1 Answers to revieuwer 1

Author Response

Dear reviewer,
Thank you very much for the time to review this manuscript and the great suggestions you made. Below is a an overvieuw on the implementation in the paper to adres your suggestions.

1.1 Mayor comments

Reviewer Comment

1) While I recognize that this is a complex model and study design, the manuscript is on the long end, and there may be opportunities to streamline the text (particularly in 2.2-2.7) to avoid repetition and allow the core messages to come through more clearly. The authors could consider using a Supplemental Information Section to present some details of the model assumptions and parameterization, as well as for some supporting figures (e.g., Figure 12). In addition, the manuscript would benefit from additional review for typos and readability. Some have been flagged below in minor comments.

Author Response

Thank you for your insights. We moved the model specifics such as the exact parameterization to the appendix and addititional visualization to the supplement section. Table 2 and Table 3 are moved to the appendix. It is essential to show the exact parameterization used, but the large tables distract from the core message of the paper. Figures 4, 5, 6, and 12 are moved to the supplement. We decided this as they do not contribute to the core message of the paper but showing more detailed model output might be valiable for a reader interested in more a more detailed understanding of the seasonal progression. We also goes the paper an additional proofreading and shorted some segments by removing some repetitive parts.

Reviewer Comment

2) It could be helpful to provide a brief summary of the drivers of spatial and temporal variation in the results, as some of these details may be contained in the cited original model papers and therefore less clear to a reader. For example, for seasonality: to what extent is temperature dependence also considered in the bioaccumulation and toxicokinetic modeling, in addition to biomass modeling? For spatial variation: What determines the spatial distribution of higher trophic levels? Is migration relevant and, if so, how is it considered? If not, what additional implications could this have for the spatial dynamics?

Author Response

Thank you for your comment. We expanded on this in several ways. I will first discuss in the MERCY and ECOSMO sections respectively what spatial and temperol drivers control biomass and Hg speciation and then add a segment to the discussion. In section 2.2.3 we added:

Implementation

Line 109
In the MERCY v2.0 model, several drivers are incorporated to model the spatial temporal variability of Hg speciation. All Hg species are treated as tracer variables and thus move with the movement of water. Light is used to estimate the photolytic reduction rate (Hg^0 + photon → Hg^{2+}), the photolytic oxidation rate (Hg^{2+} + photon → Hg^0), and photolytic demethylation ($MMHg^+$ + photon → Hg^{2+}, DMHg + photon → Hg^{2+}, and DMHg + photon → $MMHg^+$). Temperature is used to estimate the temperature-dependent dark reduction of Hg^{2+} (Hg^{2+} → Hg^0). Furthermore, the air-sea exchange of Hg in the MERCY v2.0 model is based on the approach used in Kuss (2014) and Kuss et al. (2009), which uses the temperature and salinity dependent Henry's law to estimate the equilibrium between atmospheric and marine Hg^0 concentrations and a wind-speed-dependent transfer rate.

Author Response

In the ECOSMO E2E section we added:

Implementation

Line 210
The ECOSMO E2E and the MERCY v2.0 model interact in multiple ways. First, light absorption by phytoplankton, detritus, and DOM decreases available light, affecting light-dependent Hg speciation and photosynthesis in deeper water. Ecosystem variables are treated as tracers and move with water flow, with the exception of fish 1 and fish 2, which both have no movement. Thus, if water currents carry biological variables, they also transport bioaccumulated Hg. Detritus is the only biological component with intrinsic movement, sinking at 5 m d^{-1}.

Author Response

In the discussion section we added:

Implementation

Model limitations

1.2 Movement of higher trophic level

Line 745
In our model, phytoplankton and zooplankton are treated as tracers without implicit movement, while fish have no movement at all. This means that plankton and its bioaccumulated Hg^{2+} and $MMHg^+$ are transported by currents but do not move themselves, while fish remain stationary. For most biological variables, this implementation is a reasonable simplification. However, in the case of fish, it might influence the model results. Moving fish could transport Hg; if fish move around, they could accumulate Hg in areas with high Hg while releasing it in areas with lower Hg, thus spreading the Hg around. Implementing migration for fish provides an interesting direction for further model development, but in the current implementation, it would likely not cause major differences. This is because migration would only be relevant to implement in the 3D setup, but this setup is focused on plankton and only includes mid trophic levels of fish, which are less migratory than larger higher trophic level animals.

1.3 Decreased complexity in detritus-Hg interactions

Another limitation in the model is the limited complexity of the detritus-Hg interactions. In our model, there is only one functional group for detritus. When biota die, they instantly release all bioaccumulated Hg^{2+} and $MMHg^{+}$, which is then in constant equilibrium with the detritus. The sinking rate of detritus bound Hg is estimated based on how much Hg is bound to detritus on a given time step. This approach is a reasonable simplification for small biota, such as phytoplankton and microzooplankton. But it introduces two limitations in higher trophic levels. First of all, if a larger animal such as fish dies and sinks, it might transport some of its bioaccumulated Hg with the carcass. That is not accounted for in the model. Additionally, the equilibrium between detritus bound and dissolved Hg is based on small particulate organic carbon, as this is the most common form of detritus in marine water. But in the model, predators consume detritus, and the detritus consumed by higher trophic level predators would mostly be larger particles that could have different Hg binding characteristics.

1.4 Limitations in the modeled ecosystem complexity

As discussed when analyzing the modeled and observed trophic levels, the ecosystem model uses a functional group approach to constrain the complexity of the ecosystem. This does, however, introduce limitations in the model's ability to model bioaccumulation. Several animals might be classified under the same functional group while affecting Hg dynamics in different ways. An example is the observations that some Baltic Sea zooplankton can have *in vivo* Hg speciation (Gorokhova et al., 2020). Increasing the modeled ecosystem complexity could improve the model's performance by accounting for these differences.

1.5 Physical drivers of bioaccumulation

In the current parametrization, physical drivers such as temperature and light influence the biomass and the Hg speciation, but the bioaccumulation is purely based on the concentration of bioaccumulative Hg species, a biota functional group specific bioaccumulation rate, and the biomass of the biotic functional group. The only temperature dependent driver directly influencing bioaccumulation is the temperature dependent respiration rate of fish. As the temperature increases, so does the respiration rate of fish and consequently they release Hg faster, as this is coupled to their respiration rate. As shown by Garcia-Arevalo et al. (2024) the bioaccumulation of $MMHg^{+}$ in phytoplankton is also dependent on cell dependent drivers, such as the availability of membrane transport channels. These cell dependent drivers might be different during different stages of the phytoplankton bloom due to altering bloom composition or the physiological state of phytoplankton. While these changes could not be incorporated in our current model due to lack of our understanding of nuanced drivers of bioaccumulation, seasonal changes could influence bioaccumulation at every trophic level.

Reviewer Comment

3) See below for some places where clarification of some methodological details could be beneficial, potentially in supporting material (e.g., in model-obs comparison for 1D, initial conditions).

Author Response

We went over the detailed comments and implemented that. In addition we expanded the model evaluation segment to discuss this better. Additionally, now that the paper is accepted with minor revision we will also direclty publish the full model code including workable setups that include yaml files that can be used to replicate the exact output. We also added the following description of the initial conditions to the model description part:

Implementation

Line 477

Initial conditions

The model is initialised with uniform conditions throughout the water. The model was initialised with low biomass of 0.1 mgC m^{-1} for all phytoplankton, 0.01 mgC m^{-1} for zooplankton, and 8.0E-5 for fish 1 and 8.0E-6 for fish 2. The model was spun up for 10 years to allow the model to simulate biological fluxes, Hg cycling, and bioaccumulation to stabilise. A difference between the stations was that initial conditions for nutrients of 7.5 μM nitrate and 0.47 μM phosphate were initialized in the Northern North Sea, while in the Baltic Sea .5 μM nitrate and 0.31 μM phosphate were initialised to better represent local conditions. The exact initial conditions used for all three setups are available via the YAML files that are provided and can be used to replicate the model output.

Reviewer Comment

4) The authors may have the opportunity to deepen the reflection on next steps and future directions, given the importance of the call to better represent ecosystem effects in models. Some questions I am particularly curious to get their thoughts on are: a) is model coupling the only way to do this, is it reasonable to do a back-of-the-envelope adjustment factor that is regionally specific; b) how much trophodynamic complexity is needed — does capturing the base of the food web get most of the effect or do fish 1 and 2 shift the patterns; if so, what might be missing in this current simplified representation of the ecosystem

Author Response

We added the following part to the discussion segment

Implementation

Line 792

The required ecosystem complexity to capture Hg dynamics

As discussed, a main conclusion of this paper is showing that the ecosystem is an integral part of Hg cycling, and that this should not be overlooked. However, there is nuance in how the ecosystem should be implemented in Hg cycling models, and there is a trade-off between keeping the model simple and ensuring key drivers are implemented.

High trophic levels as a reservoir of MeHg

In most marine ecosystems, the annual average biomass of primary producers is relatively low; rather, there is a very high turnover rate of primary producers during the bloom period. While the exact numbers vary depending on the location and seasonality, high trophic levels can make up a major component of the total ecosystem biomass, especially in winter. As high trophic levels have the most $MMHg^+$ per biomass, they can form a major reservoir of $MMHg^+$. Our model, however, shows that this does not have a major effect on the tHg concentration. This indicates that the inclusion of high trophic levels such as fish might be necessary to correctly estimate the $MMHg^+$ budget, but the inclusion of fish is not necessary to correctly model tHg fluxes. One point of uncertainty here is that this conclusion is based on our implementation of the ecosystem. As discussed in the model limitation segment, several drivers, such as fish migration and the transport of Hg to deep water by sinking carcasses, are not accounted for, and these drivers could still prove to be an essential component of Hg cycling.

Bentho pelagic coupling

A key component where the ecosystem is essential for a correct understanding of Hg cycling is the bentho-pelagic coupling. In coastal areas, the consumption of pelagic detritus and Hg bound to it by macrobenthos can be a major flux of organic carbon from the pelagic to the benthic system. The sediment is identified as a key area for Hg methylation; this increased transport of Hg^{2+} from the pelagic to the benthic due to biotic consumption of detritus would constitute a source of $MMHg^+$. Of course, in some areas, sediment is not resuspended, and then increased transport of Hg to the sediment can result in additional burial of Hg. As the macrobenthic influence on the bentho-pelagic coupling, the burial and resuspension are spatially and temporally variable; this cannot be accounted for by a "back of the envelope" estimation. Rather, the inclusion of a realistic bentho-pelagic coupling is essential for Hg speciation models in coastal areas.

key biota for Hg cycling

In an important aspect of estimating the role of the ecosystem in Hg cycling is understanding that not all biota affect Hg cycling in a similar way. The clearest of the ecosystem interactions with Hg cycling is the removal of Hg by biota when biomass is high and the release when biomass is low. This is extensively analyzed in this study. However, several biota might have an unexpectedly high impact on Hg cycling. The first example of this, which is also evaluated in this paper, is Baltic Sea cyanobacteria, which can facilitate biogenic reduction. But beyond this, it is demonstrated that some animals of zooplankton and cephalopods can have *in vivo* Hg speciation (Gente et al., 2023; Gorokhova et al., 2020). Another example is that sponges are demonstrated to have very high inorganic Hg levels, suggesting an important role in the bentho-pelagic coupling (Orani et al., 2020).

1.6 Detailed comments

Reviewer Comment

L22: Number of parties now exceeds the number of signatories (over 150), so could update the number https://minamataconvention.org/en/parties

Author Response

The link provided still states that indeed has 152 parties we updated the statement as below to show both:

Implementation

Line 21
Due to the consumption of polluted marine wildlife, more than 1000 people died, and more were permanently disabled (Harada, 1995). Efforts to control Hg emissions culminated in the Minamata Convention on Mercury, which is a pledge to reduce Hg emissions(Outridge et al., 2018). It has 152 parties and is currently signed by 128 countries.

Reviewer Comment

L52: "In summary, there are three fractions... in our model." Read as confusing as the model hasn't been introduced yet.

Author Response

I agree that this is incorrect to refer to this in the model before the model is introduced. I also think the rest of the alinea could be clearer. It is rewritten as below:

Implementation

Line 50
Under anoxic conditions, Hg^{2+} binds with S^{2-} to form cinnabar (HgS), which is considered a sink due to its low solubility (Oliveri et al., 2016). In seawater, the abundance of chloride ions causes Hg^{2+} and $MMHg^{+}$ to exist mainly in the form of inorganic chloride complexes. The speciation of Hg with organic carbon in the marine ecosystem, such as detritus and DOM, is a complex interaction that can influence the speciation, solubility, mobility, membrane permeability, and toxicity of Hg (Ravichandran, 2004). In this study, we refer to three distinct fractions of both H^{2+} and $MMHg^{+}$: 1) dissolved species not bound to organic material, including species such as $HgCl_2$ and MMHgCl, collectively referred to as Hg^{2+} and $MMHg^{+}$, 2) Hg^{2+} and $MMHg^{+}$ bound to dissolved organic matter (DOM), referred to as Hg-DOM and MMHg-DOM, and Hg^{2+} and $MMHg^{+}$ bound to detritus, referred to as Hg-detritus and MMHg-detritus.

Reviewer Comment

L137: As defined in the first sentence, isn't this bioconcentration only?

Author Response

That is indeed badly phrased. It changed as below:

Implementation

Line 145
Bioaccumulation is the increase in Hg^{2+} or $MMHg^{+}$ in the biota relative to the concentration of the surrounding water.

Reviewer Comment

Fig. 1: Typos in title and Scenario C. Could consider overlaying the 1-D vs 3-D component too so that it captures that aspect of the design as well. Could incorporate a map of locations as a side panel for the global audience.

Author Response

I would update the image to the updated images shown in 1 Line 197

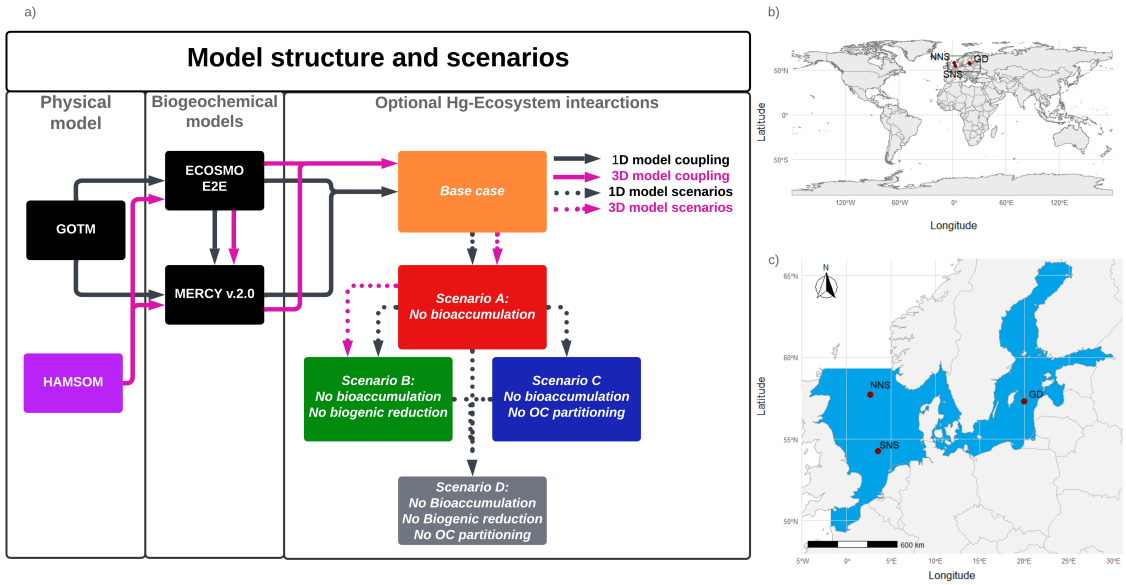

Figure 1: a) Schematic of the model setup. The black lines indicate the 1D setup where GOTM drives the ECOSMO E2E Ecosystem model and MERCY V2.0 Hg speciation model. These are used to simulate a base case and four scenarios with varying Hg–ecosystem interactions. The impact of the ecosystem is evaluated by comparing the base case to a scenario without: bioaccumulation (Scenario A), bioaccumulation and biogenic reduction (Scenario B), bioaccumulation and partitioning to detritus and DOM (Scenario C), and all mentioned ecosystem interactions (Scenario D).The purple lines show the 3D setup, where the HAMSOM model drives ECOSMO E2E and MERCY V2.0 models. The base case, Scenario A, and Scenario B are simulated in the 3D setup. b) Global map with the regional domain highlighted. c) Regional map of the North and Baltic Sea region. The 3D HAMSOM-ECOSMO-Mercy model domain is marked in blue. The three 1D setups, Northern North Sea (NNS), Southern North Sea (SNS), and Gotland Deep (GD), are labeled and marked with red points.

Reviewer Comment

Section 2.4: Include grid resolution for the 3D models (may have missed this)

Author Response

That was indeed not specified in this paper but only in the original paper. Section 2.4 i updated as below:

Implementation

Line 284
The 3D HAMSOM-ECOSMO-MERCY domain covers the Baltic Sea and the North Sea with open boundaries at the English Channel and at 63°N, where the North Sea is connected to the Atlantic Ocean, as shown in Fig. 1. The resolution of the model is about 10 x 10 km^2 on a spherical grid with a vertical resolution of 20 layers. The upper four layers are 5 m thick, while the deepest layer reaches a thickness of up to 250 m. The maximum water depth is 630 m. The first 4 years are used as spin-up and the final year is used for the analyses. The model is run in its default setup, without bioaccumulation or biogenic reduction. The effect of bioaccumulation on both tHg and tMeHg is visualized by plotting the relative difference in tHg and tMeHg caused by the ecosystem. The data is visualized using the cartopy package in Python version 3.11.2.0.

Reviewer Comment

L299: pre-dated?

Author Response

That is indeed wrong and should note have the -. Corrected it to:

Implementation

Line 381
Fish 2 is at the top of the food chain and is therefore not predated upon in the model.

Reviewer Comment

L312-316: A bit more detail on this model tuning/calibration process — what informed the choice of lowered value

Author Response

We added the following part at the end of section 2.6, replacing the part from line 316 onward with the expanded explanation below:

Implementation

Line 401
The modeled trophic levels are shown in Table 2. In the Gotland, Deep macrobenthos is absent because of the anoxic conditions. Except for fish 2 in the Northern North Sea, all functional groups have trophic levels that are lower than observed in the North and Baltic Seas. This was compensated for by reparameterizing the uptake efficiency of carbon used in the previous version of the ECOSMO E2E model. The uptake efficiency of carbon, known as assimilation efficiency, is a key parameter for biomagnification. Biomagnification occurs if the organic material is absorbed less efficiently or respirated more efficiently than a pollutant, as this would result in an increase in this pollutant compared to organic material in the organism compared to its diet. The assimilation of carbon can be seen as two components; the first absorption refers to all carbon that is used by the fish and not directly excreted via faeces, whereas the assimilation refers to the carbon that is built up into the tissue of the fish. Fish are typically shown to have a 91-92% absorption efficiency, but only a 30-49% assimilation efficiency (Shelley & Johnson, 2022).

Due to uncertainty, we parameterized the higher trophic-level fish with a lower assimilation efficiency than in the previously published ECOSMO E2E version, down to 45% in fish 2. To compensate for the decrease in carbon intake, the mortality and respiration rate of zooplankton is decreased to a value that is lower than in Daewel et al. (2019), but still within the range used in previously published models (Cruz et al., 2021). The phytoplankton is parameterized as shown in Table A1. To compensate for the increased zooplankton grazing, the growth rate is increased compared to the previously published ECOSMO E2E version, but remains within the experimentally observed range (Stelmakh & Kovrigina, 2021). All other values are the same as in (Daewel et al., 2019). This was done to tune the model to better reproduce higher $MMHg^{+}$ bioaccumulation, which is in line with observations. These interactions remain uncertain in the model, but replicating bioaccumulated concentrations is essential to estimate the bioaccumulation feedback on Hg speciation, which is the core focus of this study.

Reviewer Comment

L468-472: What are the observed values for biomass? Not sure if I missed their reporting somewhere. Could they also be put on Figure 2 for comparison?

Author Response

We updated the model evaluation segment to show this and made some changes to Fig. 2 by showing chlorophyll-a in the surface water, rather than the fully depth intergrated values. Most measurements measure the concentrations, and hence showing this increases the comparibility to observations. Additionally, we changed Fig. 2 to make it not based on the last year of the observations but the daily mean of the last 10 years of the simulation to remove the change the plots are influenced by outliers and give a better overview of the behavior of the model. I think the comparison between the model and observations is a bit too nuanced to allow an easy integration of the results into Fig. 2. Because of this, I would suggest adding the Table 1. Then I would add the update Section 3.1 to:

Implementation

Line 494

Evaluation of carbon fluxes

To evaluate carbon stocks and fluxes of the ECOSMO E2E model, we compared the modeled primary and secondary production to the production in the validated 3D ECOSMO E2E version in Daewel et al. (2019). In addition, we compared the model with observations for surface chlorophyll-a and zooplankton concentration, and the total fish and macrobenthos biomass. This comparison evaluated if our simplified 1D models remain consistent with a realistic ecosystem and is shown in Table 1.
The 3D ECOSMO E2E model estimates total primary production between 50 and 90 gC m^{-2} y^{-1} in the open North Sea and between 30-50 gC m^{-2} y^{-1} in the open Baltic Sea. The phytoplankton is initially driven by diatoms and succeeded by flagellates in the North Sea and a mix of diatoms, flagellates, and cyanobacteria in the Baltic Sea. Secondary production is estimated between 20-40 gC m^{-2} y^{-1} in the North Sea and 10-30 gC m^{-2} y^{-1} in the open Baltic Sea (Daewel et al., 2019).
The chlorophyll-a and biomass simulated in our 1D model are presented in Fig. 2. The

total yearly primary production in our model is 50, 62, and 61 gC m^{-2} y^{-1}, and the pelagic secondary production is 24, 42, and 29 gC m^{-2} y^{-1}. This means that the primary and secondary production of the 1D model are in line with the previously published and validated 3D version of the model.
The average plankton concentration during the bloom is taken. The phytoplankton spring bloom period is selected as 1st of April - 30th of June and the zooplankton bloom period as 16th of April - 31st of October to select the majority of the bloom. The average chlorophyll-a concentration and zooplankton biomass in the surface (0-10 m) are compared to observations.
Chlorophyll-a levels in the Baltic Sea display significant variation. During bloom periods, the Northern Baltic Sea typically has values of 1-2 mg m^{-3}, whereas in the Southern Baltic Sea, values can reach 6 mg m^{-3}, with a basin-wide average of 2.64 mg m^{-3} (OSPAR, 2017). During autumn, cyanobacteria can become the dominant taxa, but there is a large variety in the intensity of the bloom and the relative importance of different taxa (Hjerne et al., 2019). Our average modeled chlorophyll-a concentrations in the Baltic Sea of 0.92 mg m^{-3} better resemble the Northern Baltic Sea than the Southern Baltic Sea. While the modeled chlorophyll concentrations in the North Sea are within the range of observations.
Zooplankton concentration ranges between 50 and 200 mgC m^{-3} in the Northern North Sea and 0-50 mgC m^{-3} in the Southern North Sea (Krause & Martens, 1990). Observations from the coast of Estonia, near the Gotland Deep, report 50 mgC m^{-3} in measurements furthest from the coast (Ojaveer et al., 1998). The average concentration of zooplankton biomass during the bloom in our model falls within these ranges for all setups.
Total fish biomass in the North Sea is estimated to be between 15 and 23 g wet weight m^{-2} for both the North Sea by Sparholt (1990) and Baltic Seas by Thurow (1997), or between 2.25 and 3.45 gC m^{-2} assuming the earlier presented conversion rates of wet weight to carbon content of fish. This means that the modeled North Sea fish stocks are in agreement with observations, while the modeled fish population in the Baltic Sea is 7% higher than observed. The 7% can indicate the model overestimates fish in the Gotland Deep, but it is low enough that it can originate from uncertainty in the biomass estimate or be caused by uncertainty in the conversion from wet weight to carbon.
The peak and mean macrobenthos biomass is 12.3 and 6.84 gC m^{-2} in the Southern North Sea and 3.4 and 0.99 gC m^{-2} in the Northern North Sea, while macrobenthos biomass estimations range from 1.1 to 35.5 grams of carbon for the open North Sea, with the highest values closer to the coast (Daan & Mulder, 2001; Heip et al., 1992). So the macrobenthos biomass in our model aligns with observations. The Gotland Deep has anoxic deep water, so there is no macrobenthos in the Gotland Deep in our model, which matches observations (Kendzierska & Janas, 2024).

Overall the model produces biomass consistent with observations and the previously validated 3D version of the model. The only notable deviation is the Chlorophyll-a concentration of the Gotland Deep which closer resembles the Northern than Central Baltic Sea and the fish in the Baltic Sea is above the estimation made by Thurow (1997). This deviation, however, is only 7% which can also be caused by uncertainty in the original estimation or the conversion of the estimated wet weight to the modeled dry weight.

Reviewer Comment

L508: How is “high quality” defined?

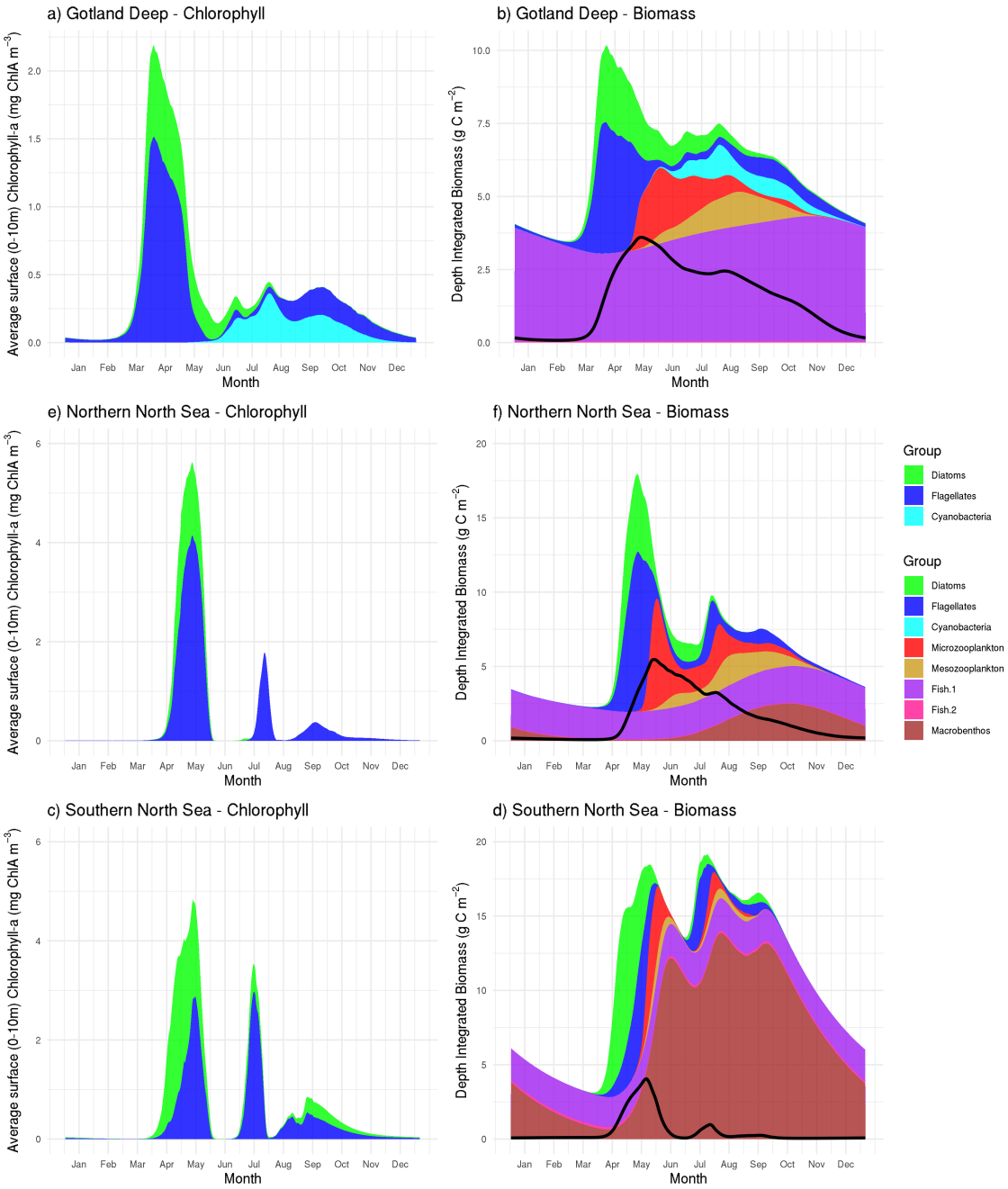

Figure 2: Modeled chlorophyll concentration (left) and organic matter concentration (right). The daily average values over are shown averages over the last 10 years of the simulation (Jan 2001 to Jan 2011). All living organic material is stacked, and detritus and DOM are plotted in the black line on top. Peak spring bloom chlorophyll concentration varies with location. Gotland Deep (a) has 2.2 mg m^{-3} chlorophyll with succession from diatoms to flagellates to cyanobacteria. The Northern North Sea (c) has 5.6 mg m^{-3} chlorophyll and is dominated by flagellates while the Southern North Sea (e) has 4.8 mg m^{-3} chlorophyll and is initially dominated by diatoms and later succeeded by flagellates. All locations have a succession of zooplankton after phytoplankton which microzooplankton and is taken over by mesozooplankton. Fish biomass is stable in the Northern North Sea (fish 1: 1.8-2.6, fish 2: 0.044-0.054 g C m^{-2}), the Southern North Sea (fish 1: 2.0-2.2, fish 2: 0.14-0.16 g C m^{-2}), and the Gotland Deep (fish 1: 3.0-4.3, fish 2: 0.40-0.44 g C m^{-2}). Macrobenthos biomass fluctuates seasonally: Northern North Sea (0.050-2.5 g C m^{-2}), Southern North Sea (0.64-13.8 g C m^{-2}), while macrobenthos is absent in Gotland Deep due to anoxic bottom water.

Table 1: Comparison of modeled and observed values for key ecosystem indicators across three regions.

	Gotland Deep		Northern North Sea		Southern North Sea	
	Modeled	Observed	Modeled	Observed	Modeled	Observed
Surface Chlorophyll ($mg\,m^{-3}$)	0.92 ± 0.11	1–6	1.78 ± 0.12	0.8–2.5	1.68 ± 0.057	0.8–2.5
Surface Zooplankton biomass ($mg\,m^{-3}$)	40.60 ± 4.60	∼50	50.12 ± 3.60	50–200	34.63 ± 5.40	0–50
Fish biomass ($g\,m^{-2}$)	3.70 ± 0.16	2.25–3.45	2.29 ± 0.17	2.25–3.45	2.27 ± 1.16	2.25–3.45
Macrobenthos biomass ($g\,m^{-2}$)	0	0	0.99 ± 0.33	0.6–17.5	6.84 ± 1.16	0.6–17.5

Author Response

That is indeed badly phrased. There are many more studies in the Baltic Sea that analyze Hg cycling than in the North Sea. Because of this, we reframed this too:

Implementation

Line 563
The modeled Hg bioaccumulation for biota in the Gotland Deep was compared to observations, because the Baltic Sea is studied more extensively for Hg cycling, and studies such as performed by Kuss (2014) provide the opportunity to validate Hg cycling, while studies such as Nfon et al. (2009) allow the validation of low-trophic-level biota, while data on Hg cycling and bioaccumulation in low-trophic-level biota are extremely limited in the North Sea. For this evaluation, we only used model output where the biomass exceeded 100 mg carbon m^{-2} and took the modeled values of the <5 m for phytoplankton and <20 m for other biota to ensure that our bioaccumulation values resemble the modeled values in surface water where biota are active. A comparison between our modeled and observed bioaccumulation is shown in Table 4. Additionally, year-round bioaccumulation of Hg^{2+} and $MMHg^{+}$ in the Gotland Deep, the Northern North Sea, and the Southern North Sea is shown in the supplementary information in Figures S1, S2, and S3 respectively.

2 Answers to reviewer 2

Author Response

Dear reviewer,
Thank you very much for comments. We answered already how we would suggest to implement your recommendations but after proofreading some formulations have slightly changed. Below we have summarised how we have implemented all your suggestions in the new version of the manuscript.

Reviewer Comment

Line 5: I find this expression peculiar "Our results show that bioaccumulation can increase total methylmercury (tMeHg) in coastal pelagic waters from 0.059 to 0.092pM, a 44% increase.". Bioaccumulation is a process enhancing concentrations in biota, not in water, which you also state in lines 138-139.

Author Response

This is refrased as below to ensure that it is clear that the increase is the difference in tMeHg between model runs with and without bioaccumulation.

Implementation

Line 5
Incorporating bioaccumulation into the model leads to a 44% increase in total methylmercury (tMeHg) concentrations in coastal pelagic waters, from 0.059 to 0.092 pM, compared to a model without bioaccumulation. Bioaccumulation and binding of Hg to organic matter contribute to elevated Hg levels in surface waters.

Reviewer Comment

Line 31: Regarding "This can lead to insufficient data to understand the cycling and bioaccumulation of marine Hg at the base of the food web,": I would argue that undoubtedly, measuring Hg only in biota is insufficient to understand Hg cycling. It may be useful to monitor the ultimate effectiveness of the Minamata treaty but certainly not to understand the observations. I suggest to reformulate this discussion.

Author Response

This is refrased as below:

Implementation

Line 32
While measuring Hg in biota can help evaluate the risk of $MMHg^+$ pollution to humans, and thus support the effective evaluation of the Minamata Convention, fully understanding Hg cycling requires further research. Understanding the link between Hg in the atmosphere and the risk posed to humans by $MMHg^+$ via the consumption of seafood requires studying the factors linking this, including the link between marine Hg cycling and the bioaccumulation of $MMHg^+$ at the base of the food web. Modeling studies are an important tool to improve our understanding of these complex interactions and can help evaluate the effectiveness of Hg reduction strategies.

Reviewer Comment

Line 41-42: I suggest to replace the work equilibrium since the Hg2+ reduction and Hg0 oxidation are largely mediated by different, independent mechanisms including photochemical and biotic processes.
Line 43: replace the term "double methylated DMHg" with "dimethylmercury DMHg"

Author Response

That is updated as below:

Implementation

Line 44
The dominant species of Hg in surface water is inorganic Hg^{2+}. Hg^{2+} and Hg^0 are in dynamic redox cycling, but in aquatic environments this favors the oxidized form, Hg^{2+}. Although Hg^0 can evaporate, Hg^{2+} can be methylated into two forms of organic Hg, monomethylmercury ($MMHg^+$) and DMHg.

Reviewer Comment

Line 70: The discussion on Hg uptake mechanisms here do not harmonize with he discussion of passive diffusion uptake in line 49.

Author Response

I see the conflict indeed. We removed the link between chloride and bioaccumulation in line 49 and merging to two observations in line 70 as follows:

Implementation

Line 72
As mentioned before, the dominant form of Hg^{2+} and $MMHg^+$ in the marine environments is $HgCl_2$ and MMHgCl respectively. These compounds can diffuse through cell membranes due to their lipophilic nature or bind to organic matter (Zhong & Wang, 2009). This diffusion is mainly dependent on the surface area of organic membranes that are in contact with water and is therefore dominated by microorganisms such as phytoplankton (Mason et al., 1996). Recent work has expanded on this basic understanding of bioaccumulation and has shown that while these lipophilic compounds can diffuse through the cell membrane, total uptake into phytoplankton is a complex two-step process in which Hg binds first to the phycosphere before it is absorbed into the cell. Recent data suggest that $MMHg^+$ uptake is influenced by cell-dependent factors such as phycosphere thickness and availability of transmembrane channels for $MMHg^+$ transport, while this is not the case for Hg^{2+} (Garcia-Arevalo et al., 2024). This suggests that Hg^{2+} only bioaccumulates due to its lipophilic nature, whereas $MMHg^+$ both bioaccumulates due to its lipophilic nature and is actively transported by the cell.

Reviewer Comment

Line 15: not only by "marine" microorganisms.

Author Response

The sentence is updated by removing marine microorganisms, it is indeed correct that also other processes can form MeHg.

Implementation

Line 15
Mercury (Hg) is a toxic pollutant that poses significant risks to marine ecosystems and human health as a result of bioaccumulation. Despite its known hazards, the processes that govern Hg bioaccumulation within the marine food web are poorly understood. This study examines the role of the marine ecosystem in Hg cycling in highly productive coastal seas.

Reviewer Comment

Line 34: I suggest to replace "are a perfect tool to" (which is hardly true) with "are an important tool to".

Author Response

I fully agree that they are not perfect tool and it is is updatd as below:

Implementation

Line 33
Understanding the link between Hg in the atmosphere and the risk posed to humans by $MMHg^{+}$ via the consumption of seafood requires studying the factors linking this, including the link between marine Hg cycling and the bioaccumulation of $MMHg^{+}$ at the base of the food web. Modeling studies are an important tool to improve our understanding of these complex interactions and can help evaluate the effectiveness of Hg reduction strategies.

Reviewer Comment

Line 35: "Because MeHg formation and subsequent bioaccumulation in seafood are the dominant source of Hg exposure to humans,... ". The sentence is grammatically incorrect, formation and bioaccumulation are not sources, seafood is the source.

Author Response

We updated that sentence as below:

Implementation

Line 38
Because seafood consumption is the dominant source of Hg exposure in humans, due to the formation and subsequent bioaccumulation of MeHg in marine organisms, Hg levels in the world's oceans are of special concern.

Reviewer Comment

Line 45: the following statement is grammatically incorrect: "Since only MMHg+ bioaccumulates, the term MeHg, in this paper, refers to the total methylated fraction of Hg in seawater.".

Author Response

That is rewriten as below:

Implementation

Line 48
In this paper, MeHg refers to all methylated Hg in seawater, this includes both $MMHg^{+}$ and DMHg. Of these two Hg species, only $MMHg^{+}$ is known to bioaccumulate.

Reviewer Comment

Line 48: replace "inorganic chlorine complexes" with "inorganic chloride complexes".

Implementation

Line 52
In seawater, the abundance of chloride ions causes Hg^{2+} and $MMHg^{+}$ to exist mainly in the form of inorganic chloride complexes.

Reviewer Comment

Line 54: avoid using the term "species" for microorganisms as it can be confounded with chemical species (which is discussed in the preceding lines).

Author Response

That is indeed confusing. We replaced the biological terms as below::

Implementation

Line 58
Bioaccumulation of Hg occurs when biota take up Hg at a rate higher than that at which it is excreted (Bryan, 1979).

Implementation

Line 67
Biomagnification can be estimated in nature by sampling stable carbon and nitrogen isotopes with Hg to assess both the Hg content and the trophic position of a series of animals (Lavoie et al., 2013).

Implementation

Line 87
Their research showed that certain genera of cyanobacteria in the Baltic Sea (notable Synechococcus and Aphanizomenon) can also react with Hg by reducing dissolved Hg^{2+} to dissolved gaseous Hg^{0}.

Implementation

Line 231
Diatoms can dominate at the start of the bloom, but other phytoplankton taxa take over once the silicate is depleted.

Implementation

Line 379
It is mainly representative of large demersal fish, such as cod (*Gadus spp.*), but would as a functional group also include other large benthic taxa such as whiting (*Micromesistius poutassou*) or haddock (*Melanogrammus aeglefinus*).

Implementation

(Caption Table 2) Dimensions, shape, and maximum growth and mortality rates of most common phytoplankton taxa in the North and Baltic Seas, to resemble ECOSMO E2E functional groups and the conversion ratio of mg C to cm^2 cell membrane and dm^3 cell volume.

Implementation

Line 304
The surface area is estimated from the most common phytoplankton taxa in the three phytoplankton functional groups for the North and Baltic Seas. The taxa and dimensions are shown in Table 2.

Implementation

Line 328
The groups representing phytoplankton taxa with a smaller size and therefore a higher uptake rate also have a high Hg^{2+} release rate. As a result, all phytoplankton groups reach equilibrium at similar Hg^{2+} concentrations.

Implementation

Line 512
During the autumn, cyanobacteria can become the dominant taxa with a biomass of up to 50 mg C m^{-3}, but there is a large variety in the intensity of the bloom and the relative importance of different taxa (Hjerne et al., 2019).

Implementation

Line 228
The constant mixing allows macrobenthos to feed directly from the phytoplankton bloom, leading to a high macrobenthos stock (Heip et al., 1992). 41.5 m is also deep enough to support larger fish, such as herring and cod.

Implementation

Line 387
This means that certain interactions of the marine ecosystem that could biomagnify Hg, such as predation of organisms within the same functional group or even cannibalism,

which do not alter nutrient fluxes or organic matter stocks, are not explicitly specified in the model (Arrhenius & Hansson, 1996; Montagnes & Fenton, 2012; Schrum et al., 2006).

Implementation

Line 369
The fish rates are based on a study investigating the uptake, release, and turnover rates in the Indo-Pacific fish Harry hotlips *Plectorhinchus gibbosus* (Wang & Wong, 2003).

Implementation

Line 367
Water fleas are abundant in the Baltic Sea, but not in the North Sea.

Implementation

Line 835
It is important to note that we wanted to implement realistic bioconcentration and trophic transfer rates to not over-tune the model. Several interactions, such as, for example, cannibalism within the functional group, can increase bioaccumulation in ways that are not captured by the model, resulting in both increased bioaccumulation and trophic levels.

Reviewer Comment

Line 65: "Biomagnification can be estimated in nature by sampling stable carbon and nitrogen isotopes with Hg" – isotopes and Hg are not sampled, they are measured.

Implementation

Line 67
Biomagnification can be estimated in nature by measuring stable carbon and nitrogen isotopes with Hg to assess both the Hg content and the trophic position of a series of species (Lavoie et al., 2013).

Reviewer Comment

Line 80-81: avoid to use "reduce" both for chemical reduction and decrease , in particular in the same discussion.

Author Response

Thank you, we replaced all mentions of reduced with decreased in the manuscript where appropriate. That is, in lines 80-81 as shown below. Additionally, I would replace the word reduce by decreased, in the abstract, in line 22, 281, 283, 493, 570, 583, 637, 640, 2x in the caption Fig. 9, 664, 665, 675, 682, 683, 685, 707, 2x in the caption of Fig. 13, 712. This way it is consistently clear that I refer to a chemical reduction or a decrease.

Implementation

Line 87
Their research showed that certain species of cyanobacteria in the Baltic Sea (notable Synechococcus and Aphanizomenon) can also react with Hg by reducing dissolved Hg^{2+}

to dissolved gaseous Hg^0. Since Hg^0 is volatile and can evaporate, increasing the fraction of Hg^0 can decrease the Hg. This process is referred to as biogenic reduction.

Reviewer Comment

Lines 138-139: an element cannot "undergo speciation". Speciation is not a process it is the distribution of an element among different chemical forms. An element may undergo changes in speciation.

Author Response

This is indeed used incorreclty, we update this as below:

Implementation

Line 145: When Hg is bioaccumulated, it can no longer evaporate, undergo photolysis, or participate in chemical reactions that change its speciation; instead, it is transported with the organism that accumulated it.

Author Response

I also update that in the sentence in line 139 and update it as below:

Implementation

Line 147
Thus, bioaccumulation removes aquatic Hg^{2+} and $MMHg^{+}$, which can otherwise participate in chemical reactions that alter their speciation.

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