

Biogeosciences Discuss., author comment AC2  
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## Reply on RC2

Laura Balzer et al.

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Author comment on "Role of formation and decay of seston organic matter for the fate of methylmercury within the water column of a eutrophic lake" by Laura Balzer et al.,  
Biogeosciences Discuss., <https://doi.org/10.5194/bg-2022-170-AC2>, 2022

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*We would like to thank reviewer 2 for the constructive and helpful comments.*

### General comments.

**This paper builds on several prior studies that show that the water column of lakes and oceans can be an important site for MeHg formation. It differs from most water column studies by focusing on a eutrophic urban lake and by specifically targeting MeHg abundance in bulk seston at different depths and dates for clues about formation and decay mechanisms. Unfortunately, the sampling technique lumped zooplankton in with seston, potentially introducing bias due to biomagnification. And the sampling scheme was spatially inconsistent, which makes the comparison of depth profiles on different dates difficult. The reason that the entire water column was sampled on one date and only the upper water column on most other dates is unexplained, and it compromises the authors' conclusions about what's going on as particles sink (especially in the hypolimnion since it was rarely sampled). Among other things (below), the authors need to justify their sampling methods and revisit the interpretation of changes in Hg speciation across depth and time. They also need to reconsider conclusions about links between climate change, productivity and bioaccumulation. This will require major revision.**

*We agree with reviewer that there might be potential bias in the interpretation of our MeHg depth profile. Our focus was on bulk seston and to our knowledge zooplankton is part of seston. We also believe, that the distribution of zooplankton alone cannot explain the THg and MeHg depth profiles in our lake. The separation of phyto- and zooplankton is useful in studies on biomagnification, which was not our topic. In this case, a qualitative separation of both fraction in small amounts is sufficient. In case of bulk seston sampling, eg. by means of a pump-and-sieve/filter system (0.45 µm?) during algae blooms as suggested by the reviewer such qualitative separation is nearly impossible (agglomeration) if larger sample volume is needed. Moreover, we believe that our interpretation of the distribution of MeHg and THg in the water phase is supported by our data on algae biomass, (Chl 1, pH) is sound. We regret that our sampling was imperfect, we made a lot of effort to gain in all cases sufficient material, however, this is a natural system with sometimes unpredictable changes of conditions. The reason why sampling is inconsistent through time and space is that we could not get sufficient material from the hypolimnion within the possible pumping time (~ 2 h per layer). We already mentioned*

*this in the text but will explain this in more detail (see L254).*

*We will tone down on our conclusion regarding the link between climate change, productivity and bioaccumulation. We assume that the reviewer based this comments on his assumption that we mainly see biodilution. We have commented on this below.*

### **Specific comments.**

**The term “endogenic” should be reconsidered. It means “within the system”, which for lakes technically includes sediments. “Water column” would be better, unless they mean “within the seston” – in which case the title and text need to be re-worded**

*This term has been introduced in other studies. For example, in Gallorini et al. 2021: We will define it within the manuscript as “production within the water column”.*

### **Line 89 is an incomplete sentence**

*Will be changed*

**Line 90: why a 25µm net? It would allow many cyanophytes and chlorophytes to pass through, and bias collection toward zooplankton (which are not “seston”). Why not a clean pump-and-sieve/filter system instead?**

*We agree with the reviewer that some of the small fraction of phytoplankton might have got lost during water pumping through a 25 µm net. We tried a pump-and –sieve filter system before. However, this took too long to gain sufficient material from each water layer to do the solid phase analyses needed here at a resolution of 1 m within a single day (filter clogging etc.). Because of this, we decided to pump the water through a 25 µm net. Although it would have been the best option to sample all phytoplankton fraction, we believe that the lack of the fraction < 25 µm has no significant influence on the overall results and conclusions of this study (it just means more phytoplankton). To our knowledge zooplankton is part of the seston, too.*

**L220-225. The seston samples collected on those dates are not really much closer to the sediment surface. There’s just one hypo sample and it’s directly beneath the RTZ. You’d need to sample more depths to justify. Revise.**

*The reviewer is right, the deepest sample was taken just below the RTZ in those months. We will revise this statement. As explained above. We were not able to gain sufficient material from deeper layers in those months within the possible pumping time (2 h)*

**L235. But peak concentrations of MeHg in seston occur in the suboxic RTZ on 4 of the 5 dates when the lake was strongly stratified. On the remaining date, seston MeHg concentrations are highest in the upper hypolimnion. During stratification, MeHg is never highest in the oxic epilimnion.**

*We agree that this sentence is misleading and will clarify this section.*

**If anything, these findings suggest that MeHg production is associated with microbial respiratory pathways that are less energy efficient than O<sub>2</sub> reduction (e.g. sulfate reduction, Fe reduction). Revise.**

*Not clear what the reviewer means here and what should be revised. We discussed in the ms that MeHg formation is related to redox conditions in the water column specifically to Mn reduction (similar to what has been shown by Petersen et. al 2020(in a lake) and by*

Kohler et al., 2022 (in the arctic ocean) Fe reduction is of minor importance in this lake and sulphate reduction does not take place in the water column (compare Fig. S3). Data on eutrophic lakes are rare and to our knowledge not available yet at similar high resolution. A major aim of this study is to show changes in MeHg in seston at this comparatively high temporal and spatial (depth) resolution to understand the evolution of MeHg and THg concentrations and proportion during sinking through the water column.

**L240-245. Alternatively, low MeHg during high productivity may reflect biodilution in the larger phytoplankton biomass (i.e. parental seston). Lacking sound data, one can't distinguish zooplankton bias from biodilution in microplankton, and neither necessarily point to sestonic microniches. Revise**

We agree with the reviewer that the variation in MeHg concentration might be a result of biodilution, zooplankton influence and/ or the formation of sestonic microniches. Based on pH, O<sub>2</sub> and chlorophyll a concentration, we have classified the sampling days of May and November as periods of lower productivity and the sampling days in April and between June to September as periods of higher productivity (L. 181-190). Our data shows that the time of lower productivity is likely accompanied by higher amounts of zooplankton (higher N concentrations and lower C/N ratios in the upper two meters) and the time of higher productivity is likely accompanied by lower amounts of zooplankton (lower N concentrations and higher C/N ratios in the upper two meters) (L 238-246; Fig. 4). But we agree with the reviewer that we cannot fully rule out that the lower MeHg concentrations in times of higher productivity are caused to a certain extent by biodilution and vice versa the higher MeHg concentrations in times of lower productivity by bioconcentration. But we believe that biodilution/bioconcentration is not the major process in our lake as our data point to zooplankton influence between the sampling days. If biodilution/bioconcentration would be the the major process determining the MeHg concentration in our seston samples, we would also see a dilution or an increase in seston THg concentration, which we do not. The THg concentration do not change in the same way as the MeHg concentration. So there has to be another process than biodilution/bioconcentration controlling the MeHg concentration. However, as discussed in the ms the distribution of zooplankton -alone cannot explain the THg and MeHg depth profiles in our lake (L267-275). Obviously, we cannot completely exclude that zooplankton contributes to the maximum MeHg concentration within the RTZ. Our idea is that more organic matter rich seston (as a result of algae blooms) favours MeHg formation in sestonic unoxic microniches as oxic microbial pathways of MeHg formation are not yet known and MeHg get enriched during decay of the phytoplankton-dominated organic material. We will revise this section to make our point clearer.

**L255-263. They could also be explained by the presence of free-water microbes that possess the methylation gene pair hgcAB and occupy the O/A boundary. DOM rather than POM could be their carbon source. Revise.**

We discussed this point in L 266. It is likely that free-water microbial Hg methylation occurs, specifically because there is predominantly easy accessible DOM in the water column. But to our knowledge, oxic microbial pathways of MeHg formation are not yet known. Many papers point to methylation within anoxic microniches. However, our data suggest that free-water microbial Hg methylation is rather not the dominant process here as high MeHg concentration only occur during times of a pronounced RTZ (compare April when production is already high but MeHg is low because redox-zonation is not yet established) We will try to make this point clearer in the ms.

**L275-284. Sestonic MeHg in the 20% range is not atypical for unpolluted temperate lakes. What's unusual is the very low %MeHg in April**

We can tone this statement down. However, we think that 20 % is a lot regarding the high

*biomass and that there is no influence from soil derived DOC/MeHg-rich inflow, which is typical for many oligotrophic lakes. We believe that the low MeHg proportions and concentrations in April (compared to the summer months) supports our conclusion that the MeHg is predominantly formed in the water phase along redox-gradients/micro-niches and the role of the RTZ. In April, algae biomass and productivity as indicated by high Chl a and high pH is already high, but the redox gradients are only weakly developed (only weak Mn-reduction) (Fig. S1), most likely because organic matter decomposition in the water column is still low. If most MeHg is originated from the sediment we should see this MeHg in the seston also in April.*

*We will try to make this point clearer in the ms.*

**L346. Actually, this was first shown in Little Rock Lake, which is only 10m deep (but the eutrophic part may be right).**

*We will refer to this point in the ms. However, we actually pointed out that our study is focused on eutrophic lakes, where data is rare.*

**L346-end. Note that the range of Hg and MeHg in the seston of this eutrophic lake is on the low end of seston data reported for mesotrophic to oligotrophic North American lakes, both for MeHg concentration and %MeHg. High productivity is not necessarily conducive to abnormally high rates of MeHg accumulation in bioseston. In fact, most data suggest the opposite due to biodilution. It may be true that higher amounts of OM decomposition in eutrophic lakes does indeed exacerbate O<sub>2</sub> depletion and enhance methylation in suboxic water, but that was not measured here. It seems that the most you can say with the data presented here is that the opposing forces of high biodilution and high decomposition need to be reconciled before addressing the impact of climate change. Revise**

*We will revise this section, although it is not clear to us what exactly the reviewer will tell us here. We speculate that there would be much lower MeHg in this lake without high productivity regarding the total amount of MeHg formed in this lake. See answer above regarding biodilution. We want to show the distribution and changes in THg and MeHg concentration and proportion in seston with depth and how they are controlled by redox-conditions and decomposition under eutrophic conditions. How many studies on North American lakes show the distribution of THg and MeHg in seston at high temporal and spatial resolution including redox conditions, Chl a data etc.?.*