Authors Response to Referee 1

We would like to thank professor Lamborg for the objective review of our manuscript and constructive comments/questions, which will improve the manuscript. Our responses to comments are shown below.

An excellent manuscript describing biogeochemical data associated with the accumulation of Hg in silica-rich sediments of the Antarctic margin. The fluxes of Hg are enormous on a per area basis, and if they extent beyond some very narrow band of continental shelf will beg a re-evaluation of sources and sinks in the marine Hg cycle. The data are of high quality and the authors carefully "game out" what the results might mean without straying beyond their dataset. I only have a few very minor presentation comments. These include:

1) To save one figure and facilitate "wiggle matching," the authors could include Hg data in Figures 2, 3, 4 and 5 (and dropping Figure 6).

This is a good suggestion. We have done that.

2) Line 56, "...the re-emission flux of Hg0 from productive regions will be lower..." should be modified with the caveat that all other factors being equal. To be sure, there are some high productivity areas, such as regions of equatorial upwelling, where productivity is high and Hg(0) fluxes to the atmosphere are high as well.

There are different assumptions about Hg behavior in the equatorial upwelling. Some suggest low Hg^0 concentration in seawater due to the removal of Hg by sinking particles and some suggest high Hg^0 and associated evasion due to biological reduction. The availability of Hg in seawater is a factor that controls the magnitude of Hg evasion. Therefore, in upwelling regions, where Hg sedimentation is high (due to enhanced removal of Hg by scavenging and particle sinking), the availability of Hg for reduction and evasion will be low. This hypothesis is also supported in equatorial upwelling regions (Soerensen et al., 2014; Munson et al., 2015). In a study by Soerensen et al., 2014 different regions with different productivity levels showed significantly different concentrations of atmospheric and aquatic Hg^0. Atmospheric and aquatic Hg^0 concentrations were lower in equatorial upwelling regions (with high productivity) compared to northern regions (with low productivity). Mason et al., 2017 also reported lower Hg evasional flux in a region with higher nutrient levels compared to regions with lower nutrients.

However, we have modified this sentence in the manuscript.

3) Materials and Methods-is the age-depth model presented in Escutia et al., 2011? Make clear where this comes from and give a sense of the depth-in-core resolution of the samples in addition to the time resolution already presented.

This information has been added:

Age data and age model (which is based on Compound-specific ^{14}C dating) were obtained from Yamane et al. (2014).

"Samples in the upper core (3.2–25.05 mbsf) were taken at ~ 40 cm intervals (a resolution of ~ 20 years) and ~ 400 cm intervals (a resolution of ~ 200 years) in deeper sections (25.05–170.35 mbsf), resulting in a total of 78 samples."

4) In the last paragraph before Conclusions, the authors contemplate how it is that such large fluxes of Hg could be found, and conclude that it is possible that with 100% scavenging of Hg "below" a diatom bloom on just a few occasions could account for all the Hg observed in the sediments. How about on the other end of the spectrum? If there were no scavenging below the bloom and given our meager knowledge of Hg in diatom concentrations, how much Hg could be buried just by diatom uptake and sinking? Since we are in the early days of thinking about Hg scavenging, having these two bounds on the data might be useful.

This is a very interesting question. But unfortunately, we have analyzed the sediments and we cannot estimate it from our data. But this should be a subject of future researches by studying fresh algae.

5) Interesting to observe that in the PCA, Hg does not appear on the same factors as other chalcophilic elements such as Cu and Zn. Could the authors speculate?

Unfortunately, we have no water column measurements from Adélie Basin, but we speculate that essential elements for diatoms such as Cu and Zn are probably re-released to the water phase during organic matter decay and sinking.

Authors Response to Referee 2

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.

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.

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

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

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

107 *We are a bit surprised about this comment. On one hand, the reviewer agrees that Hg is not coming*
108 *together with lithogenic material to the Adélie Basin. On the other hand, the reviewer does not agree with*
109 *our hypothesis as well as our suggested Hg mass balance, without giving any further suggestion or*
110 *indication about other possible processes or sources of Hg in our sediments. We also do not understand*
111 *if the reviewer questions just our suggested mass balance hypothesis/explanation or our data in general*
112 *(this idea has been previously published in Science and reviewed critically).*

113 *Hg concentrations in Adélie Basin sediments are actually in a similar range to other studied sediment*
114 *cores in the oceans. The specific situation at Adélie Basin is that algae blooms are so intense and*
115 *sedimentation rates are so high (~ 2 cm/year). While algal bloom is a local event, the exchange of water*
116 *masses could refill the Hg inventory. Scavenging is a continuous process and our model simply provides*
117 *an explanation of how a Hg mass balance could be obtained (in lakes the strong decrease in Hg*
118 *concentration can be observed). The Hg pool in the water column is not static but continuously "refilled"*
119 *by mixing with water from areas that are not affected by algae blooms (blooms are a local/regional*
120 *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 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., 2 014.

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 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.

Authors Response to Referee 3

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.

The paper present very important aspect of mercury fluxes in Antarctic region. There are very unique and valuable data. There are very interesting conclusions but firstly

1) (I) "to scavenge all water column Hg." – but this is not directly supported by the data. This is a rare situation that "Hg in the water column is removed through scavenging during diatom blooms" can you prove it? do you know situation that mercury concentration in that region was 0 pg/L? do you know literature about this phenomenon? This is opposite to PCA results where mercury don't correlate with Cp2 appears to reflect phytoplankton blooms and export of biological materials or with Cp1 - load of lithogenic elements.

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). The idea that mercury concentrations become 0 pg/L is too simplified. 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 concentration 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. Scavenging does not only affect Hg but also other elements. For instance, accumulation rates of Pb in our sediments are about 166 times higher than in other pelagic sediments. However, we made this point now clearer in the manuscript. We cited several papers on this topic in the introduction of the manuscript and in particular we 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.

209 *There is a simple reason why Hg does not correlate with biogenic silica (Cp2) in our PCA and this is not*
210 *opposed to our hypothesis. Biogenic silica appears and varies in the percent range and Hg concentration*
211 *range in the lower ppb range (and is the limiting factor based on the amount of Hg in the water column).*
212 *So it cannot be expected that there is a correlation between biogenic silica and Hg. Moreover, that Hg*
213 *does not correlate with lithogenic elements is important information indicates that the Hg source is not*
214 *lithogenic but atmospheric.*

215 2) Cp4 including Hg described 1.35 % of the variance which is marginally. Did you try to make PCA for
216 three factors? I supposed it could be more useful.

217 *We do not understand the meaning of this suggestion. The percentage of variance covered by Hg is of no*
218 *evidence here (it cannot be expected that Hg concentrations control the amount of biogenic silica). Why*
219 *should a PCA with three factors be more "useful" here?*

220 3) (II) Otherwise there are lack of comment whether these Hg values are high, toxic or not? why you
221 didn't account a Vulcanic sources in this region? Describe Hg sources to the water in this region.

222 *Explanation about how toxic are these concentrations is outside the scope of our study and would be*
223 *rather speculative. The concentrations we found (especially in the upper core) are about 2-fold elevated*
224 *compared to the (variable) natural background in the Adélie Basin, but we will not speculate how this*
225 *affects the Antarctic marine food chain.*

226 *Our PCA does not give any indication, that there is a significant volcanic Hg source in this area. Volcanic*
227 *eruptions are usually short-term events and would appear as peaks in Hg records (see e.g. Pérez-*
228 *Rodríguez et al., GCA 2019). The Hg record we found is characterized by periodic-like changes in*
229 *concentrations that cannot be explained by a volcanic source.*

230 4) (III) Why Hg and As correlated (Cp4)? Maybe this is explanation of Hg sources to the sediment?

231 *Up to now, we do not understand this relatively weak correlation. We probably see similar scavenging*
232 *processes here. What does the reviewer suggest regarding the same source of Hg and As in the sediments?*

233 And some specific comments:

234 5) - In the abstract (shortly, at least one sentence) and in the introduction section is lack of information
235 why mercury is so important, why do you research Hg?

236 *We had mentioned in the introduction that Hg studies are of concern due to its toxic effects: "Mercury*
237 *(Hg) is a metal of environmental concern due to its ability to be transported from source to background*
238 *regions (predominantly in the atmosphere) and be transformed into highly bioaccumulative and toxic*
239 *methylated forms." However, we will add a sentence to the abstract: This has been added: "Due to its*
240 *toxic nature and its high potential of biomagnification mercury is a pollutant of concern."*

241 6) - I suggest more detail map of sampling station (Fig. 1)

242 *Thanks for the suggestion but we think the map clearly shows the location of the core in Antarctica.*

243 7) - Describe more detailed preparing the core to analysis: how do you determine the age of the sediment
244 (sediment layers)? The 0-3.2 mbsf is the mixing layer?

245 *The information regarding sample preparation before analysis is given in the Materials and Methods*
246 *section. "All samples were freeze-dried and ground using a glass pestle prior to geochemical analysis."*

247 *We added a reference to where we got the age model: Yamane et al. (2014).*

248 *The 0-3.2 mbsf (meters below seafloor) is not water depth therefore it is not a mixing layer. This indicates*
249 *depth in the sediment core. The core was taken during an IODP ship cruise, extensive data is available*
250 *in the related reports and some published papers.*

251 8) - How do you determine accumulation rates

252 *The accumulation rates were calculated from concentration (ng g^{-1}) and dry mass sediment accumulation*
253 *rate (bulk density of a sediment layer (g cm^{-3}) and the interval represented by the section (cm), divided*
254 *by the time duration (yr)).*

255 9) - Too long title of 3.1 section –

256 *The title has been modified to: Geochemical processes controlling the distribution of the elements in*
257 *sediments.*

258 10) Line 148 "which includes positive loadings of lithogenic elements," – write which one

259 *These elements have been mentioned in the previous paragraph: "The first component (Cp1), explains*
260 *33 % of the variance and shows large (> 0.7) positive loadings of Mn, Ti, Rb, Zr, K, and Y and moderate*
261 *positive loading of Fe."*

262 11) - figure 9 is missing in the text
263 *We referred to the figure in the manuscript.*

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Biogeochemical processes accounting for the natural mercury variations in the Southern Ocean diatom ooze sediments

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Abstract. Due to its toxic nature and its high potential for biomagnification, mercury is a pollutant of concern. Understanding the marine biogeochemical cycle of mercury is crucial as consumption of mercury enriched marine fish is the most important pathway of human exposure to monomethylmercury, a neurotoxin. ~~, which is an extremely toxic form of mercury uptake by humans.~~ However, due to the lack of long-term marine records, the role of the oceans in the global mercury cycle is poorly understood. ~~and w~~We do not have well-documented data of natural mercury accumulations during changing environmental conditions, e.g. sea surface conditions in the ocean. To understand the influence of different sea surface conditions (climate-induced changes in ice coverage and biological production) on natural mercury accumulation, we used a continuous ~ 170 m Holocene biogenic sedimentary record from Adélie Basin, East Antarctica, which mainly consists of silica-based skeletons of diatoms. We performed Principal Component Analysis and regression analysis on element concentrations and corresponding residual~~s of element concentrations~~, respectively to investigate the link between sediment mercury accumulation, terrestrial inputs, and phytoplankton productivity. Preindustrial mercury ~~accumulation~~ in the remote marine basin ~~pristine marine Antarctica~~ show~~ed~~ extremely high accumulation rates (median: 556 µg m^{-2} yr^{-1}) that displayed periodic-like variations. Our analys~~es~~is show~~s~~ that the variations in total mercury concentrations and accumulation rates are associated with biological production and related scavenging of ~~available~~ water phase mercury by rapidly sinking algae or algae-derived organic matter after intense algae blooms. High accumulation rates of other ~~studied~~ major and trace elements further reveal~~ed~~ that in regions of high primary productivity, settling of biogenic materials removes a large fraction of dissolved or particulate bound ~~many other~~ elements from free water phase ~~ocean surface~~ (through scavenging or biological uptake). ~~In conclusion, t~~The link between mercury cycling and primary production will need to be considered in future studies of the marine mercury cycle under ~~future~~ primary production enhancement through climatic, temperature, and nutrient availability changes.

1 Introduction

Mercury (Hg) is a metal of environmental concern due to its ability to be transported through the atmosphere from industrial point source~~s~~ to ~~background~~ remote regions ~~(predominantly in through the atmosphere)~~ and its transformations ~~be transformed~~ into highly bioaccumulative and neurotoxic methylated forms. In the global biogeochemical cycle of Hg, the ocean, as the

311 dominant physical feature of our planet Earth, is of specific concern. A substantial amount of Hg (~ 80 %) which is emitted to

312 the atmosphere from natural and anthropogenic sources reaches the ocean (Horowitz et al., 2017; Schartup et al., 2019) and

313 ocean sediments ~~have been~~are considered ~~as~~ to be ~~an~~the ultimate sink of Hg on a timescale of tens of thousands of years

314 (Fitzgerald et al., 2007; Selin, 2009; Amos et al., 2013). Despite the ~~critical~~ important role of marine sedimentation ~~this sink~~

315 ~~process plays~~ in the global Hg biogeochemical cycle, little is known about the rates or amount of Hg accumulation in marine

316 sediments, especially in the open ocean. In contrast to the well-studied Hg cycling in terrestrial environments, knowledge

317 about the temporal and spatial distribution of Hg in the marine environment is limited to model estimations (Mason and Sheu,

318 2002; Sunderland and Mason, 2007)~~, and~~ water column measurements (Cossa et al., 2011; Lamborg et al., 2014; Canário et

319 al., 2017), and very few sediment measurements (Kita et al., 2013; Aksentov and Sattarova, 2020). A main reason for our

320 limited understanding of the fate of Hg in the oceans is the lack of high resolution marine sedimentary records, especially from

321 the deep ocean (Zaferani et al., 2018).

322 Hg input to the ocean is primarily through atmospheric deposition (Mason et al., 1994; Driscoll et al., 2013). After deposition,

323 as either mercuric ion (Hg^{2+}) or elemental Hg (Hg^0), Hg can be ~~(i)~~ reduced to Hg^0 and evaded to the atmosphere, or ~~(ii)~~

324 scavenged from the water column by particulate matter and eventually buried in deep-sea sediments~~, or (iii) methylated to~~

325 ~~either monomethylmercury (CH₃Hg⁺) or dimethylmercury ((CH₃)₂Hg)~~ (Mason et al., 2012; Lamborg et al., 2014). ~~It is~~

326 ~~estimated that almost 96 % of the deposited Hg to the ocean is lost through evasion from the surface, and only 30 % of the Hg~~

327 ~~flux that reaches the deep ocean is preserved in sediments (Mason and Sheu, 2002).~~ Most marine surface waters are believed

328 to be close to equilibrium between Hg deposition and evasion or saturated in Hg^0 due to biologically mediated (Mason et al.,

329 1995; Rolfhus and Fitzgerald, 2004; Whalin et al., 2007) and photochemical ~~processes~~reduction (Amyot et al., 1997; Mason

330 et al., 2001) followed by re-emission of Hg^0 to the atmosphere. It ~~is~~has been estimated that almost 96 % of the deposited Hg

331 to the ocean is lost through evasion from the surface, and only 30 % of the Hg flux that reaches the deep ocean is preserved in

332 sediments (Mason and Sheu, 2002). However, other studies indicate that the ocean surface waters become a sink for

333 atmospheric Hg at the high nutrient levels and related high primary productivity (Soerensen et al., 2016).

334 There are contradictory statements about the Hg deposition and evasion to/from different surface waters. Early works suggest

335 that Hg evasion is high in productive upwelling regions of the ocean due to enhanced biological reduction (Fitzgerald et al.,

336 1984; Mason and Fitzgerald, 1993). More recent studies, with higher spatial and temporal resolutions, suggest lower Hg

337 evasion in productive regions (O'Driscoll et al., 2006; Qureshi et al., 2010; Soerensen et al., 2013; Soerensen et al., 2014).

338 Measurements of Hg from these recent studies show relatively low concentrations of atmospheric and surface water phase Hg^0

339 in regions with high productivity compared to areas with lower productivity. These studies related their observation to sorption

340 and scavenging of Hg by suspended organic particles. They suggested that removal of Hg^{2+} associated with sorption and

341 scavenging by suspended organic particles in productive regions reduces the amount of available Hg^{2+} for reduction and re-

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342 emission. ~~However~~Therefore, ~~available data sets on Hg fluxes to the deep ocean and accumulation rates in deep ocean~~
343 ~~sediments are limited and~~ applying the model estimates across the entire ocean introduces substantial uncertainty and ~~.O~~one
344 area in particular that highlights this uncertainty is the underestimation of the role of biological productivity as a major vector
345 of Hg sedimentation in the oceans.~~in the global Hg cycle.~~

346 The marine biogeochemical cycle, especially sedimentation of many elements (Fowler and Knauer, 1986; Morel and Price,
347 2003; Schlesinger and Bernhardt, 2013), including Hg (Kita et al. 2013; Lamborg et al., 2016; Zaferani et al., 2018), in ~~seawater~~
348 the ocean is controlled directly and indirectly by biological productivity. Biogenic particles control the distribution of elements
349 through primary production, sinking, and decomposition (Fowler and Knauer, 1986). Besides direct uptake across cell
350 membranes through active metabolism, phytoplankton and sinking biogenic particles can scavenge and remove many other
351 elements from the dissolved phase and transport them to the deep sea. Sinking speed of biogenic particles plays an important
352 role in the final fate of those elements. Rapidly sinking particles such as diatom agglomerates transfer elements to the deep sea
353 (Fowler and Knauer, 1986; Smetacek et al., 2012), whereas elements associated with the slowly sinking particulates will
354 release back to the water phase through remineralization (Fowler and Knauer, 1986). In areas where pronounced seasonal
355 blooms take place, phytoplankton species appear to reach the deep-sea floor relatively fast and intact. Seasonal blooms in the
356 surface waters will also cause temporally variable fluxes of elements in the deep ocean (Fowler and Knauer, 1986; Michel et
357 al., 2002; Pilskaln et al., 2004).~~Regions of high biological productivity play an important role in the downward transport and~~
358 ~~burial of biologically essential and nonessential elements in the sediments of the deep sea (Schlesinger and Bernhardt, 2013).~~

359 For Hg, ~~T~~these ~~assumption~~findings ~~for Hg is~~are supported by water column ~~measurements~~(Lamborg et al., 2014) and marine
360 sediment measurements (Kita et al., 2013; Aksentov and Sattarova, 2020). Lamborg et al. (2014) described a nutrient-like
361 distribution of Hg in the water column of oceans. This study indicates that similar to carbon (C) and phosphorus (P), Hg shows
362 higher concentrations in the deep water due to its release during organic matter decomposition. Kita et al. (2013) found a
363 positive correlation between Hg and the absolute abundance of phytoplankton species in sediments of the Caribbean Sea. Hg
364 in these sediments was assumed to be a result of Hg binding by phytoplankton depositing Hg-bearing organic matter in the
365 photic zone. A similar conclusion was reached by Aksentov and Sattarova (2020) who used a thermo-scanning technique to
366 detect Hg forms. They found that biological productivity controlled the Hg burial in Northwest Pacific bottom sediments and
367 that the forms of Hg depended on the diatom content.

368 These observations ~~This~~ can be due to Hg—phytoplankton interactions and ~~taking~~ up-take or binding of Hg from the water
369 ~~phase~~ by phytoplankton (Le Faucheur et al., 2014; Mason et al., 1996). This interaction controls the flux of Hg from the water
370 column to sediments and facilitates the downward flux of Hg to the seafloor (Soerensen et al., 2014; Soerensen et al., 2016;
371 Lamborg et al., 2016; Zaferani et al., 2018), which, as mentioned, has traditionally been considered to be slow in its nature.

the flux of Hg from the water column to sediments in several ways. removal of dissolved Hg from the water column by algae and other particulate matter facilitates the downward flux of Hg to the seafloor (Lamborg et al., 2016), which, as mentioned, has traditionally been considered to be slow in its nature. Scavenging of Hg from surface water by particulate organic matter reduces the availability of Hg^{2+} for reduction to Hg^0. Therefore, the re-emission flux of Hg^0 from productive regions will be lower (Soerensen et al., 2014; Soerensen et al., 2016; Zaferani et al., 2018). Second, Hg scavenging by algae removes Hg from the dissolved phase and may shift the flux of Hg between the atmosphere and the ocean towards the dissolved phase by changing the dissolution equilibrium (Biester et al., 2018; Zaferani et al., 2018). Third, removal of dissolved Hg from the water column by algae and other particulate matter facilitates the downward flux of Hg to the seafloor (Lamborg et al., 2016), which, as mentioned, has traditionally been considered to be slow in its nature. Thus, underestimating the role of biological productivity in the marine biogeochemical cycle of Hg may lead to an overestimation of re-emission fluxes from surface water and underestimates an underestimation of the Hg flux to deep-sea sediments.

Commented [C6]: **Reviewer2:** This can be condensed

In this context, the Southern Ocean is of particular interest due to its high concentrations of nutrients and related elevated primary productivity (Arrigo et al., 1998). In the Southern Ocean, diatoms are major primary producers (Crosta et al., 2005). Their siliceous cell walls preserve well in sediments and form diatom ooze (Fütterer, 2006). The sedimentation rate of diatom ooze is high, estimated to reach up to 2 cm yr^{-1} (Escutia et al., 2011),. This high sedimentation rate makes making diatom ooze deposits around Antarctica a unique geochemical archive to study the influence of primary productivity as well as natural and anthropogenic changes on the marine biogeochemical cycle of Hg.accumulation of Hg as well as to entangle changes in the natural and anthropogenic marine biogeochemical cycle.

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Despite providing a unique geochemical archive, studies on Hg cycling in the Southern Ocean, and particularly in the Antarctic region, are generally limited to water column (Cossa et al., 2011; Nerentorp Mastromonaco et al., 2017b; Canário et al., 2017), surface water/air (Nerentorp Mastromonaco et al., 2017a; Wang et al., 2017), and ice core analyses (Vandal et al., 1993). Cossa et al. (2011) showed a nutrient-like distribution of Hg that Hg concentrations in water column that ranged between 0.6 and 2.8 pmol L^{-1}. in open water, 1.15 ± 0.22 pmol L^{-1} in Antarctic Intermediate Water, and 1.35 ± 0.39 pmol L^{-1} in Antarctic Bottom Water between the Antarctic continent and Tasmania. The observed variations in the vertical distributions of Hg were attributed to air-sea exchange and the affinity of Hg to bind to planktonic and inorganic particulate matter in the biologically productive zone. Nerentorp Mastromonaco et al. (2017b) found higher total Hg concentration than Cossa et al. (2011) with no significant vertical variations. Both studies reported seasonal variations in Hg concentrations and related them to seasonal variations of atmospheric Hg deposition (Cossa et al., 2011; Nerentorp Mastromonaco et al., 2017b) as well as the Hg inputs from melting sea ice and snow (Nerentorp Mastromonaco et al, 2017b). Total Hg concentrations in the Atlantic sector of the Southern Ocean obtained during a study by Canário et al. (2017) were also, in general, comparable to those obtained by Cossa et al. (2011) except for some stations that showed higher total Hg concentrations. Canário et al. (2017) attributed these differences to the

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different stages of phytoplankton bloom during the sampling. This 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 (Canário et al. 2017). Measurements of gaseous elemental mercury (GEM) and dissolved gaseous mercury (DGM) in surface water showed spatial and seasonal variations as well (Nerentorp Mastromonaco et al., 2017a; Wang et al., 2017). These studies related the increase of DGM and GEM concentrations to the presence and absence of sea ice. Sea ice that could prevent Hg evasion to the atmosphere, could initially lead to an increase in Hg emissions to the atmosphere when diminishing. Hg concentrations in an ice core, covering the past 34 Kyr, varied between 0.0005 and 0.0021 µg kg^{-1}, corresponding to depositional fluxes of 0.009 and 0.031 µg m^{-2} yr^{-1} during the Holocene and the Last Glacial Maximum, respectively (Vandal et al., 1993). Vandal et al. (1993) attributed the observed enhanced Hg flux during colder periods to marine biological productivity and emissions of volatile Hg compounds from the ocean. Given the different results of the existing studies point to the gaps in our understanding of Hg behavior in productive remote areas which warrants further investigation in the Southern Ocean.

In a previous paper, we discussed the accumulation of anthropogenic Hg in ~~Adélie Basin~~ sediments of Adélie Basin, offshore East Antarctica. The ~ 2-fold increase in Hg concentrations and accumulation rates in the upper ~ 2.80 m depth of the core was attributed to the onset of the industrial revolution and the strong increase in coal burning at ~ 1850 CE (Zaferani et al., 2018). Here, we discuss the natural processes (e.g. changes in biogenic and terrestrial material fluxes) that controlled Hg accumulation in the same sediment core prior to 1850 CE throughout the past 8600 years. We investigated ~~the~~ a continuous ~ 170 m long Holocene laminated diatom ooze sediment record from the Adélie Basin ~~offshore East Antarctica~~. Covering almost the entire Holocene, the core allows the determination of natural variations ~~in~~ of Hg accumulation rates in these sediments prior to major anthropogenic influences. Our main objective was to investigate the influence of different Hg sources, climate-induced changes in biological productivity and terrestrial fluxes (through melting of glacier ice), which have controlled the sequestration of Hg in these sediments. To evaluate the influence of different biogeochemical processes on the Hg accumulation in sediments, with an emphasis on the role of changes in planktonic productivity, we combined the data on Hg accumulation with data derived from multi-element analyses.

2. Materials and methods

2.1 Study site and core collection

Sediments of the Adélie Basin were collected during the Integrated Ocean Drilling Program (IODP) Expedition from ~~the~~ hole U1357B 318 in 2010. ~~The hole~~ U1357B is located on the continental shelf off Wilkes Land at the Mertz Glacier polynya (region of open water surrounded by sea ice), Antarctica (66°24.7990′ S, 140°25.5705′ E) at about 1021.5 m water depth

432 (Escutia et al., 2011) (Fig 1). The total length of the recovered core is 170.7 m, corresponding to nearly the entire Holocene

433 (Escutia et al., 2011). The core was sliced by 5cc plastic scoops to (1 cm slices~~wide samples~~). Samples in the upper core (3.2–

434 25.05 mbsf) were taken at ~ 40 cm intervals (a resolution of ~ 20 years) and ~~at~~ ~ 400 cm intervals (a resolution~~s~~ of ~ 200 years)

435 in deeper sections (25.05–170.35 mbsf), resulting in a total of 78 samples. Age data and age model (which is based on

436 Compound-specific ^{14}C dating) were obtained from Yamane et al. (2014).

437 The sediment core is characterized by light and dark laminations which are undisturbed by sea-level changes or glacial erosion

438 (Denis et al., 2006; Escutia et al., 2011). Light laminations correspond to spring seasons when light and high nutrient levels

439 promote intense phytoplankton blooms and are mainly composed of biogenic materials~~, i.e.~~ (mostly diatom with minor

440 abundance of silicoflagellates, sponge spicules, radiolarians, and foraminifers). ~~Whereas d~~Dark layers correspond to the

441 summer/autumn season when sea ice has retreated, and nutrient levels are low. Dark laminations are composed of a mixture

442 of biogenic and terrigenous materials resulting from summer production in open water, with ~~and~~ glacial and subglacial inputs,

443 respectively. High levels of primary production in surface water of this region, coupled with rapid fluxes of biogenic debris,

444 directly to the seafloor, led to high sedimentation rates of up to 2.0 cm yr^{-1} during the past 10,000 years (Escutia et al., 2011).

445 **2.2 Analyses of mercury and major and trace metals**

446 All samples were freeze-dried and ground using a glass pestle prior to geochemical analysis. Total Hg was determined by

447 thermal decomposition followed by pre-concentration of Hg on a gold trap and CVAAS Hg detection using a Milestone DMA-

448 80 analyzer (US EPA Method 1998). The quality of the analysis was ensured by including a certified reference material (CRM)

449 (Canmet LKSD-4 = 190 ± 17 ng g^{-1}) alongside the analyzed samples. The average measured concentration for LKSD-4 was

450 197 ± 11 ng g^{-1}. Replicate analyses (n = 20) were always within an RSD of 10 % of the certified value.

451 The samples were analyzed for concentrations of silicon (Si), titanium (Ti), zirconium (Zr), sulfur (S), calcium (Ca), potassium

452 (K), aluminum (Al), yttrium (Y), manganese (Mn), strontium (Sr), iron (Fe), lead (Pb), copper (Cu), zinc (Zn), arsenic (As),

453 bromine (Br), nickel (Ni), chlorine (Cl), and rubidium (Rb) by ~~means of~~ energy dispersive X-ray fluorescence (ED-XRF). The

454 calibration method, accuracy, and precision are described in detail in Cheburkin and Shotyk (1996). The CRMs (Canmet

455 LKSD-4, NRC/CNRC-PACS-2, NRC/CNR-Mess-3, and NCS-DC75304) and replicates were measured in each set of samples

456 for accuracy and precision control. Repeated analysis of CRMs gave relative standard deviation (SRD) less than 10 % for Si,

457 Al, Ca, Y, Sr, Zr, Br, and Rb, 6–15 % for Ti, K, Zn, S, Fe, Mn, and Pb, 6–19 % for Cl, 10–20 % for Ni, 9–14 % for Cu, and

458 14–22 % for As.

2.3 Statistical analyses

Principal component analysis (PCA) was applied to the major and trace element concentrations to identify processes controlling the variability of elements in the sediments. When there is a complex set of variables, PCA is used to reduce a large number of variables to a new set of artificial variables, called principal components. Each component includes variables with a similar down core pattern. The principal components are then interpreted in terms of relevant geochemical processes that can control the variability of the major and trace elements in the sediments. The derived interpretation from PCA was then combined with the Hg data to examine the processes that could affect Hg accumulations. The analysis was performed on the standardized concentration data using Z-scores (expressed in terms of standard deviations from their means).

Regressions analysis of the corresponding residuals was used, too to establish the important elemental relationships between the abundance of elements, by considering Si concentration as an independent variable and other element concentrations as dependent variables. Correlation analysis and PCA were performed using the statistical software SPSS 25.0.

3. Results and discussion

3.1 Elemental composition of the diatom ooze sediments and geochemical processes controlling their distribution Geochemical processes controlling the distribution of the elements in the sediments

Concentration profiles and accumulation rates of Si, Al, K, Ti, S, Ca, Zn, Fe, Br, As, and Cl are shown in Fig. 2-5. and discussed in supplementary materials. The preindustrial geochemical record of Adélie Basin sediments is generally characterized by periodic-like variations in the relative abundance of major and trace elements. The records of element accumulation rates largely follow those of periodic-like variations of concentrations and show no significant trend with depth (except Cl).

Si has the highest concentration of all elements in the sediments. On one hand, Si is associated with the flux of terrestrial derived mineral components and on the other hand with siliceous phytoplankton, protozoans, protists, plant phytoliths, and sponge spicules (Croudace and Rothwell, 2015). Si is mainly biogenic in origin in Adélie Basin sediments, dominated by diatoms (Escutia et al., 2011), and contribution of terrigenous Si is low. Therefore, it is used as a proxy for diatom abundance. The record of Si concentrations shows periodic-like variations by a factor of ~ 2 between 21 % and 50 %, with a median of 33 %, corresponding to 70 % SiO_2 or biogenic silica. Concentrations of Al, K, and Ti (as indicators of changes in the flux of lithogenic materials) range between ~ 1.6–7.3 %, ~ 0.37–1.11 %, and ~ 716–1778 mg kg^{-1}, respectively. S and Ca concentrations, which are associated with the biogenic productivity, vary between ~ 0.13–0.87 % and 0.72–1.49 %, respectively. Ca concentration indicates that calcite producing microorganisms are of minor importance in the Adélie Basin. Concentration of Zn, an important micronutrient for marine phytoplankton (Morel et al., 1994), ranges between ~ 96 and 216 mg kg^{-1}. Fe is another essential micronutrient for marine primary production (Smetacek et al., 2012) and biochemical processes

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488 of phytoplankton such as photosynthesis, respiration, and nitrogen fixation (Lohan and Tagliabue, 2018). Concentration of Fe
489 varies between ~1.05 and 3.46 %, which is similar to other siliceous sediments but lower than the reported concentration in
490 other ocean sediments (Chen et al., 1996). Fe concentrations increase at 66.45 m depth to the top of the core by a factor of ~
491 1.6 (from a median of ~ 1.50 % below the 66.45 m depth to ~ 2.40 % above the 66.45 m depth). This is attributed to the upward
492 transport of Fe under anoxic conditions. Chlorine was found to be another major component in these sediments. Cl can go
493 through biological pathways (incorporation into algae) and reach the sediments by the fast-sinking detritus (Leri et al., 2015).
494 Concentrations vary between ~ 1.3 and 19 %, with a median of median 4.5 %, and show a decrease from the top to the bottom
495 of the core which is likely attributed to the increasing mineralization of organic matter with age and the release of chloride
496 through reductive dechlorination.

497 The PCA resulted in five components, explaining almost 82 % of the total variance (Table 1). The first component (Cp1),
498 explains 33 % of the variance and shows large (> 0.7) positive loadings of Mn, Ti, Rb, Zr, K, and Y and moderate positive
499 loading of Fe. The second component (Cp2), which explains 20 % of the variance, is characterized by large positive loadings
500 of Al, Si, S, and Cl and moderate positive loading of K and Ca. The third component (Cp3) explains 17 % of the variance and
501 shows large positive loadings of Zn, Cu, and Ni and moderate positive loading of Fe. The fourth and fifth components (Cp4
502 and Cp5) account for 7 and 5 % of the variance, respectively. Cp4 is characterized by high positive loadings for Hg and As
503 and moderate negative loading of Pb. Cp5 shows positive loadings for Sr and Ca.

504 In general, results of PCA imply that opening and closing of the polynya and biological production are the most important
505 factors influencing sedimentation in the Adélie Basin. This has been shown by loadings of elemental proxies for terrigenous
506 and biological material inputs. Briefly, Cp1, which includes positive loadings of lithogenic elements, represents the variability
507 of terrigenous inputs. Melting of ice releases trapped lithogenic material into the water and leads to the sinking of lithogenic
508 particles and their sedimentation. Cp2 comprises loadings of elements of both biogenic and terrigenous sources. This
509 component appears to reflect phytoplankton blooms and export of biological materials. After ice melt, when the ice is opening,
510 favorable conditions for biological productivity lead to phytoplankton blooms and export of biogenic materials to the seafloor
511 (Denis et al., 2006). Biogenic material is mainly ~~biogenic silica~~opal because diatoms are a major component of blooms in the   **Commented [C11]: Reviewer2:** Reword
512 Adélie Basin (Escutia et al., 2011). The sinking of diatoms from the surface and their sedimentation can cause scavenging of
513 elements during bloom time. Al shows positive loadings in this component rather than in Cp1. This, other than association
514 with the flux of aluminosilicates material can also be attributed to the scavenging of Al by diatom particles (Moran and Moore,
515 1992). Cl also shows loading in Cp2. The possible explanation for the observed covariation is that marine phytoplankton is
516 rich in polyunsaturated lipids and can account as chlorination substrates (Leri et al., 2015). However, the organic C content of
517 Adélie Basin sediments is generally low (between 1 and 2 wt %), and we hypothesis that some of the Cl must be in an inorganic
518 form trapped in sediments owing to high sedimentation rates. Cp3 is mainly characterized by elements that are associated with

the organic fraction of diatom cells. This component appears to reflect the remineralization process like decomposition of organic particles during sinking. Trace elements associated with organic parts of cells can be released back into the water column during decomposition. Therefore, cellular locations of elements, i.e. opal frustules of diatoms or organic matter of diatom cells, created different components of Cp2 and Cp3. Cp4 consists of organic particle reactive metals, e.g. Hg and Pb. The possible explanation for not having these two particle reactive metals in Cp2 is that some of these two metals begin to enter the system after ice melting, while Cp2 shows scavenging of elements by diatoms that are already present in the water column. Since Pb and Hg are negatively correlated, this component cannot reflect a pollution signal. The reason for that could be the characteristic of Pb that decreases its impact in remote areas such as Antarctica. Atmospheric Pb is associated with particles and therefore is rapidly removed by wet and dry deposition. Moreover, Southern Ocean circumpolar vortex that isolates Antarctica from the other continental landmasses in the Southern Hemisphere will further limit Pb transport to Antarctica. Covariation of Ca and Sr in Cp5 represents sedimentation of planktonic foraminifera, which appears to be of minor importance here.

Cp1 explains 33 % of the variance and accounts for much of the variability/process which controlled the geochemical composition of these sediments. However, the high concentration of Si and low concentrations of terrigenous elements imply that in an environment like Adélie Basin, with extremely high productivity, input of lithogenic materials is changing while different diatoms taxa are always present in the system, e.g. as sea ice-associated and open ocean diatom (Escutia et al., 2011).

It has been shown before that seasonal blooms in ocean surface waters result in temporary variable fluxes of elements to the deep ocean (Fowler and Knauer, 1986; Michel et al., 2002; Pilskaln et al., 2004). Although the core was not sampled at a one-year resolution in our study, we suggest that the fluctuations of elemental concentrations are likely related to the seasonal blooms and variation of sea surface conditions like ice melting and freezing and its subsequent biological or terrestrial materials exports. Sampling in light (associated with spring seasons) or dark (associated with summer/autumn seasons) laminae, which contains different amounts of biogenic or terrestrial materials can cause the observed variations (see Sect. 2.1).

The element concentrations are comparable to other published sediment data, while the accumulation rates are much higher than other reported values. The existence of rapidly settling particles in the Adélie Basin can explain the high element accumulation rates. When nonessential elements and essential elements show high accumulation rates, it is tempting to suggest that The high accumulation rates suggest that most elements in the water column of Adélie Basin are subjected to removal by intense phytoplankton blooms through consumption or scavenging. This agrees with the study of Fowler and Knauer (1986) that demonstrated the role of large particles in the transport of elements through the oceanic water column. Aggregation of diatoms, which creates large particles (Turner, 2015), and their sinking to the seafloor and can create a space in which elements

548 can be trapped (Shanks and Trent, 1979). This enhances removal of elements from the water column and their sedimentation
549 as well.

3.2 Holocene record of mercury concentrations and accumulation rates

551 In the preindustrial period, i.e. from the bottom of the core at ~ 170 m to 2.80 m depth (8600 years ago to ~ 1850 CE), the Hg
552 record shows no obvious trend with depth but rather periodic-like variations. Hg concentrations fluctuate by a factor of about
553 2 between 12.6 and 21.1 µg kg^{-1} within 170–137 m depth and between 21.7 and 44.6 µg kg^{-1} within 137–2.80 m depth of the
554 core, with two more pronounced peaks at around 9.99 and 8.20 m depth (Fig. 2 and 3). The lower concentration of Hg within
555 170–137 m depth of the core is probably attributed to the cooler conditions in Adélie Basin (Crosta et al., 2007) and sea ice
556 cover during this period. Hg accumulation rates in the preindustrial period (Fig. 4 and 5) largely follow the Hg concentration
557 record, with periodic-like variations and a median of 556 µg m^{-2} yr^{-1}, which largely surpass the reported Hg deposition rates
558 to the oceans (Mason and Sheu, 2002).

559 The high preindustrial Hg accumulation rates in Adélie Basin sediments cannot be explained by preindustrial atmospheric Hg
560 depositions alone, which did not exceed 20 µg m^{-2} as recorded in an Antarctic ice core (Vandal et al., 1993). Therefore, non-
561 atmospheric sources, such as dissolved water phase Hg or terrestrial inputs, are needed for the observed high Hg enrichment
562 in these sediments.

563 To identify driving forces behind the variations in Hg accumulation we used our PCA results. PCA demonstrated that two
564 main processes, i.e. biogenic productivity and lithogenic inputs, controlled the flux of elements to Adélie Basin sediments.
565 The component scores, which illustrate the depth dependency of the extracted components, are characterized by see-saw
566 patterns throughout the entire core. This indicates different contributions of biogenic and terrigenous inputs most likely
567 associated with spring and summer/autumn seasons, respectively.

568 The variance of Hg was not captured by Cp1, Cp2, or Cp3. Hg instead forms a group on Cp4 together with positive loading of
569 As and negative loading of Pb. The absence of significant loading of Hg on Cp1, Cp2, and Cp3 (Table 1) and the lack of
570 similarity between component scores and Hg concentrations (Fig. 7 6 and 8 7), in the preindustrial period, indicates that Hg
571 fluxes are not significantly influenced by changes in lithogenic inputs through ice melting. These results further indicate that
572 changes in the contribution of biogenic material also do can not directly explain the variation of in Hg accumulation in the
573 sediments.

574 Although the high Hg accumulation rates observed in the Adélie Basin sediments could not be explained by atmospheric Hg
575 deposition, but we expect that in the Adélie Basin the Hg flux from the atmosphere will probably increase during algae blooms

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576 attributed to the removal of the dissolved Hg phase through Hg scavenging by algae. Hg removal from the upper water column

577 by diatom organic matter will also likely decrease Hg re-evasion to the atmosphere as previously assumed in model studies

578 (Soerensen et al., 2014; Soerensen et al., 2016).

579 We have hypothesized that the high Hg enrichment in Adélie Basin sediments has been caused by scavenging of dissolved

580 water phase Hg by a large amount of fast sinking algal debris. To proof this hypothesis we To calculated the maximum amount

581 of Hg which could be at maximum scavenged by a single bloom event we useding the Hg concentration of 271 ± 78 pg L^{-1} in

582 Antarctic Bottom Water, as suggested determined by Cossa et al. (2011). The amount of Hg in aA water column of one m^2

583 and 1000 m depth would then amount to 271 ± 78 µg. This means that only about 2-3 algae blooms and scavenging events per

584 year are necessary to obtain the average Hg accumulation rate in Adèlie Basin diatom ooze sediments, i.e. 556 ± 137 µg m^{-2}

585 yr^{-1}. This appears to be likely taking into account that bloom events are frequent during Antarctic summer and that the sinking

586 speed of diatom agglomerates at Adélie Basin is high has been reported to reach (100-400 m per day) (Jansen et al., 2018).

587 Formation of Antarctic bottom water which is linked to the polynyas (Ohshima et al., 2013) The annual cycle of water mass

588 transformation beneath the Mertz Glacier polynya system (Williams et al., 2008) and the exchange of water masses can rapidly

589 "refill" the Hg inventory in the water column after a scavenging event. While algal bloom is a local event at the surface water

590 of the Adèlie Basin, the exchange of water masses, which have not been affected by algae blooms, could refill the Hg inventory

591 in the water column (Fig. 8).This calculation suggests the high Hg accumulation rates in the Adélie Basin sediments can be

592 solely explained by scavenging of water column Hg and does not need an additional atmospheric or terrestrial source.

593 Nevertheless, we assume that similar to CO$_2$, Hg fluxes from the atmosphere will increase during algae blooms as a result of

594 continuous removal of dissolved phase Hg by diatom particles and the resulting shift of the dissolution equilibrium towards

595 the dissolved phase which should additionally increase the Hg flux from the atmosphere into the water. Hg removal from the

596 upper water phase by diatom organic matter will also likely decrease Hg re-evasion to the atmosphere as previously assumed

597 in a model study (Soerensen et al., 2016).

598 The main reason for not finding any statistical relation between Hg and biogenic materials is that the amount of algal material

599 during algae blooms is always large and therefore not a limiting factor for the scavenging of Hg. There has always been excess

600 algal material within or passing through the water column to scavenge all water column Hg. Thus, we assume that nearly all

601 Hg in the water column is removed through scavenging during diatom blooms, but that Hg scavenging events do not or less

602 frequently occur less frequently during winter and summer/autumn seasons when primary productivity is lower and open ice

603 expansion is at its maximum. Similar to other elements, the periodic-like variations observed in the preindustrial Hg record.

604 The influence of periodically climatic changes of phytoplankton activity on periodical changes of Hg content was suggested

605 before for the Caribbean Sea (Kita et al., 2013). It is likely, that the periodic-like variations observed in the Adèlie Basin Hg

606 record are also likely attributed to the seasonal export of biological materials.different contents of biogenic and terrestrial

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607 materials, attributed to the sampling variability at seasonal scales and laminae, which have different content of biogenic and
608 terrestrial materials. This can affect Hg concentrations. There is also evidence, based on Hg levels in water (Canário et al.
609 2017) that relevant to different development stages of a phytoplankton bloom, different amounts of dissolved Hg can be taken
610 up by phytoplankton. However, investigation at seasonal resolution is needed to further confirm our observations and
611 hypotheses.

4. Conclusions

613 Investigation of Antarctic biogenic bottom sediments revealed that biological productivity and related scavenging of water
614 phase Hg by rapidly sinking algae or algae-derived organic matter controlled preindustrial Hg accumulation in Adélie Basin,
615 Antarctica. Our study suggests that the periodic-like variations in total Hg concentrations and accumulation rates are likely
616 associated with the opening and closing cycle of polynya opening and closing and its related changes in biological productivity.
617 Although the high Hg accumulation in diatom ooze does not represent the Hg sedimentation process across all the world's
618 oceans, our data shows that Hg scavenging by algae or fast sinking algae-derived organic matter is a key process controlling
619 the sequestration of Hg in marine sediments; and therefore the marine biogeochemical cycle of Hg in general. This can be
620 similar to the association between very high benthic organic C fluxes with diatom production at the surface water which can
621 be accelerated by aggregation (Sachs et al., 2009).

622 Our observations also suggest that re-emission of Hg from ocean surface waters as a result of reduction of Hg (II) might be
623 reduced due to Hg scavenging by algae, at least in areas of high primary productivity ecosystems comparable to Adélie Basin
624 ecosystem setup. Moreover, Hg fluxes to marine sediments might be higher than previously assumed in global model
625 estimations. More data from marine sediments is needed to support this assumption. A future increase in marine productivity
626 including algae blooms especially in coastal areas and semi-closed shallow seas will likely increase the Hg flux to bottom
627 sediments. The model study of Soerensen et al. (2016) for the Baltic Sea might serve as an example for changes in marine Hg
628 cycling caused by eutrophication. The area of the seafloor which is covered by remains of calcareous phytoplankton needs to
629 be considered in future studies as well owing to the role of calcareous species on Hg content (Kita et al., 2013).

630

631 *Author contributions.* S. Zaferani carried out the analyses. H. Biester planned the study and supervised the findings of this
632 work. The manuscript is written by both authors.

633 *Competing interests.* The authors declare that they have no conflict of interest

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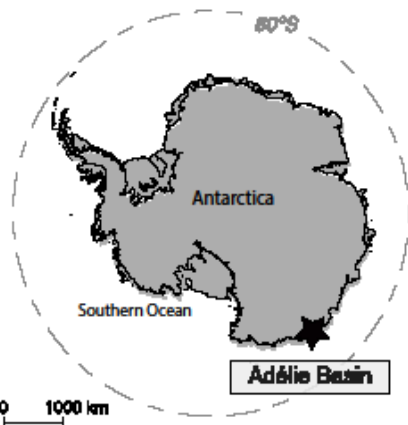
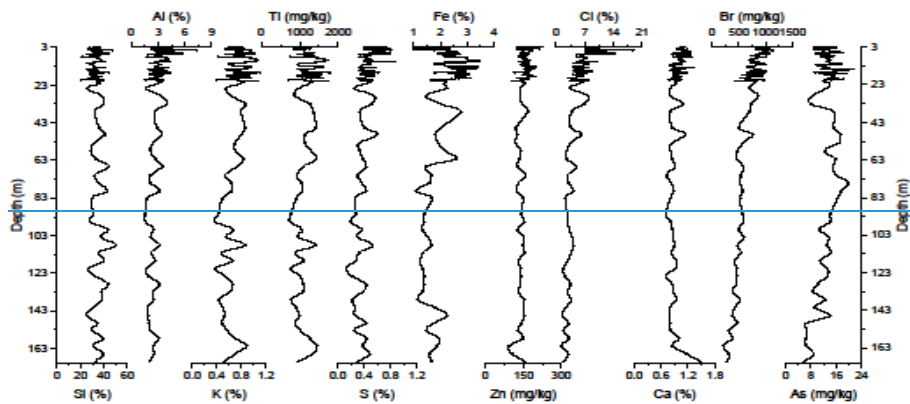
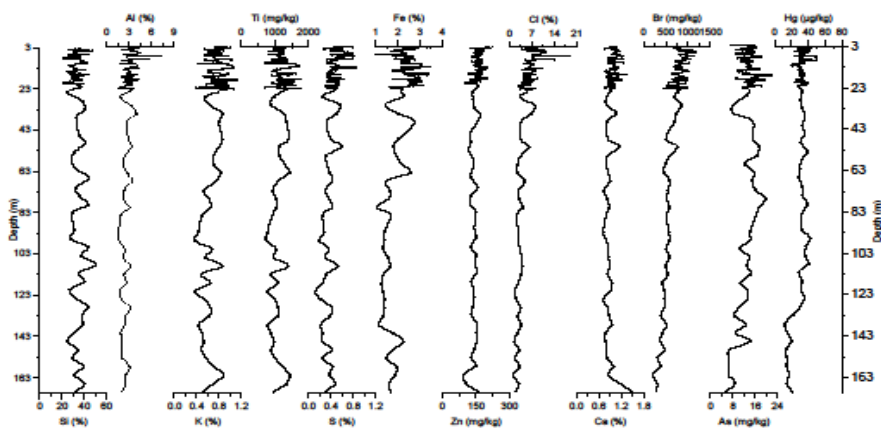
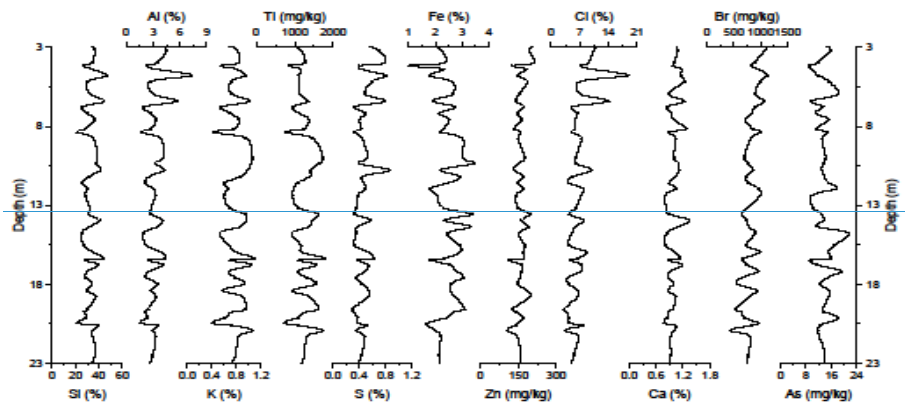

792

793 Figure 1: Map of Antarctica with the coring location of the IODP318-U1357B in Adélie Basin (Source: figure modified from
794 Zaferani et al. (2018)).

795

796

797 Figure 2: Down core records of Si, Al, K, Ti, S, Fe, Zn, Cl, Ca, Br, As, and ~~As~~ Hg concentrations of Adélie Basin sediments.

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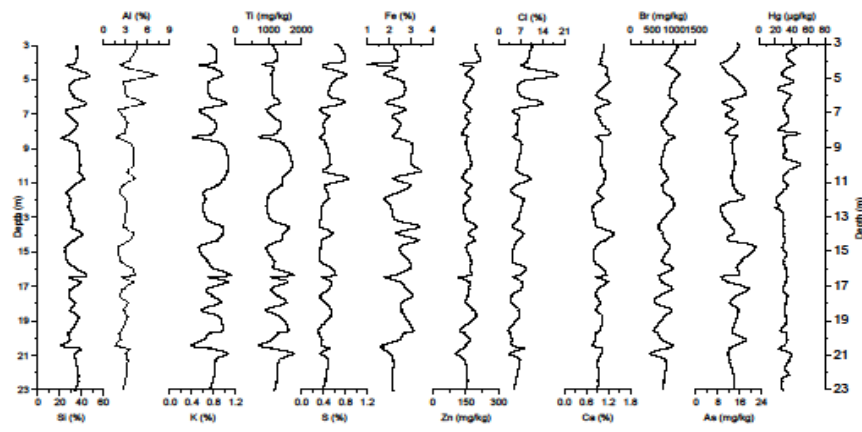
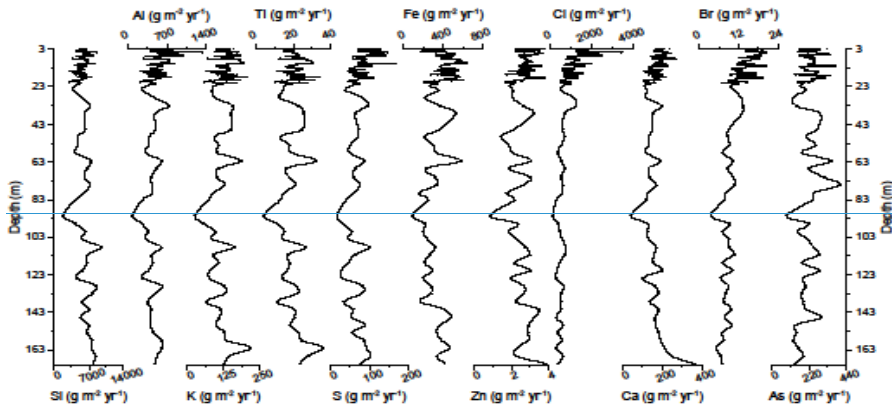

799

800 Figure 3: Down core records of Si, Al, K, Ti, S, Fe, Zn, Cl, Ca, Br, As, and As Hg concentrations of Adélie Basin sediments
801 for the top 23 m of the core.

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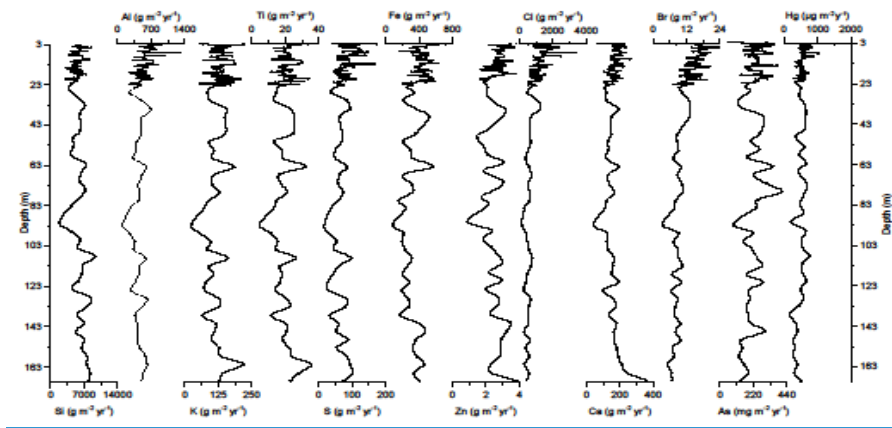
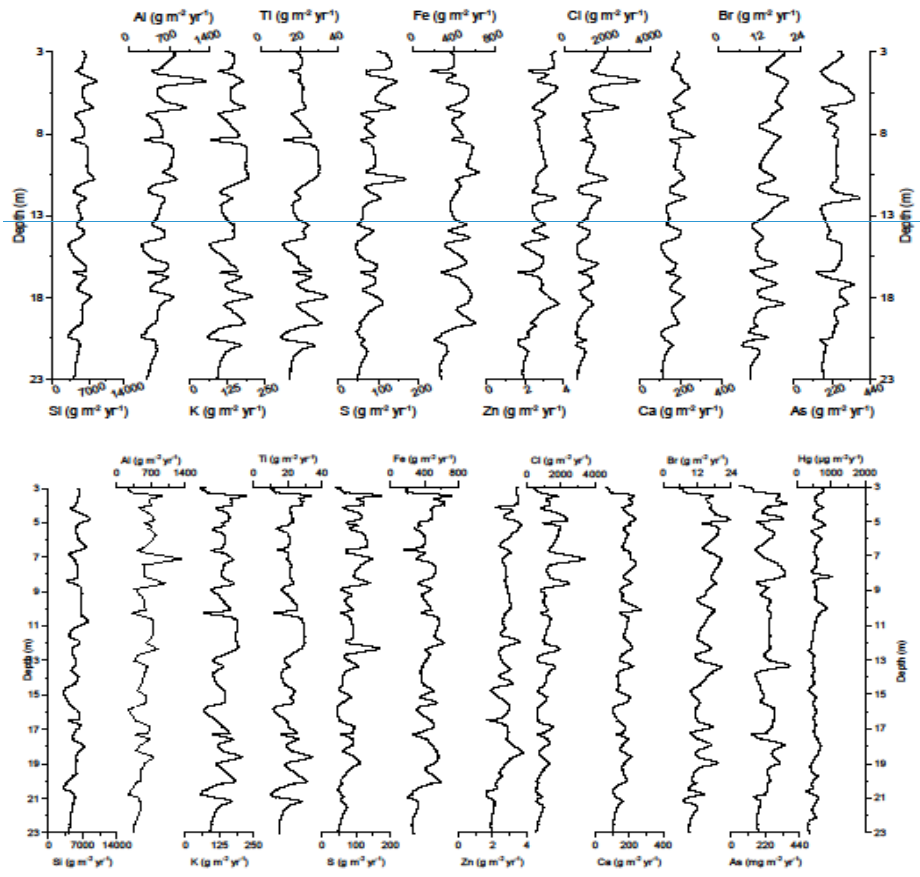
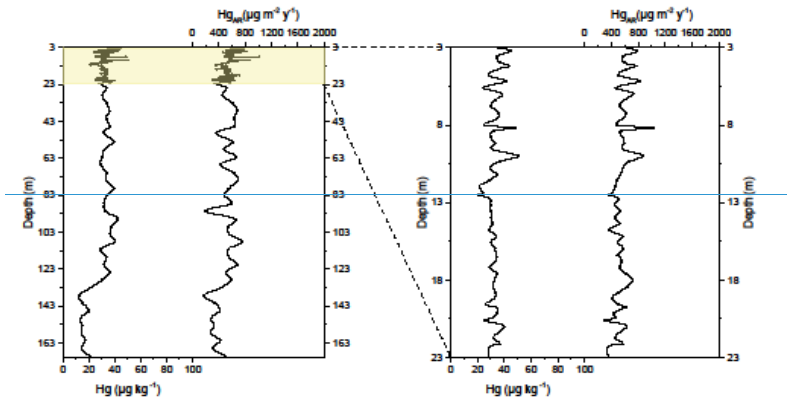
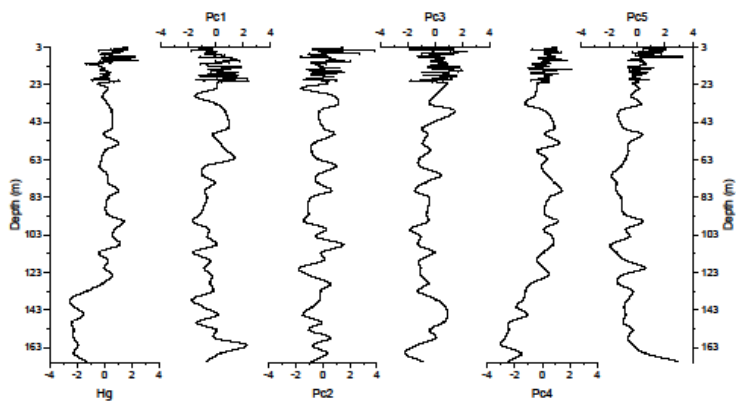
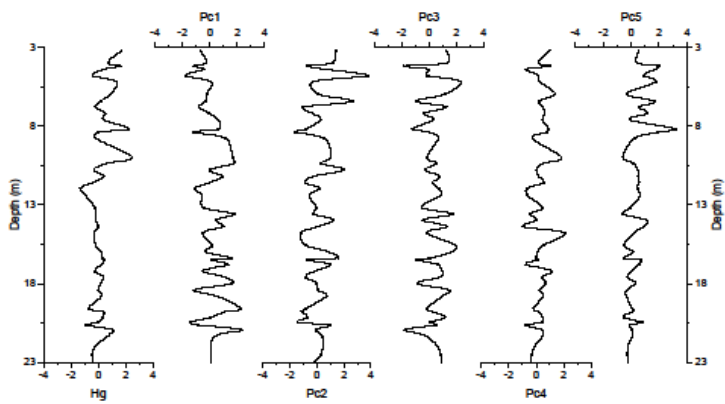

803

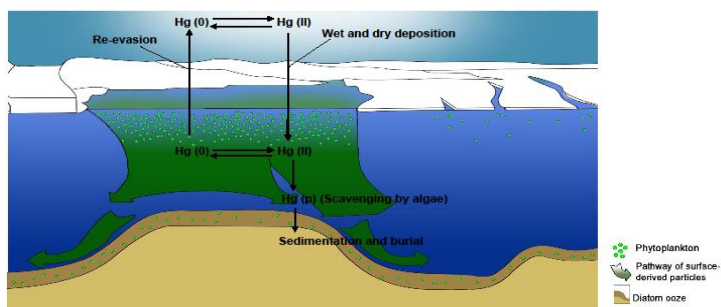
804   Figure 4: Down core records of Si, Al, K, Ti, S, Fe, Zn, Cl, Ca, Br, As, and As Hg accumulation rates of Adélie Basin sediments.

805

806

807   Figure 5: Down core records of Si, Al, K, Ti, S, Fe, Zn, Cl, Ca, Br, As, and As Hg accumulation rates of Adélie Basin sediments
808   for the top 23 m of the core.

809

810 Figure 6: Down core records of Hg concentrations and accumulation rates of Adélie Basin sediments.

811

812 Figure 76: Depth records of scores of the principal components extracted by PCA on the elemental composition of the
813 sediments along with Z-score of Hg of the Adélie Basin sediments.

814

815 Figure 87: Depth records of scores of the principal components extracted by PCA on the elemental composition of the
816 sediments along with Z-score of Hg of the Adélie Basin sediments for the top 23 m of the core.

817

818   Figure 98: Adélie Basin schematic Hg cycle model indicating the processes controlling Hg deposition and accumulation under
819   high primary production. Fast-sinking diatom particles remove dissolved water phase Hg from the water column through
820   scavenging. Hg removal from the dissolved phase by diatom particles will also decrease the Hg re-evasion to the atmosphere
821   (Figure is adapted from (Jansen et al., 2018).

822 **Table 1: Factor loadings for the five significant components extracted by PCA from Adélie Basin sediment samples.**

Elements	Components				
	1	2	3	4	5
Mn	**0.89**	0.40	-0.05	-0.05	0.01
Ti	**0.89**	0.43	0.01	0.04	-0.02
Rb	**0.84**	0.03	0.39	0.10	0.20
Zr	**0.83**	-0.22	0.05	-0.15	0.08
K	**0.73**	**0.66**	0.05	0.08	0.10
Y	**0.73**	-0.34	0.10	-0.09	0.05
Al	0.12	**0.93**	0.06	0.05	0.18
Si	0.03	**0.84**	-0.28	-0.11	-0.28
S	0.01	**0.84**	0.16	0.12	0.26
Cl	-0.16	**0.76**	0.25	0.22	0.42
Zn	-0.05	0.15	**0.78**	0.15	0.11
Cu	0.31	0.03	**0.76**	0.31	0.21
Ni	0.11	-0.08	**0.75**	-0.10	0.00
Fe	**0.62**	0.03	**0.64**	0.20	0.19
Br	-0.23	0.35	**0.50**	0.46	0.48
Hg	0.17	0.09	-0.04	**0.79**	0.14
As	-0.01	-0.09	0.30	**0.73**	-0.26
Pb	0.35	-0.11	-0.10	**-0.62**	-0.18
Sr	0.25	0.09	0.37	0.16	**0.83**
Ca	0.22	**0.52**	-0.03	-0.13	**0.67**
Eigenvalue	6.62	3.95	3.34	1.35	0.98
% variance	33.1	19.7	16.7	6.7	4.9

823