Answers to reviewer 2

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

I would suggest to refrase it as below, that it is clear that the increase is the difference in tMeHg between model runs with and without bioaccumulation.

Suggested edit

Incorporating bioaccumulation into the model leads to a 44% increase in total methylmercury (tMeHg) concentrations in coastal pelagic waters, from 0.059 to 0.092pM, compared to a model without bioaccumulation.

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

I would refrase that as below:

Suggested edit

While measuring Hg in biota can help evaluate the risk of $MMHg^{+}$ pollution to humans, and thus support the effective evaulation 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 $MMHg^{+}$ in fish 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.

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

I would suggest to edit line 41-43 as below to address both of the above comments:

Suggested edit

The dominant species of Hg in surface water is inorganic Hg^{2+}. Hg^{2+} and Hg0 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, monomethyl mercury ($MMHg^+$) and dimethylmercury (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. I would suggest removing the link between chloride and bioaccumulation in line 49 and merging to two observations in line 70 as follows:

Suggested edit

As mentioned before, the dominant form of dissolved Hg^{2+} and $MMHg^+$ is $HgCl_2$ and MMHgCl. These compounds can diffuse through cell membranes due to their lipophilic nature or bind to organic matter (Zhong & Wang, 2009). 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 suggests 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

I would suggest to update that sentence by removing marine microorganisms, it is indeed correct that also other processes can form MeHg.

Suggested edit

Mercury (Hg) is a naturally occurring toxic element that is extremely persistent in the environment (Driscoll et al., 2013). It can be methylated to methylmercury (MeHg), a dangerous neurotoxin that can bioaccumulate in the marine food chain (Mason et al., 1995; Trevors, 1986).

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 better to say an important tool.

Suggested edit

This can lead to insufficient data to understand the cycling and bioaccumulation of marine Hg at the base of the food web, although these processes are essential in linking Hg emissions to (Me)Hg concentrations in seafood. 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

I would suggest to update the sentence as below:

Suggested edit

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

I suggest to rewrite as shown below:

Suggested edit

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".

Suggested edit

In seawater, the abundance of chloride ions causes Hg^{2+} and $MMHg^{+}$ 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. I would replace the biological terms throughout the paper, and I would suggest updating line 54 as follows:

Suggested edit

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

Author Response

I would update the term species in line 66 with animals, so it becomes the sentence below:

Suggested edit

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).

Author Response

In line 79 I would replace species with genera:

Suggested edit

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.

Author Response

In line 209, 212, 296, 320, 343, 460 and Table 2 I refer to phytoplankton and fish species, here I would replace species in both the captian, the table and the text with taxa.

Suggested edit

(Line 209) Diatoms can dominate at the start of the bloom, but other phytoplankton taxa take over once the silicate is depleted.

Suggested edit

(296) 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*).

Suggested edit

(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.

Suggested edit

(Line 320) 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.

Suggested edit

(Line 343)The taxa representing phytoplankton species with a smaller size and therefore a higher uptake rate also have a high Hg^{2+} release rate.

Suggested edit

(Line 460) 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).

Author Response

In line 207, 278, and 380 I can remove the word species while leaving the rest of the sentence with the intended meaning:

Suggested edit

(Line 207) 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.

Suggested edit

(Line 278) 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).

Suggested edit

(Line 380) 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).

Author Response

In line 377 i refer to the species of Daphnia Pulex. I would replace that as:

Suggested edit

This organism is abundant in the Baltic Sea, but not in the North Sea.

Author Response

In line 700 I mention cannabilism within the same species. I would rewrite that mentioning it is within the same functional group:

Suggested edit

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.

Suggested edit

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, I 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.

Suggested edit

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

I see that I indeed used this term incorreclty, I would suggest to update this as below:

Suggested edit

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:

Suggested edit

We are interested in this because the bioaccumulation of Hg removes aquatic Hg^{2+} and $MMHg^{+}$, which can otherwise participate in chemical reactions that alter their speciation.