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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Using ship-borne observations of methane isotopic ratio in the Arctic Ocean to understand methane sources in the Arctic" *by* Antoine Berchet et al.

Anonymous Referee #1

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The presented manuscript describes an attempt to improve the characterization of the spatio-temporal variability of methane isotope ratios from various sources in the high Northern latitudes. The analyses are based on ship-borne measurements of total CH4 and isotope ratios during summer 2014, which are interpreted with a chemistry transport model and simple atmospheric inversions. As opposed to using isotope ratios to better constrain regional methane emissions and their source types, the authors test herein whether or not the assumption of homogeneous and constant methane isotope ratios for specific source types is plausible. Their findings indicate that there are indeed regional scale gradients in isotope ratios from wetland emissions, and also sources from the East Siberian Shelf appear to be heterogeneous.

The authors postulate that methane emissions from the high northern latitudes are a highly important component of the global greenhouse gas budget, and that they are likely to become more important with Arctic warming in the future. At the same time, the observational infrastructure in the region is very limited, so that isotope observations could become a valuable addition to existing data in order to better constrain the types of methane emission sources, and their location of origin. I fully agree with these statements, therefore I consider their presented attempt to better constrain the spatio-temporal variability in methane source isotope ratio signatures as very important to the research field. Unfortunately, there are some shortcomings in the presentation of the methodology. Also, sensitivity studies on some components of the study setup should be added to facilitate an evaluation of the quantitative results. Detail on these major comments are listed below:

1.) As much as I appreciate a concisely written paper, in this case the description of the methodology ended up being too short, so some characteristics of the optimization setup remain unclear. What exactly is optimized in this approach? In the setup, there are four different source types given for terrestrial areas, plus ESAS and boundary conditions (Table 1), and there are 24 regions differentiated (Figure 1). I assume that the authors optimized only wetland emissions from each terrestrial region, plus ESAS emissions and the 5 boundary conditions, which would make 30 free parameters to constrain (they discuss only 3 optimized parameters/source distributions in the results section ..). However, this is nowhere clearly documented, and given the available combinations of source types and regions, the total number could as well be 102. Also, the source of the starting values for the isotope ratio is given, but not the source of the uncertainty ranges. It may be the same, but this should be documented more clearly.

2.) This paper presents some evidence that the source signatures from wetlands is deviating between different Arctic terrestrial regions, but the accuracy of the results is not tested adequately. The authors provide a short statement that, given the good agreement between observations and simulations of total methane, they assume that

the transport model is correct here (Section 2.3) Also, they provide fixed uncertainty ranges for prior isotope ratios, but fail to clearly document where these are coming from, and how trustworthy they are .The latter may be an easy fix, while the former is clearly reducing the impact of these results. I therefore request to add sensitivity studies where the role of transport and prior uncertainties on the outcome of the study is tested and quantified. It shouldn't be too much extra work to perturb the transport, and check if this results in major shifts in optimized isotope ratios for different regions. The same holds true for the assignment of prior uncertainties.

Overall, this short and (in part too) concisely written paper highlights the need for a better characterization of isotope ratios in different source types and source regions for methane around the Arctic. The overall message, i.e. that there is a large potential in isotope ratio observations to help constrain methane sources, but we need to do a better job in characterizing them, is both important in clearly articulated here. The use of figures and tables is adequate. I therefore recommend the publication of this manuscript in Atmospheric Chemistry and Physics, once the major and minor (see below) comments I raised herein have been addressed.

ADDITIONAL COMMENTS: The introduction could be strengthened by additional material on existing attempts to constrain Arctic CH4 sources in different regions, with different approaches. Where does the controversy on ESAS emissions stem from, and where are the knowledge gaps? How exactly will this campaign contribute to filling these gaps?

As mentioned above, the Methods section is very short, sometimes too short. For the interested reader more details on the methodology (e.g. on the CHIMERE model) should be provided either in an appendix, or in an SI section. In Section 3.3, more details on the optimization target and strategy need to be provided also in the main text (see details above).

Figure 2 raises the question why the cumulative contributions of top and 4x side com-

ponents are so stable over time, given the pronounced variability in contributions from different sides of the model domain, which is also discussed in Section 3.1? Maybe it would be helpful to show the mean concentrations from the global model that were taken as a reference for the 5 boundary values applied here.

One confusing part of Section 3.2 is the mentioning of 'temporal variability' - what are the authors referring to? Obviously, there was no temporal variability in the source isotope ratios detected, or at least none is shown (or discussed) in any of the results. The only temporal variability shown is that of the retrieved isotope signal in the SWERUS data (Figure 2, bottom), but here the variability has been largely attributed to shifts in the origin of air masses, and accordingly source regions. The statement on temporal variability is repeated also in the conclusions. Please clarify.

Finally, please make sure your reference list is up-to-date (old discussion papers cited).

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