1 Answers to revieuwer 2

Reviewer Comment

The introduction mentions "MMHg+is a topic of serious concern because MMHg+is a dangerous neurotoxin that can bioaccumulate to levels. that are dangerous for human consumption in fish that are often consumed as seafood." Could this phrase be a little more concise?

Author Response

I would suggest that we rewrite the sentence as follows:

Suggested edit

The element mercury (Hg) is presently included in the World Health Organization's list of the 10 substances of greatest concern (WHO, 2020). This is due to the capability of Hg to be methylated into monomethyl mercury ($MMHg^{+}$), a potent neurotoxin generated by microbial methylation of inorganic Hg. $MMHg^{+}$ biomagnifies within aquatic food webs, accumulating in predatory fish to concentrations that can impair human neurological development upon consumption.

Reviewer Comment

The introduction mentions "Since DMHg is susceptible to photodegradation, we can assume that it plays an important role in the coastal water investigated in this study, until better observational studies confirm or correct this assumption." There are problems with logic

Author Response

My apologies, that is indeed incorrect. I would correct it to:

Suggested edit

Given the rapid photodegradation of DMHg in natural water, DMHg is assumed not to significantly bioaccumulate in biota in the coastal area investigated in this study (West et al., 2022).

Reviewer Comment

The introduction mentions "the bioconcentration process is complicated and recent studies show that the bioconcentration of MMHg+is influenced by cell-dependent factors, such as the thickness of the phytosphere, while this is not the case for Hg^{2+}." How does the thickness of the phytosphere affect the bioconcentration of MMHg+? Why is Hg2+ not affected by this?

Author Response

Thank you for this comment. There is a lot of uncertainty about these processes and they are somewhat out of the scope of this modeling paper. Therefore I will suggest to enhance the unclear sentence with the expanded sentence belows, in order to create more clarity about this interaction while not distracting from the focus of the paper

Suggested edit

However, the bioconcentration process is controlled by a variety of factors, and recent studies show that the bioconcentration of Hg^{2+} is constant when normalized for cell density, while the uptake of $MMHg^{+}$ is affected by changes in cell density and biomass. This suggests that $MMHg^{+}$ uptake is influenced by cell-dependent factors, such as the thickness of the phycosphere and the availability of transmembrane channels, while this is not the case for Hg^{2+} (Garcia-Arevalo et al., 2024).

Reviewer Comment

The references cited in the manuscript are somewhat outdated and may benefit from incorporating more recent studies to ensure the relevance and accuracy of the presented information.

Author Response

I agree that especially in the segment about other models, there are new advancements made that should indeed be incorporated, and the novelty of this model against these papers should be evaluated. I would suggest to rewrite this section (line 62) as follows:

Suggested edit

Multiple models have been developed to explain $MMHg^{+}$ bioaccumulation in marine ecosystems. Key examples include trophic transfer (Schartup et al., 2018), base-level accumulation (Zhang et al., 2020), planktonic bioaccumulation in the Mediterranean Sea (Rosati et al., 2022), MeHg dynamics on the Beaufort Shelf (Li et al., 2022), and speciation plus accumulation in the North and Baltic Seas (Bieser et al., 2023).

In all previous models, the bioconcentration of $MMHg^{+}$ is included because it is an essential driver. These models, however, do not include higher trophic level animals such as fish. It is concluded in Schartup et al. (2018) that the bioconcentration of $MMHg^{+}$ in zooplankton is not a major contributor and contributes less than 15% of total MeHg bioaccumulation. Consequently, in later models such as presented by Rosati et al. (2022) this interaction is not included because their model focuses on the base of the food web. The study performed by Li et al. (2022) includes the process of bioconcentration for invertebrates, but it is not included for vertebrates. This means that our model would be the first model to include bioconcentration at every trophic level.

The bioaccumulation of Hg^{2+} is much less studied and not incorporated in any of the above-mentioned models. This is because Hg^{2+} is much less toxic than $MMHg^{+}$ and therefore comparably understudied. While data is limited, this raises the speculative question if the link between the bioaccumulation Hg^{2+} and $MMHg^{+}$ is not underestimated as Hg^{2+} and $MMHg^{+}$ are in active equilibrium in the water.

The ECOSMO-MERCY coupled system, which is used by Bieser et al. (2023) and Amptmeijer et al. (2025) is the only coupled model that models the bioaccumulation of Hg^{2+} and $MMHg^{+}$ at higher trophic levels such as fish, while incorporating bioconcentration at every trophic level.

Author Response

Additionally, I would suggest to add the following statements based on modern references into the introduction. I would suggest to add the following statement to line 21:

Suggested edit

For example, it is estimated that the consumption of MeHg contaminated seafood contributed to 61,800 premature deaths and caused economic damage of up to 2.87 trillion USD (Chen et al., 2025). This issue is expected to become even more significant as antrhopgenic Hg emissions are projected to increase in the near future (Maria Brocza et al., 2024).

Author Response

And the below statement to line 45 in the manuscript:

Suggested edit

Additionally, it has been shown by Tesán-Onrubia et al. (2023) that plankton communities in the southern Mediteranean Sea have lower $MMHg^+$ concentration than plankton in the northern Mediterranean Sea, they linked this to changes in environmental conditions affecting bioconcentration.

Author Response

I also wanted to expand the theorical uncertainty of this interaction. Here I would also introduce some more up to date references. I would suggest to add the belows part to the model limitation section at line 302.

Suggested edit

The results of our model represent just one possible outcome based on a regional setup representing the North and Baltic Seas, and the importance of bioconcentration can vary greatly depending on the bioconcentration factors of all species in the trophic chain. We can asses expected range of importance of consumer level bioconcentration by developing theoretical maximum and minimum values based on observational studies. We can estimate that direct bioconcentration in zooplankton may account for up to 50%, based on Lee and Fisher (2017), and similarly for mid-trophic level fish, based on Wang and Wong (2003).
We can use this to estimate the maximum expected contribution of consumer level bioconcentration on bioaccumulation by making two assumptions: (1) bioconcentration in both copepods and fish lies between 0 and 50% and is equal across all trophic levels, and (2) the food chain is linear, meaning that trophic level 3 feeds exclusively on trophic level 2, which feeds exclusively on trophic level 1. Under these assumptions, we can estimate the percentage of $MMHg^+$ in the diet of a given trophic level that originated from bioconcentration in primary producers as:

$$\text{PBC\%}_n = (1 - \text{BC})^{n-1} \times 100\% \tag{1}$$

where:

- $\text{PBC\%}_n$ is the percentage of $MMHg^+$ in the diet of trophic level n that originates from bioconcentration at the primary producer level,
- BC is the fraction (0–1) of $MMHg^+$ at each trophic level originating from bioconcentration.

This is ofcourse a simplification. Huo et al. (2025) for example demonstrated that the

BMF varies with prey type, indicating that the % $MMHg^+$ from bioconcentration is not uniform across trophic levels. Furthermore, McClelland et al. (2024) observed higher $MMHg^+$ levels in animals feeding on benthic rather than pelagic invertebrates which is further supported by Liu et al. (2024) which found significantly higher $MMHg^+$ concentration in demersal fish compared to pelagic fish. Even if the BCF remains constant across different species, a higher BAF due to increased biomagnification would lessen the relative impact of bioconcentration. So the assumptions made in the above presented basic model are not true in nature, but despite this it can help us understand the strong importance of consumer level bioconcentration. This simplified model demonstrates that a 50% bioconcentration rate in trophic level 1, 2, and 3 leads to only 12.5% of $MMHg^+$ in the diet of a trophic level 4 fish being derived from primary producers, with the remaining 87.5% originating from consumer-level bioconcentration. Even a low estimate of 10% bioconcentration in trophic level 1,2, and 3 still results in 27.1% of $MMHg^+$ in the diet of trophic-level 4 fish originating from consumer-level bioconcentration.

The degree to which this interaction contributes to overall bioaccumulation depends on numerous additional factors that are not yet fully understood, including the size distribution of phytoplankton at the base of the food web, the trophic structure, consumer metabolic and respiration rates, and the assimilation efficiency of $MMHg^+$ from the diet. This complexity makes it difficult, if not impossible, to provide a definitive estimate of the importance of consumer-level bioconcentration and the uncertainity of the interaction. However, based on the bioconcentration rates provided in the current literature, we conclude that this process plays a key role in the bioaccumulation of $MMHg^+$ in higher trophic levels.

Reviewer Comment

The paper is very detailed about the basic knowledge and the content of the preliminary research, but the description of the later model establishment and the data obtained from the model is relatively brief, which can be further supplemented. For example, "The contribution of bioconcentration in zooplankton of 3.97-10.07%...... and the contribution of bioconcentration in fish between 8.14-21.82%......"

Author Response

Thank you for you comment. Based on suggestions of other researchers I have also expanded the interpretation of the results by analyzing the seasonality and performing sensitivity analyses. I would suggest to rewrite to start of the result (section 3 line 211) where I discuss the raw result. As below:

Suggested edit

The biocentration of $MMHg^+$

The direct contribution of bioconcentration varies greatly by functional group groups and setups. In zooplankton, the bioconcentration is minimally significant (4–10%), which is consistent with the model findings by Schartup et al. (2018) and the laboratory results by Lee and Fisher (2017), indicating that the bioconcentration in copepods represents up to 10% of total $MMHg^+$ bioaccumulation. In our model, the bioconcentration is highest in middle-level consumers: fish 1 (13–16%) and macrobenthos (14–25%), due to lower biological loss rates and relatively low dietary $MMHg^+$ concentrations. This is consistent with the study of (Wang & Wong, 2003) showing that bioconcentration accounts for 10–

50% of $MMHg^+$ in Sweetlips. In the modeled high-trophic level, fish 2, bioconcentration is a smaller relative component of total bioaccumulation, at 8—- 14%. Interestingly, mid-trophic level species show the highest relative bioconcentration contributions. The lower relative contribution in low-trophic-level animals is driven by a lower life expectancy and a higher organic turnover rate, which enhances the relative importance of dietary uptake. At the same time, the elevated $MMHg^+$ concentration in mid-trophic animals boosts dietary uptake in high-trophic level animals, compensating for reduced organic turnover in the top predators. On average of the three setups, the $MMHg^+$ uptake due to bioconcentratoin is 0.0053 ng Hg mg^{-1} for fish 1 and 0.0055 ng Hg mg^{-1} for fish 2. This means that fish 2 has a higher bioconcentration rate of $MMHg^+$ in absolute terms, but because its diet is richer in $MMHg^+$ compared to fish 1, the relative importance is lower.

Author Response

I would suggest to further enhance the result by discussion and visualing the seasonal component of the results as follows:

Suggested edit

Seasonality of the difference in $MMHg^+$ bioaccumulation

The seasonality of the difference in $MMHg^+$ bioaccumulation caused the bioaccumulation of Hg^{2+} and the bioconcentration of $MMHg^+$ in consumers is shown in Fig. 1. To reduce the influence of interannual variability, we calculated a multi-year average of daily mean values over the last 10 years of the simulation. For each calendar day (January 1st, January 2nd, etc.), the modeled daily values from each of the last 10 years of the simulations were averaged. The resulting time series represents an annual cycle of average daily conditions. From the producers functional groups only the diatoms are shown as the reaction is not group specific but rather caused by changes in dissolved Hg^{2+} and $MMHg^+$ which means the diffference caused for all phytoplankton groups was the same. This shows that, while the scale depends on the setup, there are interactions that consistently occur. In low trophic levels such as phytoplankotn and microzooplankton the bioaccumulation of Hg^{2+} causes a seasonal respone in the $MMHg^+$ bioaccumulation in phytoplankton which is consequently observable in low trophic level biota such as microzooplankton. While this reduction in $MMHg^+$ would compound into higher trophic levels, its effects in higher trophic level animals dwarves in comparison to the difference caused by incorporating the bioconcentration of $MMHg^+$ in consumers and it does not cause a difference larger than 3% in either fish 1 or fish 2 in any of the setups.

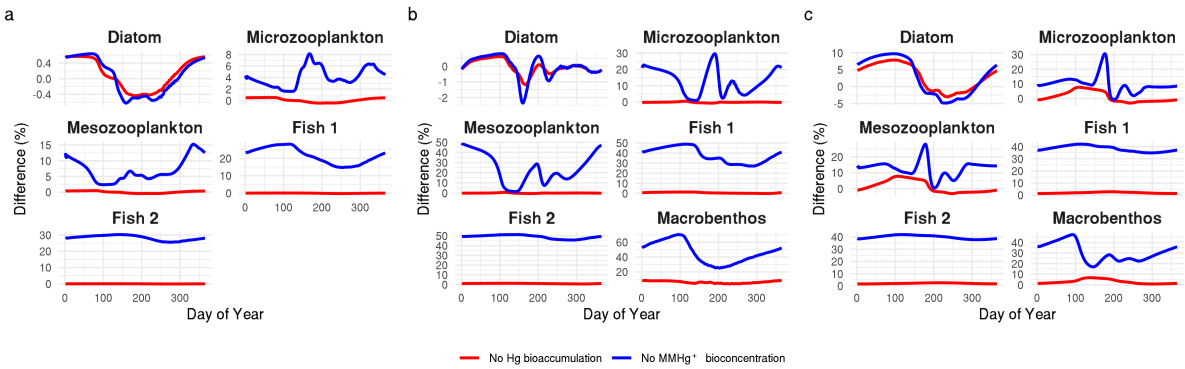

Figure 1: The seasonality of the difference in the bioaccumulation of $MMHg^{+}$ caused by the bioaccumulation of Hg^{2+} and the bioconcentration of MMHg in consumers for a) the Gotland Deep, b) the Northern North Sea and c) the Southern North Sea. In high trophic level such as fish 1 and fish 2 there is low seasonality and the effect of the bioconcentration of $MMHg^{+}$ in consumers is high while the effect of the bioaccumulation of Hg^{+} is low. In low trophic levels, notably diatoms and microzooplankton there is strong seasonal component. The bioaccumulation of $MMHg^{+}$ is up to 5% lower in diatoms in the Southern North Sea if the bioaccumulation of Hg^{2+} is modeled in late summer when biomass is high. But the bioaccumulation of Hg^{2+} does not lead to a notable ($> 5\%$) difference at any moment in fish.

Author Response

Then I would expand the method section to introduce the senstivity study to expand the results as follows:

Suggested edit

In order to further investigate how bioconcentration in consumers affects bioaccumulation of $MMHg^{+}$, we performed a sensitivity analysis on the key drivers: the bioconcentration rate of consumers and the bioaccumulation rate of producers. To this extent, two sensitivity studies are performed. In the first sensitivity study, the bioconcentration rate in all consumers is multiplied by a scaling factor that is between 0.2 and 2.0 with 0.2 intervals. The effect of this on the bioaccumulation in fish 2 for the Gotland Deep is shown to visualize the impact. Then the relative contribution of bioconcentration in consumers on the bioaccumulation of $MMHg^{+}$ in fish 2 is shown for all three setups. For the second sensitivity study, the same approach is used but the bioconcentration rate of producers is multiplied by a scaling factor.

Author Response

The results of this senstitivity study I would present in the results as follows:

Suggested edit

Sensitivy of the consumer bioconcentration rate

The results of the first sensitivity study, in which the bioconcentration rate of consumers is altered, are shown in Fig. 2. Figure 2a illustrates that the $MMHg^{+}$ contribution from bioconcentration in consumers is linearly related to the consumer bioconcentration rate scaling factor. Thus, altering the bioconcentration rate by half or double yields the same relative effect on fish 2's $MMHg^+$ content from direct bioconcentration. Based on Table 1, we can see that in the Gotland Deep, the difference between the simulation with and without consumer bioconcentration is 0.0183 ng Hg mgC^{-1}. This means that picking a bioconcentration double the real rate would result in a 0.0183 ng Hg mgC^{-1} overestimation of $MMHg^+$ bioaccumulation in fish 2, while selecting bioconcentration rates half the true values would result in a reduction of 0.00915 ng Hg mgC^{-1}. However, the relative contribution of bioconcentration to total $MMHg^+$ bioaccumulation follows a non-linear pattern, as shown in Fig. 2b. This non-linearity occurs because the total $MMHg^+$ in fish 2 is influenced by both bioconcentration in consumers and bioconcentration in producers. When the consumer bioconcentration scaling factor is 0, bioconcentration in consumers makes no contribution to fish 2's $MMHg^+$ levels. Conversely, this contribution can never reach 100% because bioconcentration in producers and consequent biomagnification from lower trophic levels always contributes to the total $MMHg^+$ burden in fish. In the same way as in the results shown in Table 1, the relative importance of bioconcentration is consequently highest in the Northern North Sea, followed by the Southern North Sea and lowest in the Gotland Deep.

Sensitivy of the producer bioconcentration rate

The results of the second sensitivity study are shown in Fig. 3. Here, rather than the consumer bioconcentration rate, the producers' bioconcentration rates are multiplied by a scaling factor. Again, the effect of this scaling on the bioaccumulation in all trophic levels is visualised in Fig. 3a, and the effect of this scaling on the relative importance of consumer bioconcentration on $MMHg^+$ bioaccumulation is shown in Fig. 3b. If the bioconcentration scaling factor is 0, there is still $MMHg^+$ bioaccumulation in fish 2, both from direct bioconcentration and from bioconcentration in consumers and consequent biomagnification. The increase in fish 2 $MMHg^+$ per step of 0.2 in the scaling factor is 0.0083 ± 0.00030 ng Hg mg^{-1}. The relative contribution of consumer bioconcentration on $MMHg^+$ bioaccumulation is shown in Fig. 3b. An important note here is that while we scaled the bioconcentration factor of producers and consumers, $MMHg^+$ can also be bioaccumulated via the partitioning to dissolved organic matter (DOM) detritus and consequent biomagnification. This is especially important in the Northern North Sea. In the seasonally stratified water column, macrobenthos cannot feed directly off the phytoplankton bloom; thus, the dying and sinking of particles is an important flux that is consumed by the benthos. Benthos, in turn, is an important food source for fish 2. So scaling the producer bioconcentration rate has less effect in the Northern North Sea. In the Gotland Deep, the opposite is true; because the deep water is anoxic, there is no macrobenthos in the model. This means that the entire ecosystem is pelagic and detritus is less important than direct consumption of the phytoplankton bloom.

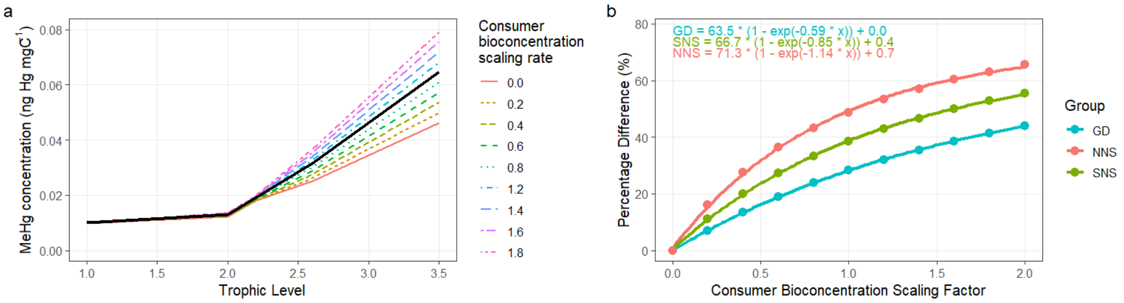

Figure 2: a) show the effect of the bioaccumulation of $MMHg^+$ per trophic level in the Gotland Deep. This shows an increase 0.0036 ± 0.00010 ng Hg mg^{-1} in fish 2 $MMHg^+$ bioaccumulation for every 0.2 step increase in the consumer bioconcentration scacling factor. 2b) shows the percentage difference due to bioaccumulation with different consumer bioconcentration scaling factors in all setups. GD refers to the Gotland Deep, NNS to the Northern North Sea and SNS to the Southern North Sea. When the consumers bioconcentration scaling factor is 0, the percentage difference due to bioconcentration is 0 %. As this increase the percentage increases. The relationship between the consumers bioconcentration factor and the percentage difference due to consumer bioconcentration is plotted assuming an saturating exponential relationship.

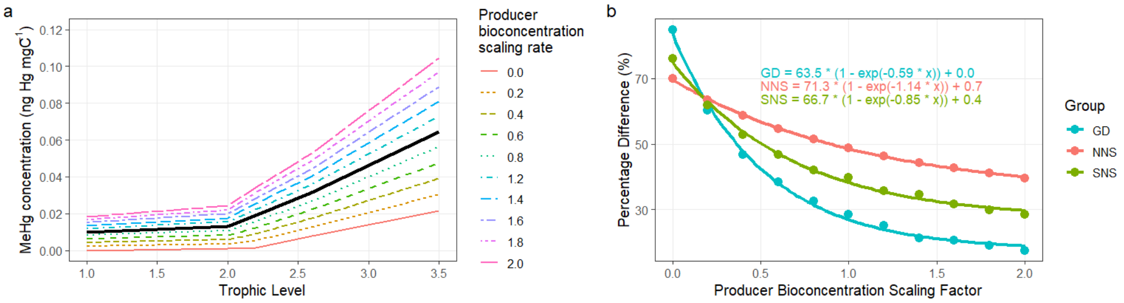

Figure 3: a) illustrates the influence of scaling the producers bioconcentration rate of $MMHg^+$ on the $MMHg^+$ bioaccumulation at each trophic level in the Gotland Deep. This shows an increase of 0.0084 ± 0.00032 ng Hg mg^{-1} in fish 2 $MMHg^+$ with every 0.2 increase in the producers scaling factor. 3b) highlights the difference due to consumer bioconcentration with different primary producers scalings factors across all setups. The relationship between producer bioconcentration scaling factor and the percentage difference in $MMHg^+$ bioaccumulation in fish 2 due to consumer bioconcentration is plotted using an expenontial decay function. This shows that in all cases the percentage difference is high when the producer bioconcentration factor is 0, and that this percentages decreases with an increasing scaling factor.

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