Comments on Berchet et al. for ACPD (ms # doi:10.5194/acpd-15-25477-2015)

We thank Örjan Gustafsson for his comment on our manuscript. We note that his comments were already used (with the very same typos) as reviews in an AGU journal (GRL) our manuscript was submitted to earlier this year. The comments have only been superficially edited since then and do not account for our efforts to improve our manuscript (especially the uncertainty quantification) for the new submission to ACP. Örjan Gustafsson's comments are reported in regular font with our replies in bold embedded in the text.

This manuscript has earlier been rejected from an AGU journal and the current version here submitted to ACPD is only superficially edited relative to the earlier rejected version.
We do not agree. Örjan Gustafsson may not have thoroughly read through our manuscript. The differential with the original text is important, we brought the required precision on the model description, we proposed new figures (figures 3 & 5, and supplementary material) and refined ESAS emission estimates by moving from a yearly analysis to a monthly analysis with a comprehensive quantification of uncertainties, thanks to reviewer's comments.

There is a lack of recognition concerning the limitations of their own approach and the huge uncertainty overall of these systems.
We estimate a range for ESAS emissions accounting for uncertainties. We performed systematic sensitivity tests on our setup, providing a range for the ESAS emissions from 0.5 to 4.3 Tg/yr. These tests do not change the main message of the paper but allow addressing the question of uncertainties in a complete and rigorous manner. The Taylor plot of figure 2 is now shown as an illustration of the more complete statistical computation.

I remain convinced about the value of atmospheric inversions, not the least as one approach to estimate current methane emission from the East Siberian Arctic Ocean, yet this study fails to provide the type of firmly supported and important insights that would be required for a journal like ACP.
This sentence is general, not adapted (we do not claim to perform a formal inversion here, but we test the compatibility of a forward scenario of ESAS emissions with continuous atmospheric methane observations) and relies on no scientific evidence.

The "new" submission to ACPD has now been studied. Below, a number of key considerations are detailed that at present hinders this study from constituting a valuable contribution, yet are meant to support the authors in their coming efforts.
As already noticed, these "new" considerations are actually mostly the same as those on the former and different manuscript submitted to GRL a few months ago. However, we answer them point by point.

1. The database for the top-down evaluation

The authors keep insisting that there are >20 pan-arctic observatories able to address the current issue of atmospheric methane fluxes from the subsea system on the East Siberian Arctic Shelf (ESAS). The study then limits itself to the big five – Zeppelin, Pallas, Alert, Barrow and Tiksi, yet a closer scrutiny shows that the actual data for the claimed year-round 2012 atmospheric observations is even smaller.
We only keep continuous sites as explained in the text in order to capture synoptic variability, not present in the bi-weekly flask sampling of other sites.

They acknowledge that ZEP and PAL are minimally influenced by CH4 emissions from the ESAS due to their distant locations; and there is apparently no methane data for ZEP-2012 (see fig. 2 and 4). While far from sufficiently acknowledged at key locations (e.g., Fig. 1 and 4 captions), the BRW 2012 record is completely absent for the key summer-fall period, thus severely limiting its value (see e.g., fig. 4).
The interior ms text further acknowledge that "*ESAS methane releases cannot be obtained from TIK alone…*". It must also be noted that the TIK methane program is not yet published anywhere, preventing thorough scrutiny, incl. suitability of local site/meteorology.
We have to do with the available observations and we are dependent of observations issues such as technical problems or maintenance of instruments. We did not use ZEP $^{12}CH_4$ observations because the instrument was changed in the middle of the year; BRW measurements failed after the middle of 2012 but were ok for the first semester. If one has to wait that no failure occurs in experimental work, one would never publish anything. We chose PAL because it was little influenced by ESAS to show that modelled boundary conditions were good and that the model was able to reproduce synoptic variation at Arctic sites. We think we provide in the text the necessary elements on TIK observations, which are closely linked to the well-reported and calibrated NOAA observations.

Taken together, this leaves us with Alert as the one key site to "nail down" the ESAS methane emissions, located some 2500 km away with complicated and non-ideally constrained high-Arctic meteorology. This by itself severely undermines the reliability of the definite conclusions articulated in this submission.
This is not true. For the analysis we use ALT, TIK and BRW (when available). The fact that TIK can hardly be used <u>alone</u> does not mean we do not use this site. We show in the paper with a detailed footprint analysis and previous literature that the fast horizontal transport in the Arctic links within days ESAS to remote locations such as ALT or BRW or even ZEP for some periods (^{13}C data). Again the comment of the reviewer "complicated and non-ideally constrained high-Arctic meteorology" **relies on no scientific evidence or peer-reviewed reference.**

2. On skills of a model

The authors claim to achieve good agreement between their top-down estimates and summer fluxes reported by Shakhova et al., while winter fluxes did not show agreement as they seem over estimated by Shakhova et al. Let's give it a closer look. It is stated that CHIMERE demonstrates a very good skill in winter in representing the atmospheric methane mole fraction variability at high latitude sites (they report correlation r=0.89). However, in winter, contributions of 2 of the 4 simulated sources (terrestrial emissions and fire-inducted emissions) are equal to zero while anthropogenic emissions are still very poorly constrained.
The reviewer mixes different things here. The r=0.89 at PAL in winter relies on emissions from regions others than ESAS (also mid latitudes through the boundary conditions of the model) as PAL is not influenced by ESAS emissions. It is used to show that our basic scenario reproduces in a good way the observed synoptic concentrations in the Arctic. Basically, it gives confidence that our high-resolution model is able to reproduce the synoptic variability at high northern latitudes.

The anthropogenic emission inventory presents in a single number the annual emissions from the whole country. How did they distributed these emissions over the Russian north, if there is a single numbers that goes to the entire territory without apportioning either particular territories or attributed to particular seasons? Thus, their model works best (with r=0.89), when two sources do not contribute at all and the most uncertain one contributes "mysterically"? Is this situation called good model skill?
We use state-of-the-art annual gridded anthropogenic emissions from EDGAR at 0.1°. This is the work of the EDGAR database to spread national totals into a 0.1° grid. We provide the reference of the EDAR database and this is beyond the scope of the paper to enter in the detailed statistical approach of EDGAR. As already explained, in any case the PAL agreement is due to the 2 Arctic sources. We find the two last sentences of this comment provocative and not very scientific.

3. On the implications of "higher resolution"

Insisting on "higher resolution" modelling, they actually replace high resolution, in the true meaning of higher accuracy, with high resolution, in this work only meaning a smaller model grid. There cannot be any high accuracy achieved by using the same data sets as global models use by only reducing the size of the grid – the amount of data on methane remains the same any way! No improved meteorology can help to make up for lack of data on the modelled tracer itself (methane).
We do not agree that a refined resolution and improved meteorology cannot bring more accuracy (again an affirmation with no scientific justification). Using a more refined grid than current global models we take full benefit of the resolution of the emission scenarios (0.1 or 0.5 °), of meteorological data (0.15°), and of the topography over the continents. Doing so, we have the objective to provide the most suited model as possible to reproduce atmospheric methane observations. Of course as we only use one model, it is not possible to demonstrate that we do better than with a global model. However, we know from Geels et al., 2007 (JGR) that global models

tends to smooth synoptic variations compared to regional models (with higher resolution) because of their crude resolution. Therefore, the use of a high resolution model limits the risk of underestimating synoptic variations at atmospheric sites which is important for our study.

4. **The "all seats taken" argument:** Why is the disagreement btw modelled and observed CH4 mixing ratios purely "blamed" to ESAS bottom-up estimate as opposed to bottom-up estimates of other sources and boundary conditions implicit in the model?

This is a key aspect of the whole study approach. The ms only pay lip service to the huge uncertainties of both the global and the terrestrial+marine Arctic methane numbers (e.g., Ciais et al., 2013).
Given the massive field observational programs that apparently are underlying the Russian estimates of the bottom-up ESAS methane emissions, in comparison with the database for bottom-up terrestrial wetland emissions in northeast Siberia, may it not be possible that the terrestrial wetland component is overestimated? That component, and all other flux vectors but ESAS, are here taken as "given" and untouchable. This approach is flawed and biased.
This very comment explicitly proves that Örjan Gustafsson did not pay a close attention to the current version of the manuscript, as we largely clarified this point in the version submitted to ACP.
In this paper we test a scenario of ESAS emissions and show that the synoptic peaks produced by these emissions in winter at atmospheric stations are not compatible with the continuous methane observations. In summer, more compatibility is found in magnitude but there is the possibility that wetland emissions are not well represented in our scenario. We test this hypothesis in extensive sensitivity tests perturbing our model set-up and the representation of transport. Thus, we provide a robustly estimated range of emissions from ESAS according to this uncertainty in summer. In any case, the sensitivity of wetland emissions at atmospheric sites was found to be less than ESAS emissions because wetland are a more diffuse source all around the Arctic.

The same is likely true not only for the Arctic scale but also for the global scale, where limited number of actual observations today serve as the base for the strongly held paradigm that tropical wetland CH4 emissions (where much lower conc is found than over the Eastern Arctic Ocean) is the globally-dominant natural source.
Authors are asked to consider why global atmosphere methane models fail to reproduce observed pole-to-pole gradients in mixing ratios (also relative to that of other gases). Decreasing tropical emissions and increasing Arctic emissions, in the model description of emissions would make this fit better. All of this affects the baseline/boundary condition of the model run in the current submission.
We think that this comment is irrelevant to the work presented here as we focus on the Arctic and on synoptic variations of atmospheric concentrations. We do not address global scale, tropical emission, north-south gradient, which are important questions but not relevant here.

The highest atmospheric mixing ratios of methane are found over the eastern Arctic – how can methane from the tropics (of lower mixing ratios) build up this phenomenon? It goes against basic entropy. Arguments of different mixing heights do not work as then all other passive gases would also be "compressed"; the opposite is observed. Arguments of strong anthropogenic methane import to the eastern Arctic does not gibe with this feature existing also before the Anthropocene. It is time to reconsider basic paradigm/assumptions of methane emissions and their model construct. Until that is done, any "new" sources will just be ill-treated as "overfilling the cup".
What is the scientific reference showing that atmospheric mixing ratios of methane are the highest in Eastern Arctic? Compared to which regions? Again, this is one affirmation without any reference. The Arctic atmospheric concentrations rely on local emissions, transport of remote emissions, fast (low) horizontal (vertical) transport. This is not only a matter of local emissions as claimed by the reviewer. The "overfilling" argument is not correct. In winter, there is nothing such as large peaks in the continuous observations. No source producing such peaks can therefore be accommodated by observations. In summer at TIK, such peaks are visible so there is space for an additional source compared to our reference scenario, either ESAS or local/regional wetlands.

5. **The insistence on the notion of large emissions from terrestrial coastal wetlands in northeast Siberia**

The pre-conceived notion of sizeable emission from the very shallow and rather dormant surface soil of NE Siberia, competing with ESAS coastal emissions (also terr wetlands, buried beneath a much warmer ocean – a thermal regime more prone to thawing) seems to lack scientific basis. The authors respond that they like to "reinforce their assertions" but offer no observational basis for their bias.
As authors ought to be aware of, this Oyagosski Yar region is subjected to intense erosion of coastline and subsea permafrost (these are not what is meant by terrestrial wetlands) – for a start, look at Günther et al (2013 Biogeosciences),Vonk et al (2012 Nature) and Shakhova et al. 2010 (JGR).
Olefeldt et al. (2012) recently collated information to show that the pan-Arctic terrestrial methane emissions from an area of >20 million m2 is covered by 300 sites (i.e., >65 thousand km2 per site). In contrast, I gather from the work reported by Shakhova et al (2010) that their emission estimates from the 2 mill km2 ESAS area is based on at-sea measurements at about 1000 sites, giving 2 thousand km2 per station). Furthermore, the NE Siberian wetlands, particularly deprieved in N. Yakutia with continuous permafrost, covers only <20% of the Siberian wetlands and <10% of the ESAS area, and are covered only by very few actual flux observations.
We thank the reviewer for this precise and documented comment, which shows that he is a specialist of these questions, more than on atmospheric issues. By the way, it is to be noticed that Örjan Gustafsson is co-author of the Shakhova 2010 and 2013 papers. As mentioned in the comment, we can hardly provide observational constraints for terrestrial wetlands if observations do not exist or are not available. This is why we chose to perform a sensitivity test with the magnitude of wetland emissions in order to determine the sensitivity of our results to our assumptions. It does not change the main message of the paper about the magnitude and seasonality of ESAS emissions

that are compatible with atmospheric observations.
We do not question here the numerous observations performed by the Shakhova group but our new estimate (0.5-4.3 TG/yr) questions the extrapolation method they used to upscale their observations to all ESAS region. In the end, the continuous observations around the Arctic cannot accommodate the extrapolated value of this flux.

6. On the transport of methane from ESAS to Alert

Some of the key underpinning assumptions articulated in Locatelli et al (2013) – a co-author of the current ms – is not met and remain to be addressed.
Although it is not very precise, we try to answer to this comment. The atmosphere is a well-known integrator of information. Emissions are mixed into the atmosphere and generate increased atmospheric mixing ratios downwind emissions zones. In particular, in the Arctic, atmospheric transport is very fast and links emission zones to atmospheric stations very efficiently (within days) even for long distances (see supplementary material). For example, European, North American and Chinese emissions can be quickly imported to the Arctic and thus significantly influence its atmospheric composition, ten thousands kilometres away.
Therefore, insights on some emission areas can be deduced from long-distant observation sites such as BRW and ALT. In this sense, Figure 1 clearly illustrates that concentration enhancements from ESAS of more than 100 ppb are found all around Arctic continents, from hundreds to thousands of kilometres away from the emission area. The little number of stations used here is compensated by the integrative capacity of the atmosphere and by the hourly frequency of the measurements, which allows sampling air masses coming quickly from ESAS all year long with limited risks of miss-detection.

Furthermore, is the model accounting for uptake of methane from the atmospheric boundary layer to the ocean water/snow/ice during its 2000-3000 km long transport (much longer if not transported on a straight line between ESAS and Alert)?
Given the much lower methane concentrations in AO interior surface waters, snow/ice and the very long passage, there is ample time for this fugacity gradient to take its toll on the BL methane levels and deliver a lower mixing ratio once the air parcel finally arrives to Alert. How is this addressed in the employed model?
We would be interested to read a scientific reference showing the uptake of methane by the ocean on such a short time scale? Again no reference is provided by the reviewer. We do not consider this hypothetical process in the paper.

There are many more questions and aspects that the current state of this manuscript is raising than what is reasonable to put down in this review.
Again a general sentence with no precision, only to bring a negative impression on our work.

For instance, if existing atmospheric observatories are so well placed to provide the type of clear constraints and conclusions claimed in this submission, why is there such urgent and prioritized efforts going into establishing major new atmospheric observatories around the ESAS, such as at Ambarchik/Kolyma mouth (MPI) and Bolshevik Island of Severnaya Zemlya (AARI and FMI)? These ESAS bordering observatories will be really well placed to address methane emissions from the ocean in the near future.
We never claim that atmospheric sites are optimally located but we just use what we have to produce scientifically-based studies. Developing new sites and making them available for the community is still a key issue to refine this kind of work and further reduce the uncertainty on regional methane emissions.

Why is the western part of Laptev Sea not marked as part of the ESAS source area in Fig 1, and the source area called Laptev Sea instead of ESAS (incl. the vast East Siberian Sea) at many locations? What source area is really considered?
This is a mistake and we will correct the source name to ESAS in the revised version of the manuscript. The domain is the one of the Shakhova (2010) paper, which we consider as the best qualitative in situ documentation for this region.

No matter if today's methane emissions from the ESAS are 4, 8 16, or 32 Tg/yr, that is still a minor portion of global methane emissions into an uncertain global methane budget. This is very likely hard to rule out given the large uncertainties of almost all natural methane sources (Ciais et al., 2013), if not taking the rest of them for granted and instead subjecting all sources to equal scrutiny.
Any study of this sort ought to recognize that the bigger question is how this system, hosting many 100s Pg C-CH4 in potentially vulnerable reservoirs may develop in the coming decades-centuries. Top-down atmos observations of mixing ratios will be able to record such changing trajectories but deeper geophysical/geochemical investigations will be needed to provide the system understanding to make such predictions. This is a useful context and the current approach is one piece to our common puzzle. The present submission is not a suitable contribution but the general approach will be a useful window in the future.
We do not agree at all. The Arctic is a very sensitive region of the methane cycle and quantifying today the amount of methane coming out of ESAS is therefore very important to track a possible destabilization of the zone. We show in the paper that this is not yet the case but it does not mean that it cannot happen. We are aware of the long-term "big" questions but we feel that is it also important to document the present and demonstrate that atmospheric stations could detect any strong destabilization of the Arctic methane, even more if the atmospheric network is reinforced in the upcoming years.