Introduction

Below I will describe for each of the reviewers' comments how they were implemented. However, several reviewers asked for similar changes, of which some resulted in the rewriting of sections of the manuscript. I will post the rewritten manuscript sections here above and then reference that in the specific reference answers to not have to overinflate the document by posting the same rewritten sections several times.

Implementation

Line 41

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

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 calculating the ratio. 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,

$$\mathrm{BAF}_i^{\mathrm{MMHg}^+} = \text{The bioaccumulation factor of } \mathrm{MMHg}^+ \text{ for organism } i\,[\mathrm{L}\cdot\mathrm{kg}^{-1}] \tag{2}$$

$$C_i^{\mathrm{MMHg}^+} = \text{The concentration of } \mathrm{MMHg}^+ \text{ in organism } i\,[\text{ng Hg}\cdot\mathrm{kg}^{-1}] \tag{3}$$

$$C_w^{\mathrm{MMHg}^+} = \text{The free concentration of } \mathrm{MMHg}^+ \text{ in water}\,[\text{ng Hg}\cdot\mathrm{L}^{-1}] \tag{4}$$

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. Because of 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 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). Bioconcentration is typically defined by the bioconcentration factor (BCF). The BCF for $MMHg^+$ in organisms i can be calculated as

$$\mathrm{BCF}_i^{MMHg^+} = \frac{BC_i^{MMHg^+}}{C_w^{MMHg^+}} \tag{5}$$

In which,

$$\mathrm{BCF}_i^{\mathrm{MMHg}^+} = \text{The bioconcentration factor of } \mathrm{MMHg}^+ \text{ for organism } i\,[\mathrm{L}\cdot\mathrm{kg}^{-1}] \tag{6}$$

$$BC_i^{\mathrm{MMHg}^+} = \text{The concentration of } \mathrm{MMHg}^+ \text{ in organism } i \text{ due to direct uptake from water}\,[\text{ng Hg}\cdot\mathrm{kg}^{-1}] \tag{7}$$

$$C_w^{\mathrm{MMHg}^+} = \text{The free concentration of } \mathrm{MMHg}^+ \text{ in the water}\,[\text{ng Hg}\cdot\mathrm{L}^{-1}] \tag{8}$$

Here, Hg could either refer to Hg^{2+} or $MMHg^+$. Note that for consumers this would define the theoretical BCF. In nature it is typically only possible to measure the BCF in primary producers, as in consumers 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 $2 \cdot 10^4$ L kg^{-1} and $6.4 \cdot 10^6$ 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 concentration 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:

$$\mathrm{BMF}_{i,j}^{MMHg^+} = \frac{C_i^{MMHg^+}}{C_j^{MMHg^+}} \tag{9}$$

In which,

$$\mathrm{BMF}_{ij}^{\mathrm{MMHg}^+} = \text{The biomagnification factor for trophic consumption of organism } j \text{ by } i\,[\text{unitless}] \tag{10}$$

$$C_j^{\mathrm{MMHg}^+} = \text{The concentration of } \mathrm{MMHg}^+ \text{ in organism } j\,[\text{ng Hg}\cdot\mathrm{kg}^{-1}] \tag{11}$$

$$C_i^{\mathrm{MMHg}^+} = \text{The concentration of } \mathrm{MMHg}^+ \text{ in organism } i\,[\text{ng Hg}\cdot\mathrm{kg}^{-1}] \tag{12}$$

The biomagnification factor of $MMHg^+$ is extremely high, Lavoie et al. (2013) estimates the diet-weighted average BMF in marine samples for $MMHg^+$ as 7.0 ± 4.9 while it is below 1 for iHg in most cases (Lavoie et al., 2013; Seixas et al., 2014). 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* Hg speciation** occurs when Hg is transformed from one form of Hg, such as $MMHg^+$ into another form of Hg, such as Hg^{2+} in organisms. Although the existence of this process has been demonstrated in specific organisms such as cuttlefish, it is poorly understood and only recently gained 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. But the relevance of *in vivo* Hg speciation cannot be excluded.

Overall the dominant pathway of bioaccumulation, the bioaccumulation of $MMHg^+$ is the bioconcentration of $MMHg^+$ in phytoplankton and consequent biomagnification. The importance of this 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 (DOM) content and oligotrophy.

Implementation

Line 108

Current models

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 and bioaccumulation in the North and Baltic Seas (Bieser et al., 2023).
In all of the previous models, bioconcentration of $MMHg^+$ is included as it is an essential driver. It is concluded in Schartup et al. (2018) that the bioconcentration of $MMHg^+$ in zooplankton 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) 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. The version used by Amptmeijer et al. (2025) expands on this by adding a higher-trophic-level fish. Because of this, the ECOSMO-MERCY coupled system, as described by Amptmeijer et al. (2025) is used in this analysis.

Implementation

Line 260

The implementation of bioaccumulation is discussed and validated in more detail in Amptmeijer et al. (2025), but the core equations are discussed 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 biological loss (respiration and mortality) and predation. The change in pollutant p due to bioaccumulation can then be calculated using the following equation:

$$\frac{dC_{g,p}^{BC}}{dt} = b_g \cdot C_p^{env} \cdot r_{g,p}^{bc} - C_{g,p}^{BC} \cdot (r_{g,p}^{rel} + r_g^{bl} + \sum_{z=1}^{n_z} r_{z,g}^{pred}) \tag{13}$$

$C_{g,p}^{BC}$ = Bioconcentrated pollutant p in group g [ng Hg m^{-3}]
b_g = Biomass of functional group g [mgC m^{-3}]
C_p^{env} = Environmental concentration of pollutant p [ng Hg m^{-3}]
$r_{g,p}^{bc}$ = Bioconcentration rate for group g and pollutant p [m^3 mgC^{-1} d^{-1}]
$r_{g,p}^{rel}$ = Release rate of pollutant p from group g [d^{-1}]
r_g^{bl} = Biological loss rate for group g (mortality, respiration) [d^{-1}]
$r_{z,g}^{pred}$ = 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 biomagnification is also dependent on the predation and concentration of pollutants from both bioconcentration and biomagnification 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:

$$\frac{dC_{g,p}^{BM}}{dt} = \sum_{s=1}^{n_s}(r_{g,s}^{pred} \cdot a_{s,p} \cdot (C_{s,p}^{BC} + C_{s,p}^{BM})) - C_{g,p}^{BM} \cdot (r_{p,g}^{to} + r_g^{bl} + \sum_{z=1}^{n_z} r_{z,g}^{pred}) \quad (14)$$

$C_{g,p}^{BM}$ = Pollutant p concentration in group g from biomagnification [ng Hg m^{-3}]
n_s = Number of prey groups consumed by g
s = Index for prey functional groups of g
$r_{g,s}^{pred}$ = Predation rate of group g on prey group s [d^{-1}]
$a_{s,p}$ = Assimilation efficiency of pollutant p from prey s [unitless]
$C_{s,p}^{BC}$ = Pollutant p concentration in group s from bioconcentration [ng Hg m^{-3}]
$C_{s,p}^{BM}$ = Pollutant p concentration in group s from biomagnification [ng Hg m^{-3}]
$r_{p,g}^{to}$ = Turnover rate of pollutant p in group g [d^{-1}]
r_g^{bl} = Biological loss rate for group g [d^{-1}]
$r_{z,g}^{pred}$ = Predation rate of predator z on group g [d^{-1}]

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

$$C_{(g,p)} = C_{(g,p)}^{BC} + C_{(g,p)}^{BM} \quad (15)$$

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

$$C_{(g,p)}^{bg} = \frac{C_{(g,p)}}{b_g} \quad (16)$$

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).

Implementation

Line 330

Sensitivity analyses

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.

Implementation

Line 335

Results and discussion

The model output is shown in Table 1. The % bioconcentrated is calculated as Bioconcentrated (%) $= \frac{\text{Bioconcentrated}}{\text{Bioaccumulated}}$ and the difference (%) is calculated as Difference (%) $= \frac{\text{Scenario}}{\text{Default}}$. The thick values in the difference category indicate when the scenario causes a change larger than 10%. The values are based on the last 10 years of the simulation and the top 20m of the water column, to create an average value that we can compare between the setups.

Bioaccumulation of Hg^{2+}

The effect of Hg^{2+} bioaccumulation on the bioaccumulation of $MMHg^+$ is shown in Table 1. All results are derived from model simulations. To quantify the influence of consumer level bioconcentration 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 $MMHg^+$. The differences are low between 1% and -6%. This is statistically evaluated, and the results are shown in Table 2. Wilcoxon's signed rank test shows that bioaccumulation of Hg^{2+} has no significant impact on the bioaccumulation of $MMHg^+$ ($p = 0.67$). Furthermore, the Bayesian t-test shows that the data are 2.9 times more likely under the null hypothesis of no effect than under the alternative hypothesis. This shows that Hg^{2+} bioaccumulation does not have a significant effect on $MMHg^+$ bioaccumulation (BF_{10}=0.40).

Bioaccumulation of $MMHg^+$

The $MMHg^+$ bioaccumulation for all biota functional groups in the different setups and scenarios and the percentage of bioaccumulated $MMHg^+$ originating from bioconcentration are shown in Table 1. These results show that the relative contribution of bioconcentration on the $MMHg^+$ content is low in microzooplankton (4–6%) while it is higher in mesozooplankton (5–10%) higher in fish 1 (13–22%), while lower in fish 2 (8–14%) and higher in macrobenthos (14–25%). The relative contribution of direct bioconcentration on

the $MMHg^+$ bioaccumulation in zooplankton, especially microzooplankton, is lower than in higher trophic levels of animals. In our model, this occurs because of the extremely high turnover rate of zooplankton. This "grow fast, die young" approach results in less $MMHg^+$ bioconcentration with higher relative contributions due to feeding caused by the high feeding rate of zooplankton.
In longer-lived fish, we see higher contributions of bioconcentration. Although these contributions are higher, they align with the experiments of (Wang & Wong, 2003) and the observations of 15% by Hall et al. (1997). Both fish 1 and fish 2 have the same bioconcentration and release rates, so it is in line with expectations that the relative contribution of direct bioconcentration in fish 2 is lower than in fish 1 since it gets more $MMHg^+$ from its higher trophic level diet.
There is a great difference in the importance of bioconcentration of $MMHg^+$ in macrobenthos between the Southern and Northern North Sea. This difference is especially notable in the direct bioconcentration in macrobenthos, which is 25% of the total bioaccumulated $MMHg^+$ in the Northern North Sea and only 14% in the Southern North Sea. This difference is caused by the low intake of $MMHg^+$ from food by macrobenthos in the Northern North Sea. Since the water column is stratified during spring and summer, macrobenthos cannot directly feed on the phyto- and zooplankton bloom. Because of this, they are dependent on sinking detritus. The detritus has a lower $MMHg^+$ content than living material and consequently, the $MMHg^+$ intake in Northern North Sea macrobenthos is lower, and thus the relative importance of bioconcentration is higher.

Implementation

Line 378

Sensitivity analyses

Sensitivity 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. 3. Figure 3a illustrates that the $MMHg^+$ contribution from bioconcentration in consumers is linearly related to the consumer bioconcentration rate scaling factor. Thus, in bioaccumulation modeling, 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 parameterizing a bioconcentration rate 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. 3b. 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 **??**, 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. 2. 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. 2a, and the effect of this scaling on the relative importance of consumer bioconcentration on $MMHg^+$ bioaccumulation is shown in Fig. 2b. 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. 2b. 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 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.

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. **??**. For each calendar day (January 1^{st}, January 2^{nd}, 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 difference 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 phytoplankton and microzooplankton, the bioaccumulation of Hg^{2+} causes a seasonal response 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 dwarf 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.

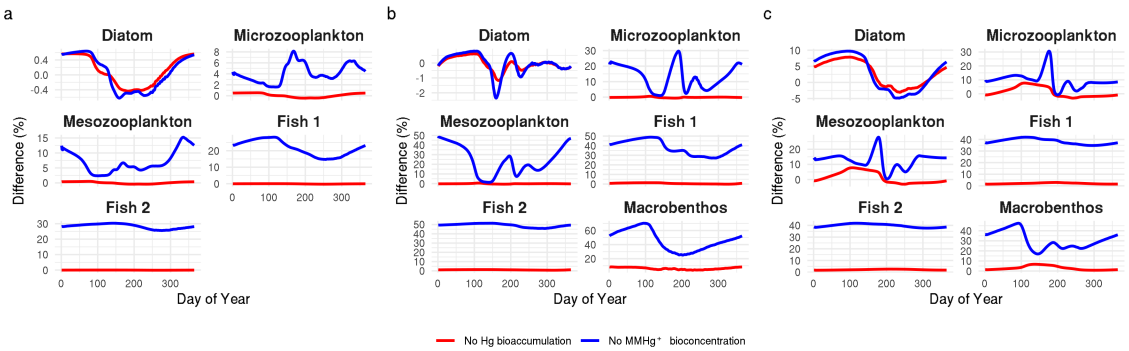

Figure 1: a) show the effect of the bioaccumulation of $MMHg^{+}$ per trophic level in the Gotland Deep. This shows an increase 0.0036 ± 0.00010 ng Hg mg^{-1} in fish 2 $MMHg^{+}$ bioaccumulation for every 0.2 step increase in the consumer bioconcentration scacling factor. 3b) shows the percentage difference 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.

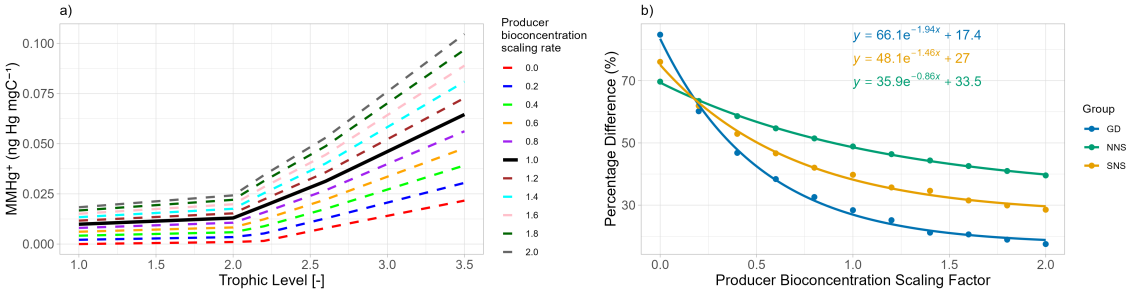

Figure 2: a) illustrates the influence of scaling the producers bioconcentration rate of $MMHg^{+}$ on the $MMHg^{+}$ bioaccumulation at each trophic level in the Gotland Deep. This shows an increase of 0.0084 ± 0.00032 ng Hg mg^{-1} in fish 2 $MMHg^{+}$ with every 0.2 increase in the producers scaling factor. 2b) highlights the relative significance of bioconcentration across all setups. The relationship between producer biococnentration 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.

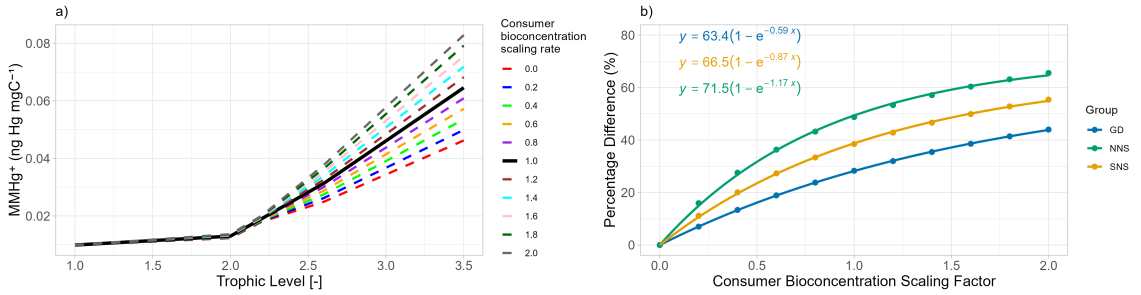

Figure 3: a) show the effect of the bioaccumulation of $MMHg^+$ per trophic level in the Gotland Deep. This shows an increase 0.0036 ± 0.00010 ng Hg mg^{-1} in fish 2 $MMHg^+$ bioaccumulation for every 0.2 step increase in the consumer bioconcentration scacling factor. 3b) shows the percentage difference 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.

Implementation

Line 589

Uncertainty of the conclusion

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 assess the 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:

$$\mathrm{PBC}\%_n = (1 - \mathrm{BC})^{n-1} \times 100\% \tag{17}$$

where:

- $\mathrm{PBC}\%_n$ 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.

Although this is a simplification, it illustrates that a high bioconcentration estimate of 50% results in only 12.5% of $MMHg^+$ in the diet of a trophic level 4 fish originating from

bioconcentration in primary producers, meaning that 87.5% originates from consumer-level processes. Even a low estimate of 10% still results in 27.1% of $MMHg^+$ in the diet of the same high-trophic-level 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 uncertainty 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.

Implementation of the comments of reviewer 1

Reviewer Comment

"Line 40-50. The authors mentioned the "bioconcentration", "bioaccumulation" and "biomagnification". For instance, the statement "bioconcentration is the most important step in bioaccumulation" lacks a clear distinction from biomagnification, risking confusion for readers unfamiliar with the terminology. What's the differences between bioaccumulation and biomagnification? In the subsequent manuscript, these two words were also used in confusion. The authors should explain and clarify them. "

Author Response

This distinction should indeed have been clarified better, especially since it is essential for understanding the paper. We have expanded the text starting at line 41 as described above to ensure clarity of the terms used and describe them in detail. Then we added a segment at line 260 to describe in details the equations used in this study to model bioaccumulation, including the different equations for bioaccumulationg originating from bioconcentration of biomagnification.

Reviewer Comment

"Line 79-85. The second hypothesis is confused. This hypothesis lacks evidence and references, making it appears speculative."

Author Response

I agree that the second hypothesis lacked references. It is mostly supported by the work of Wu et al. (2019), so we rewrote it as below to increase clarity.

Implementation

Line 140

The majority of $MMHg^+$ present in higher trophic levels is derived from their dietary intake (Lavoie et al., 2013). It is often assumed that $MMHg^+$ bioconcentration is not crucial for its bioaccumulation at higher trophic levels based on results such as those presented by Schartup et al. (2018), it is, for example, omitted from several Hg cycling and bioaccumulation models such as the model presented by Rosati et al. (2022), or not incorporated into higher trophic levels, as is the case in the model presented by Li et al. (2022). However, this assumption overlooks that bioconcentration occurs at all levels of the trophic chain. For example, if microzooplankton and mesozooplankton acquire 5% of $MMHg^+$ through bioconcentration, mesozooplankton will have 5% less $MMHg^+$ from its diet, which consists of microzooplankton, and another 5% less due to the absence of bioconcentration, leading to a total reduction of 10%. **The second hypothesis is that $MMHg^+$ bioconcentration in consumers significantly elevates $MMHg^+$ levels at higher trophic levels.** This concept has been previously suggested and studied by Wu et al. (2019). Their research found that the BCF in fish spans 3 to 7 orders of magnitude and greatly differs across studied sites; yet, they did find a strong correlation between BCF and $MMHg^+$ concentration in fish. Thus, we are not the first to suggest that direct water uptake is a significant factor in $MMHg^+$ bioaccumulation; rather, this study extends this understanding by quantifying the role of bioconcentration in all consumers

on $MMHg^+$ bioaccumulation in fish at higher trophic levels.

Reviewer Comment

"Line 140: As mentioned, "Quantifying the importance of the bioaccumulation of Hg2+ and bioconcentration of MMHg+ in consumers on MMHg+ bioaccumulation". The authors should clarify whether prior models ignored multi-trophic bioconcentration, and then highlight the novelty of this work."

Author Response

The section described above from line 108 has been updated to give a fair summary of previously published models and how they incorporated bioaccumulation and why the modelling framework used is selected.

Reviewer Comment

"Line 226 Table 1. How to calculate the bioaccumulation and bioconcentration difference (%) ?

Author Response

We added additional formula on line 339 and in the caption of Table 1.

Implementation

Line 339

The % bioconcentrated is calculated as Bioconcentrated (%) $= \frac{\text{Bioconcentrated}}{\text{Bioaccumulated}} * 100\%$ and the difference (%) is calculated as Difference (%) $= \frac{\text{Scenario}}{\text{Default}} * 100$. The values in red in the difference category indicate when the scenario causes a change larger than 10%. The values are based on the last 10 years of the simulation and the top 20m of the water column, to create an average value that we can compare between the setups.

Implementation

Caption of Table 1

The bioaccumulated $MMHg^+$, the percentage of bioaccumulated $MMHg^+$ that originates from bioconcentraton, and the bioaccumulated $MMHg^+$ in the scenario without bioaccumulation of Hg^{2+} and the bioconcentration of $MMHg^+$ in consumers and the difference to the default scenario. The % bioconcentrated is calculated as Bioconcentrated (%) $= \frac{\text{Bioconcentrated}}{\text{Bioaccumulated}} * 100$ and the difference (%) is calculated as Difference (%) $= (1 - \frac{\text{Scenario}}{\text{Default}}) * 100$.

Reviewer Comment

Line 316 "15% per trophic level". It is recommended to supplement sensitivity analyses.

Author Response

We supplemented the paper with a sensitivity study as described above. It is introduced in line 330 in the Methods sections and the results are shown in line 378 in the result section.

Implementation of the comments of reviewer 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

We have changed that sentence as described below:

Implementation

Line 15
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 to form 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

That is indeed incorrect, and we corrected that as described below:

Implementation

Line 29
However, given the rapid photodegradation of DMHg in natural water and that it is generally not assumed to bioaccumulate, DMHg is assumed not to significantly bioaccumulate in biota in the coastal area investigated in this study (Morel et al., 1998; 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

There is a lot of uncertainty about these processes and they are somewhat out of the scope of this modeling paper. Therefore I will 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

Implementation

Line 56
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

The sections described above (Especially the implementation at line 108) have updated rerences. Additionally some updated references are added below.

Implementation

Line 19
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).
Line 34
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.

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

The results and discussion are rewritten as is shown above starting from (the segment from line 335) to be more preciese about the results. Additionally, in the 376 the seasonality from the modeled data is also plotted and evaluated to further expand on evaluating the model data.

Implementation of the comments of reviewer 2

Reviewer Comment

- Section 2.2: Are there any equations for the parameterization schemes in the bioaccumulation of Hg in the model? Model equations are important for understanding the critical processes of the substance. Meanwhile, what are the critical parameters and coefficients for the critical processes in the model? The details of this model are not clarified in the method.
- Significantly, model performance should be evaluated against observations, which are deficient in this study. The literature Amptmeijer et al. (2025) is very important for this study. However, we cannot access to the paper because it is in preparation.

Author Response

My apologies. The publication of a key paper for this model was delayed. These details of this model are referenced in Amptmeijer et al. 2025, which is currently available with https://doi.org/10.5194/egusphere-2025-1486 I believe this is the main concern of the 3 points above. Here, the key equations and validation of both carbon fluxes and bioaccumulation are discussed in more detail. I addition to this paper now beign available I would discuss in more detail exactly which equations we use. This is described at the beginning of this document, and starting from line 38 to clarify the terminology and 260 to clarify the equations in the updated manuscript. Additionally, the below described summary of the performance of the model from Amptmeijer et al. (2025) is added on line 302.

Implementation

Line 302

Performance of the GOTM-ECOSMO-MERCY model

The model performance is discussed in more detail in Amptmeijer et al. (2025), but the key metrics are summarized below. The model is generally consistent with observational data and the previously validated 3D ECOSMO E2E model in terms of biomass. Minor exceptions 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 in phytoplankton, zooplankton, and fish 1 accurately, and the $MMHg^{+}$ bioaccumulation in fish corresponds well with trophic interactions. A deviation is seen in the trophic level fish 2, which has a trophic level between 3.5 and 3.7 in the model, below the expected level for Atlantic Cod (between 4.0 and 4.2). Nonetheless, this level remains high, making fish 2 representative of high-trophic-level animals. The $MMHg^{+}$ bioaccumulation in fish 2 is consistent with the observed bioaccumulation for its trophic level. Thus with the above-discussed minor exceptions, the model simulates biomass, Hg speciation, and bioaccumulation in line with observations.

Reviewer Comment

The setup of the two scenarios is a sample. In my opinion, sensitivity analysis for critical parameters or uncertainty analysis of the results is needed.

Author Response

I agree that the results of the paper where too limited and underexplored. As described above, in order support the manuscript a sensitivity study was performed. The text is at the top of the document, but the results of the senstivity and study start form line 378 in the manuscript.

Reviewer Comment

For the results and discussion, the illustrations are concise, and I cannot gain much in-depth discussion and thinking.

Author Response

In addition to the sensitivy study we also performed a seasonality analyses on the model data (at line 386 in the manuscript) and expanded the discussion starting form line 589 as is described above. I hope this provides a deeper layer of desired depth to the manuscript.

Implementation of the comments of reviewer 4

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. The method section is expanded, notably from line 41 to better describe the used terminology and from line 260 to show in detail the equations used in this study. I hope thas makes clarifies the model descriptions.

Reviewer Comment

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

Implementation

Line 6
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.

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

Moved this to the Methods section at **line** 183

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 expansion of the methods section starting at line 41 in the manuscript and 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.

Implementation

Line 343

Results and discussion

The model output is shown in Table 1. All results are derived from model simulations. To quantify the influence of consumer-level bioconcentration 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 $MMHg^{+}$. The % bioconcentrated is calculated as Bioconcentrated (%) $= \frac{\text{Bioconcentrated}}{\text{Bioaccumulated}} * 100\%$ and the difference (%) is calculated as Difference (%) $= \frac{\text{Scenario}}{\text{Default}}$. The thick values in the difference category indicate when the scenario causes a change larger than 10%. The values are based on the last 10 years of the simulation and the top 20m of the water column, to create an average value that we can compare between the setups.