Author Response

Thank your excellent feedback. Below, I will discuss how I would implement all of the suggested comments.

Reviewer Comment

This study employed different models to identify the contributions of bioconcentration and biomagnification to Hg and MeHg bioaccumulation. The primary concern is that the description of the model applied and the data sources lack clarity.

Author Response

Before going into the specific comments, I would answer your general feedback. The model does not use field data and is solely based on a model. The core model cited is presented in Amptmeijer et al. (2025). Which is discussed and evaluated here in more detail: https://doi.org/10.5194/egusphere-2025-1486 The model presented by Amptmeijer et al. (2025) is then used to run with and without bioaccumulation of Hg^{2+} and consumer-level bioconcentration of $MMHg^{+}$ to estimate the importance of these interactions. As such, the core message of the paper is aimed at showing the importance of these interactions on the outcome of the model, which shows that these interactions should be included in MMHg+ bioaccumulation models. I would expand the Methods section to include the exact bioaccumulation equations used as follows:

Suggested edit

Used terminology: bioaccumulation, bioconcentration, biomagnification, and *in vivo* Hg methylation

Bioaccumulation in the marine environment refers to the total increase in pollutants in biota compared to that in the water. This can be quantified in nature by measuring the concentration of pollutants in both water and biota and estimating the difference. This is typically expressed as the bioaccumulation factor, BAF. For example, the bioaccumulation of $MMHg^{+}$ in organisms i can be calculated based on observations as:

$$\mathrm{BAF}_i^{MMHg^+} = \frac{C_i^{MMHg^+}}{C_w^{MMHg^+}} \tag{1}$$

In which,

$$\begin{aligned}
\mathrm{BAF}_i^{\mathrm{MMHg}^+} &= \text{The bioaccumulation factor of } \mathrm{MMHg}^+ \text{ for organism } i\,[\mathrm{L}\cdot\mathrm{kg}^{-1}] \\
C_i^{\mathrm{MMHg}^+} &= \text{The concentration of } \mathrm{MMHg}^+ \text{ in organism } i\,[\text{ng Hg}\cdot\mathrm{kg}^{-1}] \\
C_w^{\mathrm{MMHg}^+} &= \text{The free concentration of } \mathrm{MMHg}^+ \text{ in water}\,[\text{ng Hg}\cdot\mathrm{L}^{-1}]
\end{aligned}$$

Since the BAF can be based on field measurements, it is a commonly used metric to estimate the link between the concentrations of pollutants in seawater and those in biota. In this study, we are interested in separating the bioaccumulation into separate pathways: the direct uptake from the water (bioconcentration) and the increase in pollutants due to trophic interactions (biomagnification).

Bioconcentration, is the increase in the concentration of Hg in biota directly due to uptake from the water. Because the process of bioconcentration relies on the exchange of Hg between the dissolved phase and an organism, it depends on the surface area of the organic material that is in contact with the water. Due to this, small organisms, such as

bacteria and phytoplankton, have a greater ability to bioconcentrate Hg (Mason et al., 1996; Pickhardt et al., 2006). However, 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 phycosphere and the availability of transmembrane channels, while this is not the case for Hg^{2+} (Garcia-Arevalo et al., 2024). Bioconcentration is typically defined by the bioconcentration factor (BCF). The BCF for $MMHg^+$ in organism i, can for example be calculated as

$$\mathrm{BCF}_i^{Hg} = \frac{BC_i^{Hg}}{C_w^{Hg}} \tag{2}$$

In which,

$\mathrm{BCF}_i^{\mathrm{Hg}}$ = The bioconcentration factor of Hg for organism i $[\mathrm{L} \cdot \mathrm{kg}^{-1}]$
BC_i^{Hg} = The concentration of Hg in organism i due to direct bioconcentration $[\mathrm{ng\ Hg} \cdot \mathrm{kg}^{-1}]$
C_w^{Hg} = The free concentration of Hg in the water $[\mathrm{ng\ Hg} \cdot \mathrm{L}^{-1}]$

Here, Hg could either refer to Hg^{2+} or $MMHg^+$. Note that this defines the theoretical BCF. In nature it is typically impossible to directly measure the BCF, as it would be impossible to separate between $MMHg^+$ that is taken up directly from the water and $MMHg^+$ that is ingested via food. Bioconcentration is the most important step in bioaccumulation and phytoplankton can have a BCF of $MMHg^+$ between 2E4 L kg^{-1} and 6.4E6 L kg^{-1} (Gosnell & Mason, 2015).

Biomagnification is when $MMHg^+$ reaches higher concentrations at progressively higher trophic levels. The biomagnification factor, the fractional increase in $MMHg^+$ with each trophic level, is estimated to be 7.0 ± 4.9 (Harding et al., 2018; Lavoie et al., 2013). This means that in addition to the high increase in $MMHg^+$ in phytoplankton, there is a large increase in $MMHg^+$ at every consecutive trophic level. Many seafoods consist of high-trophic animals, such as cod, tuna, or marlin, which can have trophic levels between 4 and 4.8 (Nilsen et al., 2008; Sarà & Sarà, 2007). Biomagnification can increase the already high levels of $MMHg^+$ in phytoplankton by up to another factor $11.9^{4.8} \approx 145420$. This is typically defined by the biomagnification factor, BMF, which can be calculated assuming steady state for organism i, preying on organism j for $MMHg^+$ as:

$$\mathrm{BMF}_{i,j}^{Hg} = \frac{C_i^{Hg}}{C_j^{Hg}} \tag{3}$$

In which,

$\mathrm{BMF}_{ij}^{\mathrm{Hg}}$ = The biomagnification factor for trophic consumption of organism j by i [unitless]
C_j^{Hg} = The concentration of Hg in organism j $[\mathrm{ng\ Hg} \cdot \mathrm{kg}^{-1}]$
C_i^{Hg} = The concentration of Hg in organism i $[\mathrm{ng\ Hg} \cdot \mathrm{kg}^{-1}]$

The biomagnification factor of $MMHg^+$ is extremely high, Lavoie et al. (2013) estimates the diet-weighted average BMF for $MMHg^+$ as 8.1 ± 7.2 while it is only 4.7 ± 4.7 for Hg^{2+}. This combined with the higher toxicity of $MMHg^+$ is the reason why the bioaccumulation of $MMHg^+$ is of much higher concern than the bioaccumulation of Hg^{2+}.

In vivo methylation occurs when animals take other forms of Hg and transform it into $MMHg^+$ in organisms. Although the existence of this process has been demonstrated in specific organisms such as cuttlefish, it is poorly understood and only recently gaining

attention (Gente et al., 2023). There is no direct evidence of in vivo methylation in the animals that we model, so it is not implemented in this model.
Overall the dominant pathway of the bioaccumulation of $MMHg^+$ is the bioconcentration of $MMHg^+$ in phytoplankton and consequent biomagnification. The important route is quantified by Wu et al. (2019) using a meta-analysis. They find that the concentration of MeHg at the base of the food web predicts 63% of the observed variability in high trophic level fish, while the remaining 37% is controlled by factors such as the dissolved organic matter content and oligotrophy.

Author Response

I would suggest to expand section 2.2 in the methods section (line 184) to explain the exact equations used in this paper to asses bioconcentration and bioaccumulation.

Suggested edit

The implementation of bioaccumulation is discussed and validated in more detail in Amptmeijer et al. (2025), but the core equations are discussesed here as well for clarity.
The increase in bioconcentrated pollutant (Hg^{2+} or $MMHg^+$) per day for a functional group is calculated based on the biomass concentration of the group, the uptake rate, and the concentration of the pollutant, while it is reduced with a rate that is the sum of the release rate of the pollutant and the loss of biomass from group g, from both biologicall loss (respiration and mortality) and predation. The change in pollutant p due to bioaccumulation can then be calculated using the following equation:

$$\frac{dC^{BC}_{g,p}}{dt} = b_g \cdot C^{env}_p \cdot r^{bc}_{g,p} - C^{BC}_{g,p} \cdot (r^{rel}_{g,p} + r^{bl}_g + \sum_{z=1}^{n_z} r^{pred}_{z,g}) \tag{4}$$

$C^{BC}_{g,p}$ = Bioconcentrated pollutant p in group g [ng Hg m^{-3}]
b_g = Biomass of functional group g [mgC m^{-3}]
C^{env}_p = Environmental concentration of pollutant p [ng Hg m^{-3}]
$r^{bc}_{g,p}$ = Bioconcentration rate for group g and pollutant p [ng Hg mgC^{-1} d^{-1}]
$r^{rel}_{g,p}$ = Release rate of pollutant p from group g [d^{-1}]
r^{bl}_g = Biological loss rate for group g (mortality, respiration) [d^{-1}]
$r^{pred}_{z,g}$ = Predation rate by predator z on group g [d^{-1}]
n_z = Number of consumer groups feeding on group g
z = Index for consumer groups (predators) of g
t = Time [d]

While the change in pollutant p due to biomagnfication is also dependent on the predation and concentration of pollutants from both bioconcentration and biomagniciation in the prey. Additionally pollutant p is released via the turnover rate rather than the release rate as is the case for bioconcentration, the change in pollutant p due to biomagnification can then be calculated as follows:

$$\frac{dC^{BM}_{g,p}}{dt} = \sum_{s=1}^{n_s} (r^{pred}_{g,s} \cdot a_{s,p} \cdot (C^{BC}_{s,p} + C^{BM}_{s,p})) - C^{BM}_{g,p} \cdot (r^{to}_{p,g} + r^{bl}_g \sum_{z=1}^{n_z} r^{pred}_{z,g}) \tag{5}$$

So the total concentration of pollutant P in ng Hg m^{-3} is:

$$C_{(g,p)} = C^{BC}_{(g,p)} + C^{BM}_{(g,p)} \tag{6}$$

Since this tracks the pollutants per volume of water, the total bioaccumulation per biomass in ng Hg mgC^{-1} is then calculated as

$$C^{bg}_{(g,p)} = \frac{C_{(g,p)}}{b_g} \tag{7}$$

This is then converted to the bioaccumulation per dry weight based on an assumed ratio of carbon to dry weight of 0.2 for diatoms, 0.33 for flagellates and cyanobacteria, and 0.5 for zooplankton and fish based on Walve and Larsson (1999) and Sicko-Goad et al. (1984).

Author Response

In addition, I would provide the key take conclusion of the model evaluation of this paper.I would place this after section 2.2 so at line 196 in the manuscript.

Suggested edit

Performance of the GOTM-ECOSMO-MERCY model

The model is generally consistent with observational data and the previously validated 3D ECOSMO E2E model in terms of biomass. Minor exception are that the Chlorophyll-a concentration in the Gotland Deep matches the Northern instead of the Central Baltic Sea, and that the fish biomass in the Gotland Deep is overestimated by 7% compared to Thurow (1997). The model also predicts tHg content and Hg^{2+} and $MMHg^{+}$ levels in phytoplankton, zooplankton and fish 1 accurately, with MMHg+ bioaccumulation corresponding well with trophic interactions. A deviation is seen in the trophic level fish 2, which has a trophic level of 3.5–3.7 in the model, below the expected level for Atlantic Cod (4.0—4.2). Nonetheless, this level remains high making fish 2 representative of a high trophic level animals. The $MMHg^{+}$ bioaccumulation in fish 2 is consistent with the observed bioaccumulation for its trophic level. Witthout the above discused minor exceptions, the model simulates biomass, Hg speciation and bioaccumulation in line with obsersations.

Reviewer Comment

Lines 6-9: This sentence is too long and not clear to me

Author Response

I would suggest that I rewrite the sentence as follows:

Suggested edit

In this study, we use a fully coupled 1D water column Hg bioaccumulation model to quantify how total bioaccumulation of Hg^{2+} and uptake of $MMHg^{+}$ from the water (bioconcentration) in consumers affects the bioaccumulation of $MMHg^{+}$ in high trophic level fish. The study is performed in three setups representing hydrodynamic conditions representative of the North and Baltic Seas.

Reviewer Comment

Line 110: Descriptions about the Modeled region in the Introduction section is weird. I suggest moving it to the MM section.

Author Response

I can move the modeled regions section to the beginning of the introduction. I would put it at the beginning at line 149.

Reviewer Comment

The model section is not clear to me. How to divide bioconcentration and biomagnification. Is there any data collected from in-lab measurements?

Reviewer Comment

Table 1: What is the source of the data provided in this table?

Author Response

I would address these comments together as they addres the same issue. This study is purely model-based. Of course, previously published data collected from in-lab measurements are used to estimate the bioconcentration rates and assimilation efficiency, which drive the processes in our model. This is discussed in detail in the referenced model paper Amptmeijer and Bieser, 2025. Which is available here in preprint: https://doi.org/10.5194/egusphere-2025-1486. I agree that we underexplained the difference between bioconcentration and biomagnification and how this is done in our model. I hope the suggested expansion of the metods section described above adresses this concern. Additionally I would suggest to add the below statement at the beginning of the results section at line 210 to make sure there is no ambiguity about the source of the data.

Suggested edit

The results are presented in Table 1. All results are derived from model simulations. To quantify the influence of consumer level bioconcentration and and bioaccumulation of Hg^{2+} on $MMHg^{+}$ bioaccumulation, the model was run under scenarios with and without bioaccumulation of Hg^{2+} and with and without consumer-level bioconcentration of $MMHg^{+}$