropics, where emissions from the wetlands range from 40-240 mg m^2 d^{-1}, an occurrence of downwind peaks in atmospheric CH_4 by up to 1000-6000 ppb will be observed? Am I correct in understanding that we should expect to measure atmospheric CH_4concentrations of up 8 ppm in the tropics? Why, then, are observed atmospheric concentrations of CH_4 in the tropics lower than in the mid latitudes, not to mention in the Arctic? As we are currently establishing an observatory site in the tropical wetlands, it would be very interesting for me to know your view on that.

Third, I have been making an effort to educate myself in your area, in which I obviously lacked knowledge. I discussed this question regarding upwind/downwind concentrations of trace gases with many atmospheric physicists in the A. Obukhov Institute of Atmospheric Physics, where A. Monin and A. Obukhov worked on the founding basics of atmospheric physics that are still used in atmospheric models today. None of the scientists working there could confirm that there is any physics that allows downwind concentrations to be greater than upwind ones. In your model (background scenario), it is assumed that the air masses are entering the modeled domain from mid-latitudes (you suggested China in your response) to create background or even peaks of increased concentrations to be observed in the Arctic. Could you, please, explain what physics was used in your model that allowed downwind atmospheric concentrations (measured in the ESAS) to be greater than those measured in mid-latitudes, as it is known that concentrations of atmospheric methane in miod-latitudes are lower than those in the Arctic. I expect a very clear and detailed response to this question.

B) The ability of the chemistry-transport model to simulate atmospheric processes,
CHIMERE is a model widely used to represent atmospheric trace gases in the lower atmosphere (Menut et al., 2013 as a starting point). We think that the high resolution reached by the model CHIMERE, specially developed for this paper, has the ability to model Arctic methane concentrations. The use of initial and boundary conditions is classical in atmospheric sciences and, although there are hypotheses as in most scientific work, it leads to satisfying results. If the regional domain is large enough, when mapping the synoptic variations (dayly to weeklys changes in the atmosphere), boundary and initial conditions are of less importance as compared to emissions and transport within the domain. This is why we mostly insist on emissions within the domain, although we account for initial and boundary conditions and we inquire into the uncertainties in the boundary/initial conditions in our sensitivity test.

NS: The fact that the model is widely used does not address any questions regarding the basic assumptions etc. used in that model. The term “high resolution” is not used properly; it is simply substituted by smaller grid cells used in the model. Because the major goal of the paper was to reproduce atmospheric emissions of CH_4 in the modeling domain, the correct use of the term “higher resolution” should only be an application to the atmospheric CH_4 data used for analysis. The way it is presented in the paper, I see absolutely no point in claiming higher resolution if the number of atmospheric CH_4 data remains the same (NO data exist within the modeling domain

C) The quality of prior estimates.
Methane budget is uncertain at the regional scale. The uncertainties in methane sources other than ESAS do not change the fact that ESAS emissions, with the magnitude extracted from Dr Shakhova's papers, produce too large synoptic signals at atmospheric stations. You can turn it in any direction, but at the end atmospheric observations cannot hold such large emissions from ESAS on a yearly basis. In addition, our estimate of ESAS emissions is particularly robust to changes in other emission inventories in our sensitivity test, confirming the incompatibility of huge ESAS emissions in the Arctic.

NS: This response simply contradicts common sense. You claim that the background scenario used in the global model (as well as in your model) incorporates all the uncertainties of the methane budget at the regional scale. How is it possible that it does not affect your modeling results? What changes does your model robust to if the global model cannot even reproduce the pole-to-pole gradient actually observed in the atmosphere?

The comments about the section of the paper presenting the isotopic analysis are very interesting and we will update our manuscript accounting for them. We will also try to clarify the main text for non-specialists of atmospheric processes in the revised version.
Overall, there is (at least) one fundamental difference between flux-oriented studies and atmosphere-oriented studies: it is "easier" for a field-oceanographer to get individual flux observations than to properly aggregate them to produce an integrated ESAS flux. It is "easier" for an atmospheric scientist to extract an integrated flux from a given region than to retrieve the spatio-temporal heterogeneity of emissions. This is inherent to the different nature and properties of air-sea fluxes versus atmospheric concentrations and transport. We think that most of the following comments and questions come from a fundamental (apparent) misunderstanding of this physical difference, which is understandable considering the scientific main topic of Dr Shakhova being air-sea methane measurements.

NS: You suggest that my understanding of differences between flux-oriented studies and atmosphere-oriented studies is fundamentally flawed. It is absolutely true that I do not understand why you are unable to properly retrieve the spatial-temporal heterogeneity of emissions by dealing exclusively with what atmosphere and atmospheric physics have to offer you, that being atmospheric concentrations of CH_4 measured at your stations and basic atmospheric physics. It appears that you are trying to evaluate our estimates by performing your modeling runs, while your models would not work without our estimates.

2/ Point-by-point detailed answers

I believe that modeling can serve science as a valuable tool, especially when this tool is used by a team of skilled modelers such as those listed among the co-authors of this manuscript (ms). When I found this ms posted, I anticipated that the value of this tool would be clearly demonstrated to the advantage of the entire community. While I have never seen attention paid to a particular regional source of atmospheric methane such as has lately been paid to the East Siberian Arctic Shelf (ESAS), I agree that decreasing uncertainties is a goal we all strive for while doing science. Thus, my expectations for this ms were very high. I can only guess at the original content of this ms, but the tone of the responses to Dr. O. Gustafsson's interactive comment suggested that significant improvements must have been made before the authors decided to re- submit a previously rejected ms and make it openly available to critics. While I found those responses largely irrelevant, I took time to closely examine the very essence of the points the authors tried to avoid answering. I now believe

that the results reported in the current version of the ms lack any scientific foundation and are not conclusive. Below I clarify my point of view and raise a few questions for the authors.
As explained in our answer to Dr. Gustafsson's comment, we took care to answer to all his comments, not avoiding them as written here, although most were adapted to the previous version of the manuscript (submitted to GRL) and not to this version (submitted to ACP). Important modifications were introduced in the paper, such as a more complete treatment of uncertainties, as requested by Dr. Gustafsson. We do not agree that no other region received a close attention in the recent years. For instance, US emissions linked to shale gas have been studied a lot in the past years (e.g. Howarth papers), and so are Chinese emissions (Liu, Zhang papers), and natural emissions, from South America (e.g. Basso; Gatti et al., upcoming in JGR). ESAS is not the only focus point of the methane cycle.

NS: Could you be more particular about what treatments you brought to bear on which uncertainties in the data you used to achieve major conclusions? That is, isotope data and the physics used to explain higher concentrations downwind relative to upwind concentrations.

The major goal of this ms was to evaluate fluxes previously reported by Shakhova et al., 2010, 2014; the authors questioned the method we used to interpolate methane fluxes over the ESAS. We used kriging; this is an exact interpolation procedure, which is based on interpolation between the actual data points distributed over the studied area (Surfer-8). To be able to achieve methodological refinement of the interpolation, for >10 years we have been collecting actual measurements during all-season expeditions. That allowed us to achieve representative coverage of the investigated area; this is an essential requirement of any interpolation. The data set we used to estimate the fluxes reported in 2010 included data from >1000 oceanographic stations or >5000 samples analyzed; thus we were able to achieve fairly extensive coverage of the studied area, with one station covering about 2x103 km2 of the studied area (about 45×45 km2). For our improved estimation of fluxes reported in 2014, in 2009 we performed a continuous survey along ~2000 km in the near-shore area of the ESAS to document 2.7×104 bubble plumes. We manually inspected 8203 sub-blocks of hydro-acoustical data and sorted data into classes by intensity and density, then we reported a conservative best estimate by combining estimated seep intensity and density class (for details, see Shakhova et al., 2014). I believe that we reported much better coverage of the investigated area than has been achieved for other regional sources. For example, the Arctic terrestrial community currently reports that they achieved coverage of >20 million m2 by investigating 300 sites (one site for >65x103 km2). The northerneastern Siberian wetlands were reported to be covered by data from only 4 sites (Olefdeldt et al., 2012). I have never heard that anyone has questioned the estimates performed based on these sparse data. On the contrary, the authors of the current ms vigorously cite and take for granted estimates of high-latitude terrestrial and Arctic Ocean methane emissions performed by McGuire et al. (2009).

We are aware of the large amount of data collected by your team all these years and of the large effort that it represents to maintain such experiments over such a long time. The spatial and temporal density of observations that are necessary to constrain surface emissions largely depend on the spatial and temporal heterogeneity of the flux to estimate. As nicely shown in your different papers, ESAS methane emissions are very heterogeneous both in space (hot-spots versus more diffuse zones versus low emitting zones) and in time (sporadic emissions depending on ice presence and continuity versus more permanent diffuse emissions). Such heterogeneity unfortunately requires a lot of observations to constrain a statistical approach such as kriging. Our results show that the regional emissions you obtain after kriging for ESAS are too large on a yearly basis to be compatible with atmospheric observations of

methane. Indeed your group gathered a lot of observations but, as seen from the atmosphere, it may not be enough to properly constrain the large heterogeneity of ESAS emissions.
We do not agree that continental scientists are not questioned on their ability to represent regional fluxes with too few constraints (we agree that they have even less observations than you have gathered). Indeed, wetland emissions are the most uncertain ones of the methane budget and a lot of work remains to be done to gather more observations (e.g. eddy covariance flux measurements) and to improve the models (Melton et al., 2013; Kirschke et al., 2013). Just one example at the regional scale: figure 4 of the Bohn et al. paper (2015) discussing possible reasons for the inconsistent modeled emission maps for Siberian low lands for 6 different models.
Finally, we do not "take for granted" the work from Mc Guire but just report syntheses of the literature about the Arctic methane emissions. We would be pleased to quote other syntheses (we may have missed some) but they are unfortunately not so numerous at the regional scale for the Arctic. Moreover, we do take into account the inherent uncertainties of emission estimates by carrying out an extensive sensitivity test based on 10000 different simulations with different emission scenarios.

NS: I gather from your response that you believe we did everything incorrectly while you everything correctly. Why do you believe this? Is no longer true that modeling is dependent upon observation? I acknowledge that you are well aware of data coverage achieved in our investigations, but the problem is that you have never expressed any interest in our data or any desire to learn from our study. If your modeling effort can go forward without any knowledge about the area of investigation, then please explain why we should consider your results as valuable findings to consider. If not to us, to whom is your effort addressed?

In their ms, the authors try to convince us that inverse atmospheric modeling (top- down approach) can do better than bottom-up estimates. They claim that using data from only 4 stations, NONE of which is within the modeled domain of 2×106 km2, could decrease uncertainties. I think that if using data from outside the modeling domains were possible to decrease uncertainties about other regional sources, why would emission estimates still vary by factor of 2 or more by source sector showing no progress during the last 20 years (Nisbet and Weiss 2010; Dlugokencky et al., 2013; Chappellaz at al., 1993)? As the authors suggested, I turned to Locatelli et al., 2014, where I learned that inverse atmospheric modeling is an interpolation-based technique, which requires three crucial components to be conclusive: observational data (in-situ measurements, satellite retrievals, etc.), prior knowledge of emissions, and appropriate modeling methods (which are atmospheric chemistry-transport and global climate models). Locatelli et al. (2015) explained that achieving consistency of regional emissions estimates by inverse modeling is mostly dependent on: A) the number, accuracy, and spatial-temporal coverage of observations constraining the inversion; B) the ability of the chemistry-transport model to simulate atmospheric processes, and C) the quality of prior estimates.
YES, due to the integration property of the atmosphere, it is possible to estimate a regional source with few continuous stations (please see below for details).
As explained in the paper, if our approach can be considered as a top-down method (we use atmospheric observations to constrain surface emissions), it is not a full formal Bayesian inversion as in the papers by Locatelli et al. (2014) mentioned in your comment. So it is difficult to directly compare the two papers. By the way, atmospheric inversions are not interpolation technics but optimization technics with the minimization of a cost function.

NS: I referred to Locatelli et al., and extracted citations from the text regarding suggested requirements simply because you referred to Locatelli et al. as a main source in which your model is described in all its

details. So, I do not understand where I can find all the details describing your model, starting from the very basic assumptions used in the background scenario etc. If you are building a case study, please, give a very clear description of every assumption you use, especially regarding the background scenario.

Regarding the top-down approach you claim to use in your paper, I see a crippling problem here. Your approach could not be considered to be a top-down approach because, as I point out above, to perform top-down evaluations you must deal exclusively with atmospheric measurements, trying to reproduce existing atmospheric concentrations as they are observed on the planet. Instead, you are trying to reproduce fluxes calculated by observers, fluxes that are full of uncertainties and lacking understanding of very fundamental things, as follows from your response. Why do you choose to depend on estimates given to you by observers? What if all observations are wrong and all regional sources are estimated as badly as in the ESAS? Why not take a closer look at your models? Don't you assume that your models lack very important abilities and you, yourself, lack understanding of atmospheric processes as well as of the nature of CH_4 emissions? It seems to me that we could better improve our understanding by working together.

Regarding the integration property of the atmosphere, I point out that your approach runs contrary to an old and classic macro-synoptical approach by Girs-Vangengeim, which has been used by Russian atmospheric scientists for decades. According to this approach, during the last 17 years the anti-cyclonic (Az) atmospheric mode has dominated in the Arctic. This means that air masses that originated in the "Pacific atmospheric circulation pattern" (where the Barrow observatory is located) never reach the area west of ~160°E, which is identified as the western boundary of the Pacific – derived waters in the East Siberian Arctic Shelf (Semiletov et al., 2005). The same pertains to Barrow; the air masses that originated in the ESAS do not reach the Barrow observatory in the macro-synoptical time scale. Regarding other stations, see my comments above.

My major concern goes to A) the number, accuracy, and spatial-temporal coverage of observations constraining the inversion. The authors claim that they achieved a resolution of 35 ×35 km2, which means that to cover the area of the ESAS they needed to fill in a grid network consisting of >1700 grids with data. As follows from sec 2.1 of their ms, the authors used data from only four (!) atmospheric observational sites that are from 2500 to 4000 km away from the ESAS; moreover, only two of their four sites are considered to be somehow affected by air masses moving downwind from the ESAS. How, then, can it be possible to interpolate data between >1700 grids without a single measurement within any of these grids? How could the high variability and heterogeneity of methane plumes be reproduced with accuracy that is on the order of 1 ppb? How do the authors explain the modeled heterogeneity? They stated that there were no sources along the paths of the air masses, so the air masses must have been very well mixed before entering the modeled domain. How could they claim agreement or disagreement with observations if their results are not testable, because no measured data point existed within the modeling domain?

This comment reveals a mis-understanding of the nature and characteristics of the atmosphere. It can be partly due to clarifications needed in our text and we will try to bring them in the revised version of the manuscript. The atmosphere efficiently transports and mixes air masses (especially in the Arctic with fast horizontal advection). *Downwind a given region, it is possible to get integrated information about the past emissions of the region crossed by the sampled air mass* ***if travel time is shorter than diffusion time****.* As shown and discussed in the paper (and the supplementary material), the ESAS region can be

very efficiently connected by atmospheric transport to nearby (TIKSI, within hours) and remote (ALT, BRW, ZEP, within days) atmospheric stations continuously measuring methane in the air.

NS: It is true that "Downwind a given region, it is possible to get integrated information about the past emissions of the region crossed by the sampled air mass", but the problem is that atmospheric concentrations entering the ESAS have never been reported to be higher than those we measured in the ESAS (see NOAA data). Besides, as I pointed out in my comments above, your statement runs contrary to what we observed in 2005 while surveying west-to-east along >4000 km of the coastal area in the Siberian shelf. We did not record any increases in the atmospheric levels of CH_4 anywhere further than the Laptev Sea, nowhere approaching the 100 ppb that you suggest. Besides, when back trajectories are calculated, they can only be applicable to air mass movements; they cannot be used as evidence of incoming or outgoing atmospheric CH_4 if neither upwind nor downwind concentrations are measured. Moreover, I do not know of any evidence that atmospheric CH_4 levels entering the Arctic were ever higher than the levels that we measured in the ESAS. Please, give clear explanations and examples relevant to this paper.

Atmospheric observations are very precise and accurate (1-3 ppb, see Dlugokencky et al. papers for instance). For these synoptic time scales, atmospheric signals generated by your emissions from ESAS are strong (up to several tens of ppb as shown on figure 2 and 4), even thousands of kilometers away (BRW, ALT). There are transport errors (addressed in our uncertainty analysis) but they cannot mask these large signals.

NS: Do I correctly understand that atmospheric concentrations of up to 8 ppm CH_4 should have been observed in the tropics, where fluxes are 10-100 times greater than in the ESAS? Please provide me with documentation.

Therefore, we do not need a lot of stations to claim that your integrated ESAS methane emissions are too large to be compatible with atmospheric observations on a yearly basis. No need of interpolation between model grid points to find this result. Of course, if one wanted to resolve the fine structure of ESAS emissions, I agree that we would need a lot of atmospheric stations within and around the emitting ESAS domain. This is not what we want to do in this paper and a few stations downwind the emission zone is enough to test the regional total of your emission scenario.
Indeed, the issues of our two scientific works are kind of opposed: it is "easier" for an oceanographer to get individual observations than to properly aggregate them to produce an integrated ESAS flux. It is "easier" for an atmospheric scientist to extract an integrated flux from a given region than to retrieve the spatio-temporal heterogeneity of emissions. We cannot do anything about that, this is inherent to the different nature and properties of air-sea fluxes versus atmospheric concentrations and transport.
For the last part of the comment (which we are not sure to fully understand), emission types are run separately in the CHIMERE model, which allows us to separately study their impact at atmospheric stations. Also, the atmospheric stations are located in the model domain.

NS: See my comments above.

Another big concern is boundary conditions and the reference emission scenario. As stated in sec. 2.3, CHIMERE requires boundary concentrations and surface emissions to simulate methane mole fractions within its limited domain. These boundary conditions were interpolated from global

analyses obtained by assimilating surface mole fraction measurements. From Locatelli et al. (2015) it follows that boundary concentrations were derived from 39 years of daily methane mole fraction measurements averaged at global scale over the first layers of the atmosphere (9000 m) assuming climatological emissions of 500 Tg CH4/year, climatological oxidant fields, and an initial surface mean mole fraction of 1650 ppb. From this it follows that the authors assume emissions from the ESAS do not contribute to creating boundary conditions, and therefore emissions from the ESAS should only serve to increase atmospheric concentrations of methane above the background level (>1.850 ppm). That assumption is incorrect!

The statements made in this comment are mostly erroneous. By definition, boundary conditions are conditions coming from the outside of the modeled domain. ESAS emissions are part of the modeled domain and therefore cannot be considered as boundary conditions at first order. As we try to reproduce synoptic variations (within days) of methane, we neglect possible recirculation of the emissions of the modeled domain that could come back through the boundary conditions because of inconsistencies between the global model and the regional model. This is a classical assumption made for short-term studies with a sufficiently large domain such as the one we use here.

NS: I believe that you have not applied much effort to answering this question. It is a pity, because, in my opinion, this is a major misunderstanding that was incorporated into the modeling assumptions. Let me repeat my point. I believe that it is not correct that emissions from the ESAS cannot be considered as first order boundary conditions. I would agree if you started your modeling with the emission of the entire amount of 5.6 Gt of methane to reproduce the existing burden of CH_4 in the atmosphere, but you emit only 500 Tg, which is the annual emissions of CH_4 from all sources on the planet; the ESAS is one of these sources. Thus, why are emissions from the ESAS not represented in the background? Why do you consider emissions from the ESAS to be increasing atmospheric levels of CH_4 above the background level? This is wrong.

Boundary conditions used here are not climatological (they are monthly varying over the time of the simulation). Only prior emissions (before assimilation) of Locatelli paper are climatological. Atmospheric concentration at a given point is the sum of the boundary conditions and of the contribution of the different emissions occurring within the modeled domain, including ESAS area. Any emission contributes to increase the atmospheric concentrations of the emitted gas *on the top of the initial conditions (background), which are the result of the transport of all past emissions.* We think that it is clear from figure 4 (e.g. at ALT or TIK) that emissions from the modeled domain contribute to fill the gap between contributions of the boundary conditions (black line) and atmospheric observations (grey line). As ESAS contribution largely overcomes this gap on a yearly basis, again, ESAS emissions are not compatible with atmospheric observations on a yearly basis, although some consistency is found in July-August as written in the paper and shown by figure 5.

Finally, we agree that our boundary conditions do not include a contribution from the diluted past contributions of ESAS emissions as it is not accounted for in Locatelli et al.. Accounting for this would (slightly) increase the boundary concentrations in a constant way along the year (black line on figure 4) and consequently further (slightly) reduce compatible ESAS emissions of the current year.

NS: Exactly – you think that the ESAS does not contribute to the background concentrations. Which sources do contribute to the Arctic backgrounds? Emissions from the mid-latitudes, where atmospheric concentrations of CH_4 are lower than in the Arctic? Emissions from the low latitudes, where atmospheric concentrations of CH_4 are even lower than in the mid latitudes? Please, list these sources that build up

the background concentrations in the atmosphere and explain what physics allows downwind concentrations to be higher than upwind ones.

I believe that when your model is able to plausibly reconstruct the existing atmospheric maximum of CH_4 observed above the Arctic, you will gain a different understanding of the actual emissions from the Arctic sources, including the ESAS.

For unknown reasons, atmospheric modelers are still searching for a specific signal coming from the ESAS as if the ESAS did not exist until recently. The ESAS has al- ways contributed to atmospheric concentrations of methane observed in the Arctic. In cold epochs, the ESAS contributes to methane emissions as major (90%) fraction of the East Siberian wetlands; area of these wetlands decreased by a factor of 10 after the ESAS was submerged in the warm epoch. (This is regarding the suggested significance of emissions from the East Siberian wetlands suggested by authors, p. 25488.) In warm epochs, the ESAS has emitted more significant fluxes of methane than in cold epochs. This is because the East Siberian wetlands are emitting methane only 3 months a year while the ESAS – a year-round. Besides, the area of the ESAS that emits methane in summer is 10 times greater than the area of the East Siberian wet- lands, while in winter, the area of open water in the ESAS is comparable with the area of the East Siberian wetlands. Moreover, fluxes from the ESAS are associated with progressing thermokarst, destabilization of subsea permafrost and seabed deposits of methane; these fluxes are increasing due to drastic change in thermal conditions of the seabed. I believe that during the warm epochs, the ESAS might be a major contributor to the global methane budget while long-lasting destabilization of the ESAS shelf hydrates could be a plausible mechanism to support the clathrate-gun theory (Kennett et al., 2003). The increasing role of the ESAS as a methane emitter in warm epochs can also explain why the atmospheric methane maximum above high latitudes exists only in warm epochs (this was true long before any anthropogenic emissions!) and is ab- sent in cold epochs. The contribution of the ESAS has been increasing during the last centuries owing to continuing high sea level and the positive feedback of growing anthropogenic carbon dioxide emissions.
ESAS may have emitted a lot of methane in other epochs, but this is not the subject of our paper. We demonstrate that, at present, integrated ESAS emissions are smaller, although not negligible, than estimated in your different papers. Such a work is made possible by the existence of continuous atmospheric observations, which are recent (mid 2000s).

I have no doubt that emissions from the ESAS contribute to the background levels of atmospheric methane observed in high latitudes; the ESAS is not just an emerging source emitting excessive amounts of methane that increase atmospheric levels of methane above the background levels. The bottom line is that boundary conditions as well as the reference scenario, which assume that background levels of atmospheric methane in the Arctic reflect no contribution from the ESAS and that atmospheric stations in the Arctic are not affected by its signal, are wrong. I believe this is where the authors exhibit a major misunderstanding; this misunderstanding shaped the initial assumption used in the reference scenario, was incorporated into the regional model CHIMERE, and therefore affected the whole effort.
ESAS contributes to what you call the background level of atmospheric methane as any source diluted by the atmospheric mixing but, before that, with the amount emitted in your scenarios, it also produces large synoptic variations at Arctic atmospheric stations that are not observed (blue peaks on figure 4). Therefore, yes, some ESAS emissions are likely to occur and increase background methane on the long-term but they are smaller than you estimate.
It is true that our boundary conditions do not include a contribution from the diluted past contributions of ESAS emissions as it is not accounted for in Locatelli et al. Accounting for this would (slightly) increase

the boundary concentrations in a constant way along the year (black line on figure 4) and consequently further (slightly) reduce compatible ESAS emissions of the current year.

NS: Absolutely! The ESAS is contributing to the background levels of CH_4 emissions and this should be taken into consideration, as it will change your results drastically! You present no data in your paper addressing compatibility or non-compatibility of the ESAS emissions with any estimates, especially yours.

The authors of this ms claim major disagreement with our estimates made during winter months, so I was particularly interested in learning what data they based their dis- agreement upon. Sec.2.3 states that surface emissions for the CHIMERE domain were "deduced from state-of-the-art models and inventories". What might this state- of-the-art modeling be based upon? The CHIMERE domain incorporates the following inventories: 1) the EDGAR v.4.2 inventory for anthropogenic emissions in year 2010; 2) the LPJ model for global monthly climatology of wetland emissions; 3) the GFED v.3 model for daily fire emissions, and 4) emissions from the ESAS. It is clear that in winter, there are NO emissions from either wetlands (they are dormant) or from fires (fires occur in summer). Therefore, there is only one remaining contributor – anthropogenic emissions. Recall that we are talking about northeastern Siberia, that area of the Russian Federation where population density is only 2 people per km2 (for comparison, European population density is 176 people per km2), where no industry or agriculture exists, and where the nearest and the only big city in the region (Yakutsk, 316,000 people) is >1000 km away from the coast. There are NO data in any database (including EDGAR v.4.2 FT2010) on anthropogenic emissions in this region nor on their spatial distribution. How did the authors obtain contributions from sources other than the ESAS upon which to base their disagreement with our results?
As can be seen on figure 4, in winter, atmospheric concentrations at Arctic stations are made of contribution from boundary conditions (coming from lower latitude emissions) and fossil-fuel related emissions (red areas) coming from the whole model domain (and not only ESAS area or vicinity). Indeed, EDGAR inventory includes emissions from gas leaks in Russia, Northern Europe and North America, which contribute to atmospheric concentrations at Arctic stations as seen on figure 4.

NS: I did not ask how anthropogenic emissions were constructed in the whole domain (the globe), I asked about the particular domain (the ESAS) for which you claimed the improved resolution. Please address my question.

The authors explain (in an interactive comment) that emissions can "generate increased atmospheric mix- ing ratios downwind of emission zones". It is an accepted tenet of atmospheric physics that atmospheric mixing ratios of trace gases decrease downwind and that higher atmospheric mixing ratios downwind are only possible if there is a source of emissions located downwind. I ask the authors to please explain the physics that supports their statement.
Although not in the paper, we answer to this comment of the comment. What we mean here is that emissions can generate synoptic peaks of concentrations downwind of emissions zones. Of course, the magnitude of these peaks decrease with the distance/time to the original source. So, there is no violation of physical properties here, but just a wording not precise enough in our answer.

NS: Yes, it is probable that emissions can generate concentration peaks downwind of emission zones – if there is an additional source downwind and/or if concentrations of atmospheric CH_4 entering the domain were higher than background concentrations in the domain itself. Once it is clear to me that the

ESAS serves as such a downwind source, I will ask you to provide me with evidence that atmospheric concentrations of CH_4 entering the domain were higher than those observed in the ESAS.

Regarding B) the ability of the chemistry-transport model to simulate atmospheric pro- cesses, according to Locatelli et al. (2015), this ability was studied only i) for ice-free terrestrial surfaces and ii) for short-lived passive tracers. Since the modeling effort presented in the ms concerns mostly oceanic surface, and methane represents a long- lived non-passive tracer, I question the ability of CHIMERE to provide any advantages in simulating methane emissions on a regional scale.
Again it is difficult to directly compare the work by Locatelli et al. with this work as Locatelli uses the coarse global model LMDZ in an atmospheric inversion and we use here the high-resolution model CHIMERE in forward simulation over the Arctic. We refer to Locatelli et al. (2014) for the boundary conditions as explained in the paper. CHIMERE has been widely used for tracer transport (e.g. Menut et al., 2013 and associated references) and the high-resolution version developed specially for this paper is evaluated against PALLAS observations as detailed in the paper.

NS: I do not understand how CH_4 could be treated as a tracer, because a tracer can only be a particle that is passively transported by the air mass (like dust). Please explain how CHIMERE uses molecules of gas that are constantly diffusing in all four directions as a passive tracer.

In respect to C) the quality of prior estimates, Dlugokencky et al. (2013) confirm that large uncertainties (by a factor of >2) still exist in the estimates of major methane sources and sinks. This means that uncertainties of any modeling estimates could vary by an even greater factor, because, as is said in Locatelli et al. (2015), any
"changes in large-scale transport have potentially large impacts on the derived fluxes". My impression is that the modeling results presented in this ms confirm this statement and provide no advantage – indeed, they are at a disadvantage - compared to estimates reported by our group, which were per- formed using interpolation between data points actually measured within the domain.
The citation extracted from Locatelli et al. refers to large-scale transport and not to regional transport as studied here. Also, as already explained above, we do not perform a Bayesian inversion here.
The uncertainties in other methane sources do not change the fact that ESAS emissions with the magnitude extracted from your papers produce too large signals at atmospheric stations. You can turn it in any direction, but at the end atmospheric observations cannot hold such large emissions from ESAS on a yearly basis.
We do not develop our scientific work with "impressions" but with emissions scenarios integrated into atmospheric models and compared to precise observations, with a thorough evaluation of all the uncertainty sources. We do not think that our work can be considered as a "disadvantage" for the scientific community and we think that our answers to your questions and comments demonstrate this.

NS: The citation from Locatelli is extracted because you refer to this source while explaining to your readers where they can obtain a detailed description of the model you are using. If you are establishing a case study, you should have taken time to describe your model in great detail in the body of your paper, including the reference scenario. For your information, we are not tuning CH_4 fluxes, we are improving our calculations based on improving knowledge we are accumulating year after year by working in the field, as we have done for the last 15 years. I wonder how much time you spent on your study of the ESAS and what particular advantage you think this study can provide to the scientific community?

I would also like to pay attention to another key argument the authors used in their paper – apportioning contributions of the Arctic sources by analyzing isotopic compositions of Arctic air (sec. 3.1). In this ms, they followed Fisher et al. (2011) who used a single-isotope signature (d13C) to apportion contributions of the Arctic sources. It is not clear why the authors chose a particular range for marine hydrates (-50 to -55‰ while it is very clear from Milkov et al. (2005) that the isotopic signature of marine hydrates is area-specific and varies from -39 to -74‰ Besides, why the authors chose isotopic signature of marine hydrates if their aim was to apportion the contribution of unique permafrost-related Arctic shelf hydrates, isotopic signature of which has not been reported yet? It is not clear why the isotopic signature of wetlands should vary within such a wide range of -60 to -75‰˙It is not clear how the authors managed to distinguish between the isotopic signature of hydrates (-50 to -55‰ and gas leaks (-40 to -55‰; they overlap. What is very clear to me is that there could be numerous possible mixtures of sources compatible with such wide and overlapping ranges. In addition, the authors do not seem to be aware of a major methodological limitation: applying a single-isotope analysis is inappropriate when methane from a few sources undergoes transformation and mixing in open systems (Galimov 2006). How could the authors claim that their modeling results are not compatible with the observations? We must ask: What observations? We have attempted to explain to the authors that their paper published in 2011 was misleading. Unfortunately, the same is true for their current interpretations, which have nothing to do with isotopic signatures of actual end members of Arctic sources; these are largely unknown. Regarding ESAS sources, our group is the only group in the world to possess data on the isotopic signature of methane source end-members. These data, consisting of hundreds of samples analyzed by triple-isotope analysis in the best isotopic laboratories in the world, await publication.

Thanks for this important and useful comment. We are aware that the original section 3.1 has to be improved as also noticed by reviewer#1. We proposed an updated version but we agree it still needs attention. Indeed you are right that the range of isotopic signature for hydrates is too narrow considering the literature and the facts that hydrates can be of thermogenic or biogenic origin, and that it is largely variable and unknown. We will change this in the updated version using the larger range reported in Milkov 2005. Concerning natural microbial sources, there is a variety of signature reported for high latitude ecosystems ranging from -70/-60‰ for wetlands, -75/-55‰ for tundras, -80/-60‰ for thermkarst lakes (e.g. table 2 of Fisher et al., 2011). This is why we used the quite large range of -75/-60‰ for microbial signature. We do not intend to solve a three-unknown problem (wetlands, gas, ESAS emissions) with only one equation as we agree that there is an infinite number of possibilities. We just want to raise the point that, considering the range of isotopic signature for wetlands and gas, whatever methane comes out of ESAS region, it is of biogenic origin (at the moment 13C observations were made), which is consistent with what you suggest in your previous publications about ESAS. One cannot explain a -68/-65‰ observed source signature in another way with the mixture of air sampled when the observations were made at ZEP. We cannot say more than that about whether this is hydrates or not and we will rephrase the text to be clearer on this point.

NS: Am I correct in understanding that you intend to eliminate this entire section from your paper, because it is based on invalid initial assumption (that a single-isotope analysis can identify a sources of methane), because widening the range of possible end-members would have increased the number of possibilities for mixing, making the results even more uncertain than before and because actual isotope signatures of the ESAS sources of methane are unknown to you?

Finally, the authors claim to have investigated the synoptic signal. I am afraid I do not understand how such an approach advances knowledge. How could fluxes that propagate over an area of 2×106 km2; that vary spatially

and temporarily by orders of magnitude, reflecting very complicated processes ongoing in the ESAS; and that depend on winds and numerous other conditions that are constantly changing in time and space be investigated and confirmed (or not confirmed) by a synoptic-scale systems background that represents anything but dynamics? This is contrary to the very essence of flux.
This is the beauty of atmospheric transport! It integrates the heterogeneity of emissions and before suppressing it by mixing, it produces peaks of concentrations downwind the emissions zones. This is physics and it is used every day by all atmospheric scientists working on greenhouse gases, air pollution, aerosol pollution, and event radioelement pollution, at various spatial scales from local to global.

NS: Perfect! Then please use this "beauty of atmospheric transport" to develop **a real top-down approach**, not the approach that you are using today.

I agree that it is better to light a candle than to curse the darkness, but the problem is that I see much more darkness than light from the authors' candle. Indeed, they did not demonstrate any interest in learning from our long-term investigations; instead, they seem to be working to convince us that modeling can produce a miracle, that no observational work in near proximity to the sources is required to make progress on assessing regional components of the global atmospheric methane budget. They claim they can do much better with no ground-truthing.
We leave you the image of the candle.
To verify a result, one needs to have the result. We try to verify your results with an independent and robust atmospheric approach, and we do not confirm them. As explained before, the properties of the atmosphere make this possible. This paper does not mean that we do not need terrain observations or that atmospheric modeling can do everything. We have our own uncertainties (accounted for in this version of the manuscript) but there may be something erroneous in your work and we suggest that it could be how you go from local measurements to regional ESAS emissions and underestimating the uncertainties in the kriging procedure. But this is just a suggestion.

N: I will consider your suggestion, because learning is always good.

My final questions are the following. Should we observational scientists decide that it is pointless to go into the field, with all the unavoidable risks and difficulties of field work in such a challenging environment, because we would do better to just speculate in the comfort of our offices? Should we learn the lesson that there is no value in gaining actual data, studying real processes, accumulating knowledge, and possessing unique skills? What is the value of this ms to science?
Although the tone of this last comment is (again) at the limit of what is acceptable, we answer it.
We never questioned the fact that observations are critical to understand our environment and constrain the models (one would notice that more than half of the scientists involved in our work have a strong experience in atmospheric observations in remote and difficult locations). You seem to be personally attacked by our work, as reflected by these 10 pages of comments. We try to stay professional. Again, it is the normal scientific work to face scientific criticisms. We think that it is important to know that ESAS do not emit as much methane as previously reported, as this is a key region for climate change. Our results do not mean that ESAS has never or will never emit more methane. It is just not the case at present, which does not mean that you have to stop going there to study this important climate zone.

NS: Thank you for taking time to address my comments! I hope that you will spend some more time to make the clarifications I have asked for.

Reference list: Chappelaz, J. A., I. Y. Fung, and A. M. Thompson, The atmospheric CH4 increase since the last glacial maximum (1). Source estimates, Tellus, 45(B), 228–241, 1993. Dlugokencky, E.J., Nisbet, E.G., Fisher, R., and Lowry, D. Global at- mospheric methane: budget, changes and dangers. Phil. Trans. R. Soc. A 369, 2058-2072, 2013. Fisher, R. E.; Sriskantharajah, S.; Lowry, D.; LanoiselleìA¸, M.; Fowler, C. M. R.; James, R. H.; Hermansen, O.; Lund Myhre, C.; Stohl, A.; Greinert, J.; Nisbet-Jones, P. B. R.; Mienert, J.; Nisbet, E. G. Arctic methane sources: Isotopic evi- dence for atmospheric inputs. Geophys. Res. Lett., 38, L21803, 2011. Galimov, E.M. Isotope organic geochemistry. Organic Geochemistry, V.37 (10), 1200–1262, 2006. Kennett, J. P., Cannariato, K. G., Hendy, I. L. and Behl, R. J. Methane Hydrates in Quaternary Climate Change: The Clathrate Gun Hypothesis, in Methane Hydrates in Quaternary Climate Change: The Clathrate Gun Hypothesis, American Geophysical Union, Washington, D. C.. doi: 10.1002/9781118665138.ch0, 2003. Locatelli, R., et al. Atmospheric transport and chemistry of trace gases in LMDz5B: evaluation and implications for inverse modelling, Geosci. Model Dev., 8, 129–150, doi:10.5194/gmd- 8-129-2015, 2015. McGuire, D., Leif G. Anderson, Torben R. Christensen, Scott Dal- limore, Laodong Guo, Daniel J. Hayes, Martin Heimann, Thomas D. Lorenson, Robie

W. Macdonald, and Nigel Roulet 2009. Sensitivity of the carbon cycle in the Arctic to climate change. Ecological Monographs 79:523–555. http://dx.doi.org/10.1890/08-
2025.1 Milkov, A.V. Molecular and stable isotope compositions of natural gas hydrates: A revised global dataset and basic interpretations in the context of geological settings. Organic Geochemistry. V.36 (5), 681–702, 2005. Nisbet, E. and Weiss, R. Top-Down Versus Bottom-Up. Science 328, 1241-1243 (2010) Olefeldt, D., Turetsky, M. R., Crill,
P. M., and McGuire, A. D.: Environmental and physical controls on northern terres- trial methane emissions across permafrost zones, Glob. Change Biol., 19, 589–603, 2013. Shakhova, N., Semiletov, I., Leifer, I., , Sergienko, V., Salyuk, A., Kosmach, D., Chernikh D., Stubbs Ch., Nicolsky D., Tumskoy V., and O. Gustafsson, 2014. Ebulli- tion and storm-induced methane release from the East Siberian Arctic Shelf, Nature Geosciences, doi: 10.1038/NGEO2007 Shakhova, N. et al. Extensive Methane vent- ing to the Atmosphere from Sediments of the East Siberian Arctic Shelf. Science 327, 1246-1250, 2010.

Reference list:

Locatelli, R., et al. (2014), Atmospheric transport and chemistry of trace gases in LMDz5B: evaluation and implications for inverse modelling, *Geosci. Model Dev. Discuss.*, *7*(4), 4993-5048, doi:10.5194/gmdd-7-4993-2014.

Fisher, R. E., et al. (2011), Arctic methane sources: Isotopic evidence for atmospheric inputs, *Geophys. Res. Lett.*, *38*(21), L21803, doi:10.1029/2011gl049319.

Menut, L., et al. (2013), CHIMERE 2013: a model for regional atmospheric composition modelling, *Geosci. Model Dev.*, *6*(4), 981-1028, doi:10.5194/gmd-6-981-2013.

Kirschke, S., et al. (2013), Three decades of global methane sources and sinks, *Nat Geosci*, *6*(10), 813-823.

Melton, J. R., et al. (2013), Present state of global wetland extent and wetland methane modelling: conclusions from a model intercomparison project (WETCHIMP), *Biogeosciences*, *10*(2), 753-788, doi:10.5194/bg-10-753-2013.

Bohn, T. J., et al. (2015), WETCHIMP-WSL: intercomparison of wetland methane emissions models over West Siberia, *Biogeosciences*, *12*(11), 3321-3349, doi:10.5194/bg-12-3321-2015.