

Interactive comment on “Biogeochemical processes accounting for the natural mercury variations in the Southern Ocean diatom ooze sediments” by S. Zaferani and H. Biester

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We would like to thank the reviewer for reviewing our manuscript and constructive comments/questions, which will improve the manuscript. Our responses to comments are shown below.

This paper presents more details of a study on Hg fluxes around Antarctica that was recently published and uses correlation approaches to investigate the factors controlling Hg fluxes. The PCA results are interesting and the conclusions from this support to some degree the authors conclusions about what support the high sediment fluxes measured at this location.

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1) As in the Arctic, based on air-sea exchange literature, the waters are likely a net source of Hg to the atmosphere and not a sink for Hg given the low atmospheric inputs.

This can be region dependent because Hg evasion is not uniform across the oceans. Different factors might have an influence on “considering the waters as a net source or sink for Hg”. The ecosystem setups, which are different in the Arctic and Antarctica, should be considered when comparing these regions. The model study of Soerensen et al., 2016 for the Baltic Sea also suggested that under a low nutrient baseline regime is close to equilibrium between Hg deposition and evasion, but becomes a sink for atmospheric Hg under the high nutrient scenario. Soerensen et al., 2014 give a good overview of this topic and also highlight the role of Hg scavenging by phytoplankton in the atmosphere-ocean cycling of Hg.

2) Contrary to the Arctic, there is low inputs from the terrestrial realm and so the source of the Hg being removed is a critical question to be answered, which the authors do to some degree, but I was not totally convinced.

As we can see from PCA results Hg does not show a correlation with lithogenic elements which should be expected when Hg together with other elements (dust or terrestrial derived) is released during, for instance, ice melting. In addition, the amount of terrestrial material in the sediments has been reported to be very low and sediments are nearly pure diatom ooze with little terrigenous input (Escutia et al., 2011). We regret that we cannot totally convince the reviewer. We see the research of the biogeochemical cycling of Hg in productive remote areas at the beginning and much more work needs to be done until we are convinced to understand what we see.

3) The data are presented in the figures and are not discussed in any detail in the paper and it would be good to have some more discussion of the various figures and why some regions are highlighted. This is in the text at present but not well represented.

The more detailed discussion on elements is presented in Supplementary Information. We moved parts of this information to the main text now.

C2

4) My main criticism is therefore the calculations of Hg removal that is needed to explain the sediment fluxes and the fact that these calculations do not make sense in terms of sources (ln 212 onwards). The paper states that the inputs from lithogenic material is small and I agree with this, and also the atmospheric inputs are small compared to the fluxes. The authors calculate that a few blooms per year scavenging all the Hg out of the water but then state that "Formation of Antarctic bottom water which is linked to the polynyas (Ohshima et al., 2013) can rapidly "refill" the Hg inventory in the water column after a scavenging event." How can the further formation of ABW water, which has its Hg removed because of the previous scavenging events replenish this water column Hg, and what is the timescale for this. To me, this calculation makes no sense. The external inputs of Hg are too small and the authors state that they are removing all the Hg from the water column. So, where is the Hg coming from to replenish. The authors need to make a more convincing argument and calculation to support his idea. I am not convinced, and the calculations just reinforce the idea that it is very difficult to support the calculated Hg fluxes in these sediments given the known water column concentrations and the magnitude of the external inputs.

We are a bit surprised about this comment. On one hand, the reviewer agrees that Hg is not coming together with lithogenic material to the Adélie Basin. On the other hand, the reviewer does not agree with our hypothesis as well as our suggested Hg mass balance, without giving any further suggestion or indication about other possible processes or sources of Hg in our sediments. We also do not understand if the reviewer questions just our suggested mass balance hypothesis/explanation or our data in general (this idea has been previously published in Science and reviewed critically). Hg concentrations in Adélie Basin sediments are actually in a similar range to other studied sediment cores in the oceans. The specific situation at Adélie Basin is that algae blooms are so intense and sedimentation rates are so high (~ 2 cm/year). While algal bloom is a local event, the exchange of water masses could refill the Hg inventory. Scavenging is a continuous process and our model simply provides an explanation of how a Hg mass balance could be obtained (in lakes the strong decrease in Hg concen-

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tration can be observed). The Hg pool in the water column is not static but continuously "refilled" by mixing with water from areas that are not affected by algae blooms (blooms are a local/regional effect). In the real world, scavenging is a continuous process and most intense during and shortly after algae blooms. From our data Hg scavenging by algae is the most likely process explaining the observed high Hg accumulation rates (not concentrations). As the reviewer questions this, what does the reviewer suggest as an explanation for these high Hg sedimentation rates? We used the Hg scavenging hypothesis, which has now been discussed for more than 10 years as a basis for the Hg accumulation rates found in Adélie Basin sediments. Adélie Basin is for sure a situation which cannot be extrapolated to all productive area in the world's ocean. It is here not the question if the scavenging hypothesis is directly supported by our data, it is based on our data the only process which could explain such high Hg accumulation rates. We think that direct proof of this process can only be obtained by sediment trap analysis (we actually found this in lakes).

5) Additionally, the idea that "similar to CO₂, Hg fluxes from the atmosphere will increase during algae blooms as a result of continuous removal of dissolved phase Hg by diatom particles and the resulting shift of the dissolution equilibrium towards the dissolved phase which should additionally increase the Hg flux from the atmosphere into the water" (lns 220-223) is likely not true as based on the data in the various papers, (Wang et al., 2017; Nerentorp et al., 2017a; 2017b) which the authors should read and include their conclusions in their manuscript. Overall, the results of these studies suggest that evasion is actually higher during higher productivity so removing Hg, and is therefore a loss term that the authors don't consider. Also, less ice leads to more evasion. Overall, the data do not support the contention in this section of the paper. The authors need to make a more convincing argument that there are sources of Hg to support the high fluxes they estimate.

This is still a hypothesis and should be the subject of further investigations in the areas with high phytoplankton productivity. We agree that this process alone cannot explain

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the high Hg accumulation in Adélie Basin sediments, but it may at least contribute. We will tune down this argument in the manuscript. Regarding the effect of atmospheric Hg, productivity, and water column Hg depletion see also Soerensen et al., 2014. The mentioned papers (Wang et al., 2017; Nerentorp et al., 2017a; 2017b) did not directly study or discuss the influence of productivity on Hg evasion but measured DGEM and not dissolved Hg. DGEM does not represent total dissolved Hg in the water phase and therefore we think that these results could hardly be used to discuss our findings. Moreover, these papers report significant variations of Hg between different stations. This once again emphasizes that deposition/evasion is not uniform across the ocean and the fate of Hg could be different in different ecosystem setups. Some data in the above-mentioned papers even support our assumptions. For instance, the average water column DGM concentration which reported to be higher in winter and lower in spring (Nerentorp et al., 2017) could be indicators of Hg scavenging by diatom and lower Hg in the water column in spring. Canário et al., 2017 also found different total Hg concentrations in their study. They attributed these differences to the different stages of phytoplankton bloom during the sampling which led to lower dissolved Hg in water in the middle-end stage of the bloom compare to the beginning stage of the bloom, owing to the Hg uptake by phytoplankton. Algal Hg scavenging will decrease seawater Hg concentrations through scavenging and sedimentation that could be reduced and evaded in the absence of algae (Soerensen et al., 2013; Qureshi et al., 2010; O'driscoll et al., 2006). We would like to recommend the paper of Soerensen et al., 2014 which gives a good overview of Hg evasion from the ocean and Hg scavenging at the surface layer. Their data does also suggest the Hg evasion in the ocean is reduced due to Hg scavenging by algae in productive marine areas. We cited the mentioned papers by the reviewer as well as other papers in order to give a better overview of Hg fluxes to and from the ocean. Also, it has been now emphasized that our hypothesis would be the case in regions comparable to the Adélie Basin, where productivity is high and further investigations are required to prove that.

6) I shared the paper with a colleague to help with the review process, who made

C5

additional editorial comments that are highlighted in the attached file.

We thank the reviewer for the editorial comments. In some sections, the reviewer just mentioned "rewrite" without indicating what exactly. We changed the text accordingly where it was possible.

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