

## ***Interactive comment on “Physical controls on the storage of methane in landfast sea ice” by J. Zhou et al.***

**J. Zhou et al.**

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REVIEWER 1: I like the originality of this finding as well as the experimental approach implemented by the authors to collect and interpret the reported data. They have finally shed greatly needed light on the origin of methane (CH<sub>4</sub>) measured within the sea ice, which some authors in earlier published papers attributed to biological processes involving in-situ production within the ice body (such as CH<sub>4</sub> production, including methanogenesis under aerobic conditions, and CH<sub>4</sub> oxidation). Neither of these recently published papers left me convinced about the origin of CH<sub>4</sub> measured within the sea ice. On the contrary, the authors of this study suggest that the landfast sea ice serves as a temporal storage depot for CH<sub>4</sub> and is one of the major factors controlling atmospheric emissions of CH<sub>4</sub>, acting as a physical barrier restricting these emissions

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during the period between ice formation and ice breakup. They demonstrate that the buildup of CH<sub>4</sub> stored in the sea ice (standing stock) is due to incorporation of CH<sub>4</sub> from the seawater and from gas bubbles, the formation of which, they suggest, is triggered by strong solubility changes. Most of their arguments are logical and convincing to me. Their data on subsequent evolution of CH<sub>4</sub> storage in the ice layers, which demonstrate that levels of CH<sub>4</sub> in the standing stocks are not affected by biological processes in these layers, serve to demonstrate how negligible the influences of biological processes are, compared to the physical controls of the sea ice. These findings are very important, because they improve our understanding of the role of sea ice in the exchange of CH<sub>4</sub> between the Arctic Ocean and atmosphere during the period from ice growth to ice melt. Thus, the authors are dealing with a topic of highest interest. I am generally happy with the methodological approach used by the authors. Methods used by authors allowed them to clearly show that mean concentrations of CH<sub>4</sub> as well as standing stocks were correspondingly increasing as sea ice thickness was growing while the ice remained impermeable for gases (from February to April). They further showed that both parameters decreased in June when sea ice became permeable for gases and started venting CH<sub>4</sub> to the atmosphere. The authors logically attribute the relative stability of the CH<sub>4</sub> standing stocks to the ability of impermeable sea ice to physically control CH<sub>4</sub> accumulated within the ice during its growth. They also showed that even if some biological activity could have taken place, its importance was negligible in comparison with the importance of the physical processes responsible for CH<sub>4</sub> accumulation within the ice. They also reasonably argued that levels of CH<sub>4</sub> supersaturation and rates of ebullition, where ebullition occurs, would determine the levels of CH<sub>4</sub> incorporated into sea ice. This is a very important conclusion that has not been suggested by other authors who are measuring CH<sub>4</sub> in the sea ice.

I agree with the conclusions as phrased except for those regarding biological processes that may affect the CH<sub>4</sub> in the sea ice. What would be the source of methanogenesis in those theoretically-possible anoxic micro-niches in the ice, if the authors reported that annual ice rejects a major fraction (85%) of all impurities, including most likely organic

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substrates, during its formation? In this regard, I would like to remind, that there were reports of possible methanogenesis occurring in the anoxic lenses accumulated on the bottom of the pycnocline in the highly productive tropical waters. Nevertheless, levels of dissolved CH<sub>4</sub> measured within those sub-surface maxima varied in the range from 4 nM to 9 nM. Moreover, these rates of methanogenesis required accumulation of organic pellets from the water column of >100 m in thickness! Note, thickness of landfast sea ice is only 2 m.”

AUTHORS: There is a selective incorporation of dissolved organic matter (DOM) in sea ice (Müller et al., 2013). Hence, there are more organic substrates in sea ice than it would be if DOM behaved as salt. We agree with the reviewer that methanogenesis in micro-niches may be insignificant, but the importance of such process still needs to be assessed.

REVIEWER 1: Regarding CH<sub>4</sub> oxidation, recently reported data from adjacent parts of the Arctic Ocean suggest that the dissolved CH<sub>4</sub> pool turnover time is much longer than the lifetime of the fast ice. In addition, the authors need to know that  $\delta^{13}\text{C-CH}_4$ , modernly produced in the Arctic environment, exhibits an isotopic signature much lighter than -50 ‰ or -70 ‰ *this signature could be as light as -100 ‰ or even lighter. If CH<sub>4</sub> is produced in situ in the sea ice, its isotopic signature should reflect the process appropriately. Nevertheless, I a*

AUTHORS: We agree that isotopic data will be the ultimate tool to reveal biological activity (if any), as highlighted in the conclusion. Also, we have improved the manuscript by including the turnover time and the lower isotopic signature following the comment of the reviewer. The related paragraphs now read as: “Furthermore, the turnover time for CH<sub>4</sub> oxidation in the Arctic Ocean exceeds 1.5 years (Griffiths et al. 1982 and Valentine et al. 2001), which is much longer than the lifetime of first year landfast ice. If we assume that the turnover time is similar in landfast sea ice, then we do not expect to find major CH<sub>4</sub> oxidation in our ice samples.” (Revised version in Word, p. 9, L17-20) And “In case of a higher mix of physical and biological controls on CH<sub>4</sub> concentrations in bulk ice, we would recommend to measure: (1) the carbon and hydrogen isotopes of

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CH<sub>4</sub> in sea ice, as isotopic fractionation is highly sensitive to biological processes, and (2) the same isotopes in the sources (e.g., organic matter). Indeed, previous studies have suggested that biogenic CH<sub>4</sub> within anoxic sediments may have carbon isotopic values as negative as -110 ‰ (Whiticar, 1999), in comparison to that formed by CH<sub>4</sub> oxidation (-10 to -24‰ (Damm et al., 2008; Schubert et al., 2011)), but few of them have considered that the measured isotopic values in the sediment or in seawater also depend on the isotopic composition of the sources.” (Revised version in Word, p.14, L22-29) REVIEWER 1: Regarding the manner of presentation, I would suggest that the authors re-think their presentation of data in Figures 2, 3, and 5 in order to improve readability. With these corrections, I recommend publication of this manuscript.

AUTHORS: We have removed the Y-axes on some of the sub-plots in Figure 3 and 5. This allows us to enlarge the subplots and to improve the readability. We will also ask to put the figures in “portrait” during the proof-reading process.

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Interactive comment on The Cryosphere Discuss., 8, 121, 2014.