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

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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 to me that significant improvements must have been made before the authors decided to resubmit 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.

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$\times$45 km2). For our improved estimation of fluxes reported in 2014, in 2009 we performed a continuous survey along $\sim$2000 km in the near-shore area of the ESAS to document 2.7$\times$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).

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.

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

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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?

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!

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 always 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 wetlands, 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 absent 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. 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.

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 disagreement 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? The authors explain (in an interactive comment) that emissions can "generate increased atmospheric mixing 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.

Regarding B) the ability of the chemistry-transport model to simulate atmospheric processes, 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. 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

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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 performed using interpolation between data points actually measured within the domain.

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‰Ḃesides, 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‰İt 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.

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! 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.

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?

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 atmospheric methane: budget, changes and dangers. Phil. Trans. R. Soc. A 369, 2058-2072, 2013. Fisher, R. E.; Sriskantharajah, S.; Lowry, D.; LanoiselleÌĄ, 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 evidence 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 Dallimore, 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 terrestrial 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. Ebullition and storm-induced methane release from the East Siberian Arctic Shelf, Nature Geosciences, doi: 10.1038/NGEO2007 Shakhova, N. et al. Extensive Methane venting to the Atmosphere from Sediments of the East Siberian Arctic Shelf. Science 327, 1246-1250, 2010.

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