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Stable Tuna Mercury Concentrations since 1971 Illustrate Marine Inertia and the Need for Strong Emission Reductions under the Minamata Convention

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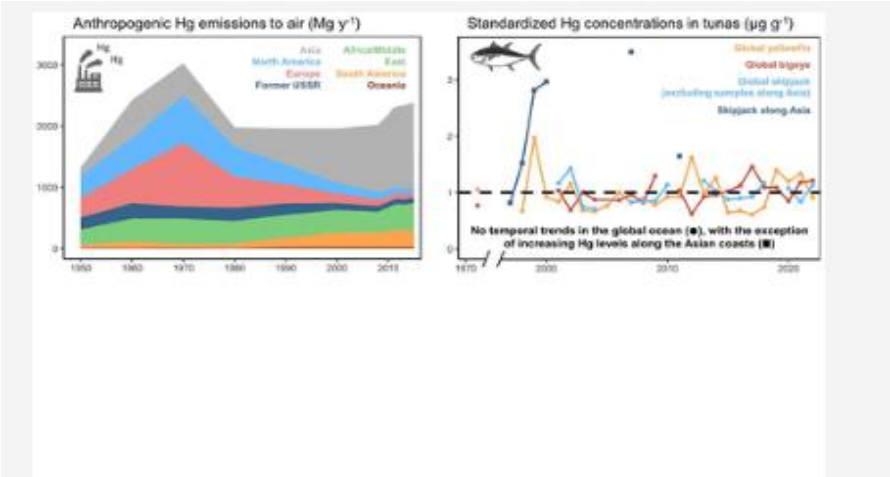
Abstract :

Humans are exposed to toxic methylmercury mainly by consuming marine fish. While reducing mercury emissions and releases aims to protect human health, it is unclear how this affects methylmercury concentrations in seawater and marine biota. We compiled existing and newly acquired mercury concentrations in tropical tunas from the global ocean to explore multidecadal mercury variability between 1971 and 2022. We show the strong inter-annual variability of tuna mercury concentrations at the global scale, after correcting for bioaccumulation effects. We found increasing mercury concentrations in skipjack in the late 1990s in the northwestern Pacific, likely resulting from concomitant increasing Asian mercury emissions. Elsewhere, stable long-term trends of tuna mercury concentrations contrast with an overall decline in global anthropogenic mercury emissions and deposition since the 1970s. Modeling suggests that this limited response observed in tunas likely reflects the inertia of surface ocean mercury with respect to declining emissions, as it is supplied by legacy mercury that accumulated in the subsurface ocean over centuries. To achieve measurable declines in mercury concentrations in highly consumed pelagic fish in the near future, aggressive emission reductions and long-term and continuous mercury monitoring in marine biota are needed.

Graphical abstract



Keywords : methylmercury, yellowfin, bigeye, skipjack, global temporal heterogeneity, anthropogenic emissions, legacy mercury, Minamata Convention

Introduction

Mercury (Hg) is a widespread contaminant that can be naturally converted into methylmercury (MeHg) in marine ecosystems[1,2]. Methylmercury is associated with neurocognitive deficits in human foetuses and children, and with cardiovascular effects in adults[3,4]. Humans are exposed to MeHg mainly by the consumption of seafood, and tunas in particular, which are amongst the most consumed marine fish worldwide[5], and have relatively high MeHg concentrations[6–8]. Global health impacts associated with MeHg exposure for the general population are estimated at $117 billion per year[9]. To protect human health and the environment from harmful effects, the UNEP Minamata Convention entered into force in 2017 with the objective of reducing anthropogenic Hg emissions.

Most anthropogenic Hg releases are estimated to have occurred during the last five centuries[10]. Cumulative releases to the atmosphere 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[11]. Anthropogenic Hg uses and emissions have considerably modified the natural global Hg cycle[12,13]. In the atmosphere, higher Hg levels are measured in the northern hemisphere, following the location of the main anthropogenic Hg emissions[14]. In the open ocean, total Hg concentrations within depths of 100 to 1,000 m are thought to have tripled globally[15]; yet, scarce Hg observation data in seawater suggest contrasted temporal trends[16,17]. It is still unclear which fraction of anthropogenic Hg is converted into MeHg in seawater, and biomagnified in marine biota. While tunas represent a major route of MeHg exposure to humans, temporal studies of their Hg concentrations are limited, showing contrasting temporal trends among species and ocean regions[18–21]. Understanding and predicting spatial and temporal Hg trends in tunas remains crucial to anticipate changes in human exposure and to evaluate the effectiveness of the Minamata Convention in protecting human health through seafood consumption.

In this study, we investigate the temporal variability of Hg concentrations in tunas from 1971 to 2022 in six regions of the Pacific, Atlantic, and Indian Oceans. We revisited existing and acquired new Hg data in tropical tunas: yellowfin (*Thunnus albacares*), bigeye (*T. obesus*), and skipjack (*Katsuwonus pelamis*). These tunas are top predators, globally distributed, highly exploited (constituting 94% of global tuna catches), and hold significant importance for human health[5]. They exhibit distinct foraging habitats (skipjack and yellowfin primarily feeding on epipelagic prey, while bigeye generally rely on mesopelagic species)[22], growth rates and lifespan[23]. Moreover, all three display relatively limited movements and show site fidelity[24,25], compared to bluefin tunas that undergo large transoceanic migrations[26–28]. Tropical tunas are thus expected to reflect MeHg patterns in surface and subsurface waters, making them valuable biological sentinel species to explore various lag times in response to changes in Hg emissions. After standardizing Hg concentrations by tuna length, we reveal overall stable tuna Hg concentrations in the global ocean, except in the northwestern Pacific where tuna Hg concentrations increased significantly in the late 1990s.

Material & Methods

Tuna sampling and data compilation

We assembled 2,910 tuna muscle Hg concentrations in six regions of the global ocean (Fig. 1; Table S1), allowing temporal analysis covering approximately 50 years from 1971 to 2022, depending on the region. We added 547 tuna Hg concentrations to the dataset of Drevnick and Brooks (2017) (Fig. S1), and 37 data points to that of Médieu et al. (2021), to re-evaluate the previously identified increasing and stable tuna Hg concentrations in the central north and southwestern Pacific, respectively. In the northwestern Atlantic, where decreasing Hg concentrations were observed in bluefin tuna between 2004 and 2012[20], we examined the temporal variability of Hg concentrations in yellowfin between 1999 and 2011. Our global compilation extends to regions such as the western Indian, the northwestern Pacific, and the eastern equatorial Atlantic where no temporal Hg data in tunas have been available to date.

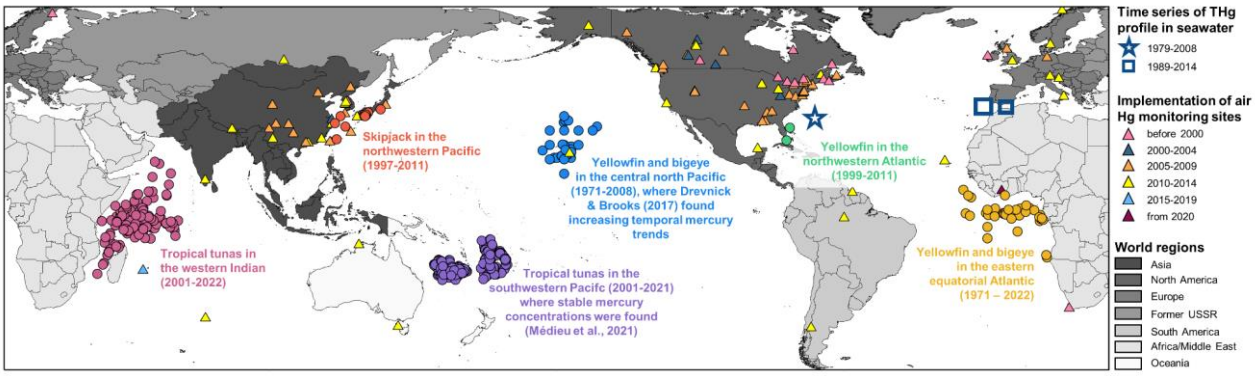


Figure 1. Origin of the tropical tuna samples. The temporal variability of mercury (Hg) concentrations was investigated in tropical tunas sampled in six regions of the global ocean. Coloured dots represent tuna catch locations, while coloured triangles show air monitoring sites according to their starting year. The blue star and squares represent the three locations where seawater concentrations profiles of total mercury (THg) are available in multiple years[16,17]. Countries are coloured following the definition of the seven world regions in the emissions inventory of Streets et al[10,11,29,30].

Total mercury concentrations and ecological tracer analyses in tunas

Total Hg concentrations were measured in tuna muscle samples in different study-specific laboratories, with laboratory-specific reference standards to ensure accuracy and traceability, and allow comparison of regional datasets. Total Hg content is expressed on a dry weight (dw) basis and is considered to reflect MeHg concentration, as most of the total Hg (> 91%) is in its methylated form in tuna white muscle[31]. As Hg is known to bioaccumulate with fish length, we calculated length-standardized Hg concentrations to remove the length effect when exploring temporal variability[8,21,32] (Fig. S2).

To account for possible confounding temporal changes in tuna trophic ecology, in particular to investigate temporal variability of tuna trophic position, muscle nitrogen ($\delta^{15}N$) stable isotope values were also compiled globally when possible (data available for more than two years, n = 1,535, Table S1).

Analytical and statistical methods are presented in the SI.

Temporal seawater concentration profiles, air monitoring sites, emissions to air inventory, and future emissions scenarios

To examine tuna Hg concentrations alongside seawater Hg concentrations, we reviewed the available temporal concentrations profiles of total Hg in seawater (Fig. 1). Similarly, air monitoring sites measuring atmospheric Hg concentrations around the globe were compiled from the literature (Fig. 1; Table S2) to explore tuna Hg data alongside atmospheric Hg concentrations. Moreover, mean anthropogenic Hg emission estimates to the atmosphere per decade for the period 1970-2010, and annually for the period 2010-2015, were taken from the emission inventory of Streets et al.[10,11,29,30]. This emission inventory discerns several world regions (Fig. 1), and includes 17 source categories.

We used a box model[33,34] to simulate the influence over time of global anthropogenic emissions on Hg masses in the atmosphere, surface ocean, subsurface ocean, and deep ocean. The model was driven by 2000 BCE to 2015 CE primary anthropogenic emissions from Streets et al.[10,11,29,30]. To simulate future emissions (2016-2100), we considered three emissions scenarios[35,36]: i) a Current Policy scenario, ii) a more stringent New Policy scenario, and iii) a Maximum Feasible Reduction scenario that leads to a dramatic decrease of annual Hg emissions (see also SI).

Results & discussion

Standardized tuna mercury concentrations

Tuna Hg concentrations were highly variable among years globally when MeHg bioaccumulation with tuna length was accounted for. Significant temporal trends of Hg levels were evidenced only in skipjack along the Asian coasts: Hg concentrations in 1997 and 1998 were significantly lower than in 1999, 2000, and 2007, suggesting an increase of skipjack Hg concentrations at the end of the 1990s in the northwestern Pacific (Fig. 2C; Table S3C). Skipjack Hg concentrations in the same area in 2011 were significantly lower than in 1999, 2000, and 2007, but comparable to the earliest years, which suggest a decrease of skipjack Hg concentrations in the 2010s, although more recent data would be necessary to validate this result. Elsewhere, in all regions, our comparison approach reveals that tuna Hg concentrations remained stable over each study period, as no significant difference between old and recent data was found (Fig. 2A-B-G-H-I-J-K-L; Table S3A-B-G-H-I).

This apparent stability of tuna Hg concentrations over multiple decades in all regions, except for skipjack in the northwestern Pacific, is consistent with reported stable tuna Hg concentrations over the past two decades in the southwestern Pacific[21], confirmed here with new data in 2020 and 2021 (Fig. 2D-E-F; Table S3D-E). Our results align with previously observed stable Hg concentrations in yellowfin in the central north Pacific, when comparing data in 1971 and 1998[37]. However, they diverge from more recent reports indicating increasing trends in yellowfin (*n* = 5yrs) and bigeye (*n* = 4yrs) during 1998-2008[19]. These discrepancies result from our larger dataset (547 data points added to the initial 422 observations), which encompasses an additional year and broader ranges of fish sizes and Hg concentrations (Fig. S1). In the northwestern Atlantic, stable Hg concentrations in yellowfin contrast with the reported mean annual decrease of 2.4% between 2004 and 2012 (*n* = 9yrs) in bluefin tuna[20].

Yet, our results align with the apparent stability of bluefin Hg levels in the same region between the 1970s and 2017 when re-investigated with additional old and recent data points (n = 13yrs)[38]. These comparisons highlight the need for collecting long and continuous temporal data on tuna Hg concentrations to be able to justify trend detection in a time series.

All species-specific Hg concentrations were of the same magnitude among regions, except for bigeye and skipjack in the southwestern and the northwestern Pacific, respectively (Fig. 2). Bigeye Hg concentrations in the southwestern Pacific were 2-3 times higher than in other regions, likely associated with the particular marine physical and biogeochemical conditions: a deeper thermocline giving bigeye access to deeper waters, and a high peak of dissolved MeHg in subsurface waters fuelling the mesopelagic food web[8,39]. In the northwestern Pacific, skipjack Hg concentrations were up to 3-4 times higher than in the southwestern Pacific and the western Indian (Fig. 2C-F-H), likely reflecting a regional anthropogenic Hg contribution from Asia[32] (see below).

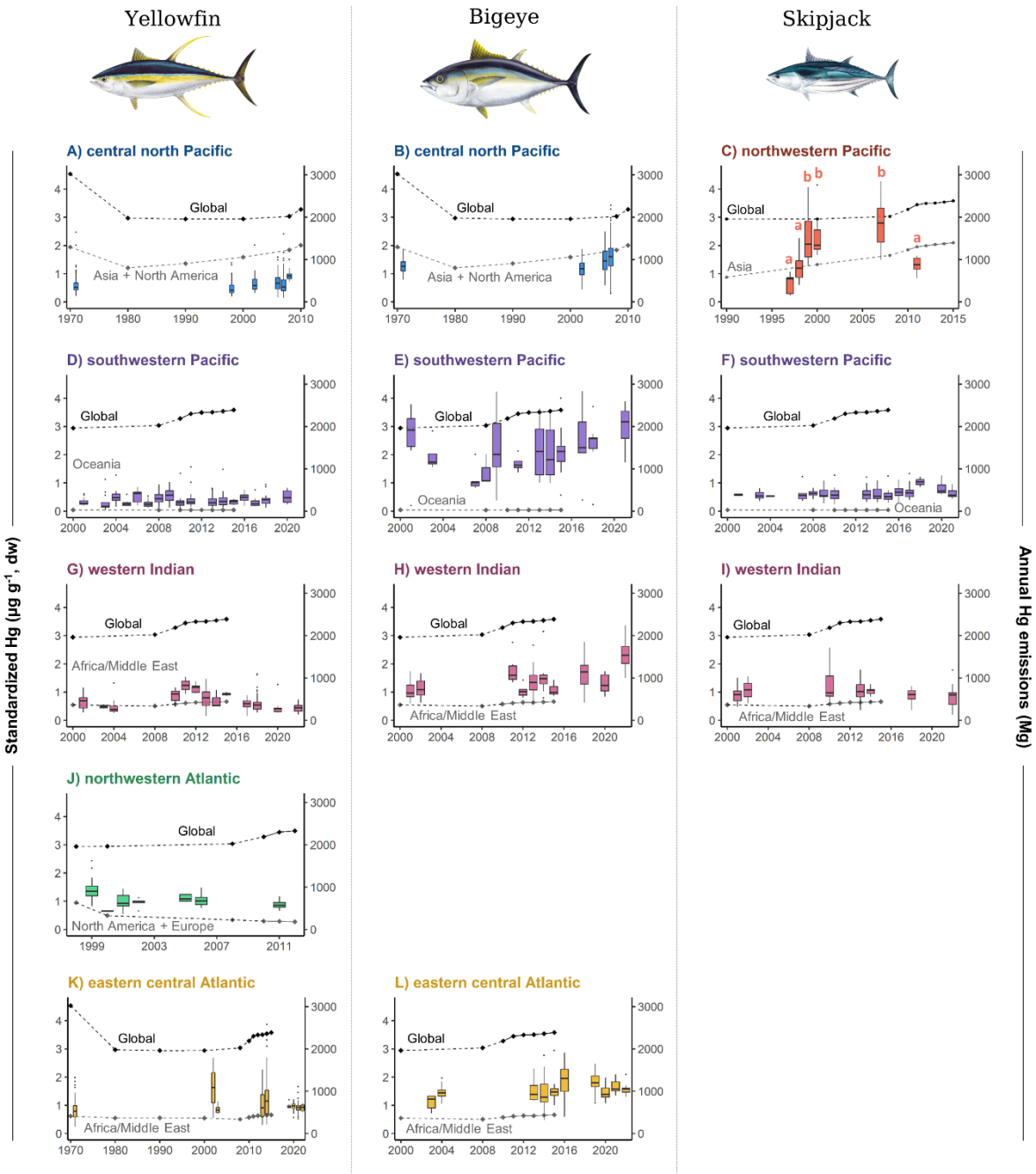


Figure 2. Temporal variability of tuna mercury concentrations and anthropogenic emissions. Boxplots of standardized mercury (Hg) concentrations (µg g^{-1}, dw) according to time in **A)** yellowfin, and **B)** bigeye in the central north Pacific (1971-2008), **C)** skipjack in the northwestern Pacific (1997-2011), **D)** yellowfin, **E)** bigeye, and **F)** skipjack in the southwestern Pacific (2001-2021), **G)** yellowfin, **H)** bigeye, and **I)** skipjack in the western Indian (2001-2022), **J)** yellowfin in the northwestern Atlantic (1999-2011), and **K)** yellowfin and **L)** bigeye in the eastern central Atlantic (1971-2022). A significant increase was found in skipjack standardized Hg content in the northwestern Pacific only, as symbolised by the letters in C). Elsewhere, the year-to-year comparison indicates non-significant difference between old and recent Hg data. Diamonds represent mean regional (dark grey) and global (black) anthropogenic Hg emissions (Mg) to the atmosphere estimated by the emissions inventory of Streets et al.[10,11,29,30]. Solid lines show annual trends of emissions for the period 2010-2015, while dashed lines show decadal trends of emissions for the period 1970-2010. Note that the x-axes (sampling years) are adjusted to each time series for greater legibility. Tunas illustrations were created by Les Hata, @SPC.

Comparison with seawater and atmospheric mercury

In the global ocean, only three temporal total Hg concentration profiles in the water column are available to explore long-term trends of seawater Hg (Fig. 1), relying on 4 sampling years and showing contrasting results[16,17]. Atmospheric Hg concentrations and deposition are extensively monitored worldwide (Fig. 1; Table S2), and exhibited in Europe a broad 2-fold decrease between 1970 and 1990 as a result of large reductions in anthropogenic emissions in North America and Europe[36,40–42]. Similarly, trends in Hg deposition inferred from remote peat sediments indicate a synchronous 2-fold decline across the planet during the late 20th century[43]. Nevertheless, we find little spatial and temporal overlap between tuna Hg and atmospheric and marine data, preventing direct comparisons (Fig. 1).

Studies of historical primary Hg emissions to air[10,11,29,30] provide an alternative to the limited local atmospheric observation data to investigate the relationship between tuna Hg trends and atmospheric Hg emissions. The increase of skipjack Hg concentrations in the northwestern Pacific in the late 1990s mirrors increasing Hg emissions to the atmosphere from Asia over the same period (Fig. 2C). This suggests a significant contribution of anthropogenic Hg release to marine Hg in this particular region, as already shown in seawater[44,45], sediment[46] and tuna[32,47] samples. The decrease in tuna Hg levels after 2011 may reflect the documented reduction in Hg emissions and atmospheric levels in East Asia since the 2010s, and/or with changes in emitted Hg speciation in China[48–53]. The inventory used in our study[11] does not reflect these recent changes in emissions (Fig. 2C). To gain a more accurate understanding of how changing local anthropogenic Hg emissions affect the marine Hg cycle in this specific region, additional Hg observations data in tunas, seawater and the atmosphere are needed, along with improved emissions inventories.

Elsewhere, stable tuna Hg concentrations between 1971 and 2022 contrast with the significant decrease of global primary anthropogenic Hg emission, concentration and deposition trends, and natural archive-based reconstructed deposition since the 1970s (Fig. 2). Such contrast has already been documented in other aquatic ecosystems[54], and likely results from i) multi-causal and local processes (e.g., marine biogeochemistry, tuna ecology) controlling MeHg net production, bioavailability and biomagnification, and ii) the large amount of legacy Hg, i.e., historically emitted Hg that continues to cycle through biogeochemical compartments and remains available for bio-uptake, leading to time lags in biotic response to reductions in primary Hg emission or release.

The surface ocean (~0-50m) is a small reservoir that is sensitive to changes in Hg loadings from atmospheric deposition, subsurface ocean Hg via upward mixing, and river runoff[34]. The subsurface ocean (~50-1,500m) has received large amounts of legacy Hg over centuries by Hg export from the surface via particle scavenging and mixing. Because of upwards mixing of this legacy Hg, the surface ocean reservoir takes years or decades to respond to changes in primary emissions[34,55], as illustrated with new Hg box model simulations forced with updated Hg emissions[10,11,29,30] (Fig. 3). Following the sharp 30% decrease of primary emissions in the 1970s, the Hg mass in the surface ocean only decreased by 2%. Subsequently, it exhibited a rapid increase over several decades, driven by legacy Hg from the subsurface ocean, which surpassed atmospheric Hg deposition to the surface ocean by a factor of two[34]. This surge was further fuelled by ongoing new anthropogenic Hg emissions and marine Hg deposition. This suppressed response in surface ocean Hg likely explains the absence of significant decreasing trends of tuna Hg levels following the decrease of emissions in the 1970s. Subsurface ocean legacy Hg is likely to buffer Hg mass in the surface ocean for decades, partly depending on future

emission scenarios (Fig. 3). Under the Maximum Feasible Reduction scenario, while Hg mass in the atmosphere is expected to decrease immediately, Hg masses in the surface and subsurface oceans are predicted to take 10 and 25 years, respectively, to start decreasing. Under the New Policy scenario, these response delays are expected to extend to 25 and 45 years, respectively. Inhabiting the surface (mainly skipjack and yellowfin) and subsurface (bigeye) waters, tropical tunas likely reflect this time lag in response between the atmosphere and the different ocean layers. Tuna Hg concentrations may therefore take several decades to decline as a result of emission reduction measures.

The box model also illustrates how the surface ocean responds faster to an increase in emissions and deposition than to a decrease. The 5-fold increase in Hg emissions in the 19th century led to immediate 4-fold and 3-fold increases in surface and subsurface ocean Hg by 1900, respectively; yet the 5-fold decrease in Hg emissions by 2050 under the Maximum Feasible Reduction scenario only induces a 7% decrease in surface ocean Hg, and even a 10% increase in subsurface ocean Hg. This explains why tuna Hg concentrations in the northwestern Pacific have reacted rapidly to the recent increase in Asian Hg emissions and releases.

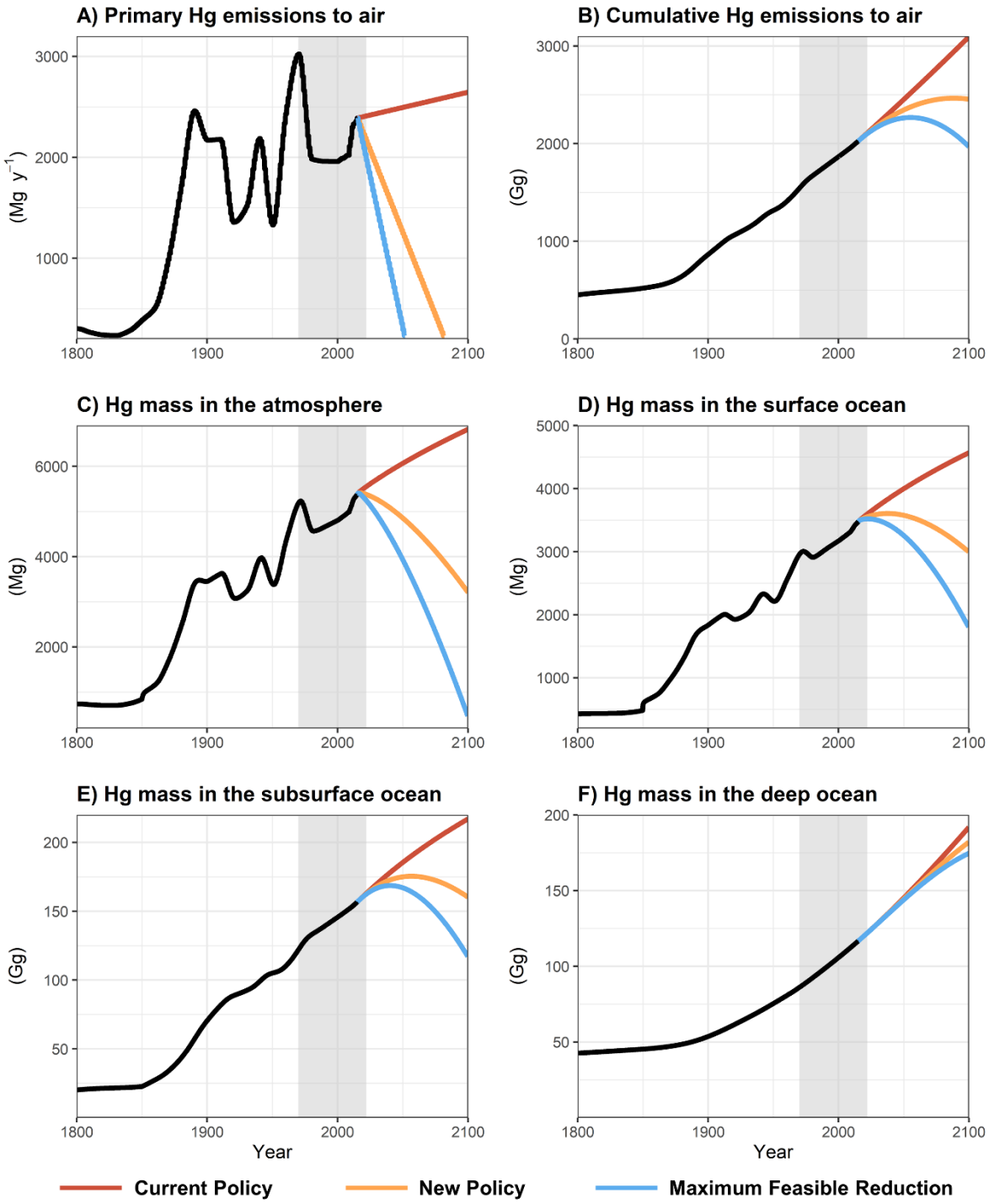


Figure 3. Simulated global anthropogenic influence on environmental mercury (Hg) loads using the box model of Amos et al.[33,34], emission inventory of Streets et al.[10,11,29,30] up to 2015, and three emissions scenarios for 2016-2100: Current Policy (red), New Policy (orange), and Maximum Feasible Reduction (blue), as defined in Angot et al.[35] and Pacyna et al.[36]. A) and **B)** show annual (Mg y^{-1}) and cumulative (Gg) primary anthropogenic Hg emissions to air, respectively. **C)**, **D)**, **E)**, and **F)** show the temporal evolution of Hg masses (Mg or Gg) in the atmosphere, the surface (extended to the base of the mixed layer, ~50m), subsurface (extending to the depth of the permanent thermocline, ~1,500m), and deep (extending to the sea floors) oceans, respectively. The grey bands indicate our study period (1971-2022).

Possible ecological and biogeochemical confounding processes

As the Hg box model lacks spatial resolution and does not include aquatic food webs, it precludes the investigation of local/regional impacts resulting from marine biogeochemical and tuna ecological processes. However, these processes play a pivotal role in governing MeHg net production, bioavailability, and biomagnification.

Since the 1970s, the oceanic environment has experienced anthropogenically-forced warming of the upper ocean[56]. Seawater warming, leading to shift in foraging habitat towards higher latitudes or deeper in the water column, is anticipated to increase MeHg concentrations in marine top predators[38]. Yet, tropical tunas were shown to display relatively site fidelity globally from 1970 to 2010s[24]. Moreover, yellowfin and bigeye trophic positions were shown to remain stable between 2001 and 2015 globally[57]. Our re-evaluation of tuna $\delta^{15}N$ values with new data in the southwestern Pacific, western Indian and, eastern central Atlantic confirms stable trophic positions for the three tropical tunas over time (2001-2022), suggesting no changes of tuna foraging depth or diet that could have biased the tuna Hg time series in these three regions (Fig. S4, Table S5). For skipjack in the northwestern Pacific, long-term trophic time series would however be necessary to fully exclude any trophic bias. Changes in ocean productivity and nutrient enrichment of coastal ecosystems may also have confounding effects on decadal trends of tuna Hg, as primary production and carbon export are key drivers of MeHg at depth[58,59]. Yet the spatial gradients in these properties are far greater than long-term temporal trends, and the signal of secular changes may be engulfed by the noise of inter-annual variability in these properties (Fig. S5). Taken together, this suggests stable long-term trends of MeHg net production but high inter-annual variability, which may in turn explain the strong year-to-year variability in tuna Hg.

In the coming decades, climate-induced changes in ocean biogeochemistry are expected to induce substantial regional variabilities in MeHg formation and bioaccumulation at the base of marine food webs[60], potentially affecting tuna Hg levels. Presently, our understanding of these regional changes is insufficient to quantify their interference with changes in Hg emissions and cycling in the environment. Consequently, extended time series of Hg concentrations and speciation in seawater, phytoplankton, and zooplankton are urgently needed to explore how climate-driven regional processes might offset or enhance measures aimed at reducing emissions.

Implications for mercury monitoring in the global ocean

With the adoption of the Minamata Convention, Hg biomonitoring in seafood is needed to evaluate the effectiveness of political decisions and reduction efforts in relation to human health[61]. With the exception of the northwestern Pacific, we show stable long-term Hg concentrations in tunas, but year-to-year variability which likely reflects natural biogeochemical and/or ecological processes. Conversely, skipjack Hg concentrations near Asia increased significantly in the late 1990s, reaching concentrations up to 4 times higher than elsewhere in the global ocean. These temporal trends suggest that at the local scale along Asian coasts close to large Hg emissions hotspots, Hg concentrations in surface tuna may integrate the elevated deposition from nearby anthropogenic Hg release through time. Yet, on a global scale, the divergence between stable tuna Hg concentration and global variations Hg in emissions and deposition likely reflects the inertia of subsurface ocean legacy Hg that continues to supply subsurface and surface ocean food webs. Taken together, our results show that tropical tunas

are able to detect concomitant increasing anthropogenic Hg emissions to the atmosphere but may take decades to decline following emission reduction measures. We estimate that only the Maximum Feasible Reduction policy scenario would lead to a detectable decrease in surface ocean tuna Hg levels and achieve the objective of the Minamata Convention in the near-future.

Supporting Information: Additional details on data compilation, analytical and statistical analyses, as well as temporal variability results regarding tuna ecology and ocean biogeochemistry.

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