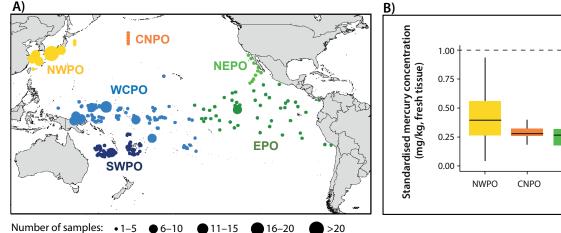
#### Tuna help to map mercury pollution in the ocean

Anaïs Médieu,¹ David Point,² Valérie Allain³ and Anne Lorrain⁴

Whether as sushi, steak or tinned, tuna is one of the world's most widely eaten saltwater fish, and yet it is known to contain toxic methylmercury, the most toxic form of mercury, which affects the nervous system, with foetuses and young children being at high risk (Box 1). The new environmental policies under the Minamata Convention<sup>5</sup> for reducing mercury emissions, and their unhealthy effects on humans, are based on scant knowledge of how such emissions affect fish mercury levels. This document provides the first detailed mercury concentration map for skipjack tuna in the Pacific Ocean, and highlights the link between anthropogenic (human-made) mercury emissions and concentrations in this species in the northwest Pacific for the first time (Médieu et al. 2022). Our study also shows that natural ocean processes heavily influence tuna mercury concentrations, especially in terms of the depth at which methylmercury bioavailability is at its highest in the water column.

In a previous study on three tuna species (yellowfin, *Thun*nus albacares; bigeye, T. obesus; and albacore, T. alalunga) in the western and central Pacific, we demonstrated that mercury concentrations were higher in the largest and deepest-diving fish, but that they also depended on species and geographical origin (Houssard et al. 2019; Lorrain et al. 2019). In this latest multidisciplinary study, funded by the French National Research Agency in the framework of the MERTOX<sup>6</sup> project, the French National Research Institute for Sustainable Development (IRD) and the Pacific Community (SPC) – assisted by a large number of partners and through access to several specimen banks<sup>7</sup> - looked into where the mercury in Pacific tuna was coming from (Fig. 1A) by focusing on skipjack (Katsuwonus pelamis), which is the most commonly eaten tuna species globally.



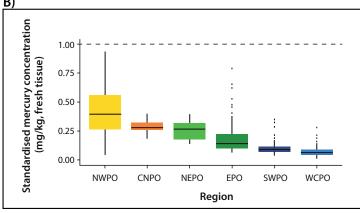


Figure 1. A) Spatial distribution of mercury-tested skipjack in six Pacific Ocean regions (NWPO = northwest Pacific Ocean, CNPO = central north Pacific Ocean, NEPO = northeast Pacific Ocean, EPO = eastern Pacific Ocean, SWPO = southwest Pacific Ocean, and WCPO = western and central Pacific Ocean). B) Standardised mercury concentrations (mg/kg, fresh tissue) in skipjack based on the six Pacific regions for a standard length of 60 cm. The dotted horizontal line indicates the maximum authorised mercury concentration (1 mg/kg, fresh tissue) for large predators such as tuna.

- PhD student, Université Bretagne Occidentale LEMAR
- Research Scientist, IRD GET
- Senior Fisheries Scientist, SPC
- Research Scientist, IRD LEMAR
- www.mercuryconvention.org
- ANR MERTOX https://www.get.omp.eu/recherche/projets-scientifiques/mertox/
- Western and Central Pacific Fisheries Commission Tuna Tissue Bank, Pacific Marine Specimen Bank managed by SPC, Tokyo University Environmental Specimen Bank, Japan, Inter-American Tropical Tuna Commission (IATTC) specimen bank and Daniel Madigan's sample collection (University of
- Author for correspondence: anne.lorrain@ird.fr

## Value of skipjack in studying mercury bioavailability in the oceans

Out of a total of 650 tested skipjack tuna specimens, none had mercury concentrations exceeding recommended health thresholds (1 mg/kg of fresh tissue; WHO and UNEP Chemicals 2008) (Fig. 1B). Although skipjack has some of the lowest mercury concentrations among the tuna species, it is still valuable for understanding and mapping ocean mercury pollution. In terms of health, it is the most heavily fished (mainly in the Pacific) and eaten tuna species, much of it in tinned form, making it a major source of animal protein worldwide. In biological and ecological terms, the species is noted for its fast growth rate and shallow vertical distribution; it migrates within the upper 100 metres of the water column, while other tuna species - such as albacore and bigeye - dive daily to depths of over 500 metres to forage. By choosing to work on this surface species, we hypothesised it could reflect mercury levels in surface waters, which are estimated to have tripled in response to anthropogenic atmospheric emissions (Box 1). The species has also been studied by a variety of research programmes and contributed to specimen banks through the efforts of onboard fishing vessel observers throughout the Pacific. As a result, we have been able to study skipjack mercury in highly contrasting areas of the Pacific (Fig. 1A), and explore various mechanisms that may affect mercury bioaccumulation in food webs. More specifically, we had access to skipjack specimens obtained off the Asian coast, where high levels of anthropogenic mercury emissions are released into the atmosphere (Box 1).

## Variable concentrations in different Pacific regions

Mercury naturally bioaccumulates during the lifetime of organisms, with older, larger fish having higher mercury concentrations. We, therefore, first standardised mercury concentrations to a given size (i.e. 60 cm), the average size of the tested skipjack. Strong standardised concentration gradients were revealed between the Pacific regions (Figs. 1B and 2); with concentrations 1.5 to 2.0 times higher in the northwest Pacific than in the north-central and eastern regions, and 4.0 to 5.0 times higher than in the intertropical regions of the western and central Pacific and southwest Pacific.

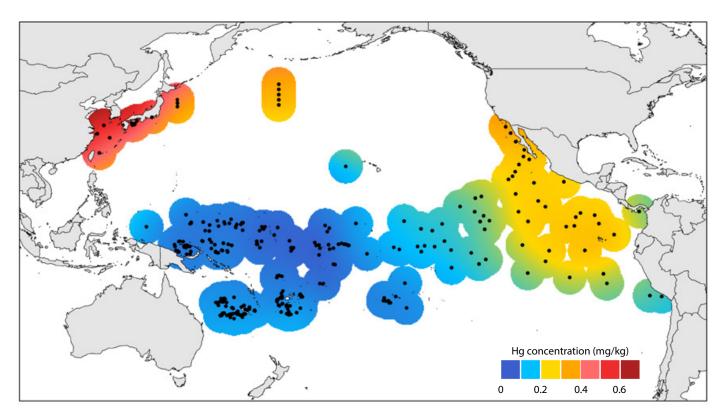


Figure 2. Spatial distribution of mercury concentrations (mg/kg, fresh tissue) in skipjack for a standard size of 60 cm. Black dots show the catch locations of the tested fish.

## Mercury gradients in tuna induced by natural ocean biogeochemical processes

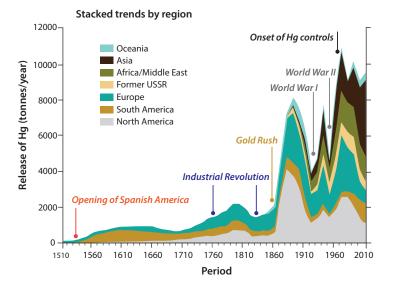
In order to attempt an explanation for such high spatial variability, we used a set of tracers to establish whether the underlying accumulation mechanisms were related to tuna dietary differences, methylmercury bioavailability at the base of the food webs, or anthropogenic mercury emissions. Using this approach, we were able to show that biogeochemical processes related to the Pacific Ocean's physical mechanisms naturally generated strong spatial methylmercury gradients in tuna. The relatively high mercury concentrations in the eastern Pacific and northwest Pacific (Fig. 2) 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 methylmercury concentrations to peak in water closest to the surface (< 100 metres) as compared with the western Pacific, where the methylmercury peak occurred in deeper water, between 400 and 800 metres. The fact that the methylmercury peak was closer to the surface and in closer contact with the food chain suggests that bioavailability was higher there. In regions where this occurs, organisms in surface food chains, including skipjack, can ingest and accumulate more methylmercury there than in the rest of the Pacific.

#### High levels of anthropogenic mercury emissions in the northwest Pacific

The very high mercury concentrations in the northwest Pacific (Fig. 2), however, may also be due to major sources of anthropogenic emissions located nearby (Box 1). They may be caused by recent atmospheric emissions linked to intensive fossil fuel use by Asian power plants. Such human-related sources (Fig. 3) add to natural biogeochemical processes that are conducive to surface methylmercury bioavailability in food webs.

## What are the implications for understanding the mercury cycle and the Minamata Convention?

For the first time ever, this study highlights the relationship between anthropogenic mercury emissions and mercury concentrations in tuna in the northwest Pacific. In general terms, even though mercury concentrations in skipjack are still below maximum authorised levels, it is vital to monitor and reduce the release of anthropogenic mercury into the environment to maintain human and ecosystem health, as required by the Minamata Convention that came into force in 2017.



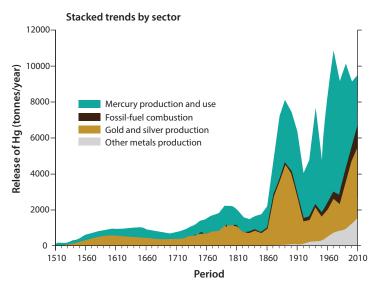


Figure 3. Temporal anthropogenic mercury release profiles from 1510 to 2010 by world regions (left) and emission source types (right) adapted from Streets et al. (2019). The "mercury production and use" category includes mercury production for use in gold and silver extraction (amalgamation techniques) and various commercial applications, such as chlorine and caustic soda production and waste processing.

By revealing the important role that biogeochemical processes play in mercury accumulation, our study also supports the assumption that climate change may affect methylmercury 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 methylmercury 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 mercury cycle is, as yet, unknown.

Our study suggests that skipjack is an effective bioindicator species for ocean mercury pollution, as it appears to reflect a given ecosystem's mercury exposure (in this case, Pacific surface waters) while also including several mercury sources on various spatial scales. Combined with mercury measurements in the air and ocean, skipjack could provide vital information for designing and implementing future large-scale mercury biomonitoring, as required for assessing the Minamata Convention's effectiveness. A comparable study on a global scale that includes the Indian and Atlantic oceans, and combines other mainstream tuna species (e.g. yellowfin and bigeye tunas), is currently underway to confirm or refute our findings.

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# Box 1. The (methyl)mercury cycle in oceans: major uncertainties regarding human activity and climate change

Mercury is released into the atmosphere as a gas by natural sources, such as volcanic eruptions, but more so by human activities (anthropogenic emissions) such as coal combustion or artisanal gold mining (Fig. A). The inorganic mercury is deposited in the oceans, where it is partly converted to methylmercury, a neurotoxin that naturally accumulates in organisms during their lives (bioaccumulation) and across the food web (biomagnification). This is why marine predators such as tunas have high methylmercury concentrations, with methylmercury representing the dominant form (> 91%) of the total mercury in tuna. Humans are then exposed to methylmercury by eating marine fish.

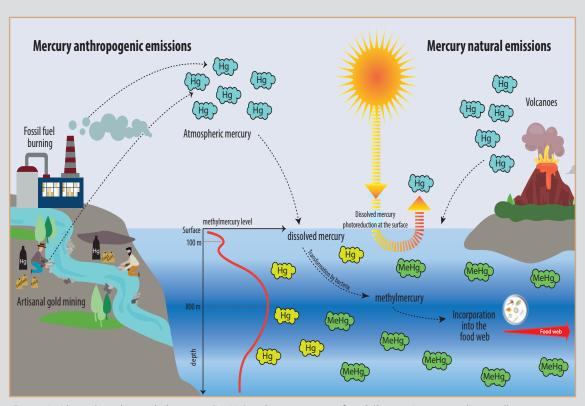


Figure A. Where does the methylmercury (MeHg) in the ocean come from? (Source: Lorrain et al. 2019; illustration Constance Odiardo)

Anthropogenic emissions began some 500 years ago in Europe and North America, but today come mainly from Asia, particularly China, where they have greatly increased over the past two decades with fossil fuel use in energy production (UN Environment 2019) (cf. Fig. 3 on page 52). Taken together, anthropogenic emissions have profoundly altered the mercury cycle and are estimated to have increased mercury concentrations by 450% over the past 20 years (Outridge et al. 2018), with rates of increase being higher in the Northern Hemisphere than in the Southern Hemisphere (Li et al. 2020)(Fig. B). In the ocean, anthropogenic emissions are said to have tripled the total mercury pool (inorganic mercury + methylmercury) (Lamborg et al. 2014), although the impacts on methylmercury concentrations in water and marine organisms, especially large predators, is undocumented.

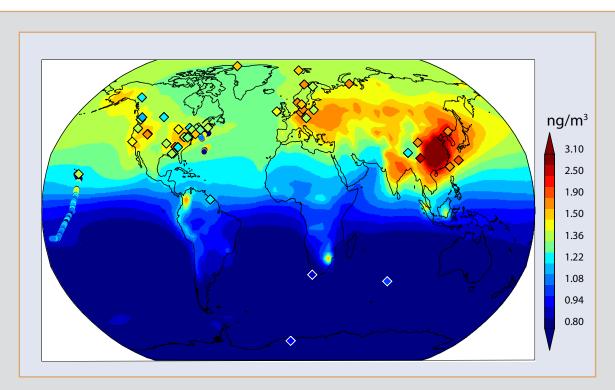


Figure B. Modelled global distribution of gaseous mercury concentrations in the atmosphere (total gaseous mercury, TGM, ng/m3), by Horowitz et al. (2017). The "diamonds" indicate gaseous mercury concentration measurements in the atmosphere and are used to confirm the modelled concentrations.

Another major uncertainty regarding the mercury cycle involves the impact of climate change, particularly on methylmercury formation and bioavailability at the base of marine food webs. It is commonly accepted that gaseous mercury dissolved in water turns to methylmercury (methylation) when broken down by bacteria in deeper, less-well-oxygenated areas of ocean (at depths of 400–800 metres). This methylmercury may be further converted into gaseous mercury in the surface layers, where it could then potentially be re-released into the atmosphere. The methylation and demethylation processes are still poorly understood, but we do know that it is the balance between the two that determines the amount of bioavailable methylmercury at the base of marine food webs. Because climate change may modify ocean circulation and productivity, and expand oxygen-minimum zones, it may also profoundly alter the mercury cycle.

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