

Interactive comment on “A Fast-response automated gas equilibrator (FaRAGE) for continuous in situ measurement of methane dissolved in water” by Shangbin Xiao et al.

Anonymous Referee #2

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The information provided with this Technical Note is clear, well presented, and sufficiently useful for the scientific community. The idea of an equilibrator for measuring dissolved gas species in situ is certainly not new, but this work may deserve consideration because it finds a clever way to make the measurement faster for dissolved methane. The set up takes advantage of the relatively new technology that led to small, fast responding gas spectrometers with quite big measuring cells and couples it to a small flow-through system to equilibrate/separate water and gas. The system is presented in a way that can be easily reproduced with affordable costs, hopefully promoting a long lasting need to resolve and map the spatial heterogeneity of dissolved methane in inland waters. It is indeed desirable for the measurement technique to achieve the

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highest surface/time ratio.

I have two remarks to be considered and few suggestions 1) I was very interested in the CH₄ profiles and - while impressed by the high vertical resolution they achieve - I understood the authors tested their system in different lakes, covering some “typical” methane concentration range. Indeed they show two profiles: one from meso-oligotrophic Lake Stechlin and one from eutrophic Lake Arend - I supposed they choose trophic state as a proxy for GHG content (Beaulieu et al., 2019). Thus I was confused seeing that Lake Arend, that they present as eutrophic, shows especially low methane, even lower than what they show for the meso-oligotrophic (Lake Stechlin). So what is the criterion behind this choice? Why picking an eutrophic lake that has even less methane than the meso-oligotrophic? Why not a typical eutrophic lake with methane building up below the oxycline during summer stratification? (They show a large literature on this issue in the Intro). For calibration the authors limited the upper range of methane to <2 micromoles L⁻¹. While for high concentrations (microM to mM range) the authors suggest dilutions (line 357) but they don't deal with the problem in this paper. I think the range they show is fair enough, but they should clarify this “lower range test”, directly relatable to surface waters but not to littoral methane rich or eutrophic lake bottom waters, AND change the sentence in the abstract “The FaRAGE is capable of continuously measuring dissolved CH₄ concentrations in the nM-to-mM range” as it may be capable of that, but is not shown here. 2) One big step forward the authors claim is the “extraordinary fast response relative to all existing gas equilibration devices” (line 274). However, when their system is used with PICARRO G2132-1 + a desiccant to measure stable isotopes of gas species, it does not get that much faster than Hartmann 2018 “High Spatio-Temporal Dynamics of Methane Production and Emission in Oxidic Surface Water” (line 107 of supplementary material). If I understand correctly then, what showed in table S4 is not entirely correct since their system response time when using PICARRO G2132-1 is obtained without any dryer. As far as I know, for using a PICARRO G2132-1 with a moist flow a dryer is absolutely necessary. How humid is the water entering the analyzer? In case the authors

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think that a dryer is needed, they should change the table S4 and the statement in line 274 “ The comparison between up-to-date previous studies and this study (Table S4) demonstrated the extraordinary fast response relative to all existing gas equilibration devices. A 53 s response time was achieved when the FaRAGE was adapted to the Picarro G2132-i, which is significantly faster than others (171-6744 s).” Suggestions When describing the set up the authors often refer to a “bubble unit”, which I suppose in the scheme (Fig.1) is called “gas-water mixing unit”. Consider harmonize Line 363- I think it would be better to rephrase the reason why CO₂ is not shown. “for simplicity” for them or for the reader? Maybe they can mention which non-simple problems do we meet when applying the system to CO₂. As to line 366 it is possible for the authors to use their system at sea enhancing the liquid to HS ratio to achieve low concentrations. I would recommend to make sure that the scientific community that works on GHG air-water exchange in oceans gets interested too (add to abstract and line 97?). For different reasons from the ones highlighted here for inland waters (to name one the massive lack of ground data to calibrate satellite inferences) this system could be applied to voluntary observing ship programs to map CO₂ and CH₄ surface concentrations. In case the authors find a major obstacle to this it would be good to mention - making a suggestion for adapting their system for oceanographic applications. Line 382- they mention how temperature should be corrected for the change along the hose- can give an example on how off can it get and does that mean to always measure temperature in situ at depth along with the profile? Line 400 replace “filthy” Line 410- wouldn’t a scrubber serve for that? Would that slow down the system RT?

References Beaulieu, J.J., DelSontro, T. & Downing, J.A. Eutrophication will increase methane emissions from lakes and impoundments during the 21st century. *Nat Commun* 10, 1375 (2019). <https://doi.org/10.1038/s41467-019-09100-5>

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