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Origin and Fate of Mercury in Tunas

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Summary

Tunas are among the most consumed seafood products but contain relatively high levels of methylmercury, the most toxic form of mercury. Limited observations suggest tuna methylmercury concentrations vary in space and time, yet the drivers are not well understood. Environmental policy under the United Nations Minamata Convention aims to protect health by reducing mercury emissions from anthropogenic activities and call for bio-monitoring programs to evaluate its impacts on methylmercury levels in the ocean and in marine fish. In this context, we built the largest mercury database in tropical tunas and albacore (n > 5,700) to explore spatial and temporal patterns of tuna Hg levels in the global ocean, and highlighted the following points:

- Global maps of tuna mercury levels highlighted strong spatial patterns among species, once accounting for natural methylmercury bioaccumulation with tuna length/age.
- Spatial patterns in tuna mercury levels are mainly due to natural marine biogeochemistry governing methylmercury profiles in seawater, and tuna foraging depth.
- Increasing tuna mercury concentrations were found in the late 1990s in the northwestern Pacific, concomitant with increasing anthropogenic mercury emissions from Asia.
- Elsewhere, stable mercury concentrations since the 1970s contrast with an overall decline in mercury emissions and deposition globally, which likely reflects the inertia of the ocean supplied by legacy emitted mercury, and calls for aggressive reduction emission measures.
- This work elevates tunas as relevant bio-monitoring tools to document changes of the marine mercury cycle in a context of reduction emission measures and climate change, and calls for large-scale biomonitoring programs.

Background

Tuna is one of the most widely eaten saltwater fish in the world, yet it is known to contain methylmercury (MeHg), the most toxic form of mercury (Hg). Methylmercury poisoning can cause neurodevelopment disorders in foetuses during pregnancy and in children, and cardiovascular risks in adults (Axelrad et al., 2007; Genchi et al., 2017), and its health and socioeconomic costs are estimated at one billion dollars annually worldwide (Zhang et al., 2021). The United Nations Environment Programme Minamata Convention aims at reducing anthropogenic Hg emissions to protect human health, advocating governments and policy makers to balance economic, environmental and health interests.

Mercury sources are both natural (e.g., volcanism, soil leaching) and anthropogenic (e.g., fossil fuel combustion and artisanal and small-scale gold mining) (Fig. 1) (Outridge et al., 2018), but their relative contributions are still poorly known. Once released, Hg transport and fate is complex and influenced by chemical, physical, biological and ecological processes. From the atmosphere, Hg can be deposited or taken up by the ocean, where an unknown fraction of Hg is microbially converted into MeHg (Mason and Fitzgerald, 1990), which is easily absorbed by organisms and can biomagnify along marine food webs (Fig. 1). This compound also bioaccumulates within individuals over time, resulting in higher MeHg concentrations in bigger fish. Humans are exposed to MeHg mainly by consuming marine fish (Sunderland et al., 2009), especially top predators like tropical tunas that exhibit high MeHg concentrations compared to the rest of the food web (Bodin et al., 2017; Choy et al., 2009), sometimes exceeding food safety guidelines (1 mg/kg fresh tissue) (FAO and WHO, 2005). Understanding the factors that drive MeHg concentrations in tunas, and how the implementation of reduction Hg emission measures will interfere in the process, is therefore critical to policy makers and, ultimately, to limiting human exposure to MeHg.

This paper summarizes 10 years of research (2014-2024) conducted in the Pacific primarily and extended to other oceans, exploring spatial and temporal patterns of tuna MeHg concentrations with the aim of tackling the yet-answered question of what controls the origin and fate of MeHg in tunas in the global ocean.

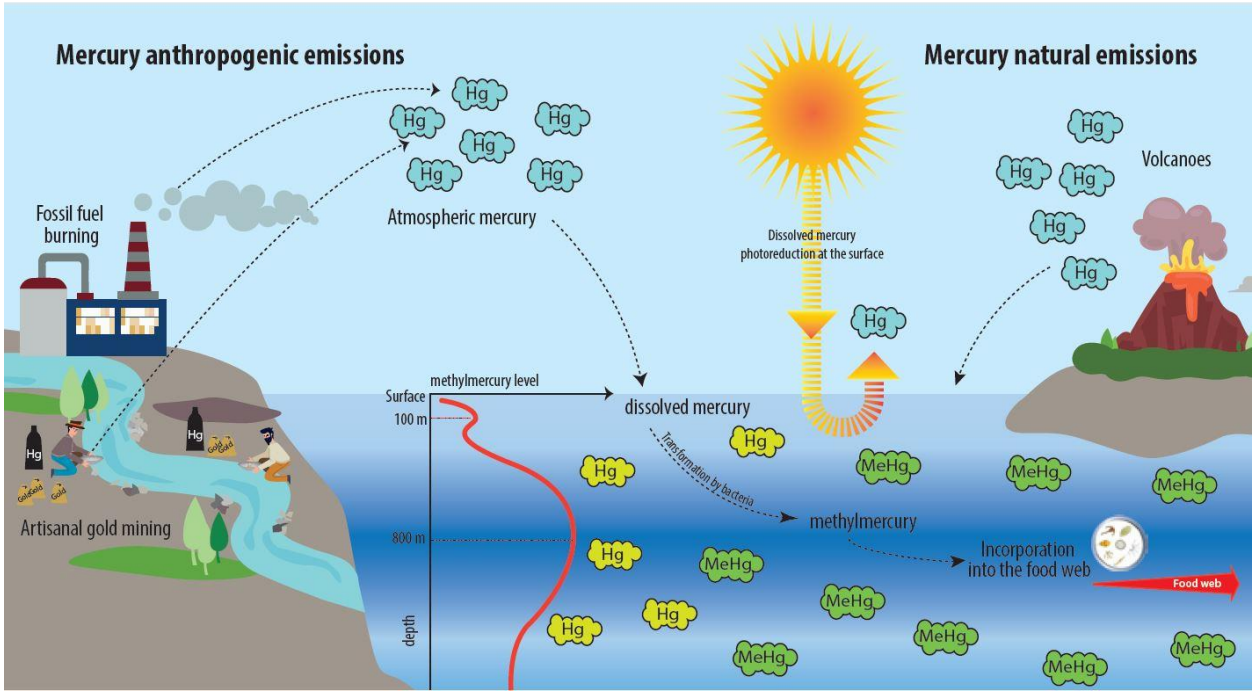

Figure 1. Where does methylmercury (MeHg) in the oceans come from? ©Constance Odiardo, Pacific Community

Material and methods

With the help of multiple partners, including the Pacific Community, and assisted by access to several tuna tissue banks including the Pacific Marine Specimen Bank, we compiled more than 5,700 total Hg concentrations measured in muscle tissue of tropical tunas (*n* = 1,422 bigeye, 2,467 yellowfin, and 1,003 skipjack) and albacore (*n* = 900) from the Pacific (*n* = 3,551), Indian (*n* = 1,218) and Atlantic (*n* = 1,023) oceans. Total Hg concentrations are considered to reflect MeHg concentrations, as most of the total Hg (> 91%) is in its methylated form in tuna white muscle (Médieu et al., 2023). The vast majority of total Hg analyses was performed at GET (Toulouse, France). For each tuna sample, precise date and coordinates of fishing operations were recorded to be able to explore spatial and temporal variability of tuna Hg concentrations at a high resolution. Individual tuna length was also recorded to account for natural bioaccumulation of Hg over time. See Médieu (2022) for additional information on tuna sampling and chemical analyses.

Inter-species differences in tuna mercury concentrations

Mercury concentrations significantly differ among tuna species, following the general pattern: bigeye > albacore > yellowfin and skipjack (Fig. 2) (Médieu et al., 2023). While bigeye, albacore, and yellowfin can display Hg concentrations exceeding the food safety guidelines of 1 mg/g fresh tissue (FAO and WHO, 2005), skipjack samples always show Hg levels below the limit. Samples with Hg concentrations exceeding the limit are generally associated with bigger individuals, illustrating the natural bioaccumulation of Hg in organisms through time, and the consequent need to consider both tuna species and fish size when addressing recommendations of food safety regarding Hg content.

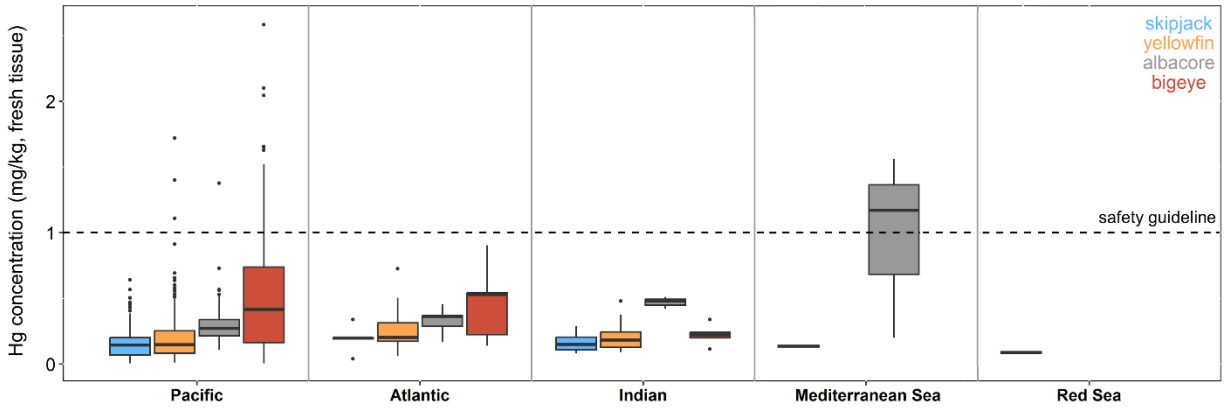

Figure 2. Inter-species variability of observed mercury concentrations (Hg, mg/g, fresh tissue) measured in white muscle tissue of skipjack (blue), yellowfin (orange), albacore (grey), and bigeye (red) caught in the global ocean, from Médieu et al. (2023). The dashed line represents the food safety guideline of 1 mg/g fresh tissue.

Tunas mirror spatial patterns of mercury bioavailability and pollution in the ocean

Despite the scientific interest, and the public health and economic needs to document tuna Hg variability in the global ocean, the vast majority of studies available to date have been conducted on a small spatial scale, relying most of the time on imprecise tuna catch locations. Intra-specific differences of tuna Hg levels were highlighted regionally in the eastern Pacific (Ferriss and Essington, 2011) and the western Indian (Chouvelon et al., 2017), and among 12 locations in the global ocean (Nicklisch et al., 2017), yet relying on spot samples acquired at a low spatial resolution, and without identifying the drivers.

Our global dataset enabled to produce broad-scale and high-resolution maps of Hg concentrations in bigeye, yellowfin, and albacore in the western central Pacific region (Houssard et al., 2019), and in skipjack in the entire Pacific Ocean (Médieu et al., 2022). This helped refining the assessments of risk exposure to Hg associated with human consumption according to tuna species and tuna catch areas.

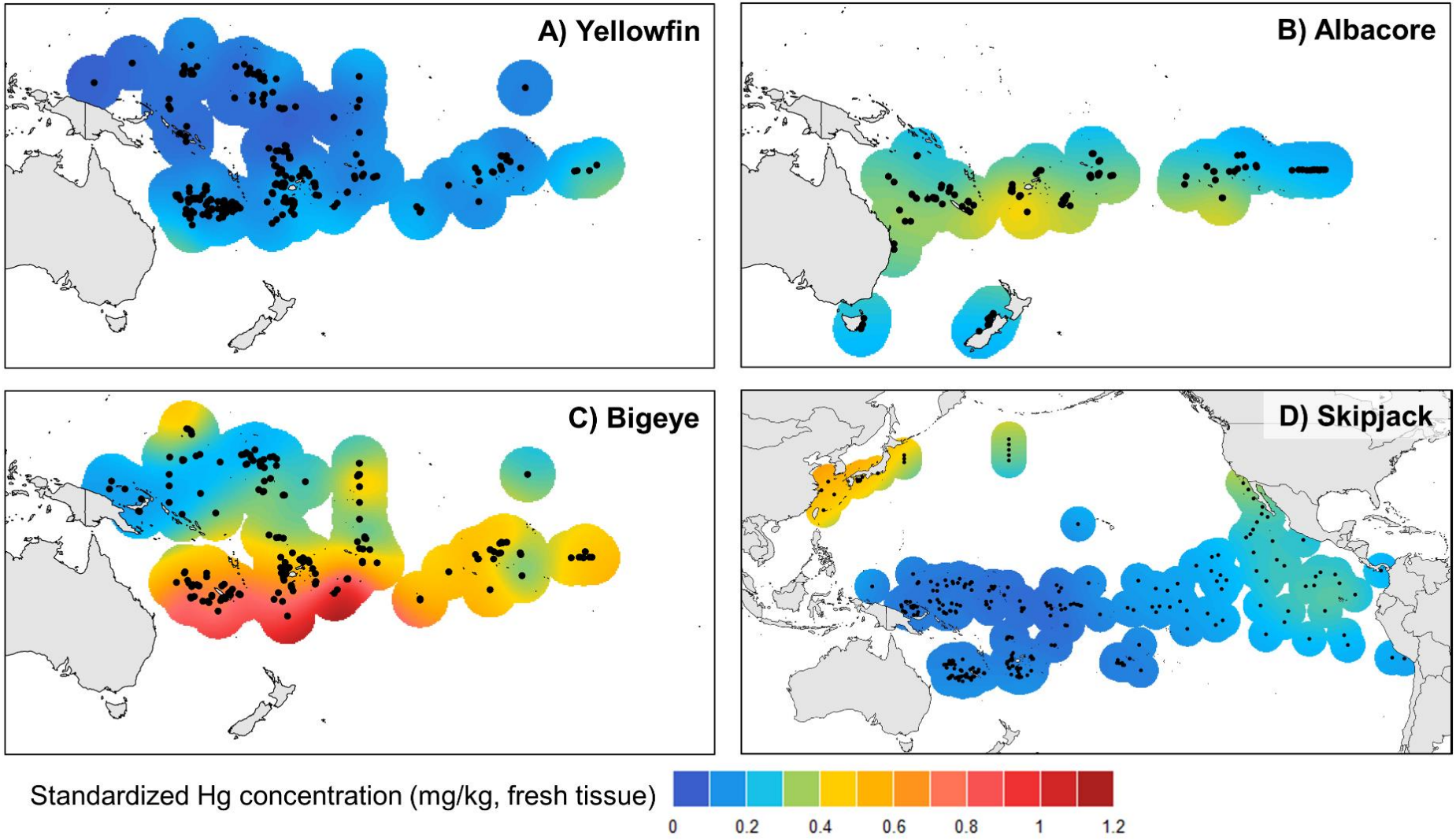

Figure 3. Spatial distribution of standardized mercury concentrations (Hg, mg/kg, fresh tissue) in A) yellowfin, B) albacore, C) bigeye, and D) skipjack, from Houssard et al., (2019) and Médieu et al., (2022). Black dots show the catch location of the sampled fish.

As Hg naturally bioaccumulates over time, leading to higher Hg levels in older/bigger individuals, we developed a method to length-standardize tuna Hg concentrations, hereafter defined as standardized Hg concentrations in contrast to observed raw Hg concentrations (Houssard et al., 2019). This enabled us to reveal and explore strong spatial patterns of tunas not explained by Hg bioaccumulation and fish size differences, but likely due instead to other processes. Within the western central Pacific region, higher standardized Hg levels were found around New Caledonia and Fiji compared to the equator, in yellowfin, bigeye and albacore (Fig. 3A-C) (Houssard et al., 2019). In skipjack, standardized Hg concentrations were 1.5 to 2.0 times higher in the northwestern Pacific than in the eastern region, and 4.0 to 5.0 times higher than in the western central Pacific (Fig. 3D) (Médieu et al., 2022).

In order to explore the drivers of tuna Hg spatial variability, we used a set of tracers to establish whether the underlying accumulation mechanisms were related to tuna dietary differences, MeHg bioavailability at the base of marine food webs, or anthropogenic Hg emissions. In the western central Pacific, this transdisciplinary approach revealed that spatial variability of standardized Hg concentrations in tropical tunas and albacore was mainly explained by both tuna foraging depth, and local biogeochemistry driving variability of seawater MeHg concentrations (Houssard et al., 2019). At the same size, higher Hg concentrations in bigeye compared to the three other species are likely due to its ability to forage deeper, in ocean layers where MeHg concentrations are generally highest (Fig. 1). Such link between MeHg concentrations and bioavailability in seawater and Hg concentrations in marine top predators is expected but rarely observed due to scarce MeHg data in the water column as they require scientific campaign at sea and specialised equipment (e.g., trace metal clean rosette) to be collected. Around New Caledonia and Fiji, we compared seawater MeHg concentrations measured from the surface down to 600 m during the 2015 OUTPACE sea campaign (Leblanc and Cornet, 2018) and Hg concentrations in muscle and blood samples of tropical tunas and albacore, four species known to exhibit contrasted foraging depths in the western Pacific according to tagging data (Evans et al., 2011; Houssard et al., 2017; Williams et al., 2015). Such comparison revealed a significant positive trend between dissolved MeHg concentrations and tunas Hg levels (Fig. 4) (Barbosa et al., 2022), confirming the importance of both marine biogeochemical processes leading to variable MeHg levels in seawater, and tuna foraging depth.

At the Pacific Ocean scale, large spatial patterns of standardized Hg concentrations in the epipelagic skipjack were also mainly explained by the natural functioning of the Pacific Ocean, likely via variations in the MeHg water concentrations and the depth at which MeHg peaks within the water column (Médieu et al., 2022). The relatively high standardized Hg concentrations in the eastern Pacific and northwest Pacific (Fig. 3D) appeared to be due to low oxygen levels in the ocean, especially in the eastern region, owing to bacteria breaking down surface organic matter. We hypothesised that such specific conditions in these areas caused MeHg concentrations to peak in water closest to the surface (< 100 m) (Bowman et al., 2020), where skipjack live and feed, as compared with the western Pacific, where the MeHg peak occurred in deeper water, between 400 and 800 m (Fig. 4).

The very high skipjack Hg concentrations in the northwest Pacific (Fig. 3D), however, may also be due to major sources of anthropogenic emissions located nearby and associated to the heavy use of fossil fuels for power generation in Asia. Such human-related sources add to natural biogeochemical processes that are conducive to surface MeHg bioavailability in food webs.

Overall, these broad-scale and transdisciplinary studies lead to the hypothesis that the spatial variability in tuna Hg concentrations is mainly explained by both tuna foraging depth and the vertical and horizontal variability of MeHg bioavailability in the ocean. These results shed light on the potential power of combining tropical tunas and albacore to investigate both horizontal and vertical variability of MeHg bioavailability in the water column, and MeHg biomagnification pathways along pelagic food webs.

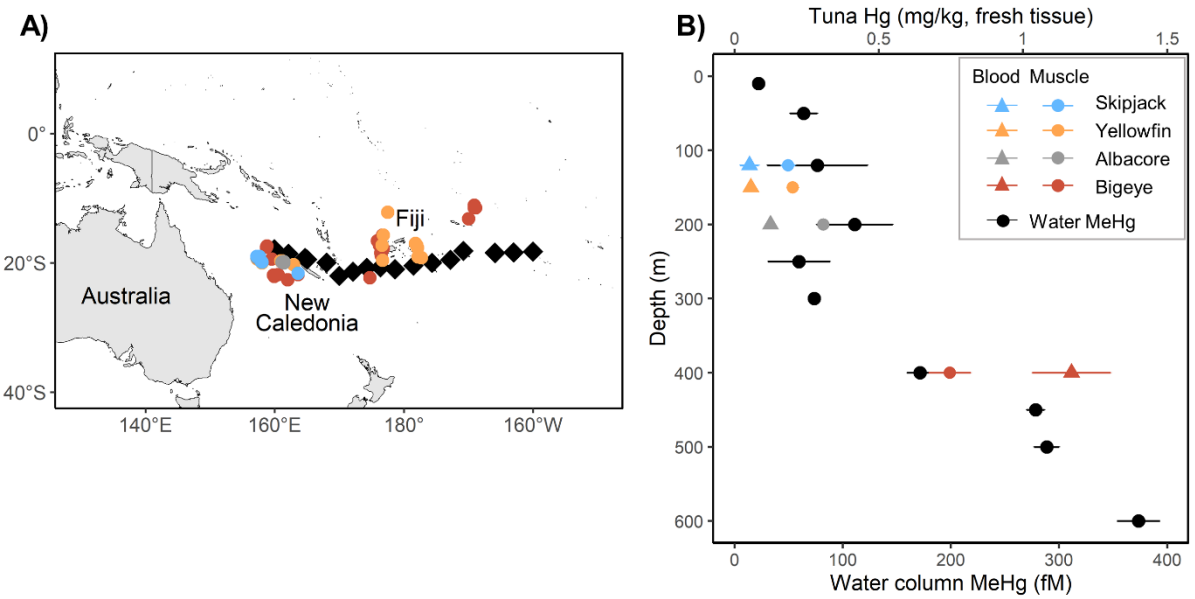

Figure 4. The importance of tuna foraging depth in the context of marine methylmercury (MeHg) depth profiles in the southwestern Pacific Ocean. **A)** Location of seawater (black diamonds) and tuna (colored dots) blood and muscle samples used to investigate the link between tuna mercury (Hg) concentrations and dissolved MeHg levels in the water. **B)** Relationship between total Hg concentrations (mg/kg, fresh tissue) in tuna species exhibiting different foraging depths, and concentration-depth profile of dissolved MeHg (femtomolar fM, concentration-depth profiles were averaged from all locations), from Barbosa et al., (2022). Species-specific foraging depths were estimated with tagging data from the western Pacific Ocean (Evans et al., 2011; Houssard et al., 2017; Williams et al., 2015).

Stable mercury concentrations in tunas since 1971 illustrates marine inertia and the need for strong emission reductions under the Minamata Convention

Another key research question regarding tuna Hg concentrations is if they are able to reflect temporal changes of Hg trends and inputs in the environment. Most anthropogenic Hg releases are estimated to have occurred during the past five centuries (Streets et al., 2019a). Cumulative releases have been largest in North America and Europe, yet anthropogenic emissions and deposition in these two regions have declined since the 1970s, following emission reduction measures. Conversely, anthropogenic Hg releases in Asia have increased since the 1980s. Anthropogenic Hg uses and emissions have considerably modified the natural global Hg cycle (Outridge et al., 2018), and are suspected to have tripled Hg levels in the open ocean (Lamborg et al., 2014). Yet it is still unclear which fraction of anthropogenic Hg is converted into MeHg and biomagnified in marine biota. While tunas represent a major route of human exposure to MeHg, temporal studies of their Hg concentrations are limited, showing contrasting temporal trends among species and ocean regions (Drevnick et al., 2015; Drevnick and Brooks, 2017; Kraepiel et al., 2003; Lee et al., 2016; Médieu et al., 2021).

In this context, we investigated the temporal variability of standardized Hg levels in tunas from 1971 to 2022 in six regions of the global ocean (Fig 5; Médieu et al., 2024), and compared them with estimated regional anthropogenic Hg emissions to the atmosphere since the 1970s (Fig 6A; Streets et al., 2019a, 2019b). Mercury concentrations in tunas were highly variable in the global ocean, but remained overall stable over the study period, except in the northwestern Pacific, off the coast of Asia (Fig. 6B). In this area, Hg concentrations in skipjack increased significantly in the late 1990s, confirming the local impact of anthropogenic Hg emissions from Asia, as explained in the previous section.

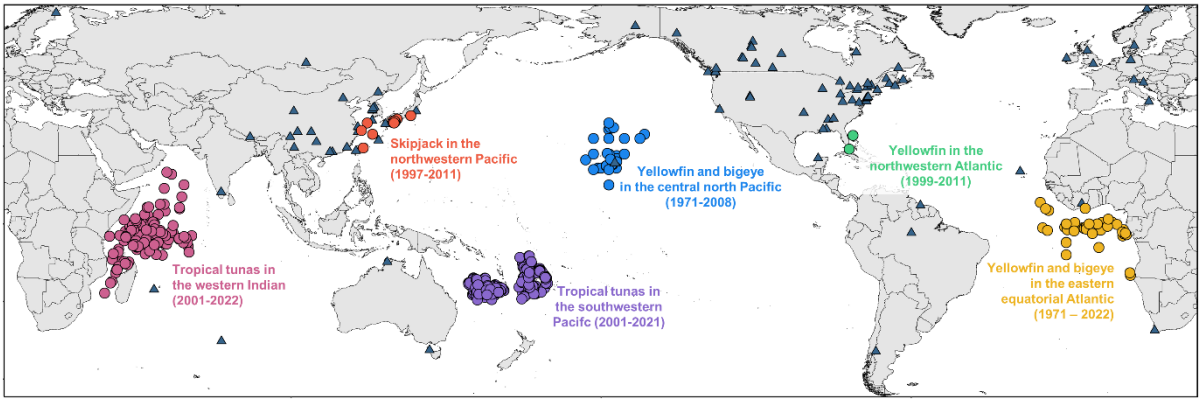

Figure 5. Spatial distribution of tropical tunas analysed for mercury (coloured circles) and atmospheric mercury level observation sites (blue triangles).

Elsewhere in the global ocean, the stability of standardized Hg concentrations in tunas contrasts with a global decline in atmospheric Hg emissions beginning in the 1970s (Fig. 6A). We assume that this stability in tunas is due to subsurface (between 50 m and 1500 m) and deep (below 1500 m) ocean inertia, and the resulting upward mixing of legacy Hg from deep ocean layers into surface waters (i.e. the uppermost 50 m of the water column). Mercury that was emitted several centuries ago continues to circulate between the different parts of the biosphere (atmosphere, vegetation, surface ocean and deep ocean), but at varying speeds. While surface waters rapidly equilibrate with the atmosphere, deeper oceanic waters have a longer residence time, so Hg that accumulated there takes longer to be redistributed to shallower waters where tuna live and feed. In those waters, Hg could have been emitted decades or centuries ago, and not yet reflect the effects of the reduction in Hg emissions into the atmosphere.

In order to better understand how emission reductions impact Hg concentrations in the different ocean layers, we used a Hg circulation model (Amos et al., 2013), featuring three emission reduction scenarios based on varying degrees of stringency (Angot et al., 2018; Pacyna et al., 2016). Such modelling illustrated that even if Hg emissions into the atmosphere were drastically reduced, it would take almost 10 years to detect a fall in Hg levels in surface waters, and approximately 25 years to detect them in subsurface waters; therefore, at least as long to see a decline in tunas. If as much Hg continues to be emitted as it is today, the model does not predict any decrease in the various ocean layers between now and 2100. Overall, we predict that the greater the reductions in emissions, the sooner we can expect to see a decline in Hg concentrations in tunas. Far from suggesting that the Minamata Convention is ineffective, our study highlights the need to continue the global effort to reduce Hg emissions more energetically.

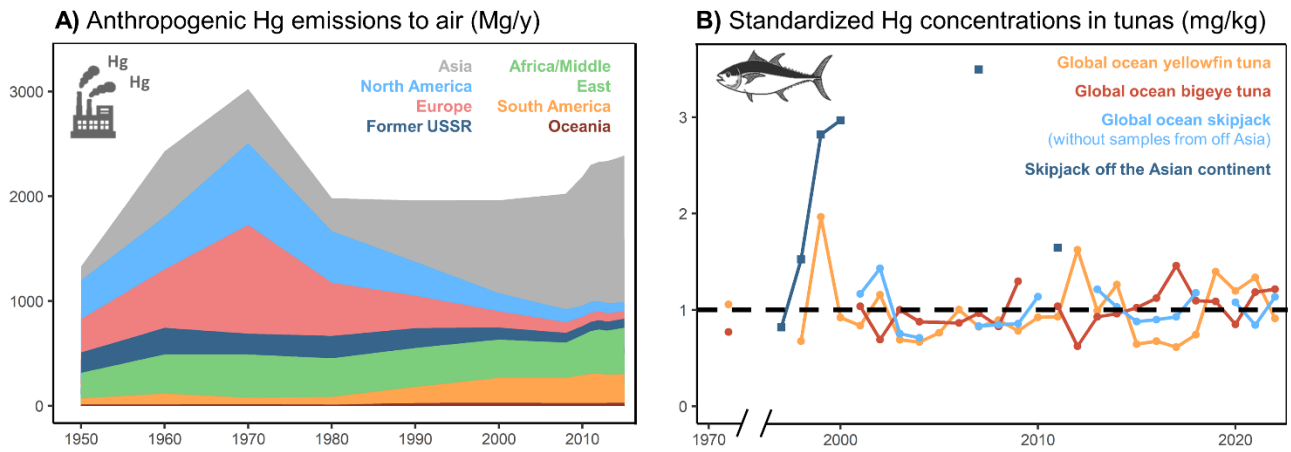

Figure 6. A) Temporal anthropogenic mercury releases from 1950 to 2010 by world region, from Streets et al. (2019b, 2019a). **B)** Temporal variability of mercury concentrations (Hg, mg/g) in tropical tunas: yellowfin (orange), bigeye (red), and skipjack (blue), from Médieu et al., (2024). The colored dots represent average annual concentrations measured in the global ocean, except in the northwestern Pacific, off Asia. In that area, average annual concentrations are represented by dark blue squares.

Implications for mercury monitoring in the global ocean

This 10-year research work provided the first high-resolution assessment of spatial and temporal patterns of Hg concentrations in tropical tunas and albacore. Through a global and multidisciplinary approach, we highlighted the combined importance of i) marine biogeochemical processes leading to variable MeHg levels in seawater, ii) tuna foraging depth, and iii) anthropogenic Hg releases, which further advances our understanding of both the distribution of Hg levels in tuna and the critical processes driving global Hg cycling. This work raises important questions about the mechanisms of marine MeHg production, and the resilience of ocean ecosystems to anthropogenic Hg releases.

In addition, by revealing the important role that biogeochemical processes play in Hg accumulation, our work also supports the assumption that climate change may affect MeHg concentrations in marine food webs. The already-observed expansion of the oxygen-minimum zone in the eastern Pacific is forecast to continue over the next few decades and may be conducive to forming MeHg and increasing its bioavailability at the base of food webs. On the other hand, changes to primary productivity and organic matter export may also counter this trend. Current ocean circulation models cannot accurately predict such biogeochemical changes, specifically in tropical areas such as the eastern Pacific, and so the effect of climate change on the Hg cycle is, as yet, unknown.

Overall, this work elevates tropical tunas and albacore as effective bioindicator species for ocean Hg pollution, as they appear to reflect a given ecosystem's Hg exposure while also including several Hg sources on various spatial and temporal scales. Combined with Hg measurements in the air and ocean, tunas could provide vital information for designing and implementing future large-scale Hg biomonitoring, as required for assessing the Minamata Convention's effectiveness. This calls for continuous and long-term global monitoring of Hg levels in tunas to explore future trends of tuna Hg levels and explore the marine Hg cycle in the context of climate change and changing anthropogenic Hg releases to the environment. Such monitoring has been carried out since 2001 in the western Pacific thanks to the Pacific Marine Specimen Bank, which enables many tuna samples to be collected yearly by onboard fisheries observers, and that can be laboratory-tested for Hg. This continuous tuna

sampling also offers perspectives to characterize complementary essential fatty acids and micronutrients in tuna muscle in order to help refining the benefits and risks associated to tuna consumption. Future analyses of selenium concentrations and speciation in particular will be performed in tuna tissues collected in the western Pacific to better understand the potential of selenium as dietary antioxidant and detoxifying agent for MeHg.

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