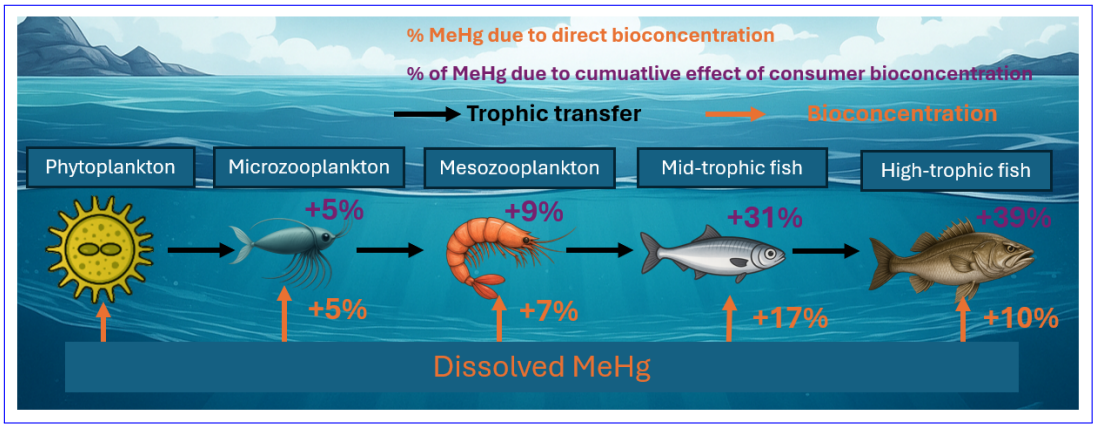

Figure 1. While the direct contribution of bioconcentration in consumers, the cumulative effect of bioconcentration in consumers increases with trophic level. This image consists of several sub images that where generated using GPT 4.1 and openArt.

Bioconcentration as a key driver of Hg bioaccumulation in ~~high trophic level~~ high-trophic-level fish

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Abstract. The ability of monomethylmercury ($MMHg^+$) to bioaccumulate in seafood is of concern due to its neurotoxic properties. ~~Understanding the bioaccumulation of $MMHg^+$ is challenging because the $MMHg^+$ content at~~ The challenge in understanding $MMHg^+$ bioaccumulation lies in the fact that its levels in higher trophic levels ~~depends on~~ result from both bioconcentration and biomagnification. Furthermore, Hg can occur in several chemical species, including Hg^{2+} and $MMHg^+$, which both bioaccumulate. Although the dominant pathway for $MMHg^+$ bioaccumulation into seafood is the bioconcentration of $MMHg^+$ in primary producers and the subsequent biomagnification to higher trophic levels, other pathways can contribute to $MMHg^+$ bioaccumulation. In this study, we ~~quantify the importance of the~~ use a fully coupled 1D water column Hg bioaccumulation model to quantify how total bioaccumulation of Hg^{2+} and ~~the~~ uptake of $MMHg^+$ from the water (bioconcentration) ~~at higher trophic levels on~~ in consumers affects the bioaccumulation of $MMHg^+$ in ~~high trophic level fish. This is analysed by running a fully coupled 1D water column Hg bioaccumulation model under 3 hydrodynamic regimes typical for~~ high-trophic-level fish. The study is performed in three setups representing hydrodynamic conditions representative of the North and Baltic Seas. We find that Hg^{2+} bioaccumulation does not influence the bioaccumulation of $MMHg^+$ but the bioconcentration of $MMHg^+$ plays an important role. Although direct bioconcentration accounts for $< 15\%$ of $MMHg^+$ bioaccumulation in cod, the cumulative effect of bioconcentration on all trophic levels increases the $MMHg^+$

content of cod by 28-48%. ~~We show that up to the highest trophic level modeled (TL = 3.7), the percentage of $MMHg^+$ that originates from consumer bioconcentration increases with an average of 15% per trophic level.~~ These results demonstrate that bioconcentration in consumers is essential to accurately model the bioaccumulation of $MMHg^+$ at higher trophic levels.

1 Introduction

~~The natural~~ The element mercury (Hg) is ~~currently on the list of~~ presently included in the World Health Organization's list of the 10 substances of ~~most concern by the World Health Organization~~ greatest concern (WHO, 2020). This is ~~because Hg can be methylated into~~ due to the capability of Hg to be methylated to form monomethyl mercury ($MMHg^+$). ~~$MMHg^+$ is a topic of serious concern because $MMHg^+$ is a dangerous neurotoxin that can bioaccumulate into levels that are dangerous for human~~ , 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. For example, it is estimated that the consumption ~~in fish that are often consumed as seafood. Aquatic foods account for more than 15% of the world's protein consumption , and preserving this as a safe and reliable food source is essential to feed an increasing population (Boyd et al., 2022)~~of MeHg contaminated seafood contributed to 61,800 premature deaths and caused economic damage of up to 2.87 trillion USD globally (Chen et al., 2025). This issue is expected to become even more significant as anthropogenic Hg emissions are projected to increase in the coming decades (Maria Brocza et al., 2024). Despite recent efforts, the bioaccumulation of $MMHg^+$ in the marine environment is a complex topic and is not yet fully understood. Part of the complexity of understanding $MMHg^+$ bioaccumulation and toxicity is that Hg can undergo speciation and occur in the environment in several chemical forms with distinct physical and chemical properties (Bieser et al., 2023). In particular, ~~there are~~ these include dissolved Hg (Hg^{2+}), dissolved elemental gaseous Hg (Hg^0), $MMHg^+$, and dissolved dimethylmercury (DMHg).

Often both $MMHg^+$ and DMHg are combined and are termed methylmercury (MeHg). The importance of DMHg is currently debated. Although DMHg is ~~uncharged, which would give it higher permeability to migrate through cell membranes, it is assumed that it does not bioaccumulate to a significant degree (Morel et al., 1998). The higher~~ a common form of Hg in deeper water, there are no measurements in the North and Baltic Seas that would differentiate between DMHg and Hg^0, and its role can therefore not be assessed in the model (Fitzgerald et al., 2007). 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 (West et al., 2022; Morel et al., 1998). The strong bioaccumulation of $MMHg^+$ ~~compared to DMHg~~ , on the other hand, can be attributed to several reasons~~;~~ : $MMHg^+$ can be absorbed by phytoplankton by cell-dependent factors, such as membrane channels (Garcia-Arevalo et al., 2024), ~~volatile DMHg can more easily migrate out of cells and evaporate from the water column, and $MMHg^+$~~ and it can strongly bind to sulfhydryl (-SH) groups in organic material, ~~notable~~ notably cysteine, which traps toxic $MMHg^+$ in the cell (Arnold et al., 1983) . Additionally, it has been shown by Tesán-Onrubia et al. (2023) that plankton communities in the southern Mediterranean Sea have lower $MMHg^+$ concentrations than plankton in the northern Mediterranean Sea; they linked this to changes in

environmental conditions affecting bioconcentration. ~~Although DMHg appears to be a common form of Hg in deeper water, there are no measurements in the North and Baltic Seas that would differentiate between DMHg and Hg^0, and its role can therefore not be assessed in the model (Fitzgerald et al., 2007). 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 (West et al., 2022) .~~

Both Hg^{2+} and $MMHg^+$ can bind to organic material and bioaccumulate in the marine food web (Mason et al., 1995). However, due to the higher toxicity and bioaccumulation potential of $MMHg^+$, the bioaccumulation of $MMHg^+$ is the most important concern and receives the most attention (Mason et al., 2012). There are ~~3 ways in which species bioaccumulate $MMHg^+$;~~ three ways the $MMHg^+$ content of biota can increase: bioconcentration, biomagnification, and ~~in vivo methylation.~~ *in vivo* Hg speciation.

~~**Bioconcentration**, is the uptake of Hg directly~~

1.1 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,

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

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

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

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~~column~~. 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~~ 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 ~~complicated~~ 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 ~~phytosphere~~phycosphere and the availability of transmembrane channels, while this is not the case for Hg^{2+} (Garcia-Arevalo et al., 2024). Bioconcentration is ~~the most important step in bioaccumulation and phytoplankton can bioconcentrate $MMHg^+$ to values between 2E4 and 6.4E6 higher than $MMHg^+$ in surrounding water (Gosnell and Mason, 2015). While the~~ 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,

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

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

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

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 $MMHg^+$ ~~content of phytoplankton is~~ that is taken up directly from the water and $MMHg^+$ that is ingested via food. Bioconcentration is the most important ~~predictor of $MMHg^+$ in higher trophic levels, it "only" predicts 63% of the variability of the $MMHg^+$ in fish (Wu et al., 2019)~~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 and 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~~ concentration in $MMHg^+$ in phytoplankton, there is a large increase in $MMHg^+$ at every consecutive trophic level. Because of this, low trophic level animals such as copepods typically bioaccumulate less $MMHg^+$ than higher trophic level animals such as predatory fish. Even though the direct uptake rate from the water by the copepod might be higher than that in fish, there are fewer trophic levels below the copepod that provide the opportunity to biomagnify $MMHg^+$, typically resulting in lower $MMHg^+$ concentration in the food of copepods than in the food of high trophic level fish, and thus a higher overall $MMHg^+$ concentration in the higher trophic level fish than in the lower trophic level copepod. 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 and 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:

$$BMF_{i,j}^{MMHg^+} = \frac{C_i^{MMHg^+}}{C_j^{MMHg^+}} \quad (9)$$

In which,

$$BMF_{ij}^{MMHg^+} = \text{The biomagnification factor for trophic consumption of organism } j \text{ by } i\,[\text{unitless}] \quad (10)$$

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

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

The biomagnification factor of $MMHg^+$ is extremely high, ~~based on Lavoie et al. (2013) we estimate that the biomagnification factor~~ Lavoie et al. (2013) estimates the diet-weighted average BMF in marine samples for $MMHg^+$ ~~is 1.5 times higher than for Hg~~as 7.0 ± 4.9 while it is below 1 for Hg^{2+}~~. This~~ in most cases (Seixas et al., 2014; Lavoie et al., 2013). 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**~~***In vivo*** **Hg speciation** occurs when ~~animals take other forms of Hgand transform it into $MMHg^+$~~ 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 ~~gaining~~ gained attention (Gente et al., 2023). There is no direct evidence of ~~in vivo~~ *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.

~~Several models model the bioaccumulation~~ Overall the dominant pathway of bioaccumulation, the bioaccumulation of $MMHg^+$ ~~. Notable models focus on the trophic transfer of Hg and $MMHg^+$ (Schartup et al., 2018), the bioaccumulation of~~ 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 ~~(Zhang et al., 2020),~~ 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.

1.2 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 ~~bioaccumulation of phyto and zooplankton in the~~ Mediterranean Sea (Rosati et al., 2022)~~and the marine~~ , MeHg dynamics on the Beaufort Shelf

(Li et al., 2022), and speciation and bioaccumulation in the North and Baltic Seas ~~by (Bieser et al., 2023), which is expanded upon in (Amptmeijer et al., 2025).~~ (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) ~~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. 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.

1.3 The hypotheses

While $MMHg^+$ is more concerning than Hg^{2+} at higher trophic levels, Hg^{2+} can form up to 98% of the bioaccumulated Hg in phytoplankton (Pickhardt and Fisher, 2007). This results in a large removal of Hg^{2+} during the phytoplankton bloom period~~(Soerensen et al., 2016). **Our first hypothesis in this study is that the bioaccumulation of Hg^{2+} can lower the bioaccumulation of $MMHg^+$ by removing Hg^{2+}, which in turn cannot be methylated and accumulated as $MMHg^+$**~~.

~~A counterpoint is that Amptmeijer et al. (2025) does not show an average change in tHg and aqueous Hg caused by bioaccumulation, including the bioaccumulation of~~ However, it is demonstrated by Amptmeijer et al. (2025), which analyzes the feedback of bioaccumulation on Hg $^{2+}$~~. While this indicates that there is no likely effect of Hg^{2+} bioaccumulation on $MMHg^+$ bioaccumulation, it does not exclude this. Most of the bioaccumulation of Hg $^{2+}$ and $MMHg^+$ takes place at the same time and space, in the surface layer during the phytoplankton bloom. While~~ cycling, that there is no change in ~~the average tHg , the results of Amptmeijer et al. (2025) show seasonal variation~~ average tHg and aqueous Hg caused by bioaccumulation, but that there is a seasonal variation in the aquatic tHg content due to bioaccumulation. This means that even if the average concentrations of tHg are not altered by bioaccumulation, there may still be an effect of Hg^{2+} bioaccumulation on $MMHg^+$ bioaccumulation, as during the phytoplankton bloom tHg is reduced which could lead to a reduction of available $MMHg^+$ for bioaccumulation. It could be theorized that as the ecosystem reduces tHg during the phytoplankton bloom, it would reduce dissolved $MMHg^+$, as this is in active equilibrium with other Hg species and therefore reduce the availability of $MMHg^+$ for bioaccumulation. Based on this, we propose **our first hypothesis that the bioaccumulation of Hg^{2+} can lower the bioaccumulation of $MMHg^+$ by**

removing Hg^{2+}, which in turn cannot be methylated and accumulated as $MMHg^{+}$.

~~Most $MMHg^{+}$ in high trophic levels originates from their diet (Lavoie et al., 2013). So it~~ The majority of $MMHg^{+}$ present in higher trophic levels is derived from their dietary intake (Lavoie et al., 2013). It is often assumed that ~~the bioconcentration of $MMHg^{+}$ does not play an essential role in high trophic level $MMHg^{+}$ bioaccumulation~~ $MMHg^{+}$ 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 ~~ignores the fact that bioconcentration plays a role at all trophic levels . If both micro and mesozooplankton, for example, have~~ assumption overlooks that bioconcentration occurs at all levels of the trophic chain. For example, if microzooplankton and mesozooplankton acquire 5% of $MMHg^{+}$ ~~originating from bioconcentration. Mesozooplankton~~ through bioconcentration, mesozooplankton will have 5% less $MMHg^{+}$ ~~in their diet, composed~~ from its diet, which consists of microzooplankton, and ~~an additional~~ another 5% less ~~from its lack of bioconcentration. This results in~~ due to the absence of bioconcentration, leading to a total reduction of 10%. **The second hypothesis ~~in this paper~~ is that ~~the bioconcentration of~~ $MMHg^{+}$ bioconcentration in consumers ~~leads to a large increase in~~ 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.

Studies that have analyzed the relative contribution of bioconcentration in the bioaccumulation of $MMHg^{+}$ in fish found that in freshwater fine-scale dace (*Phoxinus neogaeus*), the bioconcentration accounts for up to 15% of the total bioaccumulation of $MMHg^{+}$ (Hall et al., 1997). A study by Wang and Wong (2003) found that in marine sweetlips (*Plectorhinchus gibbosus*) bioconcentration in fish can dominate total $MMHg^{+}$ concentration, if they eat food with low $MMHg^{+}$ levels, while intake from food dominates total $MMHg^{+}$ uptake when fish eat food with higher $MMHg^{+}$ levels. This means that there will be an effect of $MMHg^{+}$ bioaccumulation in higher trophic levels as it is a direct flux of $MMHg^{+}$ into the organism. In this study, we want to expand this and quantify the cumulative effect of $MMHg^{+}$ bioconcentration in all consumers. This allows us to evaluate if consumer bioconcentration is indeed a small percentage of total $MMHg^{+}$, or if it is a major contributor to the total $MMHg^{+}$ concentrations.

It is important to analyze these interactions using models as they cannot be fully understood using field and laboratory studies. This is because Hg^{2+} and $MMHg^{+}$ are in active equilibrium and we cannot measure $MMHg^{+}$ in a system where phytoplankton would not absorb Hg^{2+}. The effect of bioconcentration of $MMHg^{+}$ can also not easily be measured. It is possible to measure the direct uptake and release of $MMHg^{+}$ by higher trophic levels from the water column. This is done, for example, to estimate the bioconcentration rates of Hg^{2+} and $MMHg^{+}$ in sweetlips in the earlier mentioned study by Wang and Wong (2003). The ~~complexity is that the origin~~ challenge lies in that the source (bioconcentration or biomagnification) of $MMHg^{+}$

cannot be measured in observational studies~~, and part of the $MMHg^+$ that is~~. Additionally, $MMHg^+$ consumed by higher trophic levels is bioconcentrated in ~~consumers at the~~ lower trophic level ~~, making it impossible to measure the full importance of bioconcentrationin consumers~~consumers, complicating the quantification of bioconcentration's full impact.

To test the ~~2~~ two hypotheses that Hg^{2+} bioaccumulation decreases $MMHg^+$ bioaccumulation and that $MMHg^+$ bioconcentration ~~increases it~~in consumers significantly increases $MMHg^+$ bioaccumulation in higher trophic levels, we quantify the effect of bioaccumulation of Hg^{2+} and the bioconcentration of $MMHg^+$ on the bioaccumulation of $MMHg^+$. We do this by running the fully coupled GOTM-ECOSMO-MERCY coupled system used in Amptmeijer et al. (2025) with and without the bioaccumulation of Hg^{2+} and the bioconcentration of $MMHg^+$ in consumers. Then we analyze the bioaccumulation of $MMHg^+$ at different trophic levels in these different scenarios and finally evaluate the importance of both interactions.

The model is run using ~~3~~ three idealized 1D water column setups to represent different hydrological conditions. The setups represent the coastal hydrodynamics found in the North and Baltic Seas.

~~**Modeled region**~~

2 Methodology

To quantify the importance of Hg^{2+} bioaccumulation and $MMHg^+$ bioconcentration on the bioaccumulation of $MMHg^+$ we modeled the bioaccumulation of $MMHg^+$ in three different scenarios using three idealized 1D water column models representing different hydrodynamic regimes typical for the North and Baltic Seas.

2.1 Modeled region

The first North Sea setup is the ~~**permanetely**~~ **permanently mixed- ~~southern~~ Southern North Sea** at ($54°15'00.0"N$ $3°34'12.0"E$). The 41.5 m deep location of this setup is characterized by having constant water-column mixing. This remixing of nutrients within the ~~euphoric~~ euphotic zone creates good conditions for phytoplankton growth. Additionally, since the water column is mixed during the bloom period, macrobenthos can feed directly from the phytoplankton bloom, which results in a ~~late population of macrobenthos~~ high macrobenthos biomass. This results in a high biomass turnover rate ~~, and macrobenthos are~~ and makes macrobenthos an important food source for predatory fish (Heip et al., 1992). The ~~southern~~ Southern North Sea is rich in nutrients, and the phytoplankton bloom ~~is often light limited~~can be light-limited. Diatoms typically dominate the phytoplankton bloom in spring until silicate limitations reduce their growth, and flagellates can become dominant (Emeis et al., 2015).

The second setup is the **seasonally mixed- Northern North Sea** at ($57°42'00.0"N$ $2°42'00.0"E$). This 110 m deep setup is only seasonally mixed. The ~~northern~~ Northern North Sea is still rich in nutrients, resulting in similar high phytoplankton growth, which is dominated by diatoms in spring and ~~succeded~~ succeeded by flagellates in summer, ~~as is the case in the southern~~ similar to the Southern North Sea (Bresnan et al., 2009). A key difference between the ~~southern and northern~~ Southern and Northern North Sea setups is that in the ~~northern~~ Northern North Sea setup, macrobenthos cannot feed directly on the bloom but ~~predominately~~ predominantly feed on sinking detritus. This results in lower macrobenthos biomass and lower importance

of macrobenthos in the diet of top predators. The reduced biomass of macrobenthos in the Northern North Sea is in line with observations (Heip et al., 1992).

The final ~~set~~ setup is the **permanently stratified- Gotland Deep** ($57°18'00.0"N\ 20°00'00.0"E$). This setup is different from the ~~2~~ two North Sea setups in ~~a few~~ several ways. First, the Baltic Sea, in general, is not limited by silicate, resulting in a dominance of diatoms in the phytoplankton bloom. In the Gotland Deep specifically, silicate limitation can occur, but diatoms will still be ~~more dominant~~ dominant throughout the bloom. Gotland Deep has a very low salinity (7 g l^{-1}), is strongly stratified, and can be eutrophied in phosphate. This results in ~~perfect~~ good growth conditions for nitrogen-fixing cyanobacteria that can form a major part of the total phytoplankton biomass in ~~the~~ autumn when nitrogen limitations limit the growth of other phytoplankton (Kahru and Elmgren, 2014). The presence of cyanobacteria can alter bioaccumulation because they can reduce dissolved Hg^{2+} to volatile Hg^0, which increases Hg evaporation and therefore lowers the concentration (Kuss et al., 2015). This can reduce the average Hg content by up to 8% (Amptmeijer et al., 2025). At the same time, the small size of the cyanobacteria gives them an extremely high surface: biomass ratio, resulting in a very high bioconcentration factor of $MMHg^+$ (Pickhardt et al., 2006). Finally, the Gotland Deep has anoxic water below ~~thermocline,~~ the thermocline; because of this, there is no macrobenthos (Conley et al., 2009).

~~The 1D setups are the same as those used in (Amptmeijer et al., 2025) and are described there in more detail. The physics of the setups is based on the Generalized Ocean Turbulence Model (GOTM) (Bolding et al., 2021), the biogeochemistry is based on the ECOSMO E2E (Daewel and Schrum, 2013), and the Hg chemistry is based on the MERCY V2.0 model (Bieser et al., 2023).~~

Quantifying the importance of the bioaccumulation of Hg^{2+} and bioconcentration of $MMHg^+$ in consumers on $MMHg^+$ bioaccumulation into higher trophic levels under these idealized circumstances will provide a unique insight into the drivers of the bioaccumulation of $MMHg^+$ ~~bioaccumulation~~ and increase our fundamental understanding of this process. Additionally, it is important to quantify the importance of these interactions using lighter models because their inclusion in models comes at a cost. Especially, the implementation of the bioaccumulation of Hg^{2+} is done by adding ~~1~~ one state variable to the model per biota functional group, or ~~2~~ two state variables if the biomagnified and bioconcentrated Hg^{2+} is treated as separate variables, as is done in the model used in this study. While this is feasible without much concern in the 1D water column models that we use in this study, when running ~~an~~ large earth system models, adding insignificant state variables becomes an unnecessary waste of computational resources which results in the wasteful expenditure of research funds and energy.

3 ~~Methodolgy~~

~~To quantify the importance of Hg^{2+} bioaccumulation and $MMHg^+$ bioconcentration on the bioaccumulation of $MMHg^+$ we modeled the bioaccumulation of $MMHg^+$ in three different scenarios using three idealized 1D water column models representing different hydrodynamic regimes typical for the North and Baltic Seas.~~

2.1 Model

Hypotheses are evaluated using the Generalized Ocean Turbulence Model (GOTM) (Bolding et al., 2021) that is coupled to the ECOSMO E2E ecosystem model (Daewel et al., 2019) and the MERCY v2.0 ~~mercury~~ Hg speciation model (Bieser et al., 2023). The models are coupled using the Framework for Aquatic Biogeochemical modeling (FABM) (Bruggeman and Bolding, 2014). This setup is chosen because it has been used and evaluated in previous studies to analyze the bioaccumulation and cycling of Hg in the North and Baltic Seas, and it is the only fully coupled model to incorporate the bioaccumulation of Hg^{2+} and the bioconcentration of $MMHg^{+}$ at higher trophic levels.

2.1.1 GOTM

GOTM is used to simulate the hydrodynamics of the 1D water column models. GOTM is a 1D turbulence model that computes the 1D version of the transport equation of temperature, momentum, and salinity. It does this while being nudged to observational datasets. GOTM simulations are designed using the iGOTM tool (https://igotm.bolding-bruggeman.com/). This tool compiles the observational datasets used for the GOTM simulation and estimates the water depth based on the gridded bathymetry data (1/240° resolution) (GEBCO Bathymetric Compilation Group, 2020), the ECMWF ERA5 data for meteorological data, the TPOX-9 atlas for tides (1/30° resolution)(Egbert and Erofeeva, 2002), and for salinity and temperature, it uses the ~~wold ocean~~ World Ocean Atlas (0.25° resolution) (Garcia H.E. et al., 2019). The state is solved every 60 seconds using forward Euler differential equations. The setups have 1 grid cell per meter, and the variables are exported as daily means for the post-processing analyses.

2.1.2 ECOSMO E2E

The ecosystem model used in this study is the ECOSMO E2E ecosystem model (Daewel et al., 2019). The ECOSMO E2E ecosystem model is an intermediately complex ecosystem model that uses a functional group approach to estimate the biomass and carbon fluxes in the North and Baltic Seas. The version used here has ~~3~~ three functional groups of phytoplankton~~; ;~~ diatoms, flagellates, and cyanobacteria~~, 2 ;~~ two functional groups of zooplankton~~; microzooplankton , and mesozooplankton, 2 ;~~ microzooplankton and mesozooplankton; two functional groups of fish~~, and 1 ;~~ and one group of macrobenthos. The basic version of the model is published by Daewel et al. (2019), but the version used here has some modifications to make it more suitable for bioaccumulation. ~~The modification included reducing carbon uptake efficiencies at higher trophic levels and~~ This includes the addition of ~~1 more fish functional group~~ a second functional group of fish to represent high trophic level animals, the explicit resolution of the trophic level, and tuning of some of the carbon flux parameters such as growth rates and assimilation efficiencies. This is ~~described in detail in (Amptmeijer et al., 2025)~~discussed in more detail in Amptmeijer et al. (2025).

2.1.3 MERCY v2.0

The MERCY ~~V2~~v2.0 model links atmospheric Hg to $MMHg^{+}$ in fish. It does this by estimating air-sea exchange and wet deposition of Hg based on the CMAQ-Hg model and calculating the marine cycling while taking into account marine speciation

and bioaccumulation. It uses 35 state variables to estimate Hg speciation, transport, and bioaccumulation. ~~Estimates~~ The model estimates the partitioning of Hg^{2+} and $MMHg^{+}$ into ~~dissolved organic carbon and particulate~~ DOM and detritus, and bioaccumulation based on ecosystem parameters derived from the ~~ecosystem model~~ ECOSMO E2E ecosystem model (Bieser et al., 2023).

2.2 Bioaccumulation in the model

~~The bioaccumulation of Hg^{2+} and $MMHg^{+}$ is the same as in (Amptmeijer et al., 2025). All functional groups of the biota can bioconcentrate both Hg^{2+} and $MMHg^{+}$ by default. This is implemented in the form of an active equilibrium in which the bioconcentrated Hg^{2+} or $MMHg^{+}$ on each interaction is changed by the uptake rate m^3 mgC d^{-1} multiplied by the biomass in mgC of the functional group and the concentration of Hg in ng Hg $^{-3}$. The release of Hg from the functional group is based on the respiration, mortality, and Hg release rate in d^{-1}.~~ In addition to the bioconcentration, all consumer functional groups can take up Hg from the consumption of contaminated food. The uptake of Hg^{2+} or $MMHg^{+}$ from food depends on the assimilation efficiency of the food and Hg species. After Hg has been assimilated from food, $MMHg^{+}$ is released based on the mortality and respiration rate within the functional group, while there is an additional release rate for Hg^{2+}. Since fish have a temperature-dependent respiration rate in the ECOSMO E2E model, this means that fish lose Hg from both bioconcentration and biomagnification faster in warmer water as their respiration, and thus Hg release rate, increases with temperature. The bioconcentration rates for zooplankton are based on Tsui and Wang (2004), and those for fish are based on Wang and Wong (2003).

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:

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

$C^{BC}_{g,p}$ = 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 [m^3 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 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:

$$\frac{dC^{BM}_{g,p}}{dt} = \sum_{s=1}^{n_s}(r^{pred}_{g,s} \cdot a_{s,p} \cdot (C^{BC}_{s,p} + C^{BM}_{s,p})) - C^{BM}_{g,p} \cdot (r^{to}_{p,g} + r^{bl}_{g} + \sum_{z=1}^{n_z} r^{pred}_{z,g}) \quad (14)$$

$C_{g,p}^{BM}$ = 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:

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

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

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

2.3 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.

2.3.1 Post-processing analysis

The post-processing analysis ~~analysis~~ is performed in R v4.4.1. Plots are generated using ggplot v3.5.0., and linear regression and statistics are calculated using ggpubr v0.6.0. ~~A Wilcoxen signed rank~~ The hypotheses are tested using two different tests. First, a Wilcoxon signed-rank test is performed ~~to test the significance of differences and similarities between treatments~~using the `wilcox.test` function from the stats (v4.4.2) package in base R. This is ~~done because we assume that the trophic level influences the~~ a non-parametric statistical test that determines if there is a significant difference between the ~~scenarios, which would mean that the differences between the scenariosare not normally distributed. The results are interpreted as p~~ base case and the scenarios. We accept a p-value of <0.05 ~~means a significant difference and $p > 0.05$ does not indicate~~ to indicate that the scenario has a significant deviation from the base case. The scenarios are compared using a ~~significant difference. Additionally, a~~ Bayesian t-test ~~is run using the BayesFactor~~ implemented via the `ttestBF` function from the BayesFactor package (v0.9.12-4.7~~packages in R, to assess the likelihood that the scenarios are different from or the same as the base case.~~). The Bayesian t-test computes the Bayes factor BF_{10}, defined as the ratio of the marginal likelihood of the data under the alternative hypothesis (different means) to the marginal likelihood under the null hypothesis (equal means). The analysis is performed using Jeffreys-Zellner-Siow priors (Zellner and Siow, 1980). A $BF_{10} < 1$ indicates evidence for equal means, whereas $BF_{10} > 1$ indicates evidence for a difference in the mean. Typically, a $BF_{10} < 0.1$ or $BF_{10} > 10$ is interpreted as strong evidence, and $BF_{10} < 0.01$ or $BF_{10} > 100$ as very strong evidence for equal or different means, respectively (Doll and Jacquemin, 2019).

2.4 Scenarios

The model is run in ~~3~~ three different scenarios. The "Base case", "No Hg^{2+} bioaccumulation" and "No $MMHg^{+}$ bioconcentration". The base case scenario is the same as the base case used in (Amptmeijer et al., 2025). For the "No Hg bioaccumulation" setup, all uptake rates of Hg^{2+} are set to zero. For the "No $MMHg^{+}$ bioconcentration" scenario, all consumer bioconcentrations of $MMHg^{+}$ and all Hg^{2+} uptake rates are set to zero.

~~The bioaccumulated $MMHg^{+}$, the fraction 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.~~

2.5 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. The effect of the consumer and producer bioconcentration scaling factor on the difference caused by consumer level bioconcentration on total bioaccumulation is visualised. The data for both instances are plotted with an optimal fit for each of the three stations. The consumer bioconcentration factor is shown with a best fit assuming an asymptotic growth model $y = a \times (1 - e^{-bx})$, which is fitted using the `nls` function from the R stats package (version 4.4.2). For producer bioconcentration, a best fit with an asymptotic exponential decay model $y = a \times e^{-b \times x} + c$ is assumed also using the `nls` function.

3 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.

3.1 Bioaccumulation of Hg^{2+}

The effect of Hg^{2+} bioaccumulation on the bioaccumulation of $MMHg^{+}$ is shown in Table 1. 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 ~~(1-5%)~~between 1% and -6%. This is statistically evaluated, and the results are shown in Table 2. ~~Wilcoxen~~Wilcoxon's signed rank test shows that bioaccumulation of Hg^{2+} has no significant impact on the bioaccumulation of $MMHg^{+}$ (p ~~> 0.99~~= 0.67). Furthermore, the Bayesian t-test shows ~~that the data is 2.9~~ with a BF_{10}=0.40 that the results are 2.5 times more likely under the null hypothesis of no effect than under the alternative hypothesis. ~~This shows that~~

3.2 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. ~~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 shallow 20m of the water column, to create an average value that we can compare between the setups.~~

These results show that the relative contribution of bioconcentration on the $MMHg^{+}$ content is low in microzooplankton (~~4-6~~4–6%) while it is higher in mesozooplankton (~~5-10~~5–11%) higher in fish 1 (~~13- 22%)~~13–22%), while lower in fish 2 (~~8-14~~8–14%) and higher in macrobenthos (~~14- 25~~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,

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 (%) $= (1 - \frac{\text{Scenario}}{\text{Default}}) * 100$

Gotland Deep							
	Default		No Hg^{2+} bioaccumulation		No $MMHg^+$ bioconcentration		Trophic Level
	(ng Hg mg C^{-1})	Bioconcentrated (%)	(ng Hg mg C^{-1})	Difference (%)	(ng Hg mg C^{-1})	Difference (%)	-
Diatom	0.0050	100%	0.0050	0%	0.0050	-0%	1
Flagellate	0.0095	100%	0.0095	0%	0.0095	-0%	1
Cyanobacteria	0.015	100%	0.015	0%	0.015	0%	1
Microzooplankton	0.013	5%	0.013	0%	0.012	-5%	2.0
Mesozooplankton	0.0190	5%	0.0190	0%	0.018	-5%	2.2
Fish 1	0.031	**16%**	0.031	0%	0.025	**-20%**	2.6
Fish 2	0.065	8%	0.065	0%	0.046	**-28%**	3.5
Southern North Sea							
	Default		No Hg^{2+} bioaccumulation		No $MMHg^+$ bioconcentration		Trophic Level
	(ng Hg mg C^{-1})	Bioconcentrated (%)	(ng Hg mg C^{-1})	Difference (%)	(ng Hg mg C^{-1})	Difference (%)	-
Diatom	0.0053	100%	0.0050	-6%	0.0049	-7%	1
Flagellate	0.0080	100%	0.0077	-3%	0.0077	-3%	1
Microzooplankton	0.011	4%	0.011	-3%	0.011	-7%	2.0
Mesozooplankton	0.014	6%	0.014	1%	0.013	-8%	2.5
Fish 1	0.049	**13%**	0.048	2%	0.030	**-38%**	3.2
Fish 2	0.071	9%	0.069	-2%	0.043	**-40%**	3.5
Macrobenthos	0.023	**14%**	0.023	-1%	0.017	**-27%**	2.3
Northern North Sea							
	Default		No Hg^{2+} bioaccumulation		No $MMHg^+$ bioconcentration		Trophic Level
	(ng Hg mg C^{-1})	Bioconcentrated (%)	(ng Hg mg C^{-1})	Difference (%)	(ng Hg mg C^{-1})	Difference (%)	-
Diatom	0.0034	100%	0.0034	0%	0.0034	-0%	1
Flagellate	0.0062	100%	0.0062	0%	0.0062	-0%	1
Microzooplankton	0.010	6%	0.010	0%	0.0098	-6%	2.0
Mesozooplankton	0.012	**11%**	0.012	0%	0.011	**-15%**	2.5
Fish 1	0.021	**22%**	0.021	0%	0.013	**-36%**	2.9
Fish 2	0.037	**14%**	0.037	0%	0.019	**-49%**	3.7
Macrobenthos	0.0081	**27%**	0.0081	0%	0.0050	**-38%**	2.3

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 and Wong, 2003) and the observations of 15% by Hall et al. (1997). Both fish 1 and fish 2 have the

Table 2. The results of the statistical test performed to evaluate the difference between the scenarios and the base case. The high p-value ($p = 0.67$) and below 1 Bayes Factor (BF_{10}=0.40) indicate that there is no significant difference between the mean of the base case and the scenario without Hg^{2+} bioaccumulation and that the change that the mean is equal is 2.5 times larger than the chance that there it is not. The difference between the scenario without $MMHg^+$ bioconcentration is significant ($p < 0.001$) and the change that there is a difference in the mean bioaccumulation of $MMHg^+$ caused by the bioconcentration of $MMHg^+$ in consumers is 5.96 times higher than the change that there is no difference in the mean.

	No Hg^{2+} bioaccumulation ~~does not have a significant effect on $MMHg^+$ bioaccumulation (BF~~	No $MMHg^+$ consumer bio
Wilcoxon signed-rank test	$p =$ ~~0.35).~~ 0.67	$p < 0.001$
Bayesian t-test	$BF_{10} = 0.40$	$BF_{10} = 5.96$

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~~27% 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.

3.3 Bioaccumulation of $MMHg^+$ and trophic level

The relationship between trophic level and $MMHg^+$ bioaccumulation is plotted in Fig. 2a. Since we assume biomagnification to be an exponential effect per trophic level on top of bioconcentration, the model is fitted as an exponential function with the average $MMHg^+$ content of phytoplankton as the origin.

3.4 ~~Evaluation hypotheses 1; The effect of Hg^{2+} bioaccumulation on $MMHg^+$ bioaccumulation~~

~~Based on the results of the statistical analysis shown in table 2 we can see that~~ Fig. 2b expands on this by showing the relationship between the trophic level and the effect of both investigated drivers on the bioaccumulation of $MMHg^+$. This shows that while there is no ~~significant difference ($p = 0.99$)caused by Hg~~effect of the bioaccumulation of Hg^+ on the bioaccumulation of $MMHg^+$. The setup without bioconcentration in consumers has 15% less $MMHg^+$ bioaccumulation per trophic level than the setup which does include this interaction.

3.4 Seasonality of the difference in $MMHg^+$ bioaccumulation

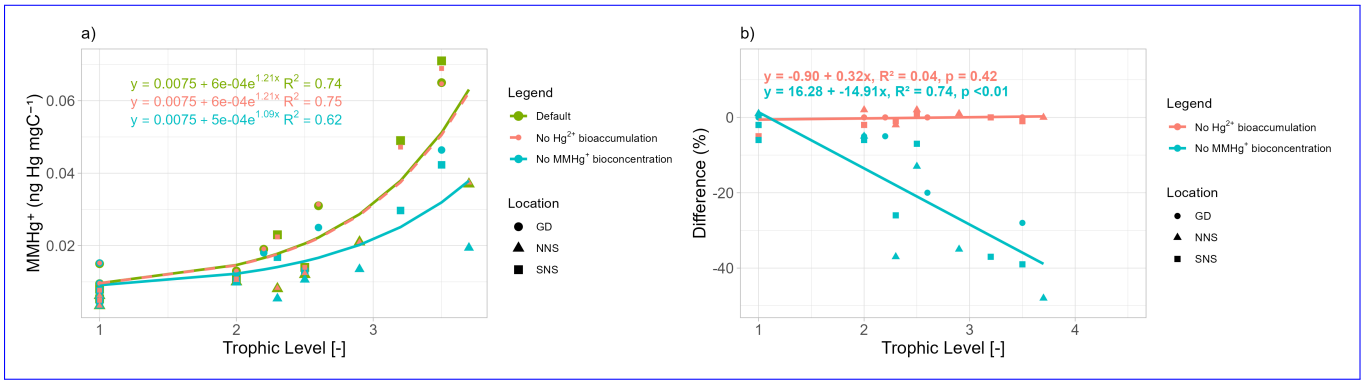

Figure 2. Figure a) shows the trophic level vs $MMHg^+$ bioaccumulation for the base case and the 2 scenarios. The base case and scenario "No Hg^{2+} bioaccumulation" have the relationship $0.0075+6E4*e^{1.2*TrophicLevel}$ and the scenario "No $MMHg^+$ bioconcentration" has $0.0075+5E4*e^{1.09*TrophicLevel}$. Figure b) shows that there is an exponential increase in $MMHg^+$ with trophic level, which is higher for the base case and the scenario without Hg bioaccumulation than for the scenario without $MMHg^+$ bioconcentration. Figure b) expands on this and demonstrates no significant effect on the importance of trophic level on the effect of the bioaccumulation of Hg^{2+} on $MMHg^+$ bioaccumulation. There is a reduction of 15% per trophic level caused by the bioconcentration of $MMHg^+$.

The seasonality of the difference in $MMHg^+$ bioaccumulation caused the bioaccumulation of Hg^{2+} ~~bioaccumulation on $MMHg^+$ bioaccumulation. Based on the Bayesian t test we estimate that the change is~~ and the bioconcentration of $MMHg^+$ in consumers is shown in Fig. 3. For each calendar day (January 1~~/0.35 = 2.86 times greater than the data, that is , there is a difference. Based on these results, we conclude that Hg bioaccumulation~~st, 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 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+} ~~does not play a major direct role in the bioaccumulation of~~ 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.

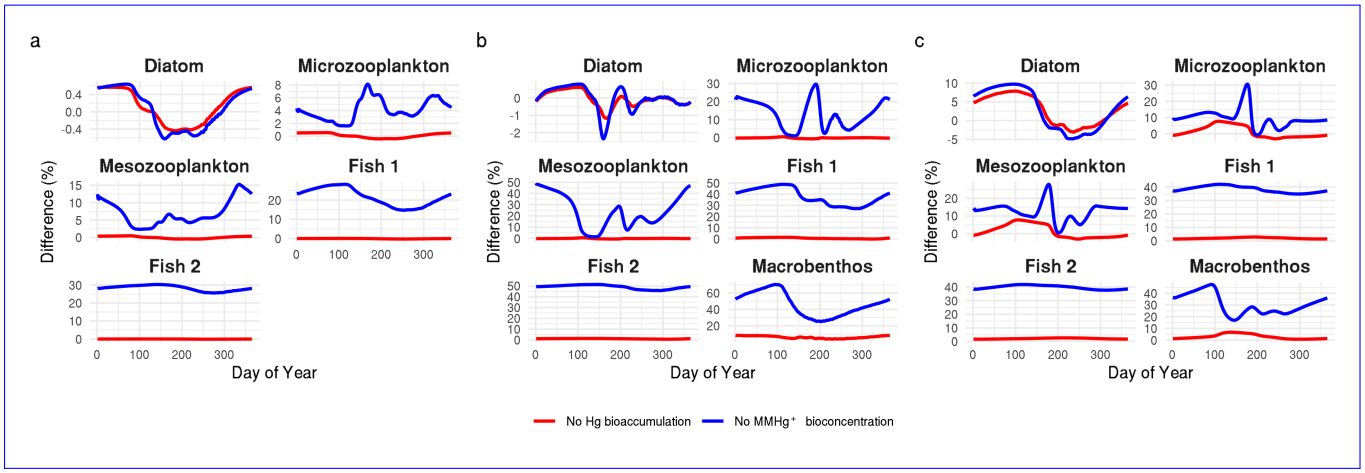

Figure 3. The seasonality of the difference in the bioaccumulation of $MMHg^+$ caused by the bioaccumulation of Hg^{2+} and the bioconcentration of MMHg in consumers for a) the Gotland Deep, b) the Northern North Sea and c) the Southern North Sea. In high-trophic-level such as fish 1 and fish 2 there 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.

4 Sensitivity analyses

4.1 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. 4. Figure 4a 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. 4b. 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, thus the percentage difference is also 0. However, the contribution of consumer level bioconcentration 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

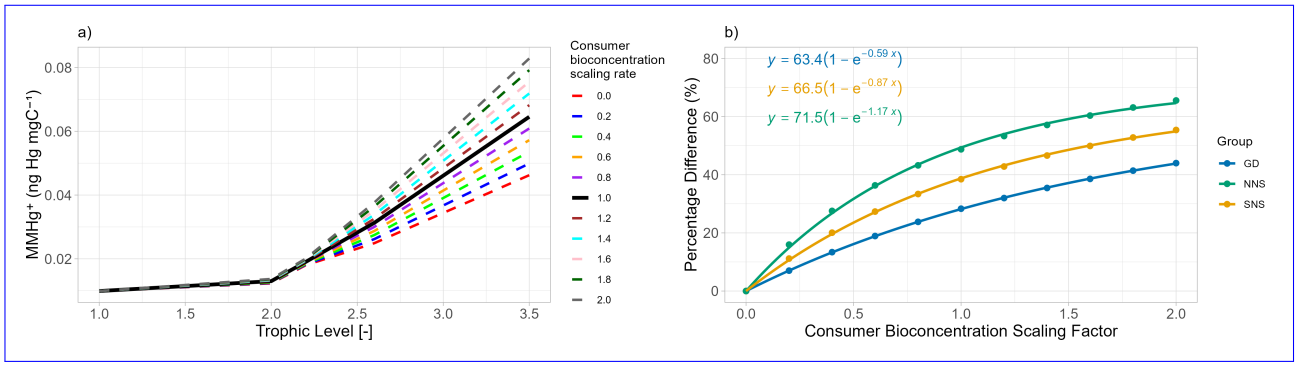

Figure 4. a) show the effect of the bioaccumulation of $MMHg^+$ per trophic level in the Gotland Deep. This shows an increase 0.0036 ± 0.00010 ng Hg mg^{-1} in fish 2 $MMHg^+$ bioaccumulation for every 0.2 step increase in the consumer bioconcentration scacling factor. 4b) shows the percentage difference due to bioaccumulation in fish 2 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.

way as in the results shown in 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.

4.2 Sensitivy of the producer bioconcentration rate

The results of the second sensitivity study are shown in Fig. 5. 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. 5a, and the effect of this scaling on the relative importance of consumer bioconcentration on $MMHg^+$ bioaccumulation is shown in Fig. 5b. 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 $MMHg^+$ ~~. However, it should be noted that bioaccumulation of Hg~~per step of 0.2 in the scaling factor is 0.0083 ± 0.00030 ng Hg mg^{2+}~~$^{-1}$ can still play a role in the MMHg~~. The relative contribution of consumer bioconcentration on $MMHg^+$ ~~content in biota by in vivo methylation. However,~~ bioaccumulation is shown in Fig. 5b. 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 ~~data suggestion that this is a major pathway, so based on the current~~

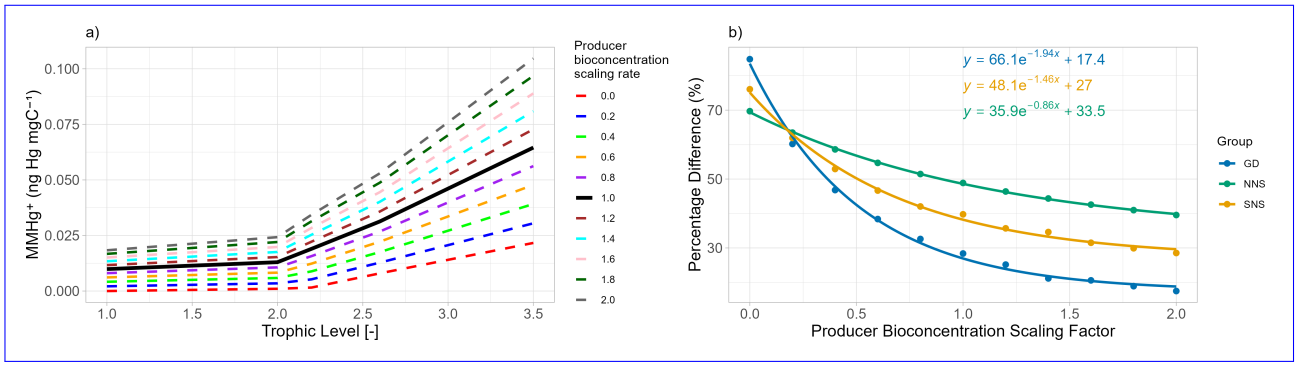

Figure 5. a) illustrates the influence of scaling the producers bioconcentration rate of $MMHg^+$ on the $MMHg^+$ bioaccumulation at each trophic level in the Gotland Deep. This shows an increase of 0.0084 ± 0.00032 ng Hg mg^{-1} in fish 2 $MMHg^+$ with every 0.2 increase in the producers scaling factor. 5b) shows importance of consumer level bioconcentration across all setups by showing the percentage difference in the bioaccumulation of $MMHg^2$ in fish 2 between the setup with and without consumer level bioconcentration. This shows that in all cases the percentage difference is high when the producer bioconcentration factor is low, and that this percentages decreases with an increasing producer bioconcentratoin scaling factor.

~~state of knowledge of $MMHg^{2+}$ bioaccumulation we conclude that the first hypothesis is incorrect and Hg^{2+}~~ macrobenthos in the model. This means that the entire ecosystem is pelagic and detritus is less important than direct consumption of the phytoplankton bloom.

5 Evaluation of the hypotheses

Our results show that the $MMHg^+$ content of high-trophic-level fish is a combination of the direct uptake of biomagnification by these high-trophic-level fish and the $MMHg^+$ they take up via their diet. The $MMHg^+$ content of their diet is, in turn, made up of the bioconcentration in each trophic level, including producers and consumers.

5.1 Evaluation hypotheses 1; The effect of Hg^{2+} bioaccumulation on $MMHg^+$ bioaccumulation

Based on the results of the statistical analysis shown in Table 2, we can see that there is no significant difference ($p = 0.67$) caused by Hg^{2+} bioaccumulation on $MMHg^+$ bioaccumulation. Based on the Bayesian t-test, we estimate that the change is 1/0.40 = 2.5 times greater that the mean is equal to that it is not. We do note that the seasonal changes in the total Hg concentration due to bioaccumulation change the bioaccumulation of $MMHg^+$ at the base of the food web, and we can see this change in phytoplankton and low trophic level consumers, but it does not cause a notable ($> 5\%$) change in $MMHg^+$ bioaccumulation in fish. Based on these results, we conclude that Hg^{2+} bioaccumulation does not play a major direct role in the bioaccumulation of $MMHg^+$ in ~~coastal food webs.~~ our model.

5.2 Evaluation ~~of hypothesis~~ hypotheses 2~~:~~ ; The effect of $MMHg^+$ bioconcentration in consumers on $MMHg^+$ bioaccumulation

Based on the statistical results shown in Table ~~2 ,~~ 2 we conclude that there is a significant difference between the base case and the scenario without consumer bioconcentration (~~$P < 0.001$). Additionally, based~~ $P < 0.001$). We base our conclusion on the Bayesian t-test ~~, that~~ the chance that the ~~data are different is 6.81~~ mean bioaccumulation is different without consumer level bioconcentration is 5.96 times larger than ~~that no difference exists.~~

the chance that there is no difference in the mean. Based on the results, we conclude that the bioconcentration of $MMHg^+$ in consumers is a significant contributor to the bioaccumulation of $MMHg^+$. ~~We quantify this significant increase in the bioaccumulation of $MMHg^+$ at 15% per trophic level in our model.~~

~~The results of the statistical test performed to evaluate the difference between the scenarios and the base case. The high p-value ($p > 0.99$) and below 1 Bayes Factor (BF=0.35) indicate that there is no significant difference between the base case and the scenario without Hg^{2+} bioaccumulation and that the change that there is no difference is 2.86 times larger than the chance that there is a difference. The difference between the scenario without $MMHg^+$ bioconcentration is significant ($p < 0.001$) and the change that there is a difference is 6.81 times higher than the change that there is no difference caused by the bioconcentration of $MMHg^+$ in consumers on the bioaccumulation of $MMHg^+$. No Hg^{2+} bioaccumulation No $MMHg^+$ consumer bioconcentration~~

~~Wilcoxen signed-rank test $p > 0.99$ $p<0.001$ Bayesian t-test BF=0.35 BF=6.81~~

6 Model limitations

6.1 ~~Model limitations~~Scope of the biogeochemical model

There are some limitations to our model. First, estimates of the biomagnification factor ~~or~~ of $MMHg^+$ range between 2-10. Our model represents the estimations of the lower end. The bioconcentration factor is probably more important in low biomagnification food webs. Another limitation is that our model stops at trophic level 3.7. This is a ~~high trophic level~~ high-trophic-level that can represent piscivorous species, but many marine species that are consumed by humans, such as tuna, ~~Great Marlin~~great marlin, and cod~~can have higher trophic levels. These higher trophic levels might be influenced even more by consumers $MMHg^+$ bioconcentration. Additionally, it must be noted that fish 2 in our model is only trophic level 3.5-3.7. Typically, large cod can have a higher trophic level of 3.7-4.2 and high trophic levels such as blue fin tuna can reach trophic levels~~ , can have higher trophic levels of up to 4.8 (Nilsen et al., 2008; Sarà and Sarà, 2007).

When the absolute concentration of $MMHg^+$ increases at higher trophic levels, the relative increase in the importance of direct bioconcentration per trophic level likely decreases. Our modeled top predator with a trophic level of 3.7 has a ~~high trophic level~~ high-trophic-level for a coastal ocean, but there is a marine biota with even higher trophic levels in our model domain, such as marine mammals. Without a dedicated modeling study to simulate the diet and bioconcentration of even higher

trophic levels, we cannot simply extrapolate our ~~finding~~ findings to predict the importance of $MMHg^+$ bioconcentration in their $MMHg^+$ bioaccumulation.

~~Overall~~

6.2 Uncertainty in paramaters

Overall, the most important driver of our model is the fraction of $MMHg^+$ that is bioaccumulated by bioconcentration for each trophic level, as this drives the relative importance of bioconcentration at the higher trophic levels. This is influenced by both the direct bioconcentration in that trophic level and the bioconcentration of animals in lower trophic levels. This relationship is quantified in the sensitivity study. The contribution of bioconcentration in zooplankton of ~~3.97–10.07~~4–11% is in line with the $<$ 20% reported by Schartup et al. (2018), and the contribution of bioconcentration in fish between ~~8.14–21.82~~8–22% is in line with the study by Wang and Wong (2003).

The main uncertainty for the fraction of $MMHg^+$ that originates from bioconcentration is the parameterization of bioconcentration and biomagnification. Both the bioconcentration and biomagnification of zooplankton are based on the work of Tsui and Wang (2004) on water fleas (*Daphia Pulex*) and for fish this is based on Wang and Wong (2003) and their work on the Indo-Pacific species ~~Sweatlips~~sweetlips. Although water fleas are common in the Baltic Sea, they are not in the North Sea, and sweetlips live neither in the North nor in the Baltic Sea. This means that the most important parameters in our model are not based on the animals they represent in our model. Although it is unfeasible to have dedicated bioaccumulation studies in every animal or functional group, there are currently not enough studies to verify whether these rates would differ between the circumstances in our model and those in the experiment. Drivers that might influence these factors are the size of the biota, physical circumstances such as temperature and salinity, or if there is a seasonal effect related to the activity of the animals. It would greatly improve our ability to model the bioaccumulation of $MMHg^+$ if more information on these different drivers were available.

6.3 Limitations due to model design

It is a deliberate choice to perform this study in 1D idealized water column models, as it allows ~~us to get~~ a clear overview of the driving processes and enables us to generalize our findings. In this way, we can provide a general conclusion based on the biomagnification and bioconcentration rates of the biota that are presented in laboratory and field studies. However, it poses limitations compared to real fish by omitting spatial variability. Locally variable circumstances, such as the seasonally dependent flow of Hg from rivers to the ocean, could cause the importance of bioconcentration on $MMHg^+$ bioaccumulation to be regionally different.

Although the model can predict the importance of $MMHg^+$ bioconcentration, it ~~can not~~ cannot evaluate the importance of the bioconcentration of gaseous Hg species, such as Hg^0 and DMHg. These gaseous Hg species are assumed not to biomagnify because they are not polar but could bioconcentrate. Because the gills of fish are optimized to facilitate the exchange of ~~gasses~~ gases between water and fish blood, these gaseous Hg species can likely bioconcentrate ~~into organisms. However, to~~ in organisms, but it is unclear to what degree. To model and evaluate the importance of this interaction, studies must be

performed first, investigating both the bioconcentration and release rates of these gaseous species and their fate in the organism. In particular, the effect of the bioconcentration of DMHg on the concentration of MeHg at higher ~~tropic~~ trophic levels will be very dependent on whether DMHg stays gaseous in the organism and is excreted quickly via the gills, or whether DMHg is demethylated in $MMHg^+$ and further biomagnified in the food chain. Since DMHg concentrations are low in the ~~north~~ North and Baltic seas, this is unlikely to play a major role in our setups, but it could influence the importance of bioconcentration on the $MMHg^+$ content of higher trophic level fish in seas with higher DMHg concentrations, such as the open oceans and the Mediterranean sea.

6.4 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.

7 Conclusion

In our paper, we used a 1D water column model to test two hypotheses. Our first hypothesis is that the bioaccumulation of $MMHg^+$ is influenced by the bioaccumulation of Hg^{2+}. We theorized that Hg~~bioaccumulation $^{2+}$~~ $^{2+}$ bioaccumulation removes a significant portion of Hg^{2+} from the water column, resulting in less Hg^{2+} that can be methylated in $MMHg^+$. As a result, we would expect that the bioaccumulation of Hg^{2+} can reduce the bioaccumulation of $MMHg^+$. Our second hypothesis is that the bioconcentration of $MMHg^+$ in consumers is a major contributor to the bioaccumulation of $MMHg^+$ at higher trophic levels. We theorized that while the direct effect of bioconcentration in ~~high trophic level~~ high-trophic-level animals is low, the cumulative effect of bioaccumulation in all trophic levels below becomes a major source of $MMHg^+$

Our results show that the bioaccumulation of $MMHg^+$ in our model with and without the bioaccumulation of Hg^{2+} is ~~the same~~not significantly different, while this is ~~not~~ the case for the model with and without the bioconcentration of $MMHg^+$. We show that the bioconcentration of $MMHg^+$ in consumers becomes more important at higher trophic levels because it is an effect of the sum of all trophic levels before it. We show that while direct bioconcentration only accounts for 8-14% of $MMHg^+$ bioaccumulation in our highest trophic level fish, the total effect of bioconcentration in consumers accounts for ~~28-48~~28-49%. This effect increases with the trophic level and the percentile contribution of the cumulative effect of $MMHg^+$ ~~biooconcentration~~ bioconcentration in consumers on $MMHg^+$ bioaccumulation is 15% per trophic level.

Because of this, we reject the first hypothesis that bioaccumulation of Hg^{2+} lowers $MMHg^+$ bioaccumulation and accept our second hypothesis that bioconcentration of $MMHg^+$ increases bioaccumulation of $MMHg^+$ in higher trophic ~~levels~~ level fish. We supplement the second hypothesis by quantifying the effect as an average increase in bioaccumulated $MMHg^+$ of 15% per trophic level.

These results demonstrate that to model the bioaccumulation of $MMHg^+$, the bioaccumulation of Hg^{2+} can be ignored to save computational resources. However, the bioconcentration of $MMHg^+$ on the other hand, is an essential interaction that should be taken into account. When modeling the bioaccumulation of $MMHg^+$ at higher trophic levels.

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Author contribution

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Competing interest

None of the authors declares any competing interest.

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This paper's readability was refined using gAI tools like ChatGPT (OpenAI), and spelling corrections were made utilizing AI-driven applications, including Grammarly and Writefull. Moreover, AI tools optimized the R and Python scripts and offered coding suggestions, all of which were manually verified before implementation. For literature research, Google Scholar and Perplexity assisted in identifying sources, which were then manually verified and cited. Sub-images comprising the graphical abstract were crafted using openART and GPT-4.1.

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