

Review of :

Significant methane undersaturation during austral summer in the Ross Sea (Southern Ocean)

Ye et al.

General Comments

This paper presents a nice data set on CH₄ water concentration profiles in the Ross Sea during the Summer 2020. This is rare data, which, *per se*, deserves publication. The data set is interpreted in terms of potential driving processes such as water mass mixing, sea ice melting, biological consumption/production and air-ocean exchange processes.

I have however reservations on several aspects of the paper, both in terms of structure of the paper, methodology and conclusions.

My major concern is that the paper argues that CH₄ profiles in the mixed layer demonstrate the impact of sea ice melting on the CH₄ dynamics in the area. First of all, I have doubts on the pertinent use of CH₄ % saturation vs. Salinity plots to demonstrate the impact of mixing processes. I believe several processes are embedded in the saturation level of CH₄: temperature, salinity changes, air-ocean exchanges biogeochemical processes and not only dilution processes by mixing. Also, nowhere in the paper is it made reference to existing CH₄ sea ice concentration data in the Antarctic (they are indeed rare, but they do exist). The latter show bulk ice concentration ranges similar to those presented in the water profiles of this paper, which make it very difficult to explain 22% reduction of CH₄ saturation in the whole (25-50m thick) mixed layer from melting contribution of 1m thick sea ice, even when using minimal sea ice values. Plotting CH₄ concentration values vs. Salinity does not show any trend from West to East along the transect, probably due to partial re-equilibration with the atmosphere (on this I agree with the authors).

The authors also make an overall budget of CH₄ air-ocean fluxes for the Ross Sea, based on a 3-days cruise flux measurements... which I think is a bit overstretched. More precaution should be taken in presenting those results (e.g. no clues on what happens in the winter!). The authors already reckon that the unbalance is rather insignificant for their observation period extrapolated to the whole summer period in the Ross Sea.

There are also a certain number of contradictions at several places in the manuscript, which I have listed more completely in the detailed comments here below.

In short, I think the paper is not yet mature for publication in Biogeosciences. My first reaction would be to reject the paper at this stage, but to encourage the authors to rethink the methodology and interpretation of the data and provide us with a new manuscript. Arguments should be presented to dissociate the impact of temperature, salinity, dilution and biogeochemical processes on the observed saturation states, and how these are disentangled from water mass mixing processes.

Detailed Comments

I. 15: Delete sentence "Simple box model...waters."...this is more like just a mass balance calculation, from the description in the methods...also there is a contradiction with the methodology where it is stated that advection is considered as negligible!(I. 125)

I. 18: add sentence: "Simple mass balance calculations further suggest that CH₄ consumption also contributes to the CH₄ undersaturation."

I. 18: Delete "Thus we argue that.." and replace by: "Both CH₄ consumption and sea ice melting are therefore important drivers of CH₄ undersaturation, which implies that the high-latitude area of the Southern Ocean act as a sink for atmospheric CH₄ in the summer."

I. 19-20: This is a summer only budget!... cannot be used as an annual budget", not knowing what happens in the winter!..

I.47-48: "storage of CH₄ in ice **crystals** is usually a minor source compared to **sea ice brines (bubbles or dissolved)** and under-ice sea water **in the Arctic** (..)."

I.53: "we propose that (i)..."

I. 54: "..our results **from a summer West-East field transect in the Ross Sea...**"

I. 54: "and (ii) sea ice melting.."

I. 47-54: This section should refer to the work of Damm et al. on in-situ sea ice CH₄ production and impact on air-sea fluxes in the Arctic

I. 56: "Hydrographic **measurements** and.."

I. 57: I am missing here the description of the actual measurements!.. CTD, precisions...actually, move material from section 2.2

I. 57-71: This is not "Method"!.. but more general statements about ocean circulation in the Ross Sea.. This should be moved to "Introduction" (after lines 32-34) , or eventually used in the discussion section

I. 57-58: Figure 2 and Tables S1 are "Results", not methods

I. 76: Table S2 ans S3 are Results, not Methods

I. 76-77: not clear what the "total volume" is in this case..

I. 78: "**CH₄** sampling and analysis"

I. 79-80: "onboard...January 2020".. move to section 2.1

I. 85-87: Move to section 2.1

I. 91: "is **calculated** by"

I. 95: "-1.9 **to** 1.4°C"

I. 95: "sampling **with the** atmospheric CH₄..."

I. 96: South Pole data.. why not use data from closer by?...Arrival Heights?..(<ftp://ftp.niwa.co.nz/CH4/arch>)

I. 101: "2.3 Flux Density Calculations"

I. 107-110: Wind data: I am sure there were true wind data recordings on the Xuelong 2, isn't it?.. It would be more accurate to use those

I. 113: "CH₄ budget in the mixed layer": Is this what you also refer to as "box model" elsewhere in the paper. should be mentioned!.. is it really a "box model"?.. it is indeed more of a mass balance calculation than a "box model"

I. 114: "In order to give a first estimate of..."

I. 123: This is a true equation for Fvd..should have the status of an equation on a separate line

I. 124-125: "advection... negligible" .. in contradiction with what is said in the abstract!

I. 127: "sink **for** the mixed layer"

I. 128: Introduce here figures 1 and 2 and supplementary table with CH₄ concentration... which should actually be grouped with all the rest of the supplementary material!...

I. 130: Figure 1 d should include a plot of $\Delta\sigma$

I. 131: "temperature gradient" .. not differences, given the units..

I. 141: Introduce also here a few words on the CH₄ concentrations, referring to the table, and indicating low variability overall (1.5-5.2 nM). Figure 1b should also show the CH₄ concentration profiles. Actually Figure 1b deserves to be an isolated figure.

I. 150: "(Figure 2b, Table S2)

I. 155: "most **heterogeneous** region"

I. 156: "was found only **closer to** the ice sheet **and at depth (Table S2)**"

I. 161: "in February **2020**"

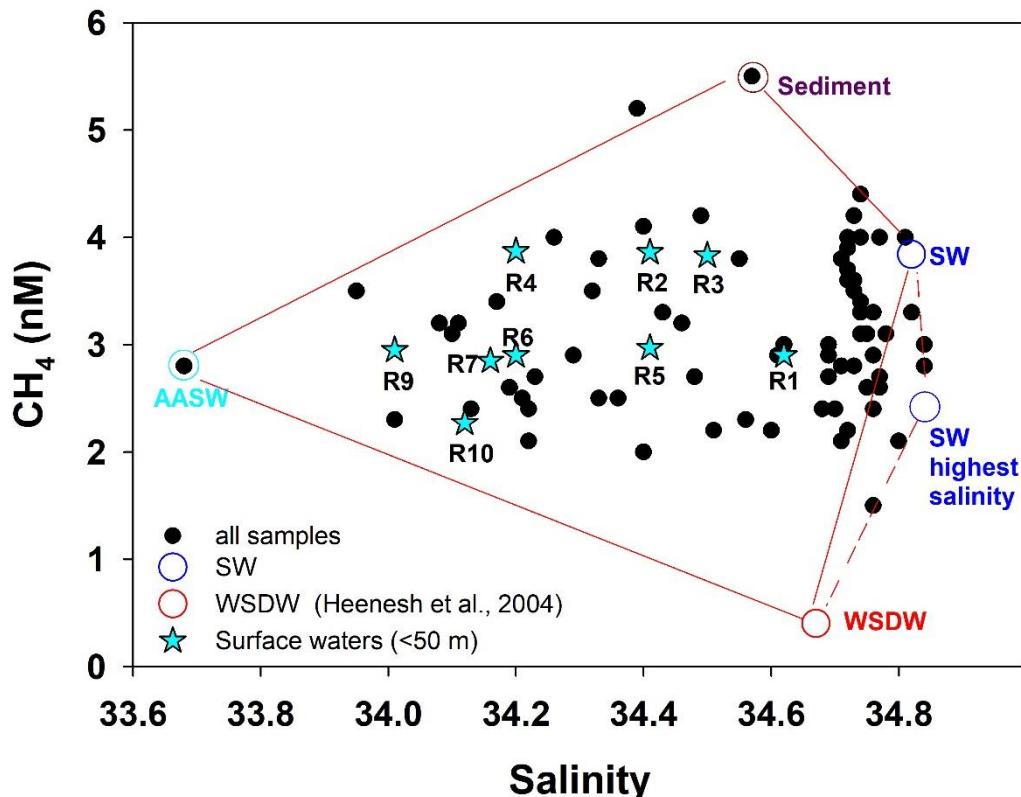
I. 164: " (**Figure 3d**, stations R1-R5..."

I. 166-172: This is already "Discussion"

I. 163-164: However, CH₄ concentrations are relatively constant in surface waters throughout (ca. 3 nM) so this trend could be S/T impact on saturation rather than dilution through mixing with melted sea ice.

I. 170-172: "We found...".. Where is the blue arrow in Figure 3e coming from?.. If I understood correctly, you are using the numbers from Figure 3d, correct?...But there, the change in saturation level could also result from the combination of other processes such as the effect of temperature and salinity on solubility (which is used to calculate the saturation) or exchanges with the atmosphere, correct?..

In fact I am a bit disturbed by the use of a % saturation vs. salinity plot to discuss mixing processes, while obviously temperature and salinity changes should also affect the saturation numbers. Why not use simply a CH₄ concentration vs. salinity plot, as shown here below (built from the values in the supplementary table, in the same way you built your Figure 3e)?



There you see a similar arrangement than in your figure 3e, but clearly the concentration of the mixed layer waters (same criteria as yours – all 50 m depth in this case), shows no trend with salinity... dynamics of CH₄ does not indicate dilution from melting sea ice... the CH₄ bulk concentration of which could be close to SW (and not negligible!).. see further comments..

- I. 173: "Contribution of Production/oxidation from mass balance calculations" ... this is not really a box model, and you are talking about those biogeochemical processes in this section
- I. 187: "Figure 3e" .. there is no MSW in Figure 3e
- I. 189: "in a **negative** CH₄ gradient.." a gradient is a slope.. I don't think you are tracking changes of slopes
- I. 190: Figure S2 is tricky to interpret.. it is strange that linear correlations come out while 4 end members are involved (?).. Is it that mixing of AASW is mainly with SW, and not CDW (which would inverse the trend)
- I. 191: "Hence, superimposed to the effect of microbial oxidation of CH₄ (section 3.4)..."
- I. 192-194: Why supersaturation, then?...and why limited to the stations R1 to R4?.. Clearly the distribution of supersaturation in Figure 1b (top 100m) "roots" into the coastal areas.. could there be enrichment from shallow sedimentary sources (e.g. triggered by tidal forcing?) Another source might be ISW (in contact with sediments upstream), but there doesn't seem to be higher CH₄ at those depths in your profiles..
- I. 196: "caused by dilution" ... but, as mentioned before, undersaturation could also result from T/S changes, where is the balance with dilution?

I. 202-205: same comment as above! You should estimate the contribution of each process to demonstrate that dilution is indeed the main factor!... Looking at the figure above, there is no trend in surface waters with the decrease of salinity... but of course, there exchange with air might have blurred the signature...

I. 211: "box model" ... rather "Mass Balance calculation"?

I. 212: "increased its contribution in the east" ...isn't this in contradiction with Figure 3d?

I. 215: "In the west..." not east

I. 216: "injected freshwater" ...If I understood correctly from previous saying in the paper, this refers to sea ice melt, correct?.. I am a bit surprised that it would have affected the whole mixed layer: sea ice is maximum 1m thick, the mixed layer is 25-50m. Sea ice CH₄ concentrations in the Antarctic are not very well documented, but they do exist. Jacques et al., 2021 report on a range of 1.5 to 7.4 nM for bulk CH₄ concentration in McMurdo Sound, with a mean around 3 nM. This value is similar to the values for waters in this paper (1.7 to 5 nM). I am therefore not convinced of the impact of dilution from melting sea ice on mixed layers concentrations. Even supposing a minimum sea ice concentration of 1 nM, it would be a factor 3 to 5 lower than SW values, while the mixed layer is a factor 25-50 thicker!...again, plotting CH₄ concentration vs. salinity does not show any trend..

I. 218: "exposition to the air"

I. 218: "30 days ice free period in the east" ... but this is where Fig. 3d shows minimum supersaturation, correct?.. contradiction?

I. 222: "suggests" .. why not only continuous dilution?

I. 223: "previous study" references?

I. 229: "the compared results among stations" ... I don't understand. Please rephrase!...

I. 231: rather "Summer CH₄ uptake in the Ross Sea"

I. 255-256: you should also insist on the fact that no CH₄ data is available today, which hampers the possibility of providing an annual flux budget!..

I. 260: "sea ice melting is likely to enhance.." .. this is actually not demonstrated by the CH₄ data in this paper..

I. 265: "which underlines the potential significance.."

Suppl. Table CH4: please use contrasting background to better define the stations data

Figure 1: should show ISW out of the Ross Ice Shelf

Figure 1b, should be a separate figure

Figure 3 caption:

- a) define "surface"
- b) Heeschen et al. do not give CDW values, if I remember correctly, but WSDW with a minimum of 0.4 nM CH₄

Figure S3: where is this scheme commented in the text?.. Maybe I missed it!..