Response to comments by Parmentier and Bruhwiler:

We appreciate the online discussion comments by Parmentier and Bruhwiler, which primarily focused on a minor aspect of the manuscript, one that we failed to mention in the study motivation section. This has been rectified to provide the proper context of the in situ data in our satellite study. Comments by Antonia Gambacorta, a well-recognized expert on satellite trace gas retrievals, are addressed in a separate response.

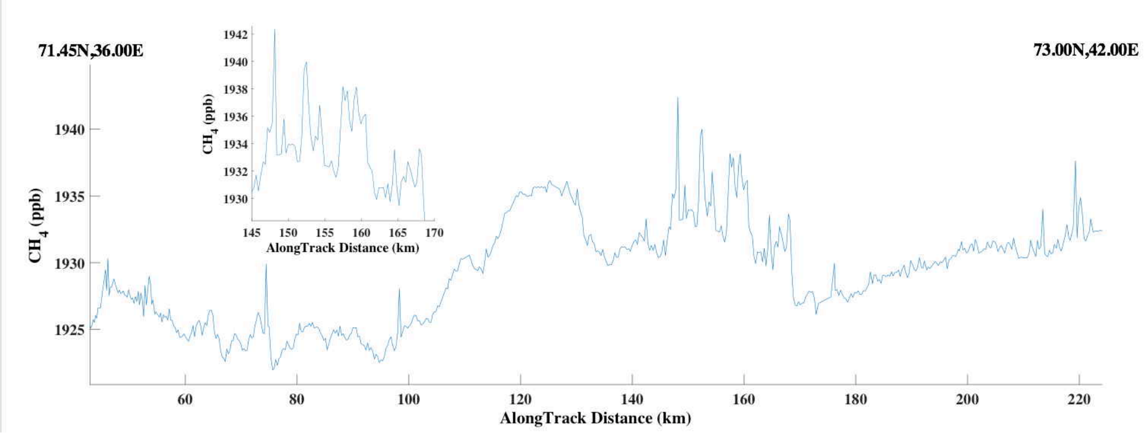
Our focus was an analysis of satellite data that show anomalies for the Barents Sea. The satellite data analysis shows statistically significant *trends* of increasing atmospheric methane over the Barents Sea, but we did not detect statistically significant trends for increasing methane off west Svalbard. In the Barents Sea there was evidence of ocean warming and reduction of sea ice during the period of increasing atmospheric methane anomalies. The documented offshore petroleum reservoirs are primarily located in Russian waters, and the satellite data provides an advantageous and first public view to examine anomalies in methane in the atmosphere over this region, which is geopolitically and logistically from highly challenging to inaccessible for in situ investigations over much of the year. This is a region for which there are few to no available weather stations, or weather data, over millions of square kilometers. Our satellite data are averaged monthly, for which prevailing wind patterns are adequate for our analysis. Detailed numerical models were not within our budget or scope. We fully agree with the comments that such investigations would be useful; however, we feel out work is a contribution to the community in *discovery* mode and to demonstrate that important hypothesis can be tested by average research groups with limited resources. Additionally, its unclear that meteorological atmosphere models would add much to our discoveries as they remain, at best, poorly validated over the region (given the absence of weather stations). To quote Berchet et al. (2015) "Any inversion has to be confronted to independent data in order to evaluate its results." Berchet et al. (2015) also discusses inversion limitations for Arctic methane sources due to uncertainty in atmospheric transport and other factors. In this regards, both our unique *in situ* data and our satellite data analysis provide important contributions.

We agree that it would have been good to have water column data from the NABOS cruise. Unfortunately, we were not afforded such an opportunity, as we were invited guests on the cruise, with no agreement on wire time. Our invitation was to collect atmospheric methane data. To our knowledge, our atmospheric methane data for these areas of the Barents Sea are the only publicly available data and we want to make these data public.

Our field data, like, any ocean cruise data, only provided a snapshot of atmospheric methane. As such, satellite data are a particularly important complement to existing field information, particularly for seldom visited, stormy, remote seas and oceans. These data help understand the fate of the Arctic seabed methane emissions. We know that some fraction of the seep methane reaches the atmosphere elsewhere in marine petroleum basins from depths of 500 to 1000 m deep, such as those mentioned by (Solomon et al., 2009) and (MacDonald et al., 2010), and its potential to enhance atmospheric methane is consistent with the extensive knowledge of geologic petroleum basins worldwide

Our field data suggest a kilometer-wide methane plume in the atmosphere. Our analysis and experience from other studies leads us to our conclusion. Specifically, an atmospheric plume's signature grows in size with distance from a source (Hanna et al., 1982), As such, small, strong anomalies typically don't materialize from distant sources. A rule of thumb for a plume is that a 1 km-wide plume likely arises by dispersion from a source within a distance of more or less 10 km (about a factor of 10 dispersion). The details depend on wind speed and atmospheric stability, faster dispersion for lighter winds and greater instability. In this case, the shallow Arctic planetary boundary layer supports our interpretation. This behavior is described by the Gaussian Plume model. See validation by our own work on plume dispersion in (Leifer et al., 2018a; Leifer et al., 2016a; Leifer et al., 2018b; Leifer et al., 2019; Leifer et al., 2016b).

Here we illustrate our results by plotting a small segment of our *in situ* data. This data segment shows fine-scale structure on kilometer length scales that are extremely improbable to have originated in Europe. Yes, there are larger scale anomalies, e.g., see the transect between 100 and 170 km. These anomalies could have arisen from northern Europe, from nearby area sources, or from transitions between airmasses. These features, however, are not part of our argument for local sources. We only point out that the small-scale features are consistent with local sources in a region absent offshore production platforms and little shipping. Note, these anthropogenic sources would be filtered from the data by their signature in the carbon dioxide data – along with exhaust from the research vessel – as noted in the manuscript.



Still, we considered the treatment of long-range atmospheric transport arguments carefully, as it was a key critique by Anonymous Reviewer #1 as presented in the first version of our manuscript. In response, we significantly refined our analysis and discussion. As such, Reviewer 1 is now satisfied and replied that we treated these concerns seriously and adequately.

In terms of whether there is a strong potential for methane seafloor seepage in the Barents and Kara Seas, our reasoning is as follows:

All petroleum reservoirs in the world leak (termed seepage) methane (Kvenvolden et al., 2001). This is especially the case in areas that are tectonically active. The Barents Sea is such an area. In our manuscript, we cite extensive geologic data that there are reservoirs of oil and gas underlying the Barents Sea, which supports our inferences. These would have been formed by the same geological processes that formed the large and famous Siberian oil and gas fields, whose methane emissions are a significant contributor to the global methane budget (Berchet et al., 2015). Additional circumstantial seabed geological data that these reservoirs leak was presented.

We note that we are familiar with the extensive literature on seepage off Svalbard, e.g., Westbrook et al. (2009), which did not exhibit strong trends in the satellite data, and which is in a highly different geological setting – unrelated to petroleum hydrocarbon reservoirs. As such, it is at best a poor analogy for potential Barents Sea seepage, and although acknowledged, it is not a focus of the manuscript. We have added a citation to hydrates (Westbrook et al. 2009) as a source of arctic methane seepage in the introduction.

We note that our paper is focused on the Barents Sea, and as such, we did not cherry pick references – we select references relevant to our study area, which is geologically distinct from the other areas that have been the focus of arctic methane to date. Bruhwiler also objects that our study did not estimate emissions. We agree that is important next step; and we encourage other researchers to work to evaluate and explore further.

We thank Bruhwiler for highlighting the importance of oxidation time scales; however, this comment seems more addressed to the first version of the manuscript – the revised version has significant discussion, and no new data has arisen for methane oxidation rates at low levels (i.e., outside of plumes) that contradicts the large body of earlier data – again, our study is a satellite study – its is not on plume scales (km), but averaged over much larger spatial scales. As such, appropriate oxidation rates are for those outside plumes, now noted in the manuscript. We are uninterested in reviewing Ruppel and Kessler (2017) here, so simply note that they did not consider methane shoaling, which although uncommon in some ocean basins, is common in the Barents Sea, as we discuss.

Parmentier correctly points out that methane bubbles lose much of their methane as they rise in the water column, citing a recent paper, Leifer et al. (2017). This is not true if they are large or oily (Leifer and Patro; Leifer et al., 2003). Indeed, although this methane dissolves into the water column, this water and its dissolved methane can still mix to the surface, where methane can escape to the atmosphere. This has long been demonstrated for atmosphere trace gases (Watson et al., 1991) and is enhanced by winds that cause wave breaking (Nightingale et al., 2000) and mixing by deep Langmuir circulation (Thorpe, 1992). Barents Sea winds often are high and storms are common (over 15 m s^{-1} for over 125 days per year, mostly (Kolstad, 2008)), as discussed in the manuscript.

We also note that underwater advection of dissolved methane into shallower depths, what we termed methane shoaling by water currents, occurs in the Barents Sea on time scales that are short compared to typical oxidation timescales (please see discussion in the manuscript). These processes would be hard to detect from standard cruises although they could be reconstructed with intensive sampling from ships, moorings, and other technologies – a costly experiment. Satellite data provides an initial basis with which to consider such efforts, as suggested by the reviewer.

Parmentier also neglects the possibility that storm or convective mixing can bring seep methane dissolved in the water column to the sea surface in the Barents Sea. Storms promote air-sea gas exchange to the atmosphere. Direct proof of the efficiency of storm sparging of water-column methane was shown in Shakhova et al. (2013). In the Arctic, multiple storms can occur within the analysis's relevant timescale of one month, increasing the average sea to air transport. Equally important is thermal convection, which can form bottom waters in the northeast Barents Sea (McClimans and Nilsen, 1993), providing a third mechanism to transport deep methane to the atmosphere. These are discussed in the manuscript in detail.

Thus, we provide clear mechanisms and geologic arguments why methane seepage affecting the atmosphere is reasonable and supports the satellite data analysis. Against, this, Parmentier argues that an inversion model of data from the Zeppelin air station on Western Svalbard is sufficient to conclude that there is no significant gas seepage in the central and northern Barents Sea. Yet, the Zeppelin air station is at an altitude that often is above the Arctic marine boundary layer, which traps surface emissions. More importantly, based on prevailing winds, Zeppelin data will seldom be downwind (monthly-averaged) of the Barents Sea areas highlighted as potential sources in the satellite data. Inversion models cannot be considered definitive when there is virtually no weather data to validate the winds and their transport (Berchet et al., 2015) – the Barents Sea only has a couple of weather stations covering 1.4 million square kilometers! As such, we argue that the potential contribution from Barents and Kara Sea geologic methane deserves further investigation and discussion.

Parementier states that "The authors claim that sea ice reduction increases methane emissions to the atmosphere (line 61-62) but no reference is given to support this". Perhaps there was a misunderstanding, but we did not conclude this, but rather pose it as a hypothesis.

The authors note that our team is not government funded, and was not government-funded at a level to include a modeling group. We are seeking to have the support to conduct such models and encourage others to pursue such research

References:

Berchet, A., Pison, I., et al., 2015. Natural and anthropogenic methane fluxes in Eurasia: A mesoscale quantification by generalized atmospheric inversion. Biogeosciences 12, 5393-5414.

Hanna, S.R., Briggs, G.A., et al., 1982. Handbook on Atmospheric Diffusion. Technical Information Center, U.S. Department of Energy.

Kolstad, E.W., 2008. A QuikSCAT climatology of ocean surface winds in the Nordic seas: Identification of features and comparison with the NCEP/NCAR reanalysis. Journal of Geophysical Research: Atmospheres 113, D11106.

Kvenvolden, K.A., Lorenson, T.D., et al., 2001. Attention turns to naturally occurring methane seepage. EOS (American Geophysical Union Transactions) 82, 457.

Leifer, I., Melton, C., et al., 2018a. Atmospheric characterization through fused mobile airborne and surface *in situ* surveys: Methane emissions quantification from a producing oil field. Atmospheric Measurement Techniques 11, 1689-1705.

Leifer, I., Melton, C., et al., 2016a. Fusion of mobile in situ and satellite remote sensing observations of chemical release emissions to improve disaster response. Frontiers in Science 4, 1-14.

Leifer, I., Melton, C., et al., 2018b. Validation of mobile in situ measurements of dairy husbandry emissions by fusion of airborne/surface remote sensing with seasonal context from the Chino Dairy Complex. Environmental Pollution 242, 2111-2134.

Leifer, I., Melton, C., et al., 2019. Husbandry trace gas emissions and exposure: Leveraging fusion of in situ and airborne and satellite remote sensing. Environmental Pollution Under Review.

Leifer, I., Melton, C., et al., 2016b. Remote sensing and in situ measurements of methane and ammonia emissions from a megacity dairy complex: Chino, CA. Environmental Pollution 221, 37-51.

MacDonald, I.R., Garcia-Pineda, O., et al., 2010. HYFLUX: Remote sensing and sea truth of CH_4 flux from the Gulf of Mexico seep system, 10th International Conference on Gas in Marine Sediments, Lake Baikal, Russia.

McClimans, T.A., Nilsen, J.H., 1993. Laboratory simulation of the ocean currents in the Barents sea. Dynamics of Atmospheres and Oceans 19, 3-25.

Nightingale, P.D., Malin, G., et al., 2000. In situ evaluation of air-sea gas exchange parameterizations using novel conservative and volatile tracers. Global Biogeochemical Cycles 14, 373-387.

Ruppel, C.D., Kessler, J.D., 2017. The interaction of climate change and methane hydrates. Reviews of Geophysics 55, 2016RG000534.

Shakhova, N., Semiletov Igor P., et al., 2013. Ebullition and storm-induced methane release from the East Siberian Arctic Shelf. Nature Geoscience 7, 64-70.

Solomon, E., Kastner, M., et al., 2009. Considerable methane fluxes to the atmosphere from hydrocarbon seeps in the Gulf of Mexico. Nature Geoscience 2, 561-565.

Thorpe, S.A., 1992. Bubble clouds and the dynamics of the upper ocean. Quarterly Journal of the Royal Meteorological Society 118, 1-22.

Watson, A.J., Upstill-Goddard, R.C., et al., 1991. Air-sea gas exchange in rough and stormy seas measured by a dual-tracer technique. NATURE 349, 145-147.

Westbrook, G.K., Thatcher, K.E., et al., 2009. Escape of methane gas from the seabed along the West Spitsbergen continental margin. Geophysical Research Letters 36.