The Causes and Effects of Mercury and Methylmercury Contamination in the Marine Environment: A Review

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Abstract

Purpose of Review The concern of mercury pollution and the impact that it poses on the marine environment were studied heavily since the case of the poison from Minamata bay in the 1960s. The present study provides an insight into the cycle of mercury and methylmercury in the marine environment and the bioindicators that reflect the exposure levels. The paper also used the driving forces, pressures, states, impacts, and responses (DPSIR) analysis to evaluate the global mercury and methylmercury contamination problem.

Recent Findings The high global budgets of atmospheric total mercury influence the ocean surface water. Therefore, the aquatic environment contamination level is in turn affected by the surrounding emission sources such as industrial and petroleum activities in addition to the transport and fate of mercury across the environmental compartments. This will increase the mercury levels in fish species and will cause an adverse risk to human health through biomagnification.

Summary This review presents a thorough description of mercury sources and emissions and their fate and transport across the different environmental compartments, despite the fact that serious mitigation measures were taken and guidelines were applied. The risk from fish consumption is still a serious concern as a result of the current mercury emissions and stability and persistent characteristics.

Keywords Mercury methylation · Wastewater · Fate and transport · Mercury pollution · Mercury measurements

Introduction

Heavy metals include metals and metalloids with high atomic weight and density that exist naturally in the environment at low levels [1]. They are persistent, stable, and non-biodegradable [2]. They are transported through the earth's spheres: atmosphere (air), geosphere (soil), hydrosphere (water), and biosphere (biotic) and recycled by biogeochemical cycles [3, 4•]. Heavy metals are classified into essential and non-essential heavy metals. Essential heavy metals are needed in small quantities by the human body to ensure their normal functioning. High quantities can become toxic and

alter the normal biochemical processes of the human body's functions [5].

Non-essential heavy metals like mercury (Hg) are not needed by the human body and can adversely affect human health [6]. Mercury (Hg) occurs as a major pollutant in the aquatic environment and is listed as one of "the ten leading chemicals of concern" [7•, 8]. In the aquatic environment, mercury is transformed into a more toxic form, methyl mercury (MeHg) (CH_3HgX, where X is Cl^-, OH^-, or organic thiols and other reduced sulfur ligands ($R\text{-}S^-$)), by microorganisms present in the sediment. Aquatic organisms readily absorb MeHg, which is then bioaccumulated and biomagnified within the aquatic food chain [9, 10•]. Bioaccumulation refers to the increase of mercury concentration through continuous feeding within the same fish. In contrast, biomagnification refers to the rise of mercury concentration in different fish species across the trophic levels of the food chain, i.e., the higher the trophic level of the fish species, the more elevated the mercury concentration [11]. Figure 1 shows the difference between bioaccumulation and biomagnification in the marine environment.

Fig. 1 Bioaccumulation and biomagnification in the marine environment
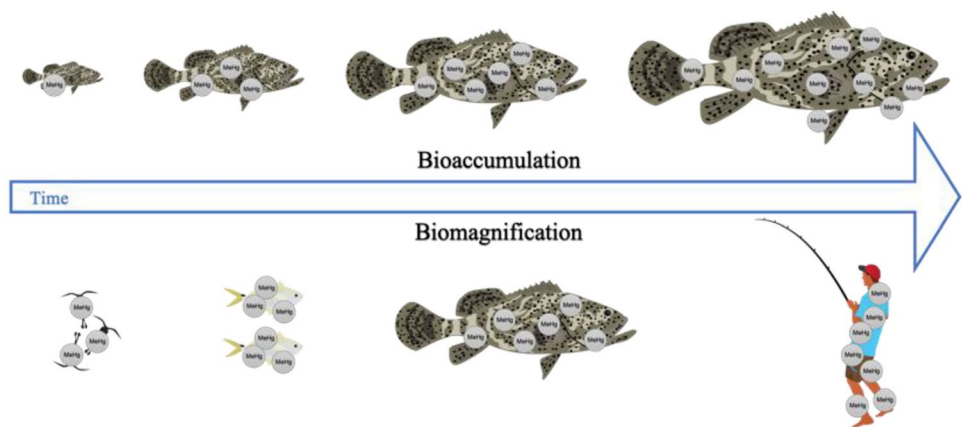
Various factors may affect the mercury and methyl mercury concentrations in fish. Some of those factors are related to the fish like trophic level, species, size, age, gender, habitat, and niche [12••]. Abiotic factors can also influence mercury levels like water column, temperature, salinity, and deposition from anthropogenic emissions from industrial and petroleum activities [13, 14].

The contamination of fish with Hg and MeHg poses a significant challenge to public health. Fish is considered one of the most important sources of healthy proteins and is a staple food item for many communities worldwide [3, 5]. In addition, fish is rich in omega 3, docosahexaenoic acid, linolenic acids, unsaturated fatty acids, micro- and macronutrients, and different vitamins [15]. Besides, fish consumption was proven to be highly effective in reducing the risk for noncommunicable diseases (NCDs), particularly cardiovascular diseases and rheumatoid arthritis, and is essential for normal neuronal development in children [3].

A major concern associated with fish consumption is human exposure to the toxic Hg and MeHg [15, 16]. A wide range of factors have affected the Hg levels in the aquatic environment around the Arabian Gulf region. The Arabian Gulf has a physiologically arid environment, hot climate, high water salinity, and poor water circulation resulting in long water residence time [10•]. It is a semi-closed Gulf that receives a limited amount of freshwater input from rivers in the northern part and low precipitation levels [17]. Furthermore, petroleum and natural gas industries form the backbone of the economy in the Gulf region. As a result, the region was exposed to multiple oil spill incidents [2]. The region also relies on water desalination plants, wastewater treatment plants, and power plants established in the coastal areas [9, 18]. This increase in mercury levels can pose a risk to the Gulf population since fish is a major food source of the region due to its abundance and easy availability that remains unaffected by the arid climate, unlike other meat sources [18]. To plan appropriate responses at both the upstream and downstream levels, it is necessary

to investigate all possible causes and effects relationships of mercury. Thus, understanding the source–receptor relationships associated with sources of mercury emissions, transformations, and cycling in each of the environmental compartments is needed [19•].

Numerous studies on Hg in the environment have focused on the Hg biogeochemical cycling in aquatic systems, the presence and bioaccumulation of Hg in seafood, and the risk of Hg exposure from fish consumption. However, limited studies are available, until now, to link and summarize all the information in one study. Therefore, this review presents the current knowledge on mercury sources and discusses the mercury life cycle starting from anthropogenic emission from air pollution or wastewater discharged and its levels. The paper investigated Hg's life cycle and emissions to link that information to Hg levels on fish species, and the impacts on human health and the economy. This review used the driving forces, pressures, states, impacts, and responses (DPSIR) framework to evaluate and analyze all possible causes and effects of mercury and methylmercury contamination in the marine environment.

Mercury Speciation and Categorization

Mercury can be categorized in three different ways; the first way is by its oxidation state. Hg is categorized through three oxidation states, namely, elemental or metallic Hg^0, mercurous $Hg(I)/Hg^+$, and mercuric $Hg(II)/Hg^{2+}$ [20]. The first oxidation state is observed only in the atmosphere due to its high volatility, and in highly polluted soil. The second oxidation state is unstable; this will make Hg^+ difficult to be observed in any environmental compartment. The third oxidation state (Hg^{2+}) is produced mainly after the dry/wet deposition from the atmosphere to form one of two types of salts, inorganic by ionic bond or organic by a covalent bond [12••, 19•].

The second way to categorize mercury is based on its chemical form, namely, elemental (natural gas), organic, and

inorganic forms. Methyl mercury (MeHg) is the most common organic form and the most toxic one [21]. The term methylated Hg consists of MeHg and dimethylmercury (DMeHg). MeHg is sometimes generally used to represent the methylated Hg, not only monomethyl mercury [22]. The final way to categorize mercury is based on its physical characteristics: volatile (elemental and dialkyl mercury), insoluble (mercury sulfide HgS and mercury selenide HgSe), and soluble (monomethyl mercury halides, dialkyl mercury, and ionic forms) [4•].

Methylation and Demethylation

The methylation processes are still considered to be ambiguous. The main locations identified for methylation are sediments, water, and some microorganisms on the macrophyte roots [23]. The methylation and demethylation in incubation studies were found to follow first-order reactions [22]. The rate of reaction depends only on the concentration of Hg or MeHg. All other surrounding conditions do not have a significant influence [22]. MeHg is a persistent form of Hg with low elimination rates and high absorption levels in the bloodstream (up to 95%) [12••, 21]. This form of Hg makes up 95% of Hg in marine species [9]. There are two main pathways for methylation and demethylation: the photo and bacterial [24••]. Bacterial methylation or biomethylation of Hg produces MeHg from the methylation/ biomethylation process of inorganic Hg by soil and water heterogeneous anaerobic bacteria [24••, 25, 26•]. The bacterial groups associated with methylation are iron-reducing bacteria (FeRB or IRB), methanogens and fermentation bacteria, sulfur-reducing bacteria (SRB), methanogens, and acetogenic microorganisms [24••, 27].

The methylation ability of SRB to convert organic matter (OM) and Hg to MeHg is highly related to the sulfur biogeochemical cycle. Sulfur-rich sediments, sulfate (SO_4^{2-}) and sulfide concentration, and other forms of sulfur will create a strong ligand bond with Hg. The sulfur cycle will activate the methylation processes. However, the level of methylation is determined by the sulfur species. In general, it was found that the increase in SO_4^{2-} concentration will enhance the methylation [27]. It was found that the methylation process accelerates in acidic pH levels. This was explained by the fact that Hg uptake into the fish tissues by the bacteria is more efficient at lower pH levels [26•, 28]. Moreover, the negativity of dissolved organic carbon (DOC) in low pH levels will be weaker. This will limit the creation of a complex bond with Hg, which will allow more methylation processes to take place [29]. However, the two main factors are the availability of inorganic Hg and the microbial community [30]. Deacidification in water was found not only to decrease the Hg level but also the SO_4^{2-} concentration. This will influence the methylation and will reduce the

assimilation of Hg into fish cells increasing the Hg^0 in water [29]. In Ziarati et al.'s study on *Carcharhinus dussumie* fish, the total Hg was 0.79 µg/g and MeHg was 0.78 µg/g; this means that MeHg makes around 99.76% of the total Hg [31]. Photomethylation occurs by solar irradiation on the surface of the water. However, the photomethylation rate is slow and the intensity of light will not influence the concentration of MeHg in the surface water [24••]. Demethylation can occur by microorganisms, photodecomposition, or abiotic processes [22, 32]. In some cases, demethylation can be processed by the SRB [23].

Photodemethylation is the process of releasing the Hg from the methyl group by UV radiation and elemental Hg will emit back through the system. The greenhouse effect will prevent and slow the demethylation processes [33]. Photodemethylation in the marine system is highly influential and has a faster rate than photomethylation. In 1972 and 1975, the first experimental study for Hg photochemical methylation was conducted. In 1972, mercury chloride solution was tested with methanol (CH_3OH), acetic acid (CH_3COOH), propionic acid ($CH_3CH_2CO_2H$), and ethanol (C_2H_5OH) under 253.7 nm wavelength for 20 h. In 1975, mercury acetate ($C_4H_6O_4Hg$) solution was tested with solid sulfur, mercury oxide (HgO), and mercury sulfide (HgS). The results identified HgO, mercury sulfate, and $HgSO_4$ as the photosensitizers. Thiols also work as photosensitizers when the thiol (R-SH) group and CH_3COOH are available and the reaction will be an intramolecular decarboxylation reaction [24••]. Mercuric acetate solution produced MeHg in an acidic medium in the dark. When the solution was exposed to light, the reaction slowed and photolysis of MeHg occurred. Radical CH_3 groups formed from acetate photolysis also reacted with the dissolved oxygen (DO), not in methylation [34].

Sources of Mercury Emissions

The occurrence of mercury can be attributed to both natural and anthropogenic sources. Natural sources include soil and the earth's crust (earth crust off-gassing process into water) [12••]. This can be highly affected by the geological location, earthquake activities, volcanic eruption, or natural seeps [4•, 12••]. These factors can influence the concentration of Hg in raw petroleum products [4•, 35]. In soil, part of the Hg comes from natural sources like volcanoes, geothermal springs, and forest fires. In natural gas, Hg is considered to be a trace metal, except when the geographic location of the soil is rich in Hg [30]. Furthermore, anthropogenic sources contribute to a total of 86×10^6 kg [7•]. These sources are highly related to industrial activities causing pollution such as mining and smelting, artisanal small-scale gold mining (ASM), coal burning, oil refining, chlor-alkali process (mainly inorganic Hg), and cement production [7•].

Heavy metal contamination in fish is a result of the consumption of toxic wastes discharged into the water [31]. According to WHO, Hg from small-scale gold mining can bioaccumulate in predatory fish that feed on non-predatory fish [36]. Periodic discharge from a chlor-alkali plant caused an increase in the Hg level in flounder fish [7•]. In Qatar, fish found near the industrial city of Ras Laffan had a higher level of total Hg compared to Al-Khor, resulting from high Hg salt (mercuric/Hg(II)) loading [9]. In the food industry, the canning process of tuna fish can introduce not only Hg but different heavy metals to the fish tissues [5]. In the Arabian Gulf, the main source of Hg is related to oil fields like oil terminals, oil spills and natural seeps, and sewage discharge. The shore of Qatar has four coastal petrochemical plants [12••]. Hg accumulation in the soil is estimated to be around 25×10^7 kg to 1×10^9 kg globally. During wastewater discharge from petroleum refineries, Hg can leak out into soil and water to result in soil and water contamination. This can be controlled and limited by using Hg removal techniques [4•].

Global warming has a definite direct and indirect impact on the water chemistry (temperature and pH), which influences the fish size, productivity, Hg level, and methylation rates in the fish [25]. Carnivorous species of fish can reflect the increase in Hg through the food chain [37]. In general, the Hg level in the Arabian Gulf fish is considered to be below the maximum allowable levels (MALs) with only 10% of fish samples being above it. Most of the fish exhibiting Hg levels within 10% of the limit are found around chlor-alkali plants. Both these plants discharged Hg-rich waste into the sea [10•].

Wastewater (WW) Discharged

The level of Hg in the wastewater discharged into the water bodies is a main global concern. According to the National Association of Clean Water Agencies, around 36% of the Hg in wastewater comes from dental clinics due to the use of Hg in dental amalgam. In 2006, the U.S. Environmental Protection Agency (USEPA) created a roadmap to characterize Hg sources and collection, and provided mitigation methods [38•]. Wastewater and contaminated liquids discharged into water bodies can have adverse effects on animals and humans. The discharge of 90,718.5 kg of Hg into rivers and fields in China from an organic chemical factory between 1971 and 1997 resulted in underweight livestock, a reduction in annual yield of grain production by 30–40%, and increased cancer cases. Furthermore, in 1980, Hg contamination in water bodies from explosives in chemical companies manufacturing pesticides in Switzerland affected France, Germany, and the Netherlands through the Rhine river [39]. In recent years, Hg removal from wastewater (WW) improved significantly. The annual Hg import from

treated WW in Guangzhou city, China in 2016 was 0.27 kg, with around an 82% decrease in Hg level [40]. The annual treated WW discharged into rivers from wastewater treatment plants (WWTPs) in Switzerland in 2017 was estimated at around 4.7 ± 0.5 kg, with around a 96.4% decrease in Hg level [41].

Even though modern WWTP can remove up to 90% of total Hg and 70% of MeHg from influent, municipal wastewaters still contain MeHg concentrations that are higher than the normal MeHg levels in water bodies. The overall Hg imports into the environment from municipal WW in China in 2015 was 160,000 kg, with a total of 23,000 kg into aquatic ecosystems [42]. This can be attributed to the presence of organic matter (OM) and ionic Hg in the effluent, which facilitates the methylation process. In the oxidation ditch process in China, the MeHg was uptaken by the microorganisms but the total Hg will end up in the sludge. Moreover, the type of process applied in the municipal WWTP Hg removal from the wastewater. Gao et al. stated that the primary sedimentation will not significantly reduce the Hg concentration and that the main factor for high total Hg removal including MeHg is pH and temperature. In general, establishing the fate of Hg in WWTP still needs further research [40].

The most famous Hg pollution case was that of Minamata Bay in Japan caused due to waste discharge by the Chisso company discharged waste (CCDW) between 1932 and 1961. Even though the total Hg concentration reduced with time, the bay still needed restoration that was started 30 years after 1961 by the Kumamoto prefecture from 1991 to 2008. However, 2 to 4% of the total Hg from the CCDW is still present in the sediment [43, 44]. Sediment samples taken from the bay in 2012 resulted in a weighted average of 2.28 mg/kg (dry) from different sediment depths (0 to 32.5 cm). The highest average of 2.96 mg/kg was recorded for the sediment surface [43]. Sediment samples from different sediment depths (0 to 23 cm) in the Yatsushiro sea in 2017 had a weighted average of 0.46 mg/kg (dry), and the highest average was for sediment surface of 2.77 mg/kg [44]. The concentration of total Hg from different years and countries is listed in Table 1.

Eutrophication can alter the geochemistry of water by enhancing the growth of the autotroph species resulting in an increase in the MeHg concentration caused by increasing the methylation process [51]. Eutrophication will result from algae bloom due to the presence of essential growth factors such as high nutrients level and exposure to sunlight [33]. Eutrophication will increase the sediment organic matter (OM) and hypoxic/anoxia conditions, and reduces pH levels [47•, 48]. In addition, the algae can bioaccumulate MeHg, and after they die, the MeHg will be released back into the water or cause biomagnification through the food chain. In Ji et al. [52], the eutrophication conditions were used to

Table 1 Total mercury (Hg) concentration in sediment from different locations in mg/kg (dry weight)

Authors	Concentration	Location	Year of publication
Burke et al. [45]	0.09055	Alaska, USA, Atqasuk lakes	2020
	0.06279	Alaska, USA, Reindeer Camp lakes	
	0.00108	Alaska, USA, Atqasuk lakes	
	0.00012	Alaska, USA, Reindeer Camp lakes	
Kreish et al. [46]	0.0239–0.179	Qatar from 8 coastal areas	1999
De Mora et al. [47•]	0.0007–0.0167	Qatar from 5 coastal areas	2004
	0.0006–0.0022	UAE	
	<0.0001–0.0112	Oman	
	0.0025–0.2202	Bahrain	
Hassan et al. [30]	0.008–0.0337	Qatar from 11 coastal areas	2019
Satheeswaran et al. [48]	0.02–9.29	Parangipettai coastal region, India	2019
Akito et al. [43]	2.28	Minamata Bay	2014
Matsuyama et al. [44]	0.46	Yatsushiro Sea, Japan	2019
	0.1	Amakusa Sea, Japan	
Elsagh et al. [49]	3.24	Bandar Abbas, Persian Gulf, Iran	2021
Song et al. [50]	0.14–44.0	China from 4 different locations	2021

mitigate and reduce the increasing levels of MeHg level by injecting eutrophic waters with nanobubbles O_2 [52]. Therefore, MeHg absorption by algae can be used under control as the MeHg mitigation method [53]. The dynamics between WW discharge, eutrophication, and MeHg level are shown in Fig. 2A. However, the dynamics between eutrophication and Hg still need further research [54]. Pharmaceutical WW, mainly from antibiotics production, contain methyl groups like tetracyclines (TC) and oxytetracyclines (OC), which enhance the production of MeHg [55••]. This will enhance the production of MeHg. This phenomenon highlights that

bio-methylation might not be the main path for the methylation of mercury [56]. However, the effect of antibiotic WW on the methylation process needs further studies. The production of MeHg from antibiotics and Hg is shown in Fig. 2B and Scheme 1.

Air Pollution

To understand the transportation of air pollutants, we need to establish a geospatial distribution for the pollutant from the source to the receptor must be established. In most cases,
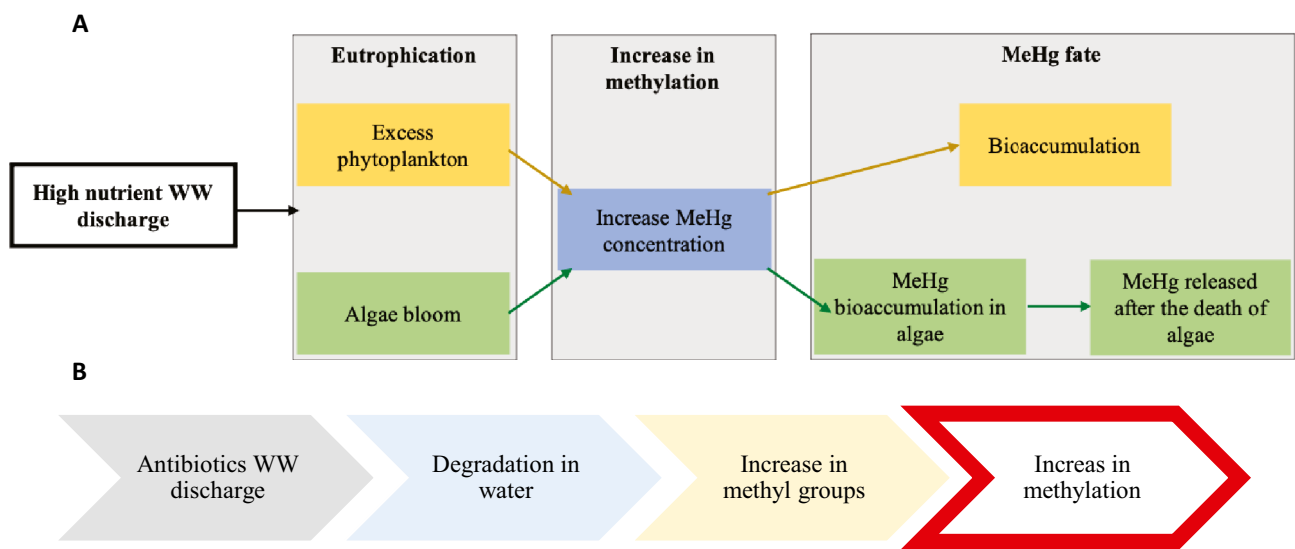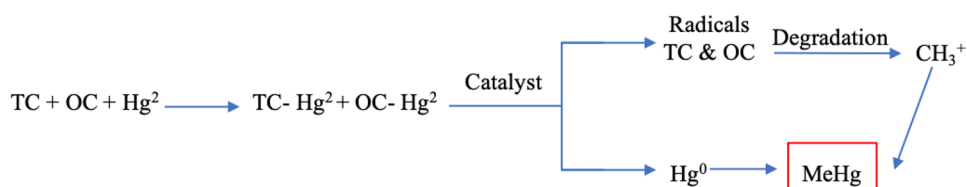

Fig. 2 **A** Two scenarios for the effect of eutrophication, and **B** antibiotics and MeHg formation

Scheme 1 The assumed antibiotics degradation to form MeHg [56]

$$TC + OC + Hg^2 \longrightarrow TC\text{-}Hg^2 + OC\text{-}Hg^2 \xrightarrow{\text{Catalyst}} \begin{cases} \text{Radicals} \\ TC \& OC \xrightarrow{\text{Degradation}} CH_3^+ \\ Hg^0 \longrightarrow \boxed{MeHg} \end{cases}$$

these are point sources. Geospatial distribution requires the exact location of the source using geographic coordinates and accurate emission values to each source or using the national emission estimates [57]. The chief pollutant sources are mainly related to industrial activities and power generation. Around 74% of atmospheric Hg comes from small-scale mining (ASM), stationary combustion of coal, and non-ferrous metals production [53, 54]. Estimating the geospatial distribution for ASM sources is difficult because of their presence in large numbers and, in many cases, transience over time [57]. The Middle Eastern region (the Asian part) is accountable for 2.4% (40,700–93,800 kg) of the global Hg emissions, of which the industrial sector contributes more than 55% [58]. Figure 3 shows the contribution of different sectors to the total Hg emissions globally and in the Middle East. In Qatar, Hg emissions mainly arise from industries using the chlor-alkali process (such as cement production), medical and municipal wastes, and natural gas production (ranging between 10^3 ng/m^3 and 200×10^3 ng/m^3) [30].

Fate and Transport of Hg in the Environment

Figure 4 shows the fate and transport of Hg species between earth's spheres: atmosphere, hydrosphere, geosphere, and biosphere.

In the Atmosphere

In air elemental, Hg^0 makes up to 95% of the mercury species and has a lifetime range from months up to 1 year. This will give it time to travel and to deposit from (dry/wet) far from the emission source [19•]. There are three types of atmospheric elemental Hg, which are named the total atmospheric mercury (TAM) and they are gaseous elemental mercury (GEM), gaseous oxidized mercury (GOM) or reactive gaseous mercury (RGM), and particulate mercury (HgP) or particulate bound mercury (PBM) [59, 60]. The GEM has the lowest deposition rate due to its low water solubility [59]. The PBM is an airborne particulate from the absorption of GEM or RGM [61]. The RGM and PBM make up only 10% of the TAM; they have shorter lifetimes that range between hours and days or weeks and have higher dry and wet deposition rates [61]. The highly volatile Hg species like
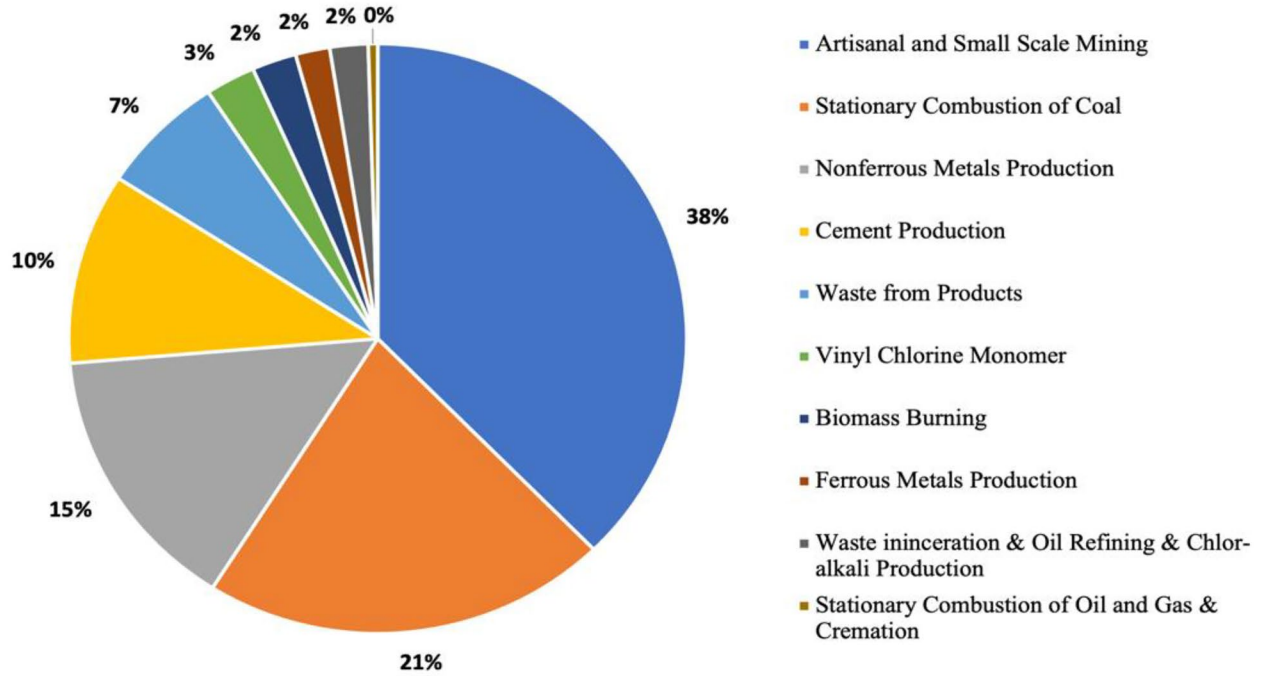
GEM and dimethyl mercury are able to go through long-range transport [62]. The mercuric mercury Hg^{2+} has a short lifetime in the atmosphere that is eventually deposited by wet deposition into the water bodies and then will go through the methylation process to form MeHg [3]. The highest Hg^{2+} wet deposition is around the equator owing to the influence of the low-pressure system and the trade winds along the line. The upper troposphere contribution makes 60% and in some areas 70% of the deposited Hg^{2+}. However, the lower troposphere is the highest contributor in the high latitude areas. The highest dry deposition will be in the subtropical anticyclone areas like the Middle East (mainly the Asian side), in high altitudes (e.g., the Himalayas), and around Antarctica. The upper troposphere is the main contributor by 79–82% except in the northern and southern poles, while the lower troposphere is the main contributor to the water [63]. Figure 5A shows the fluxes and spatial distribution of Hg^{2+} from wet, dry, and total deposition. The wet deposition for Hg^0 has a similar trend to Hg^{2+}. The total Hg^{2+} wet deposition is also similar to the total Hg^0 deposition. However, in some regions, the Hg^0 is approximately higher in concentration. For example, in the total deposition of Hg^{2+} in the regions around the equator in Africa, Hg^{2+} is less than 15 μg/m^2/year compared to 13–60 μg/m^2/year for Hg^0, and in the poles [58]. Figure 5B shows the fluxes and spatial distribution of Hg^0 from wet and total deposition.

In the Soil and Sediment

Soil is a natural Hg source and reservoir and is considered to be a Hg sink [61]. Hg contamination in soil results from the atmosphere or from the anthropogenic addition [64]. For example, during the gold extraction process [62], the atmospheric pathway occurs in two ways, namely, direct Hg deposition into the soil surface or when plants absorb the Hg and then recycle it into the soil through litterfall [64]. Mercury can also be transported from the soil to the atmosphere through volatilization, and from the hydrosphere to groundwater through infiltration [7•]. The inorganic Hg in the soil may be present in different species. One of the most stable species is Hg^{2+} sulfide or cinnabar HgS. Hg can also be transported through the food web and bioaccumulated as MeHg. Therefore, to understand the fate of Hg in the soil, we need to measure Hg using sequential extraction procedures [62]. In coastal areas, the sediment is considered to

A

Global sources of Hg

- Artisanal and Small Scale Mining
- Stationary Combustion of Coal
- Nonferrous Metals Production
- Cement Production
- Waste from Products
- Vinyl Chlorine Monomer
- Biomass Burning
- Ferrous Metals Production
- Waste ininceration & Oil Refining & Chlor-alkali Production
- Stationary Combustion of Oil and Gas & Cremation

B

Middle East sources of Hg

- **Fuel Combustion**
- **Industrial sector**
- **Intentional use (product waste)**
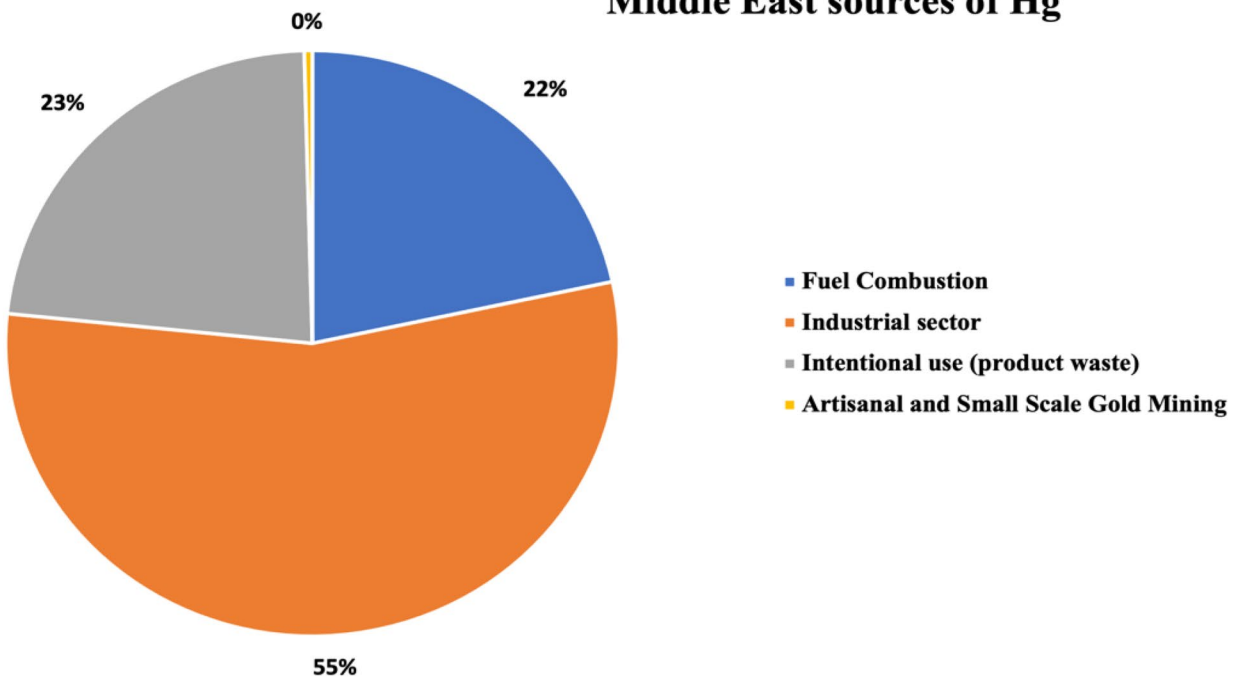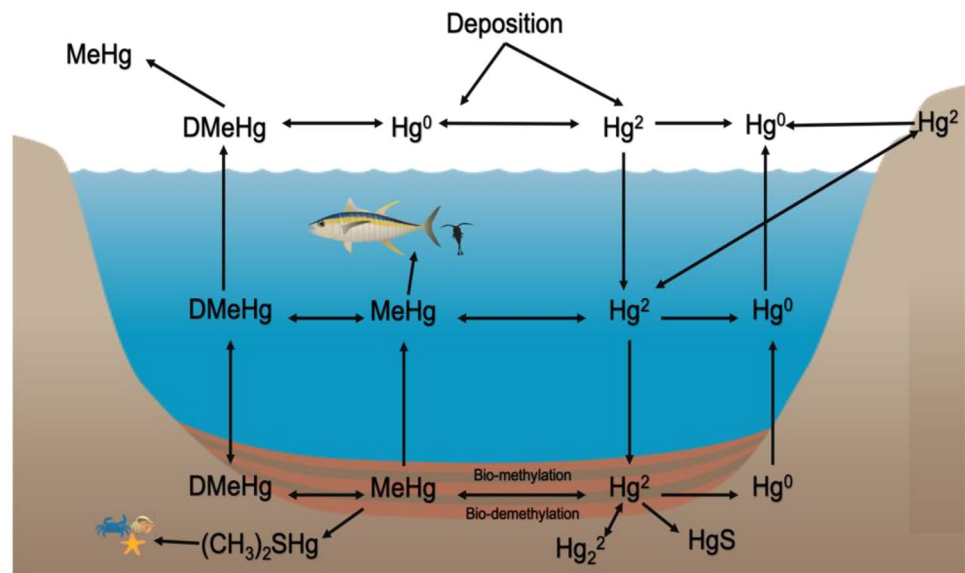- **Artisanal and Small Scale Gold Mining**

Fig. 3 Chart showing Hg emissions contribution from different sectors: **A** global Hg emission and **B** Middle East Hg emission

Fig. 4 Hg biogeochemical cycle between earth's spheres

be the main source of MeHg and it is considered as a Hg sink. However, methylation and demethylation dynamics are difficult to predict [35]. The water–sediment interaction makes predicting the concentration of MeHg and total Hg very difficult because of the advective transport, sediment characteristics, molecular diffusion, and the surrounding conditions [35]. The mangrove ecosystem is considered to be a carbon sink system. This means that the organic matter (OM) levels will be high, further enhancing the bioaccumulation of Hg in the sediment. Moreover, mangrove sediment
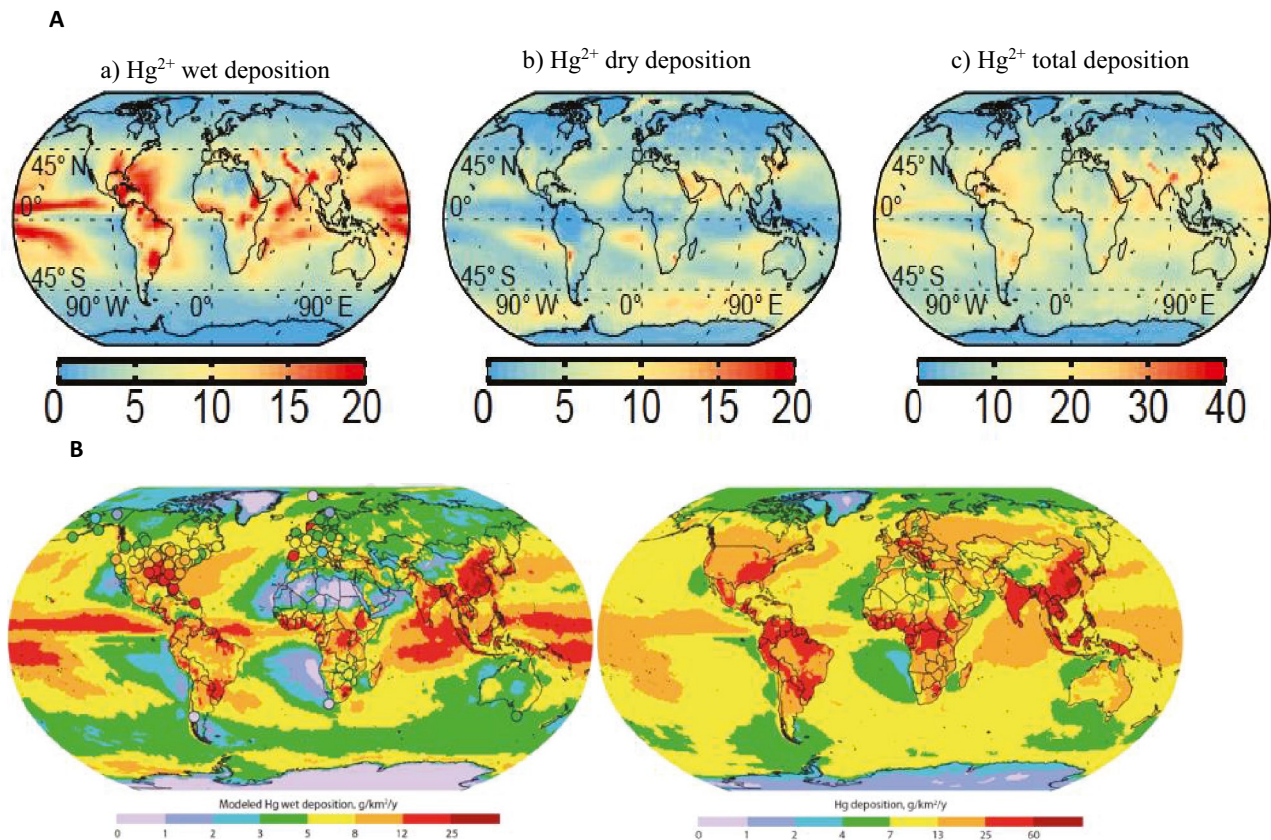

Fig. 5 A The fluxes of Hg^{2+} (*a*) wet deposition, (*b*) dry deposition, and (*c*) total (wet and dry) deposition (μg/m²/year) [63], and **B** the fluxes of Hg^0—left is wet deposition and right is total (wet and dry) deposition (g/km.²/year) [58]

is low in pH, has an anoxic surrounding, and has adequate amount of sulfate and SRB. All those characteristics will create the perfect environment for Hg methylation. The litterfall from the mangrove tree was the main source of the OM and during anaerobic conditions, the MeHg concentration increased [65].

In Hydrosphere

Zhang et al. used the ocean tracer model the OFFline global 3D ocean TRACer model (OFFTRAC) to estimate the Hg distribution in the ocean for the present time. The high atmospheric wet deposition rate increased the Hg concentration in the oceanic mixed layer in the coastal area and around the mid-latitude. The anthropogenic input contributed 80 to 100% of the overall Hg input, and it is concentrated in the middle and northern hemispheres, and lower around the poles. The overall Hg concentration increased with the increase in depth at around 1 km. However, methylated Hg is considered to be the highest subsurface water and part of the net OM [22]. The organic particle deposition and the low oxygen levels increased the Hg concentration in the Gulf of Guinea, the Arabian Sea to South Asia, and the highest (~0.401 ng/L) spreading on the American side of the Pacific Ocean between 60°N and 30°S. The anthropogenic input is the highest in the North Atlantic Ocean and between 30°S

and 60°S [66]. Figure 6 shows the Hg spatial distribution for the present-day Hg concentration and Hg from anthropogenic emissions. Rivers and estuaries are considered to be two of the main total Hg sources for the aquatic system by contribution range between 50 and 80% compared to less than 10% from terrestrial [67•].

Fish and Mercury

Fish Species and Mercury Bioaccumulation

Mercury bioaccumulates in the tissues of aquatic species through the ingestion of contaminated soil and its concentration increases through the trophic chain [3]. Fish are considered to be at the top of the trophic levels in the marine ecosystem [6]. The trophic chain starting from the bottom of the food chain to the top is as follows: heterotrophic (zooplankton and benthic invertebrates), herbivorous, and carnivorous (predatory fish). Therefore, predatory fish contain higher levels of Hg since they are placed at the top level of the trophic chain [68]. The carnivorous fish sharp nose shark (*Rhizoprionodon oligolinx*) had the highest Hg concentration 1.287 ppm compared to 0.0068 ppm for the Badah (*Gerres oyena*), which is considered an omnivorous fish [12••]. When low or mid-trophic level species have high Hg, the
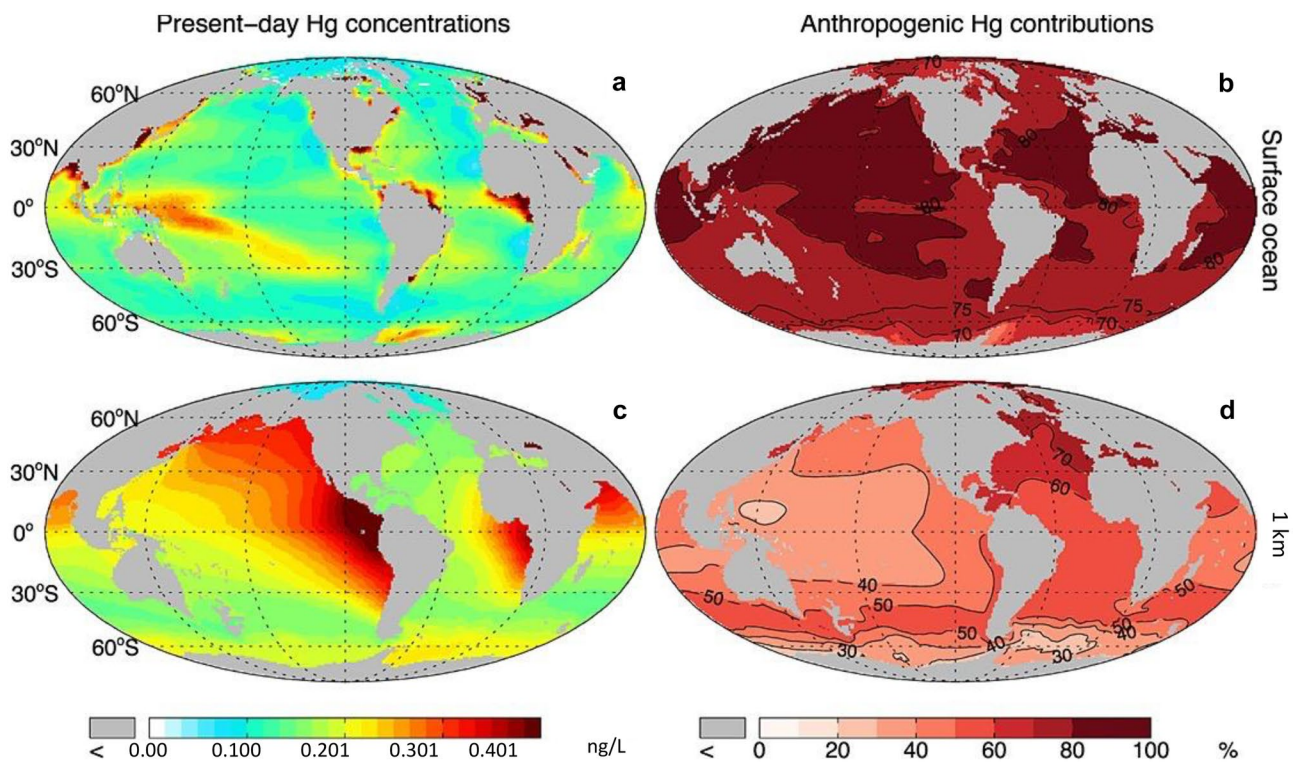

Fig. 6 Spatial distribution of Hg concentrations in the present day in nanograms per liter and percentage of anthropogenic contribution

Hg levels increase in the upper trophic level species (e.g., tuna fish) [9]. In addition, it was reported that the deeper the water column of the ocean, the higher the Hg level in the fish species. For example, benthic species have higher Hg levels than pelagic species (e.g., sardines and mackerels) [68]. A study showed that mesopelagic fish had lower Hg levels compared to epipelagic fish [37]. This was explained by the trophic level of the fish, which will reflect on the feeding sources for the fish in each layer. Since the mesopelagic fish in this study are planktivorous (*Hilsha ilisha*) and the epipelagic are carnivorous, the Hg level will be influenced by the trophic level instead of water depth [47•]. Some carnivorous, non-predatory species like *Lethrinus nebulosus* can feed on echinoderms and crustaceans instead of small fish, which would lead to lower Hg levels compared to other carnivorous species like *Epinephelus coioides* that feed strictly on small fish [9]. On the other hand, anchovy species (*Stolephorus indicus* and *Engraulis encrasicolus*) are low-trophic level fish that feed mainly on zooplankton, prawn, shrimp, and amphipods [17]. They are also prey for carnivorous, pelagic, and demersal species [69]. Another example of low-trophic level fish is sardine (*Sardina pilchardus*), a pelagic fish that feeds on phytoplankton and zooplankton. Sardines can feed in two ways, namely, filter feeding and particulate feeding, depending on the food source available [14, 67•]. Hg concentration in the parts of anchovy from United Arab Emirates (UAE) ranged between 0.05 mg/kg (w/w) and 0.18 mg/kg (w/w) [17]. Hg concentration in the muscles of sardine from Portugal ranged between 0.0016 and 0.0006 mg/kg [68]. Figure 7 shows the relation between Hg levels in different fish species based on the trophic levels, niches, and/or habits.

MeHg has the highest bioaccumulation ability in the tissues compared to other forms of Hg [12••]. Hg exists as MeHg between 70 and 100% in fish. The concentration of total Hg and MeHg in the Arabian Gulf ranged between 0.012 ppm (w/w) and 0.970 ppm (w/w), and 0.03 ppm (w/w) and 0.248 ppm (w/w), respectively. Some studies only measured the total Hg. Therefore, the highest concentration for MeHg is low compared to the total Hg concentration [12••]. The concentration of heavy metals and metalloids in fish is highly associated with the increasing size and length of the fish. It was also found that secondary carnivores have higher levels compared to fish from lower trophic levels [28]. Length and weight of the fish have a positive correlation with the MeHg concentration [9]. This can be related to the increase in the fish's life span and size [47•]. In some fish species, the warmer temperature was noted to enhance the feeding rates and in others will not [70]. The species, where the feeding was enhanced, will increase the metal bioaccumulation. However, warmer temperatures might increase the growth rate, which will dilute the content of the metal [33, 70]. Hg concentration is also related to the type of food the fish consumes and the composition of the water. The functional proteins in the muscle of the fish have the highest level of Hg [21]. Hg levels in the fish parts were observed in the liver, muscle, and finally gonad and other parts in decreasing order. This is a result of high metabolic activity in the liver compared to the other parts and tissues, which will increase the metals' accumulation ability [12••]. Moreover, female fish have higher Hg levels compared to males [9]. However, if the concentration of Hg in the muscles is 0.5 ppm and below, the high detoxification ability of the liver will maintain a lower Hg level than the muscles [12••]. The Hg level in fish during the fall season is higher than in the other seasons as a result of the thermocline effect in the summer, which brings
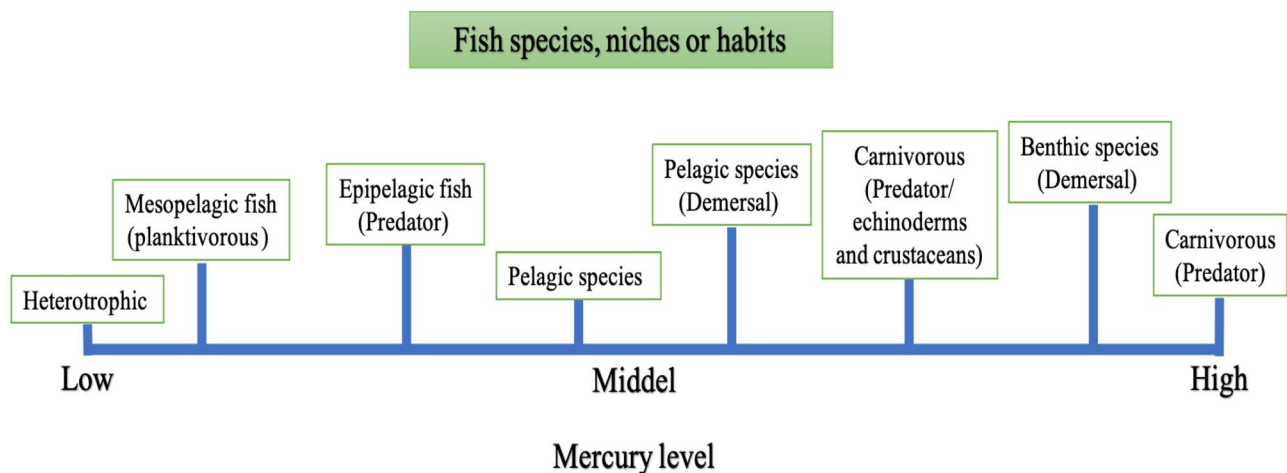

Fig. 7 The relationship between Hg concentration and the effect of species trophic levels, niches, and/or habits. Species habits based on water column (up to bottom): epipelagic, mesopelagic, and benthic

Table 2 Fish species from different trophic levels and their Hg concentration

Fish	Trophic levels	Country	Part—dry or wet	Hg concentration (ppm)	Reference
Lethrinus nebulosus	Carnivores (non-predatory)	Qatar	Liver—wet	0.773	Al-Ansari et al. [9]
		UAE	Liver—dry	1.02	De Mora et al. [47●]
		Oman	Muscle—dry	0.522	
		Bahrain	Muscle—wet	0.110	Freije and Awadh [71]
Epinephelus coioides	Carnivores	Oman	Muscle—dry	1.3	De Mora et al. [47●]
		Bahrain	Liver—dry	2.1	
		UAE	Liver—dry	4.65	
		Qatar	Liver—dry	1.28	
		Kuwait	Liver—dry	0.01–3.92	Al-majed and Preston [37]
		Kuwait	Muscle—wet	1.29	Laird et al. [72]
		China	Muscle—wet	0.056	Chen et al. [73]
		Iran	Muscle—wet	0.317–0.489	Saei-Dehkordi et al. [74]
Scomberomorus commerson	Carnivores	Malaysia	Muscle—wet	0.108	Anual et al. [28]
		Bahrain	Muscle—wet	0.126	Freije et al. [71]
		Kuwait	Muscle—wet	0.37	Laird et al. [72]
Stolephorus indicus	Herbivore	UAE	Muscle—wet	0.04–0.18	Alizada et al. [17]
Sardinella albella	Planktivores	Bahrain	Muscle—wet	0.029	Freije et al. [71]
Siganus rivulatus	Herbivore	KSA	Muscle—wet	0.02	Hakami et al. [75]
		KSA	Muscle—wet	ND*–0.004	Burger et al. [76]
Siganus canaliculatus	Herbivore	Bahrain	Muscle—wet	0.032	Freije et al. [71]
Siganus fuscessens	Omnivores	China	Muscle—wet	0.018	Chen et al. [73]
Rastrelliger brachysoma	Planktivores	Malaysia	Muscle—wet	0.025	Anual et al. [28]

*Not detected

the Hg from the ocean ground sediment into the upper layers [68]. However, a study in Qatar showed that there are no seasonal variations [9]. Tables 2 and 3 present Hg and MeHg concentrations in different fish species from different trophic levels.

Fish Preservation

Food preservation is an important process in the food industry since it makes storing food for a long time possible and preserves the nutritional content [77]. Tuna is a predatory

Table 3 Fish species from different trophic levels and MeHg concentrations

Fish	Trophic levels	Country	Part—dry or wet	Hg concentration (ppm)	Reference
Lethrinus nebulosus	Carnivores (non-predatory)	Qatar	Liver—wet	0.771	Al-Ansari et al. [9]
		Bahrain	Muscle—wet	0.032	Freije et al. [71]
Epinephelus coioides	Carnivores	Kuwait	Muscle—wet	0.55	Laird et al. [72]
		Kuwait	Muscle—dry	0.001–3.27	Al-Majed et al. [37]
		Bahrain	Muscle—wet	0.107	Freije et al. [71]
		China	Muscle—wet	0.045	Chen et al. [73]
Scomberomorus commerson	Carnivores	Bahrain	Muscle—wet	0.123	Freije et al. [71]
		Kuwait	Muscle—wet	0.23	Laird et al. [72]
Siganus canaliculatus	Herbivore	Bahrain	Muscle—wet	0.028	Freije et al. [71]
Siganus fuscessens	Omnivores	China	Muscle—wet	0.0145	Chen et al. [73]
Sardinella albella	Planktivores	Bahrain	Muscle—wet	0.028	Freije et al. [71]
Gerres oyena	Carnivores (non-predatory)	Qatar	Muscle—wet	0.0283	Elsayed et al. [12●●]
Chiloscyllium arabicum	Carnivores		Muscle—wet	0.1662	
Rhizoprionodon oligolinx	Carnivores		Muscle—wet	0.7942	

fish (top in the food chain). As a result, they have a high ability to bioaccumulate heavy metals in the tissue [78]. Canned tuna is one of the most consumed canned fish around the world [79]. Tuna and salmon make up around 9.2% of the fish captured and produced around the world [80]. Commercial handling and processing like canning increase the Hg level in tissues [79]. Exposing the fish to high temperatures while cooking or the canning process will increase the Hg concentration. This can be explained by some chemical reactions between Hg species and sulfhydryl groups forming complex compounds, the loss of water during cooking (reduction in humidity), and an increase in mercury/mass ratio due to loss in minerals and mass reduction [81]. Freezing is an important method to preserve high-protein food sources and does not affect carbon (C) and nitrogen (N) concentrations [82]. Frozen fish from the Arabian Gulf had an average of 0.79 µg/g Hg concentration. This is above the World Health Organization and Food and Agriculture Organization (WHO/FAO) non-predatory fish limit [31]. A study was done on frozen fish to see the effect of freezing on the Hg concentration. The different fish species were frozen at –20 °C temperature from 2002 to 2006 (around > 600 days). The study concludes that there is no significant difference in the Hg concentration [83]. Since food processing can increase the heavy metal concentration in fish, humans would minimize their health risks by eating varied fish species in smaller quantities [84].

Tuna is a pelagic fish with various tuna species sold as canned fish. Examples of tuna fish include species skipjack (*Katsuwonus pelamis*), bigeye (*Thunnus obesus*), yellowfin tuna (*Thunnus albacares*), albacore (*Thunnus alalunga*) [85, 86], and Atlantic bluefin tuna (*Thunnus thynnus*) [87]. Some tuna species can accumulate Hg more than others [88]. Milatou et al. showed that farmed tuna has a lower concentration than the tuna caught from the ocean [13]. Table 4 shows different Hg levels in tuna fish. The effect of canning on the heavy metal concentration in fish is affected by the processing conditions, filling medium quality, the quality of inside coating, the oxygen concentration in the headspace, pH level, and storing temperature and time [77]. The canning process can increase the heavy metal concentration in the canned fish. This means that the quality of the canned fish processing is a critical factor to be taken into consideration [89]. Tuna species have a wide range of variations in the trophic level between 3 and 4.8 as a result of bioaccumulation and biomagnification, habitat, size, and age [5]. The effect of different habitats, for example, bluefin tuna from the Atlantic, reach higher trophic levels faster compared to the Mediterranean. Bluefin tuna aged more than 1 year will feed on plankton, shrimps, coastal benthopelagic fish, and small pelagic fish regardless of size. On the other hand, age groups 6 to 9 will feed on medium and larger pelagic fish

[90]. Table 4 shows the concentration of total Hg in different canned tuna samples from different countries; 16 out of 30 samples were above the criterion level of Hg in fish at 0.3 ppm [91]. If we considered the weekly intake, this can influence human health significantly.

Mercury Bioindicators and Measurements

Fish as Bioindicator

In general, fish take up heavy metals in two ways: directly through water and feeding, and indirectly through permeable membranes [17]. The ability of metals to interrupt the vital metabolic processes in the fish tissues makes fish a good bioindicator [10•]. The best way to study heavy metal pollution is by using fish as a bioindicator for two main reasons: bioaccumulation and biomagnification [88]. The first factor is that the metals in general have strong persistent characteristics and have the ability to bioaccumulate in the fish tissues [10•]. The second factor is biomagnification, where metals that were consumed by lower trophic level species will bioaccumulate in the tissues, following the mid-trophic level species that will consume them and increase the concentration of the metal in their tissues. This process goes on until it reaches the human tissues [5].

Mercury Stable Isotope

The isotope technique (also known as a chemical tracer, or isotopic measurement) is a method used to study the biogeochemical Hg cycle and sediment deposition rate in water [99]. It is also used to study the Hg sources and transformations in rice [79, 81]. Hg has seven stable isotopes and can be divided into five different types: radioactive, enriched stable, natural abundance, light isotopes (not Hg), and ^{13}C isotopes [81, 82, 100]. Radioactive isotopes include ^{203}Hg for small marine species Hg uptake and efflux [101•]. Spiking of highly enriched stable isotopes ^{199}Hg, ^{200}Hg, ^{201}Hg, or ^{202}Hg into the study area is carried out to study the methylation and demethylation rates in soil and the Hg biogeochemical cycling in watersheds [79, 81]. Natural abundance isotope ratios ^{198}Hg, ^{199}Hg, ^{200}Hg, ^{201}Hg, ^{202}Hg, or ^{204}Hg are used for MeHg photodemethylation in water, and food chain in marine species and humans. They are also used to investigate the dry and wet deposition of Hg contribution. The last two types will use the isotopes of other elements, the light isotopes ^{13}C, ^{15}N, or ^{34}S, for food web complexities and trophic level estimation for MeHg biomagnification, and finally, the natural abundance stable ^{13}C isotope to study the carbon source for the MeHg [81, 82].

Table 4 Concentrations of total Hg in canned tuna fish wet-weight basis

Country of origin of the tuna	Tuna species	Fresh or canned	Hg concentration (ppm)		References
			Average	**Range**	
Iran	Unknown	Canned	–	$(0.01–0.05) \pm 0.01$	Rahmani et al. [5]
	Thunnus albacares	Canned in oil	0.0321	–	Mansouri et al. [92]
	Thunnus albacares and *Thunnus tonggol* (mix)*	Canned in oil	0.024		
	Thunnus albacares and *Thunnus tonggol* (mix)*	Canned in oil	0.0394		
	Thunnus albacares and *Rutilus kutum* (mix)	Canned in oil	0.0339		
Greece	*Thunnus thynnus*	Fresh	–	0.49–1.60	Milatou et al. [13]
Turkey	Unknown	Canned in oil	0.30	ND–4.13	Mol et al. [79]
Italy	Unknown	Canned in oil	–	0.050–0.057	Russo et al. [93]
Brazil	Unknown	Canned (mix oil and water)	0.256	0.071–0.588	Alva et al. [94]
	Unknown	Canned in water	–	0.05–0.46	de Paiva et al. [95]
		Canned in oil	–	0.044–0.402	
		Canned in water	–	0.041–0.46	
		Canned in oil	–	0.035–0.393	
Colombia	Unknown	Canned in water	0.77	Up to 3.1	Alcala-Orozco et al. [96]
		Canned in water	0.63	> 0.001–0.86	Alcala-Orozco et al. [97]
		Canned in oil	0.61	> 0.001–0.88	
Mediterranean Sea	*Thunnus thynnus*	Fresh	–	0.760–1.89	Kljaković-Gašpić et al. [87]
Indonesia	Unknown	Fresh	0.3174	–	Nong et al. [80]
Japan			0.1824		
Marshall Islands			0.0597		
Vietnam			0.0506		
USA	*Thunnus tonggol*	Canned in water	0.22**	–	Islam et al. [84]
Thailand	*Thunnus thynnus*	Canned in oil	0.098**		
Korea	*Thunnus thynnus*	Canned in oil	0.07**		
	Thunnus thynnus	Frozen	Dorsal = 0.18** Ventral = 0.24**		
Sri Lankan	*Thunnus albacares*	Fresh	0.48	0.07–1.60	FDA et al. [98]
Libya*	*Euthynnus alletteratus*	Fresh	1.185 ± 0.968	–	Abolghait et al. [88]
	Katsuwonus pelamis	Canned (mix oil and water)	0.373 ± 0.249	–	
	Thunnus albacares	Canned (mix oil and water)	0.163 ± 0.122	–	

ND not detected

*Different brand; **Highest level, the average or range was not mentioned; ***The study was done in the mentioned country; however, the origin is either different countries or unknown

Hair as Bioindicator

Hair can be used as a biomarker for MeHg exposure. MeHg is uptaken from blood and accumulates during hair formation. After the formation of the hair, the MeHg will be preserved as an endogenous character in the hair [102]. This method can show the time period of Hg exposure by taking the advantage of the hair natural growing process of hair by 1 cm/month [103]. Another advantage of this method is the easiness of sample collection and storage. Hg in hair is as follows: USEPA guideline is 1.0 µg/g and the Joint FAO/WHO Expert Committee on Food Additives (JECFA) is 2.3 µg/g, calculated using the reference dose from USEPA and provisional tolerable daily intake (PTWI) from [83, 104].

DPSIR Model

Driving forces, Pressures, States, Impacts, and Responses (DPSIR) is a conceptual model that can be used to assess the environmental risk for different risk sources [85, 86, 105, 106••]. Driving forces refer to high-level human needs and/or the need for the emission source [86, 87]. In turn, this would result in pressure on the environment by the use of methods to meet the needs of the driving forces. States refer to the environmental characteristics as a result of pressures [106••, 107]. The impacts are the changes or the quantitative effects on the environment and humans [85, 87]. The responses are the actions taken or implemented to prevent or mitigate the impacts [87, 88]. The DPSIR facilitates the investigation of all the possible cause–effect relationships to plan appropriate responses. Figure 7 shows the DPSIR for this review article.

Driving Forces

Mercury is an element that occurs as waste in different industries. This means that the economic growth will increase the production of those industries, which will increase the emissions and discharge them into the environment [108]. In developing countries, ASM is an important industry and it contributes 837,658 kg of Hg annually to the atmosphere [54, 89]. The main issue is that in developing countries, the ability to effectively apply mitigation measures is difficult [108]. Another major source is burning coal, which is the main energy source in China and Mongolia; this makes controlling Hg emission a major problem [109]. For years, the relationship between economic growth and the environment has been negatively correlated. In recent years, this began

to change in developed countries with stringent regulations being applied to industrial waste disposal [110]. The MeHg will accumulate in the biotic aquatic species to enter the food chain [19•]. Humans are exposed to mercury and MeHg through the consumption of contaminated food [9]. MeHg bioaccumulation can be a result of consistent contaminated fish consumption [12••]. Fish capturing around the world increased from 8.86×10^{10} kg in 2012 to 9.64×10^{10} kg in 2018 [111]. In many countries around the world, fish is considered one of the main protein dietary sources [28]. Fish consumption from the 1960s increased rapidly by 67% to 20.3 kg in 2017 [112]. Fish supply was more than 21 kg per capita in 2018 compared to around 14 kg per capita during the 1990s [111]. Table 5 presents fish consumption around for different countries world.

Pressures

Figure 8 shows the comparison between global total Hg emissions, deposition, and surface ocean from different studies. The Arctic Monitoring & Assessment Programme/ United Nations (AMAP/UN) estimations for 2019 showed a significant increase compared to 2013 levels [58]. Kawai estimations show a significant decrease compared to the AMAP/UN (2013 and 2019) estimations [58, 116••, 117]. Comparing the AMAP/UN anthropogenic emission for the three different years, a significant increase in the Hg emission can be seen. The Kawai et al. study using the FATE-Hg model shows a small decrease compared to the United Nations Environment Programme (UNEP) and AMAP/UN 2013 and 2019 modeling results which cover a wider range of data from different countries, regions, trends, and sector

Table 5 Fish consumption per capita for different countries around the world

| Country	Fish consumption	Year	Reference
International	20.5 kg per capita/year	2019	Cunningham et al. [10•]
	20.3 kg per capita/year	2017	FAO [112]
International (without China)	16 kg per capita/year	2017	FAO [112]
Asia	24.1 kg per capita/year		
Malaysia	160 g per capita/day	2018	Anual et al. [28]
Qatar	10–20 kg per capita/year	2017	FAO [112]
Brazil	10 kg per capita/year	2020	Alva et al. [94]
	175 per capita/week		Ferreira da Silva et al. [113]
KSA	Saudis: 150 g/week	2019	Cunningham et al. [10•]
	Expatriates: 397 g/week		
Mexico	250 g/week or 35.71 g/day	2021	Murillo-Cisneros et al. [114]
Greece	20 kg per capita/year	2020	Milatou et al. [13]
	68.68 g/day	2019	Sofoulaki et al. [15]
Iran	20.3 kg per capita/year	2021	Mansouri et al. [92]
	21–147 g/person/day (frozen fish)	2017	Ziarati et al. [31]
Colombia	8–10 kg per capita/year	2021	Alcala-Orozco et al. [96]
USA	18.7 g per capita/day	2018	Sunderland et al. [115]
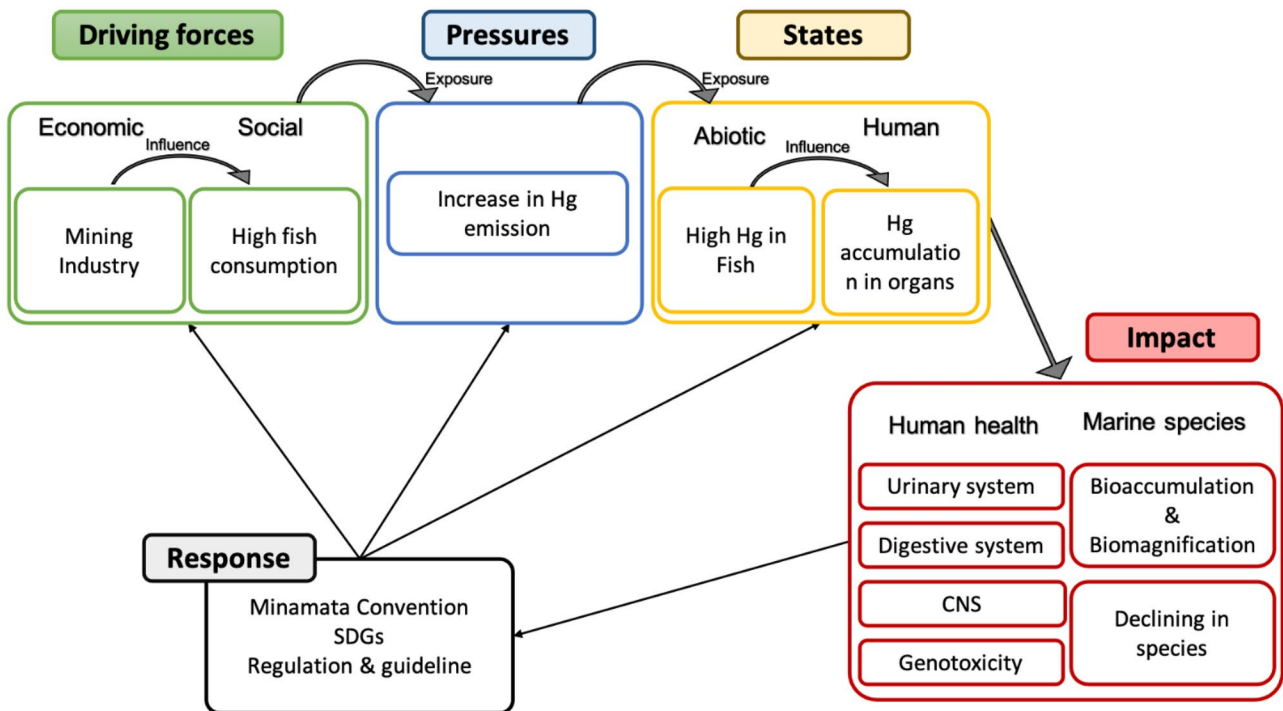
Fig. 8 DPSIR model sample diagram

[58, 118]. The pressure of increasing Hg emission increase into the atmosphere reflected the increase in the total deposition in the AMAP/UN results. However, the surface ocean in 2019 showed conflicting results. Compared to 2013, the total deposition was lower and the re-emission from the ocean was significantly lower than in 2019. The Hg level in the surface ocean in 2013 was higher compared to 2019. Typhoon season in some countries like China will enhance the air quality by distributing Hg in larger areas through the atmosphere. On the other hand, typhoons enhanced the wet deposition processes into soil and water bodies. The total gaseous mercury (TGM) concentration emitting from fluorescent lamp manufacturing in China in the industrial and residential areas ranged between 6.2 and 49 ng/m^3 and 0.57 and 5 ng/m^3 before typhoon season, respectively. During typhoon season, the concentration in industrial and residential areas ranged between 0.63 and 17 ng/m^3 and 0.37 and 2.7 ng/m^3, respectively [119]. The geochemical characteristics of Hg and weather conditions will deposit more Hg into the water bodies and sediment, which can result in an increase in MeHg in the water column [120]. Hg isotopes testing from different countries around the world have proven that mining sites discharged into rivers are the main contributor to Hg levels in water and sediments [121]. Eventually, it will be discharged into the sea through estuaries [104]. Estuarine sediments are the perfect methylation ecosystem since it is naturally rich in dissolved organic matter (DOM), and different anaerobic bacteria such as sulfate-reducing bacteria

(SRB) and methanogens bacteria. The average MeHg concentration in 70 different estuaries around the world was 880 ng/g [27] Fig. 9.

States

The gaseous elemental mercury (GEM) can travel long distances and has a lifetime ranging from 6 months to 2 years [122]. In Taiwan and during the Asian Northeastern Monsoon in winter, the Hg emissions from coal-burning in the Mongolian plateau reach the oceans and influence the ambient air quality in the coastal areas in Taiwan. The total gaseous mercury (TGM) concentration average was 4.56 ng/m^3 [59]. This is higher than the range of exposure from areas away from industries at around 2–4 ng/m^3, but lower than urban areas' exposure at around 10 ng/m^3 [123]. According to WHO, the Hg level for areas away from industries should not exceed the range between 5 and 10 ng/m^3 [124]. Soil quality criteria (SQC) are used to assess the quality of soil from polluted sites, and it differs between countries. In Finland, the SQC is 0.5 mg/kg compared to 20 mg/kg in Germany. The European Union (EU) range for Hg in polluted soil is 1 to 1.5 mg/kg. Soil quality in active mining sites in Ghana significantly exceeded all limits by 71 mg/kg and the inhabited areas near the mine site exceeded the EU by 2.7 mg/kg [125]. The transportation of Hg species into food through water or soil will influence food quality. Rice fields that are closer to industries that produce Hg as a pollutant

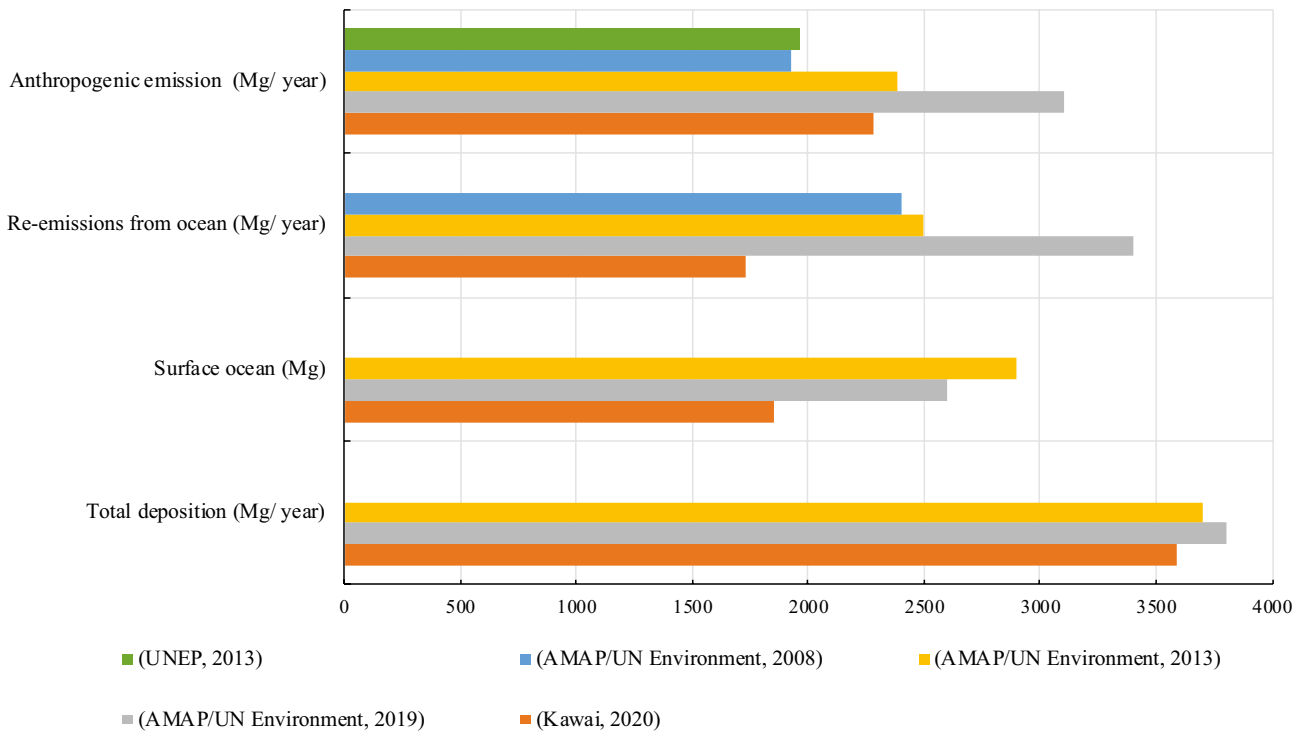The Global budgets of Total Hg concentration

Fig. 9 The global budgets of total Hg concentration from anthropogenic emissions, deposition, surface ocean, and the re-emission from the ocean to the atmosphere

will be highly contaminated as a result of the bioaccumulation ability. The bigger issue with contaminated rice is that rice is a major food export around the world; this makes it a risk to a wider population around the world [126]. In Bangladesh, Hg pollution influences the air, water, and soil quality as a result of industrial growth. This influences the quality of food products, mainly fish and rice [127]. Hg discharged from mines into the Bonsa River in Ghana resulted in serious deterioration of water quality and hence undrinkable [128]. The Office of Environmental Protection and Energy in Liestal, Switzerland, estimated that the concentration of total Hg in the Rhine River ranged between 2.7 and 4.5 ng/L from 1994 to 2017, respectively. Suess et al. estimated the total Hg concentration between 2016 and 2017, and it was ranged between 3.8 and 6.3 ng/L, respectively. Only 1.5 to 3% of the estimated total Hg was from WW and the remaining 91% was from other sources like air pollution [41]. Hg levels were measured in sediment, water, and fish species from a water reservoir in Nigeria. The maximum metal concentration in sediment was Hg. The concentration of Hg in water and sediment in the reservoir was 0.39 mg/L and 2.46 mg/g, respectively. The bioaccumulation factors (BAFs) of Hg from water in fish tissues ranged between 1.41 and 2.28. When BAF is more than 1, this indicates an accumulation of Hg in the organism (Usman, 2020). The quality

of water and sediment will be reflected in marine species based on their feeding habits. MeHg in sediment correlated positively with the total Hg concentration in mummichog fish, while total Hg in water correlated positively with the total Hg concentration of Atlantic silversides fish. This difference in correlation was related to the feeding habits [129].

Impacts

Impacts on Human Health

All chemical forms of mercury are toxic to humans. These forms differ in their degree of toxicity and the type of toxic effects they are producing. The chemical form also influences its absorption and distribution in the human body. Hg enters the human body through different routes via inhalation, ingestion, and dermal contact [21]. The absorption levels of the different chemical forms of mercury for the different routes of exposure are presented in Table 6.

Elemental Hg is rapidly absorbed through respiration in high concentrations (32 to 160 ng/day). Elemental mercury is highly lipid soluble. Therefore, once in the systemic circulation, it rapidly penetrates the red blood cells, with 95% of this form found in the red blood cells. It is further oxidized to mercuric in the red blood cells by the hydrogen peroxide

Table 6 Hg species and absorption through different routes of exposure (H: high, M: moderate, and L: low) [21, 130]

Bioavailability from exposure routes	Hg species		
	Elemental	Organic (MeHg)	Inorganic
Inhalation	H for vapor (~85%)	(~80%) H	L–M (40%)
Oral	Very L for liquid (0.01%)	H (95%)	L–M but high in infant (~15)
Skin	M for vapor (3%)	L–M (~5%)	L–M (~3)

catalase pathway [9, 115]. The elemental Hg oral pathway is mainly from dental amalgams [20]. Elemental Hg distributes through the blood and can penetrate the brain. Elemental Hg can accumulate and oxidize in the brain to mercuric forms [130]. Elemental Hg is metabolized by oxidation that converts it to mercuric ions. However, this process is slow [20, 130]. Elemental Hg is eliminated through the feces and exhaled air [130]. The half-life of elemental mercury in the human body is reported to be approximately 60 days [130]. The toxicity of elemental Hg is presented in Table 7.

Inorganic mercury can enter the human body through inhalation, ingestion, and to a lower degree through the skin [130]. The absorption rates from all routes are low. They are estimated to be approximately 40%, 15%, and 4% for the ingestion, inhalation, and dermal routes, respectively. It was found that exposure to mercuric chloride for 4 h a week (1 h per day) results in daily absorption between 37 and 44 µg/kg [130]. Inorganic forms have low lipid solubility [131]. They do not readily cross the cell membrane and do not cross the blood–brain barrier. However, once absorbed, mercuric salt binds to the cysteine group in proteins, which facilitates their transport across the cell membranes. The mercuric form can also be converted to methylmercury by microorganisms

in the intestinal tract or reduced back to elemental Hg by mitochondrial proteins, nicotinamide adenine dinucleotide phosphate (NADPH), and nicotinamide adenine dinucleotide (NADH) [130, 132]. The half-life of inorganic mercury compounds in the body is reported to be approximately 40 days. The toxicities of inorganic Hg salts are presented in Table 7.

Among all the chemical forms of Hg, the organic form, namely, methyl mercury (MeHg), is the most toxic one. The main route of exposure of MeHg is through ingestion of contaminated fish. The absorption rate from the ingestion route is estimated to be around 95%. MeHg is highly liposoluble and binds to the cysteine group in proteins yielding its rapid transportation across cell membranes. Binding to the thiol group forms MeHg-L-cysteine/S-(methylmercury)-L-cysteine ($C_4H_9HgNO_2S$) complex which imparts MeHg the ability to readily cross the blood–brain barrier [18, 117]. In contrast to the other forms of Hg, MeHg is stable and is slowly broken down through demethylation by microorganisms, photolysis, or by different abiotic conditions [22]. MeHg has a long half-life that ranges between 32 and 164 days, with the main route of elimination being through demethylation and excretion in the feces [20]. Besides, MeHg is secreted into breast milk,

Table 7 Mercury species' effect on human health

	Elemental Hg	Inorganic Hg	Organic Hg (MeHg)	Reference
Central nervous system (CNS)	Neurological dysfunction: erethism, memory loss, insomnia, nerve sensing, and motoring ability	Nerve pain, decrease in the cerebellar and the brain weight, irregular arm movements, and dysphagia	Neurological dysfunction and poor development in children and fetus Acrodynia, seizures, losing sight and hearing, language disorders, memory loss, paresthesia, and numbness in extremities and perioral area	EPA [111], EPA [116••], WHO [120]
Urinary system	Renal dysfunction	Renal transient proteinuria and failure	–	
Digestive system	Gastrointestinal dysfunction	Nausea, abdominal cramps, diarrhea, and corrosive to the gastrointestinal tract	–	
Genotoxicity	–	Chromosomal aberrations and disorders	Chromosomal aberration in lymphocytes	
Others	Chest pain, dyspnea, and reduction of pulmonary function	Corrosive to skin and eyes	Cardiovascular system: elevation in blood pressure Immunotoxicity	

which can yield to the exposure of newborns and infants [81, 130]. Methylmercury reacts with sulfhydryl groups interfering with cellular structure and function [130]. MeHg is a neurotoxin that can affect the central nervous system (CNS) and causes loss of speech, hearing, and vision; dementia; and dysarthria [1, 10•]. It can also cause damage to the cardiovascular system [28]. MeHg is highly toxic compared to the other forms of Hg [21]. The toxicity of organic Hg as MeHg is presented in Table 7. Moreover, due to its high penetration ability through plasma membranes, MeHg can also cross the placenta. Fetus exposure to MeHg can cause premature delivery and lifetime mental and physical disabilities to the newborn [133••]. Exposure of infants and young children to low levels of MeHg can have a negative effect on their development, learning abilities, and behaviors [21].

Impacts on Marine Species

Bioaccumulation of MeHg in the fish tissues, muscle, and liver is fast since MeHg has a low elimination rate [12••, 68]. In many cases, the high levels of Hg can cause death to the aquatic organisms [21]. Hg toxicity can increase the risk of oxidative damage to the fish that can be recognized by testing DNA damage and lipid peroxidation in muscles and the liver [68]. Seabirds and marine mammals that feed on the contaminated fish will be at risk of Hg toxicity [16]. The contaminated fish will reach the toxic level as a result of Hg bioaccumulation, which results in decay in a fish population [9]. This would result in a decrease in the number of marine species, particularly fish communities, and adversely impacts biodiversity in general [10•]. In addition, MeHg will bind to metallothionein protein in the liver and increase hepatic methylation. This results in increasing the MeHg concentrations in the muscles [12••, 113]. Similar to the human body, the fish body will accumulate Hg in the tissue by binding to the thiol group of proteins [87].

Socioeconomic Impact

Important environmental and socio-economic impacts are caused by Hg anthropogenic emissions. The effect that Hg

exposure has on children will influence their IQ level and lifetime. Trasande et al. assessed that losses in production in the USA in 2000 were around \$8.7 billion. The cost was estimated using the Institute of Medicine (IOM) model [134].

$$Costs = disease\ rate\ \times EAF \times population\ size \times cost\ per\ case$$

$$EAF: environmentally\ attributable\ fraction$$

Guo et al. stated that in China consumption per capita and population growth were the main factors for the increase in Hg emissions. However, for the time period from 2007 to 2012, there was a noticeable decrease in Hg emissions in some provinces as a result of improving the energy production efficiency with the exception of some developing provinces that still rely on coal-burning as an energy source [109]. A recent finding by Huang et al. for the time period from 2010 to 2019 confirmed that provinces and isolated regions like the Tibetan Plateau emit higher Hg levels and are at higher risk for Hg pollution impacts on health and the environment [135]. Illegal mining projects on the Bonsa River, Ghana, result in Hg level exceeding the EPA permissible limit and resulted in undrinkable water. This will impact the economy by limiting freshwater sources and increasing the cost of freshwater production [128]. In addition, this impacts the supply of safe fish species in Ghana. The impact of Hg pollution from mining projects on the economic value of fish species was observed. The levels of Hg in all fish species exceeded the WHO limit (0.5 mg/L). This will have a negative economic impact on fish as the main food source in this developing country [136].

Responses

Minamata Convention

In 2017, mercury is considered one of the ten main chemicals that is of concern to public health [137]. The United Nations (UN) introduced the Minamata Convention on Mercury in 2013. This convention was established to face the anthropogenic emissions of mercury. The convention takes into consideration the life cycle of mercury

Fig. 10 The Sustainable Development Goals (SDGs) related to Minamata Convention [138]

Table 8 The Sustainable Development Goals (SDGs) related to Minamata Convention [138]

SDG	Title	Relation
1	**No poverty**	Mining workers, bad living conditions, and direct exposure to coal-burning are in poor developing countries
2	**Zero hunger**	The consumption of fish is a major protein in many countries, and providing uncontaminated food is a major issue in many poor developing countries especially since Hg bioaccumulates in the body
3	**Good health and well-being**	The many Hg exposure routes to humans are a major problem and can have an adverse effect on CNC, digestive system, and immune system
7	**Affordable and clean energy**	Coal and oil burning is still the main energy and transportation producer; new renewable renovation can reduce the Hg emissions significantly
8	**Decent work and economic growth**	The main exposure threat is among the people who work in places that emit/discharge Hg or around those areas. The solution is to provide protection for workers, follow safety standards, and develop Hg-free products and processes
12	**Responsible consumption and production**	The aim of this goal reduces Hg pollution and waste by using Hg-free products, processes, and technologies that are cost-effective
14	**Life below water**	Marine and coastal areas are the nerve of life, and the negative influence of Hg pollution, biomagnification, and bioaccumulation in fish and other marine species is a major threat to human health

and waste management. The convention aims to regulate mercury compound emissions and the use of Hg in any form in the industry. The convention allowed the parties to design implementation plans within the regulation. The parties should report to the conference about Hg sources; anthropogenic emissions into the atmosphere, soil, and water; mercury amalgamation from artisanal and small-scale gold mining; and the use of Hg compound in the industry. Moreover, the convention addresses the importance of research, development, and monitoring of mercury emissions and biotic media including fish [19•, 120]. These are seven Sustainable Development Goals (SDGs) that can be linked to the Minamata Convention listed in Fig. 10 and the relation is listed in Table 8 [138].

Regulations and Guidelines

The World Health Organization and the Food and Agriculture Organization (WHO/FAO) developed a

Table 9 MeHg guideline levels (GLs) in fish

GLs	Concentration	Reference
Predatory fish (MeHg) permissible limits (PL)	1.0 ppm (wet wt)	EU Commission and FAO/WHO [122, 142]
Non-predatory fish (MeHg)	0.5 ppm (wet wt) or 2.5 ppm (dry wt)	Al-Ansari et al. and FAO/WHO [10•, 104]
PTWI (MeHg)	1.6 µg/kg	FAO/WHO [139]
TWI (MeHg)	1.3 µg/kg	EFSA [141]
PL	0.3 ppm (wet wt)	EPA [91]

research-based intake guideline for non-predatory and predatory fish [139] (Table 9). Those guidelines are based on the estimated intake of the contaminant per body weight over a lifetime without resulting in any risk to human health [140]. The guidelines also indicated the acceptable daily or weekly intakes [94]. The tolerable weekly intake (TWI) is the amount of contamination in a specific amount of food or water per unit of body weight, which a person can ingest weekly without having the risk to develop adverse health effects. Tolerable intake can also be used to address the daily intake (TDI) [8]. The provisional tolerable weekly intake (PTWI) expresses the maximum safe long-term exposure intake from the contaminant [141]. The PTWI was set at 1.6 µg/kg BW for methyl mercury. Exceeding the PTWI level is highly possible in nations where fish consumption and/or contamination levels is/are high. When pregnant women exceed the PTWI level, the Hg can influence the fetus's brain development [28].

$$\text{PTWI} = \frac{mean\ Hg\left(\frac{mg}{kg}wet\ weight\right) \times weekly\ fish\ consumption\ (mg)}{body\ weight\ (kg)}$$

The Environmental Health Criteria 101 Methylmercury estimated the intake for a different form of Hg in the general population. MeHg was 2.41 µg/day with 2.4 µg/day from fish. The inorganic Hg intake from fish was only 0.6 µg/day and 0 µg/day for elemental [143]. The WHO guideline for the maximum inorganic Hg in drinking water is 6 µg/L [8] and the EPA maximum contaminant level (MCL) is 2 µg/L [144]. The EU maximum limit for total Hg in the surface water is 70 ng/L [145].

Conclusion and Future Recommendations

The global risk from the anthropogenic atmospheric Hg emissions and wastewater discharge is still a major issue regardless of the mitigation measures and guidelines. This is a result of the resilience and accumulation ability of Hg. In the atmosphere, the gaseous elemental mercury (GEM) has a long lifetime in the atmosphere, which has risks of spreading the pollution to different regions and causing a global issue. Even though the soil is a natural sink for Hg, it naturally exists in the earth's crust. Hg in the soil can be accumulated as inorganic and organic Hg, re-emit to the atmosphere, or leach to groundwater. Water is another major sink for atmospheric Hg, but the main issue is the accumulation of Hg as MeHg in marine species. Fish is the main food product that has a high Hg concentration because of bioaccumulation and biomagnification. The risk of consuming highly contaminated fish cannot be overlooked since Hg is a neurotoxin. The use of the DPSIR framework made it possible to draw a scenario of the Hg cycle from emission to health risks. It was found that rapid economic and population growth could play a significant part in the mitigation process by following the Minamata convention regulations and guidelines. However, developed nations will continue to emit high He levels and will be exposed to a higher level of Hg in fish. The future perspective should focus on linking Hg pollution sources to Hg level in fish and the human body to be able to assess the real hazard risks. In addition, the impact on and emissions from developed nations or regions should be taken into consideration since they can reverse the global mitigation efforts. Most studies investigate Hg in the earth's sphere without linking the impact directly on human health from seafood consumption.

Compliance with Ethical Standards

Conflict of Interest The authors declare no competing interests.

Human and Animal Rights and Informed Consent This article does not contain any studies with human and animal subjects performed by any of the authors.

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- • Of major importance

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