

Interactive comment on “Evidence for methane production by marine algae (*Emiliana huxleyi*) and its implication for the methane paradox in oxic waters” by K. Lenhart et al.

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General comments Despite there are several suggestions to explain the methane (CH₄) production pathway in the oxic ocean, the accumulation of oceanic CH₄ remains enigmatic. The idea that CH₄ might be produced by phytoplankton (algae) is not a new one; however, detailed studies on this issue are still lacking. Lenhart et al. present a novel data set of CH₄ production rates from a study with a *E hux* culture. The data and conclusions presented are of high interest for anyone dealing with the biogeochemistry of oceanic CH₄ cycle. Authors: We thank the referee for their positive comment

However, and very unfortunate, the authors try to over-emphasize the significance of

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their results. The ms needs to be focused on the main conclusion (i.e. *E hux* has the potential to produce and release CH₄). Any further far-reaching speculations about the CH₄ paradox are not justified by the results presented. Therefore, I can recommend publication of the ms only after major revisions. These are my points:

1) *E hux* plays an important role in the ocean, but of course it is only one of many algae species out there. Therefore the authors should avoid giving the impression that *E hux* is representative for all algae. Their phrases ‘. . . marine algae such as . . .’ (p.20325, l. 21) or ‘Since our results unambiguously show that algae are able to produce CH₄ per se under oxic conditions . . .’ (p.20344, l. 17/18) etc. have to be rephrased. This also applies to the title. To make it short, it is not acceptable to draw the conclusion that algae generally produce CH₄. Authors: As suggested by the referee we have made some modifications to the manuscript. We have modified phrase p.20325, l. 21 to read “Moreover, the absence of methanogenic archaea within the algal culture and the oxic conditions during CH₄ formation suggest that the widespread marine algae *Emiliana huxleyi* might contribute to the observed spatial and temporal restricted CH₄ oversaturation in ocean surface waters.” And phrase p.20325, l. 21 to “Since our results unambiguously show that the common coccolithophore *E. huxleyi* is able to produce CH₄ per se under oxic conditions we thus suggest that algae living in marine and freshwater environments might contribute to the regional and temporal oversaturation of surface waters.” Furthermore we have revised the title to read “Evidence for methane production by the marine algae *Emiliana huxleyi*”.

2) Introduction: The oceanic source of CH₄ is negligible compared to other natural and anthropogenic sources of atm. CH₄ (see e.g. IPCC 2013). This is not mentioned in the introduction leaving the reader with the impression that the oceanic source is indeed significant for the global budget, which is not the case (see e.g. p.20326, l.25/26). Please modify the introduction and mention the oceanic source strength. Authors: We have removed the sentence “In order to reliably apportion the global CH₄ budget, it is essential to know all significant sources and sinks and the principal parameters that

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control emissions.” from the manuscript and we now mention the ocean CH₄ source strength: “The world’s oceans are considered to be a minor source of CH₄ to the atmosphere with approximately 20 Tg CH₄ yr⁻¹ (Etioppe, 2008).”

3) Introduction: The introduction needs a significant shortening and a focus of the main theme of the ms. There is a lot of information given which are not necessarily needed in the context of the ms. See e.g. paragraph about MPn as source of CH₄ (see p.20328, l.5-19) and other parts of the introduction. Authors: We have modified parts of the introduction and removed a few sentences (e.g. comparison with freshwater ecosystems). However, we would like to mention the potential role of MPn for methane formation in the ocean (please refer to comment by referee 2 who asked to present alternative CH₄ formation pathways). Furthermore, we would also like to keep the introduction in its greater detail for readers which are not so familiar with biospheric methane formation under aerobic conditions and the CH₄ cycle in the ocean.

4) Please avoid comparison with freshwater lakes) and terrestrial (plants) systems which are not comparable with the oceanic ecosystems at all; there are several places in the text where a comparison with results from lakes and terrestrial plants are presented. Please modify. Authors: In the revised manuscript we have omitted the comparison with freshwater ecosystems and have been also much more careful with the comparison of the emission rates with terrestrial plants.

5) I am wondering about different interpretation of the conclusions from Bange and Uher (2005). On the one hand, I read that photochemical production is ‘negligible under oxic conditions’ (p.20328, l.3). On the hand the authors cite Bange and Uher (2005) as being in line with their findings of a chemical CH₄ production found in their study which was conducted under oxic condition, I suppose. I think that this latter case is a misinterpretation of the results of Bange and Uher (2005). See also p.20342, l.25/26 where a photochemical CH₄ formation is listed as a potential CH₄ pathway in oxic surface waters. This is not correct; please modify. Authors: We have modified the sentences taking into account the referees suggestions.

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6) The section 5.4 ‘Methane paradox in oxic waters reconsidered’: This sections does not present any new results or conclusions and is way too much speculative. Therefore, it has to be omitted. Authors: We have reworded the subtitle of this discussion section to read ‘Potential implications for the occurrence of CH₄ in oxic marine waters’. As this section belongs to the discussion of the manuscript we believe that it is helpful for the readers to discuss our findings in a broader context in relation to previous findings regarding precursor compounds and potential reaction pathways.

We would like to discuss the potential meaning of algae-derived CH₄ formation in oxic waters. However, we have reworded this section and changed the title of this section to “Potential implications for CH₄ cycling in oxic waters”.

Minor comments Section 2.3 Gas Chromatography: Why are CO₂ and N₂O mentioned? These measurements are not presented in the ms. Please correct. Authors: Correction made

p.20334, l.5: ppbv is not a concentration it is a mixing ratio. Please correct. See also p.20334, l.15. Authors: We changed concentration to mixing ratio throughout the manuscript.

p.20339, l.20: Schiebel et al. (2011) is missing in the ref list. Authors: Correction made

p.20340, l.9: I could not find any information how CH₄ emissions (given in ng/gPOC/h) from the E hux culture have been converted to ng /gDW/h. Please explain. Authors: The description is provided in the materials and methods section 3.4 “Calculation of CH₄ production” (20335 Line 7-16): “In order to compare CH₄ production to literature data it was necessary to normalize to cellular particulate organic carbon (POC) quota, as opposed to cell. The POC normalized CH₄ production is termed “methane emission rate” in the following. Since it was not possible to measure cellular POC quota on a daily basis, we used a literature value determined for the same strain under similar culture conditions, i.e. 10.67 pg POC cell⁻¹ (Langer et al., 2009). We are aware of the fact that cellular POC quota is likely to change alongside other element

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quotas when approaching stationary phase, but this change is well below an order of magnitude (Langer et al., 2013). For our purpose this method is therefore sufficiently accurate to determine POC normalized CH₄ production.

The authors wish to thank the referee for their efforts in reviewing our manuscript and for the helpful and constructive comments provided. Below are our point by point responses to all issues raised by the referee. The manuscript has been revised accordingly.

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