Special Issue on Bioconversion, Bioaccumulation and Toxicity of Mercury in a Changing World

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1. Mercury in a Changing World

Mercury (Hg) is recognized as a persistent global chemical contaminant that accumulates in biota, thus being an ecological hazard, as well as a health risk to fish consumers. Human past and current activities play a predominant role in the emission and mobilization of Hg in the environment. Elemental Hg (Hg₀) and inorganic Hg (IHg) emitted in the environment are constantly cycled and recycled through Hg biogeochemical cycle, among which bioconversion by microorganisms into mono-methyl-Hg (MMHg), bioaccumulation (MMHg and IHg) and biomagnification (MMHg) in food webs is a critical aspect for Hg toxicity to biota, as well as for humans. Despite decades of research, global Hg pollution requires a deeper understanding of specific toxicity mechanisms for Hg compounds and of the mechanisms underlying its transport and accumulation in biota. Indeed, the entrance of Hg into food webs is not fully understood.

In light of the above, this Special Issue was introduced to collect the latest research on relevant topics, and more importantly to address present challenging issues with the cycling of Hg in complex systems. The accepted papers addressed various topics, mainly on bioaccumulation, food web transfer, effect and development of new methods in these contexts.

2. Uptake in Low Trophic Levels

MMHg has been repeatedly observed to biomagnify through food webs. Hg concentrations at the base of the food web seem to be the main factor controlling Hg transfer in food webs, rather than contrasted biomagnification rates. Risk reduction in top predators being poisoned via ingestion of contaminated preys appears central for Hg risk management. Three papers concerned uptake of IHg and MMHg in biota [1–3].

The paper, authored by Vignati et al. (2020), tested the use of standardized laboratory tests to infer Hg bioaccumulation in indigenous benthic organisms of Lake Maggiore (Italy) [1]. Collecting an adequate amount of biomass to assess Hg accumulation in resident freshwater benthic invertebrates is a challenge, and site intercomparison may be limited by seasonality, as well as by the availability of species at all monitoring points. Laboratory-reared organisms are attractive for bioaccumulation experiments in medium- to large-sized monitoring programs. The authors observed a good correlation of Hg concentrations in *Chironomus riparius* versus indigenous chironomids, suggesting the possibility of using linear regressions to predict Hg accumulation by these benthic invertebrates [1].

The review of Hg uptake in the freshwater macrophyte *Elodea nuttallii*, authored by Cosio (2020), demonstrates that this plant bioaccumulates both IHg and MMHg [3]. Nonetheless, IHg shows a higher affinity to cell walls and a basipetal in planta transport, whereas MMHg acropetal transport was evidenced [3]. In contrast, uptake in the absence of dissolved organic matter (DOM) in water gave highly similar enrichment factor for IHg and MMHg, resulting in similar intracellular THg concentration in *E. nuttallii*, pointing to a role of affinity to DOM in the observed difference between the fate of IHg and MMHg observed in biota in the field [3].

In the same line, the paper authored by Monperus et al. (2020) evidenced a contrasted toxicokinetic of IHg and MMHg in glass eels [2]. After rapid uptake through the gills, MMHg was transported in the heart, the liver, and the brain, and finally transferred to the skeletal muscles. IHg uptake was also mainly observed in gills but transited through olfactory bulbs with a very low transfer and storage in the other organs and a rapid depuration [2]. As previously observed in *E. nuttallii*, data supported the importance of chemical speciation on the target cells and tissues for Hg accumulation [2].

3. Food Web Transfer to Fish

Two papers addressed food web transfer of Hg to fish [4,5]. The paper, authored by Gentès et al. (2020), determined the bioavailability of MMHg in aquatic freshwater system, revealing MMHg production in plant roots and MMHg trophic transfer to *Pseudancistrus* sp. tissues and gut [4]. Authors highlight that MMHg formed in the periphyton could significantly participate in food web contamination, because aquatic plants are an important food source [4]. The paper, authored by Camacho et al. (2020), assessed the bioaccumulation of Hg in the commercial marine flat fish species *Solea senegalensis* co-exposed to dietary MMHg and climate change (warming and acidification) [5]. Fish liver exhibited the highest Hg concentration, followed by brain and muscle [5]. Warming enhanced Hg bioaccumulation, whereas acidification decreased this element's levels [5]. Hazard quotient (HQ) estimations evidenced that human exposure to Hg through the consumption of fish species may be aggravated in tomorrow's oceans, thus raising concerns for future seafood safety [5].

4. Effect of Hg in Biota

Two papers detailed effects of Hg in biota [3,5]. In the macrophyte *E. nuttallii*, IHg reduced chlorophyll, while MMHg increased anthocyanin [3]. Transcriptomics and metabolomics in shoots revealed that MMHg regulated a higher number of genes and metabolites than IHg [3]. Proteomics and metabolomics in cytosol revealed that IHg had a greater effect than MMHg [3]. In sum, MMHg and IHg showed different cellular toxicity pathways [3]. The main impact of MMHg and IHg was on the non-soluble compartment and the soluble compartment, respectively [3]. This is congruent with the higher affinity of IHg with DOM and cell walls.

In *S. senegalensis*, neuro-oxidative responses were affected by both climate change-related stressors and Hg dietary exposure [5]. Exposure to dietary MMHg reduced weight, length, catalase (CAT) and superoxide dismutase (SOD) activities in muscles and the liver, respectively, and increased catalase and acetylcholinesterase activities in the brain, as well as the lipid peroxidation (LPO) in the brain [5]. Interactions with warming and acidification were antagonistic for growth, CAT in the liver and the brain, SOD in the muscle and the liver, and LPO in the brain [5]. Authors suggested for these interactions to take place via variations of metabolic rates [5]. The three studied stressors elicited differential neuro-oxidative responses in fish muscle, liver, and brain [5]. Such differences can be mostly attributed to the different Hg concentrations accumulated in tissues (liver > brain > muscle), and to specific modes of action in target organs [5]. As such, data showed a more notorious neuro-oxidative response in the brain [5]. The data support the viewpoint that seafood safety will be likely compromised if the climate continues to change as projected. The authors stress that future legislation and recommendations regarding Hg risk assessment should account for the expected effects of warming, while advising a conscious and parsimonious consumption of wild seafood species [5].

5. Effect of Dietary Hg to Microbiota

In the past decade, the intestinal microbiota has been recognized as a major player in the health of its host as it takes part in major biological functions, including nutrition, immunity, and the metabolism of xenobiotics. As such, it is hypothesized to play a major role in the exposure of the host to dietary pollutants, including Hg. Two papers studied the impact of Hg on microbiota [4,6]. The paper authored by Brantschen et al. revealed changes in microbiota of earthworms exposed to Hg contaminated soils [6]. Data pointed out that the health of macro-organisms in the food web might be impacted

through changes in their gut microbiome, subsequently affecting their metabolism and legacy in the soil [6].

In fish exposed to roots of macrophytes, methylating bacteria were identified in the gut contents of the fish [4]. As such, authors observed not only the dietary transfer of MMHg, but also of bacteria from plants to fish [4]. Besides, Hg methylation is strongly suspected in the fish gut, potentially increasing the Hg bioaccumulation. This endogenous methylation is a current major question in the context of the environmental impact of Hg and the understanding of the Hg cycling in ecosystems [4].

6. Development of New Approaches

Two papers that presented recent methodological progress are promising for the understanding of the bioaccumulation and effect of Hg [2,3]. A multi-isotope image mapping procedure was developed by Monperrus et al. (2020) to simultaneously study the uptake and distribution of both MMHg and IHg within the organs of glass eels [2]. Cosio (2020) coupled multiomics approaches to identify cellular toxicity pathways at several levels of organization and the early impact of sublethal pollution [3].

In 2013, the nations of the world agreed on the first global treaty to mitigate the many deleterious health outcomes associated with Hg release into the environment and MMHg exposure. In this context, all the papers in this Special Issue provide innovative insights into the Hg fate in the aquatic and terrestrial ecosystems and increase the understanding of the Hg biogeochemical cycle [1–6].

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