Variation in methylmercury biomagnification in freshwater and terrestrial invertebrates: a critical review

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Abstract:

Mercury (Hg) is a highly toxic pollutant that can be found in several types of ecosystems. Methylmercury (MeHg(I)) is the form of mercury with the highest capacity to bioaccumulate and biomagnify. Organisms that occupy the low end of food webs, such as invertebrates, may be a key factor in MeHg(I) biomagnification in the ecosystems. This research consists in a systematic review of the published peer-reviewed literature related to mercury bioaccumulation in freshwater and terrestrial invertebrates. It provided insight into what type of invertebrates pose a higher risk to upper trophic levels of the food web. It also helped to clarify what types of ecosystems are more susceptible to bioaccumulation at these lower trophic levels and the associated physico-chemical conditions. This review also showed the importance of including the physico-chemical characteristics of the study sites in papers related to mercury bioaccumulation and biomagnification, due to their significant impact on the susceptibility of the food webs to MeHg(I). The outcomes can also potentially improve further mercury risk assessment analyses.

Keywords: Mercury, bioaccumulation, biomagnification, methylmercury, invertebrates

1. Introduction

Mercury is a highly toxic pollutant that can be found in remote areas due to the atmospheric global dispersion of elemental mercury (Driscoll et al., 2013). The methylation of divalent mercury (Hg(II)) produces MeHg(I), the most concerning form of mercury in food webs due to its capacity to bioaccumulate and biomagnify (Driscoll et al., 2013). Therefore, it can reach higher trophic-levels and ultimately affects human health (Sundseth et al., 2017). MeHg(I) in a food web is influenced by abiotic conditions, that influence Hg(II) methylation and bioavailability, and biotic conditions, that regard the organisms' characteristics that make them more susceptible to the pollutant (Kidd et al., 2011).

The input of nutrients is one abiotic factor that affects methylation (MacMillan et al., 2015). Birds can transport and deposit contaminants (Blais et al., 2007) and their guano has a variety of nutrients that will influence the ecosystem where they nest (Mallory et al., 2015). The over-enrichment of nutrients in an aquatic environments can also lead to algae blooms, which decrease the mercury concentration per cell and results in lower bioaccumulation from these organisms to their consumers (Pickhardt et al., 2002). Additionally, algae degradation increases the levels of dissolved organic matter (DOM) (Zhou et al., 2018). DOM is a mixture of several compounds like carboxylic acids and humic substances (Ravichandran, 2004). Research shows that DOM stimulates bacterial activity (Creswell et al., 2017), but the effects on mercury speciation are not as straightforward because it also affects mercury's complexation with ligands and photochemical processes (Klapstein and O'Driscoll, 2018). The chemical form of DOM functional groups depends on pH (Ravichandran, 2004), which is another abiotic factor that influences methylation. Most studies show an inverse relation between pH and MeHg(I) in biota (Chételat et al., 2011; Douglas et al., 2012).

MeHg(I) bioaccumulation is also affected by the ecological characteristics of the organisms (Kidd et al., 2011). Studies show that MeHg(I) concentration depends on the life stage and, generally, adults have higher concentrations (Chételat et al., 2008; Mason et al., 2000). The type of diet of the organisms also affects their MeHg(I) concentration. Generally, MeHg(I) concentration tends to increase with the trophic level (Clayden et al., 2014; Edmonds et al., 2012), but there are some exceptions to this tendency. In fact, the ecology can be more relevant to the MeHg(I) concentration of the organisms than its trophic position. An example is polychaete worms that, although considered a primary consumer, have high levels of MeHg(I) because they create oxygenated burrows walls in anoxic sediments that accumulate DOM, leading to higher MeHg(I) (Sizmur et al., 2013).

To link an organisms' diet and trophic position with its MeHg(I) concentration, many authors use stable isotopes such as $\delta^{15}N$ and $\delta^{13}C$ (e.g., Clayden et al., 2017; Kidd et al., 2012). Organisms tend to retain ^{15}N and excrete ^{14}N, so as the trophic position increases $\delta^{15}N$ values also tend to increase and the $\delta^{13}C$ can potentially assess the source of dietary carbon and its evolution through the food web (Fry, 2006).

The present study aims to address a research gap related to mercury bioaccumulation in lower trophic levels of the food web, more precisely, freshwater and terrestrial invertebrates. It aims to provide insight into what type of invertebrates pose a higher risk to upper trophic levels of the food web, which ecosystems are more susceptible to bioaccumulation and obtain information on the most relevant conditions that affect biomagnification in these systems.

2. Methods

A manual search of some databases, such as Google Scholar and Web of Science™, for relevant articles was performed using keywords such as "mercury", "methylmercury", "bioaccumulation", "biomagnification, "invertebrate" and "freshwater". The selection criteria included only studies that provided the values of MeHg(I) or THg in freshwater and terrestrial invertebrates. Papers that studied sites impacted by anthropogenic activities or by natural phenomena were kept in the analysis. Two articles on controlled environments, which were considered similar to a natural environment, were also kept. Ultimately, it was obtained data from 25 papers. From those, mean values of MeHg(I), THg and the percentage of total mercury in the form of MeHg(I) (%MeHg(I)) of the invertebrates were extracted. It was also extracted information of the invertebrates' stable isotopes $\delta^{15}N$ and $\delta^{13}C$ and information regarding the ecosystem characteristics such as pH, total organic carbon (TOC) and total nitrogen.

Three analyses were made using the software *IBM® SPSS® Statistics*, version 26. The first analysis done was to check the general pattern of MeHg(I) bioaccumulation in the tissue of freshwater and terrestrial invertebrates using the mean values of MeHg(I) and %MeHg(I). The second analysis was a comparison of the MeHg(I) concentration in invertebrates in two different ecosystems. Non-parametric Mann–Whitney *U* tests were conducted to determine the statistical significance. The third analysis conducted was a multiple regression analysis, to examine the combined influence of different variables on MeHg(I) biomagnification. MeHg(I) concentration in the invertebrates was log-transformed and regressed against selected variables regarding the water characteristics of the sites and the $\delta^{15}N$ value. The multiple regression method selected was the stepwise method and as verification of the modeling approach, model selection was made using the Akaike Information Criterion (AIC).

3. Results

Patterns of MeHg(I) Concentration in Invertebrates

The MeHg(I) concentration in each type of invertebrates is represented in Fig. 1. The Canadian MeHg(I) tissue residue guideline for the protection of wildlife consumers of aquatic biota is also plotted, which is 33 µg/kg ww (CCME, 2000), that is the same as 33 ng/g dw. Because the MeHg(I) concentrations

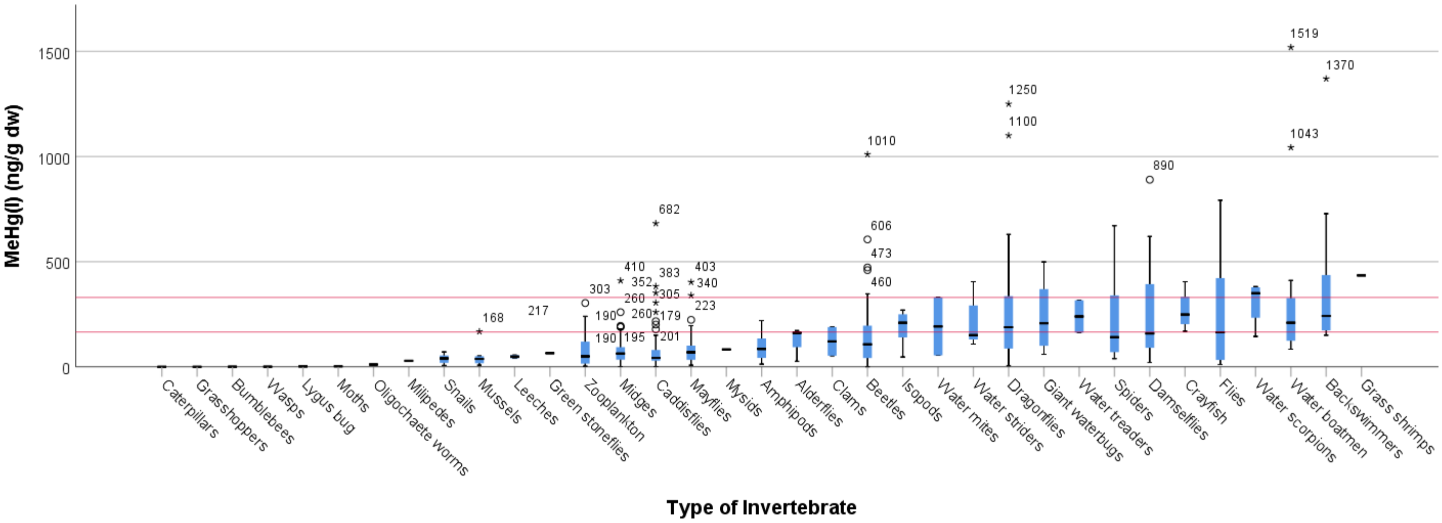

Fig. 1: Boxplots of MeHg(I) concentration in the dry tissue of invertebrates separated by common name, ordered from lowest to highest mean value. The median is represented with a black bar, the outliers with circles and extreme outliers with asterisks. The red lines represent the Canadian methylmercury tissue residue guideline for the protection of wildlife consumers of aquatic biotas (33 µg/kg ww) in dry weight assuming an 80 to 90% water content, which is 195 ng/g dw and 330 ng/g dw respectively.

represented in Fig. 1 are in dry weight it was necessary to convert the guideline value to dry weight. To do this it was considered an interval of 80% to 90% of water content in the invertebrates, which leads to an interval between 165 and 330 ng/g dw.

The type invertebrate with the highest mean value of MeHg(I) concentration is the grass shrimp (M=435 ng/g dw, N=1) followed by backswimmers (M=382.9 ng/g dw, Mdn=242.0 ng/g dw, N=15), water boatmen (M=339.5 ng/g dw, Mdn=210.0 ng/g dw, N=16) and water scorpions (M=306.4 ng/g dw, Mdn=350.0, N=6). It is important to take into consideration that the number of individual data points of each type of invertebrate (referred to as "N") varies significantly. Among all invertebrates reported, caddisflies showed the greatest variation in mean values of MeHg(I) (CV=133.7%, N=76) followed by water boatmen (CV=114.6%, N=16) and beetles (CV=112.8%, N=49).

The %MeHg(I) in the invertebrates is displayed in Fig. 2. Backswimmers had the highest mean value of %MeHg(I) (M=97.2%, Mdn=96.0%, N=5) followed by water treaders (M=Mdn=94.8%, N=2) and water scorpions (M=93.1%, N=1). The type of invertebrate with more variation regarding the mean values of %MeHg(I) mussels (CV=92.5%, N=3), midges (CV=80.4%, N=21) and mayflies (CV=57.8%, N=31).

Comparison Between Wetlands and Lakes

To compare MeHg(I) bioaccumulation and biomagnification in wetlands and lakes, it was selected a smaller data set of data containing information on only caddisflies, mayflies, dragonflies and damselflies. In Fig. 3, it is possible to see the results from the analysis.

In lakes, caddisflies were the group of invertebrates with the lowest mean value (M=54.3 ng/g dw) and median (Mdn=40.0 ng/g dw) but were the group that showed the highest variation between the mean values from the literature (CV=94.3%). Dragonflies were the group with the highest mean (M=177.5 ng/g dw) and median (Mdn=122.0 ng/g dw) and lowest variation (CV=75.7%). In wetlands, mayflies were the group with the lowest mean value, 137.2 ng/g dw (and a median value of 94.1 ng/g dw). Caddisflies showed the lowest median value, of 47.7 ng/g dw, and the highest variation between values (CV=129.3%). Damselflies were the group with the highest mean (M=385.3 ng/g dw) and median value (Mdn=340.0 ng/g dw) and lowest variation between values (CV=70. 8%).

For all four groups of invertebrates, the MeHg(I) concentrations showed to be significantly higher in wetlands than in lakes (Mann–Whitney *U* test, P=0.05, P=0.008, P=0.031, P=0.037 for caddisflies, mayflies, dragonflies and damselflies, respectively).

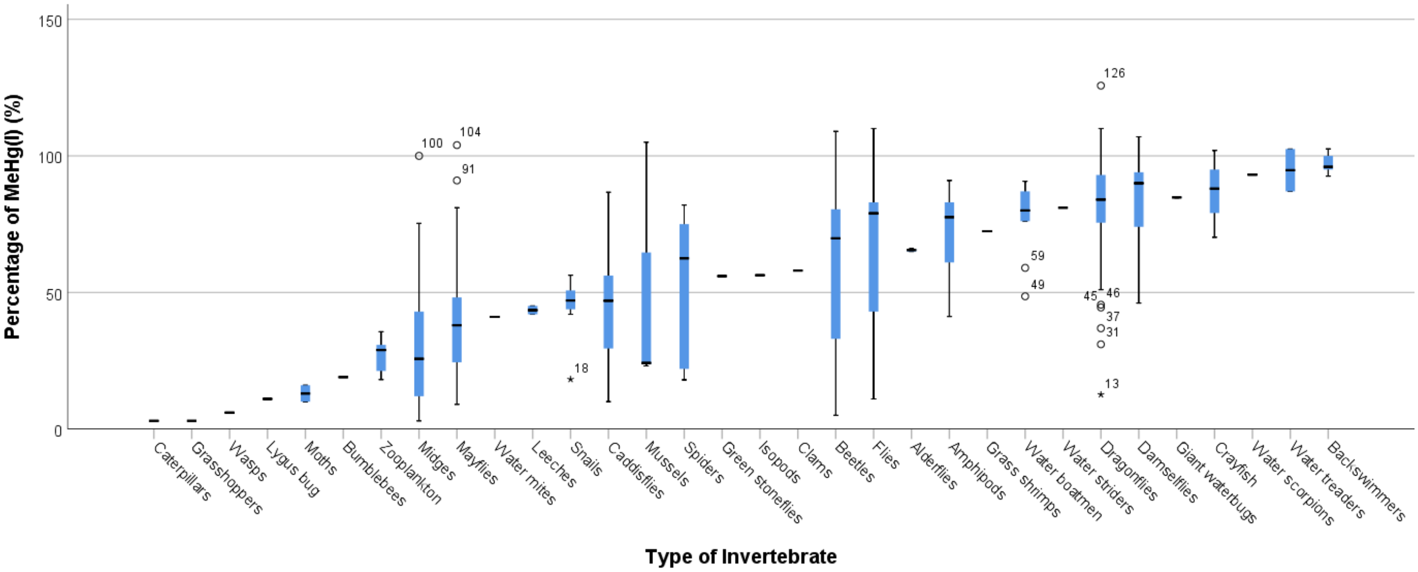

Fig. 2: Boxplots of percentage of MeHg(I) of total mercury concentration in the dry tissues of the invertebrates. In each boxplot the median is represented with a black bar, the outliers marked with circles and extreme outliers marked with asterisks. The invertebrates are separated by common name and ordered from lowest to highest mean value.

Combined Influence of Different Variables in MeHg(I) Bioaccumulation

The first multiple regression analysis was conducted using the full data set of invertebrates. The logarithm of MeHg(I) was regressed against the pH of the study sites and $\delta^{15}N$ of the invertebrates. Two models were created, one with pH as the predictor ($R^2=0.211$, $F=45.487$, $p<0.001$) and the other with pH and $\delta^{15}N$ as predictors ($R^2=0.261$, $F=29.841$, $p<0.001$). On both models, pH showed a negative partial standardized regression coefficient ($b'=-0.459$, $p<0.001$ in the first and $b'=-0.541$, $p<0.001$ in the second). On the second model, $\delta^{15}N$ showed a positive partial standardized regression coefficient ($b'=0.238$, $p=0.001$). The best-fitting model, using the AIC_c, was the one with pH and $\delta^{15}N$ as predictors.

To study the effect of other parameters it was selected a smaller data set, containing information on caddisflies, mayflies, dragonflies and damselflies, and a second multiple regression analysis was made. The logarithm of MeHg(I) was regressed against the pH and TOC of the study sites and $\delta^{15}N$. Two models were created and in both TOC was removed. One model had $\delta^{15}N$ as the predictor ($R^2=0.229$, $F=18.741$, $p<0.001$) and the other had $\delta^{15}N$ and pH as predictors ($R^2=0.622$, $F=50.950$, $p<0.001$). On both models, $\delta^{15}N$ showed a positive partial standardized regression coefficient ($b'=0.479$, $p<0.001$ in the first and $b'=0.770$, $p<0.001$ in the second). On the second model, pH showed a negative partial standardized regression coefficient ($b'=-0.691$, $p<0.001$). The best-fitting model, using the AIC_c, was the one with pH and $\delta^{15}N$ as predictors.

4. Discussion

Patterns of MeHg(I) Concentration in Invertebrates

It was found that the type of invertebrate with the highest mean MeHg(I) in tissue was the grass shrimp, followed by backswimmers, water boatmen and water scorpions. Grass shrimps, backswimmers and water scorpions are predators (Thorp and Rogers, 2011), so their high MeHg(I) concentration is expected due to MeHg(I) capacity of biomagnification (Lavoie et al., 2013). Nevertheless, some invertebrates showed unexpected positions in the Fig. 1, such as water boatmen, that are generally non-predaceous (Thorp and Rogers, 2011).

The type of invertebrate that showed the highest variability in mean values of MeHg(I) were caddisflies, followed by water boatmen and beetles. As mentioned, water boatmen presented not only high mean and median values of MeHg(I) but also a high variability. This could be related to the fact that these organisms belong to the Corixidae family, which has

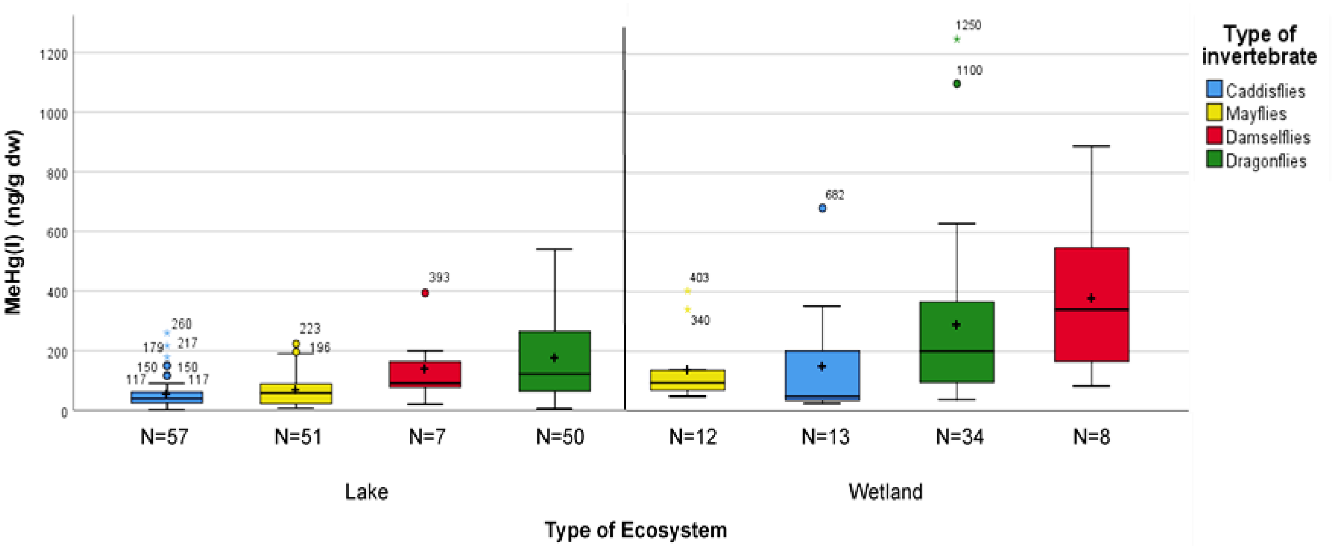

Fig. 3: Boxplots of mean values of MeHg(I) of caddisflies, mayflies, dragonflies and damselflies separated by type of ecosystem, and sorted from lowest to highest mean value. The median is represented with a black bar, the mean with a plus sign, the outliers with circles and extreme outliers with asterisks. The number of individual data points used for each boxplot (N) is also displayed.

species with significantly different ecologies and types of diet. Most water boatmen are non-predaceous (Hilsenhoff, 2001) but studies found that some species showed predatory tendencies and some scavenge dead organisms (Hädicke et al., 2017), which may increase its MeHg(I) concentration by returning MeHg(I) of dead organisms to other trophic levels of the food chain (Sarica et al., 2005).

In Fig. 1, it was observed that a large portion of the invertebrates have values that exceed, for example, the Canadian methylmercury tissue residue guideline for the protection of wildlife consumers of aquatic biotas (CCME, 2000). Also, the majority of the outliers in Fig. 1 are from studies made in wetlands and reservoirs, which poses a risk to high trophic organisms, such as birds and fishes, that feed on the invertebrates that inhabit these ecosystems. Jackson et al. (2014) assessed the THg concentration in songbirds that inhabit the eastern part of North America and concluded that there is a correlation between the type of diet and habitat with the concentration of mercury. Invertebrate-eating species had significantly higher concentrations when compared to omnivores, especially the ones that inhabited wetland habitats. So, a specific type of diet or a change in diet due to external reasons can pose a risk to these organisms and lead them to feed on invertebrates with higher MeHg(I).

Regarding the mean values of %MeHg(I), backswimmers were the type of invertebrate that showed the highest mean value, followed by water treaders and water scorpions. These three groups of invertebrates are predatory which matches with previous studies that found an increase of %MeHg(I) with trophic position (Riva-Murray et al., 2020). Backswimmers not only showed a high mean and median value of %MeHg(I) but also a low variation between the values collected (CV=4.1%). This could potentially indicate that the MeHg(I) concentration could be estimated by measuring THg, which is a simpler laboratory analysis and less expensive.

The types of invertebrates with more variability regarding the mean values of %MeHg(I) were mussels, midges and mayflies. These three types of invertebrates are mostly primary consumers, which can influence their high variability. Riva-Murray et al. (2020) findings suggest that the %MeHg(I) of aquatic primary consumers is correlated with the aqueous MeHg(I) concentration, that is influenced by several environmental conditions (Paranjape and Hall, 2017). Thus, the high susceptibility of primary consumers to MeHg(I) concentration of the surrounding environment may explain their high variability.

The high variability registered in MeHg(I) concentration and %MeHg(I) in some of the invertebrates could potentially be consequence of the decision of identifying them by common name, which implies that some groups of invertebrates include several families and others only one. But even the groups of invertebrates that only include one family showed high variability, which is the case of the water boatmen. It is plausible to ponder if this variability could possibly be reduced if all sampled organisms were identified as species, because grouping invertebrates by a higher taxonomic level could mask the utility of having species-specific values. However, the identification of invertebrate species requires time and expertise and, in fact, Gerwing et al. (2020) showed that in ecological studies there was no significant difference in the conclusions taken when identifying organisms by species or by family, in coastal ecosystems. Nevertheless, the results obtained in this review could be an indication that, for contamination studies, certain individual species behaviors may create outliers in the bioaccumulation data. In these specific cases, the identification by species could provide useful information.

Comparison Between Wetlands and Lakes

With the data extracted from the literature, it was possible to conclude that the concentrations of MeHg(I) in caddisflies, mayflies, dragonflies and damselflies were significantly higher in wetlands than in lakes. Several studies have concluded that wetlands are hot-spots for mercury methylation due to their high content of dissolved organic matter, anoxia, and low pH (Hall et al., 2008). Eagles-Smith et al. (2020) assessed mercury bioaccumulation in aquatic ecosystems in the United States using larvae of dragonflies as biosentinels. In this study, the THg in dragonflies that inhabit wetlands was higher, but not significantly, than the ones that inhabit lakes. For this assessment, it was chosen to analyze THg and not MeHg(I), which could be the reason why a significant difference was not found.

From the group of four, dragonflies and damselflies occupy higher trophic levels because the majority are predators that feed on other insects, such as caddisflies and mayflies, that are generally non-predaceous (Hilsenhoff, 2001). The results obtained also show a correlation between the MeHg(I) concentration and the feeding habits of organisms because, in both ecosystems, the predators showed higher concentrations of MeHg(I).

Additionally, caddisflies were the type of invertebrates with the highest variation between mean values of MeHg(I) in the literature. Clarke (2018) findings show that the MeHg(I) concentration in caddisflies might be correlated with the concentration of MeHg(I) in sediments. It was suggested that this could be related to the fact that the caddisflies sampled in the research were all case-makers, that use natural materials to build protective cases. Also, caddisflies biosynthesized silk to use in the construction of their cases and that can be important to remove Hg(II) from their body (Clarke, 2018). In the present review, it is not possible to conclude that all the data of caddisflies used are from families that construct these protective cases. Therefore, the high variability seen in this group of invertebrates can be related to their susceptibility to the concentration of MeHg(I) in the sediments and also due to the variability of species' ecologies in the order Trichoptera.

Another relevant outtake from this part of the results was the understanding of the clear underrepresentation of wetland invertebrates in studies when compared to lakes. For this analysis, fourteen studies presented this data in lakes and only three in wetlands. It was possible to verify that wetlands seem to pose a higher risk to organisms due to their elevated content of MeHg(I), so further research is needed to identify what factors have a higher influence on MeHg(I) bioaccumulation in this type of ecosystem.

Combined Influence of Different Variables in MeHg(I) Bioaccumulation

Bioaccumulation and biomagnification of MeHg(I) is known to be significantly impacted by the physical and chemical characteristics of the ecosystem (Sumner et al., 2020). In the first multiple regression analysis, it was used the full data set of invertebrates and the MeHg(I) concentration showed to have a positive relationship with the $\delta^{15}N$ and negative with pH, which was expected. Because MeHg(I) has the capacity to biomagnify in a food web, MeHg(I) tends to increase with $\delta^{15}N$. Additionally, studies show that acidic waters are generally correlated with an increase in MeHg(I) concentration in the organisms (Edmonds et al., 2012).

In the second analysis, using a smaller data set of invertebrates, the same relationships between MeHg(I) and $\delta^{15}N$ and pH were found. The results showed that the MeHg(I) concentration in caddisflies, mayflies, dragonflies and damselflies was significantly affected by $\delta^{15}N$, however, by adding pH to the model, the correlation between MeHg(I) and the combined effect of $\delta^{15}N$ and pH was much higher.

In the second multiple regression analysis, it was also included TOC, but it was removed from the models because it was the variable with the weakest correlation. Organic matter can have several effects on mercury speciation that depend not only on its concentration but also its composition (Ravichandran, 2004) and research shows some contradictory results regarding its effects (Jiang et al., 2018). Therefore, the weak correlation obtained in the analysis between MeHg(I) and TOC can be associated with this complexity of effects that organic matter can have.

The first analysis was executed with the data of six different articles and the second with only three, because only these provided the information needed. Consequently, the strength of the analysis was not as rigorous as desired. An important take away message from this review was the understanding that there is essential information missing in many of the articles examining bioaccumulation and biomagnification of MeHg(I), more precisely, regarding the physico-chemical characteristics of the ecosystems such as pH, the content of organic matter and the concentration of important nutrients. This information can be unnecessary for some studies but will enable further and deeper analysis of the topic.

Application to Risk Assessment

In 2017, the Minamata Convention entered into force and the Parties agreed to tackle the anthropogenic emissions of mercury (UN Environment, 2019). Although the agreement refers the importance to address atmospheric, land and water releases, a lot more attention has been put in the atmospheric emissions (You, 2015) which has a separate article in the Convention report (UN Environment, 2019). The article where water releases are referred also compromises the land releases and the requirements on these are weaker than the ones for atmospheric emissions (You, 2015). The coverage of the article is narrow and does not define specific relevant point sources and requirements for controlling them (You, 2015).

In the literature, it is also possible to see the discrepancy between the mercury inventories that have been made regarding global atmospherics emissions and direct releases to aquatic systems, that have been widely understudied (Kocman et al., 2017). This research also supports the importance to assess mercury releases to aquatic systems due to their susceptibility to be affected by MeHg(I), that bioaccumulates and biomagnifies through the food webs of these systems.

Additionally, although there has been a large decline in atmospheric mercury over the past years (Zhang et al., 2016), research shows that a discrepancy exists between this trend and the one seen in mercury in aquatic biota, due to processes that occur specifically in the ecosystems (Wang et al., 2019). This review supports the importance of internal processes of the ecosystems that makes the organisms that inhabit there more susceptible to MeHg(I).

Taking this into consideration, mercury risk assessments could potentially be performed using monitoring data of mercury concentration in invertebrates from field studies. Certain families of invertebrates are distributed globally and inhabit several types of ecosystems. Furthermore, the sampling of these organisms is much simpler than sampling fishes. It is relevant to note that bioconcentration of MeHg(I) from the water column to plankton can be a good predictor of the vulnerability of the ecosystems to MeHg(I) biomagnification (Wu et al., 2019) and also that upper trophic level organisms can be a better indicator of mercury in certain ecosystems. Nevertheless, it is important to mention that invertebrates are a key linkage in food webs and important for biomagnification in the ecosystems.

Consequently, using invertebrates as biomonitors could be a good way to track mercury in aquatic environment and if it was included the analysis of the physico-chemical parameters of the study sites, a lot of information could be added to the assessment and improve the knowledge on the specific processes that occur in each ecosystem.

As such, the best practices for the analysis of bioaccumulation and biomagnification of mercury in the invertebrates' food web are assessing the concentration of MeHg(I) and THg, the %MeHg(I) and the stable isotopes of the invertebrates and physico-chemical characteristics of the ecosystems, such as the pH value, the concentration of organic matter, the concentration of dissolved oxygen, the concentration of phosphorus and nitrogen. And the best practices to report these studies include providing the raw data of MeHg(I), THg and %MeHg(I) in the invertebrates (mean, median, standard deviation and number of samples). The lowest taxonomic level possible of each type of invertebrate should also be provided or data on stable isotopes, such as $\delta^{15}N$, because it allows better differentiation between organisms and might reduce the variability. As mentioned before, in contamination studies is also essential to be aware of specific invertebrates that, due to their behavior, can be outliers in the dataset. For these organisms, it would be important to identify them by species. The reports should also include data regarding the physico-chemical characteristics of the ecosystem and if the study includes the analysis of different sites, these parameters should be reported individually and not present a mean value of all sites.

5. Conclusions

The results of this review allowed to conclude that predatory tendencies are a key influence on the general patterns of MeHg(I) concentration in invertebrates. Also, a large percentage of the invertebrates exceeded the guideline value chosen. This confirms that MeHg(I) in the lower trophic levels of the food web is a key consideration in ecological risk assessment of higher organisms.

The high variability in mercury content of some species of invertebrates indicates that grouping by family may not be appropriate in contamination studies and that more research is needed to identified key species outliers.

The analysis also confirmed that certain types of ecosystems pose a higher risk for MeHg(I) bioaccumulation and biomagnification, such as wetlands.

It was possible to understand that, in articles examining bioaccumulation and biomagnification of MeHg(I), there is important information missing regarding the physico-chemical characteristics of the studied ecosystems.

It was also possible to conclude that invertebrates can be used as biomonitors due to the global distribution of some families and because the sampling of this organisms is significantly simpler than upper trophic organisms, such as fishes. Additionally, the assessment of the physico-chemical characteristics of the habitats from where they were sampled provides a significant increase of knowledge of MeHg(I) bioaccumulation and biomagnification through the food web in the studied ecosystem.

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