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REVIEW

Ecosystem Mercury Recovery and Health Benefit Under the Minamata Convention in a Changing Climate

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Abstract

Mercury (Hg) is a toxic metal that can be released from natural and anthropogenic processes. Gaseous elemental Hg is the main form of Hg in the atmosphere with a long residence time, allowing its long-range transport and deposition at a global scale. The deposited Hg can be transformed into methylmercuy (MeHg), which bioaccumulates and biomagnifies in aquatic ecosystems and is a known developmental neurotoxin to humans. Chronic exposure in pregnant women via fish consumption, even at the modest levels, can impact intellectual development in infants, which is considered as a global environmental health issue. To protect human health and the environment, the Minamata Convention on Mercury, a legally binding international treaty, entered into force in 2017 to reduce anthropogenic emissions and releases of Hg and Hg compounds. With the enforcement of the Convention, anthropogenic Hg emissions and releases are projected to decrease in the foreseeable future, but the recovery of the ecosystem and mitigation of health impact from Hg pollution are expected to lag behind. In this paper, we provide a critical review on the processes affecting when and to which extent Hg-affected ecosystems will recover under the Minamata Convention in the context of climate change and associated human health benefit. Our review is organized around seven major scientific questions. These seven questions covered historical Hg change in the ecosystem, major factors and processes, and future assessment. The impact of implementation of Minamata Convention and climate change on Hg emission, atmospheric transportation and deposition, biota accumulation, and human exposure should be comprehensively evaluated. The Hg stable isotope tool and process-based integrated global model should be integrated together, which also should consider future global social-economic pathways and mitigation options. We hope that the knowledge gaps and potential tools identified will assist the understanding of global biogeochemical cycling of Hg, inform policy-making on mitigation and community adaptation, and support the effectiveness evaluation and future improvement of the Minamata Convention on Mercury.

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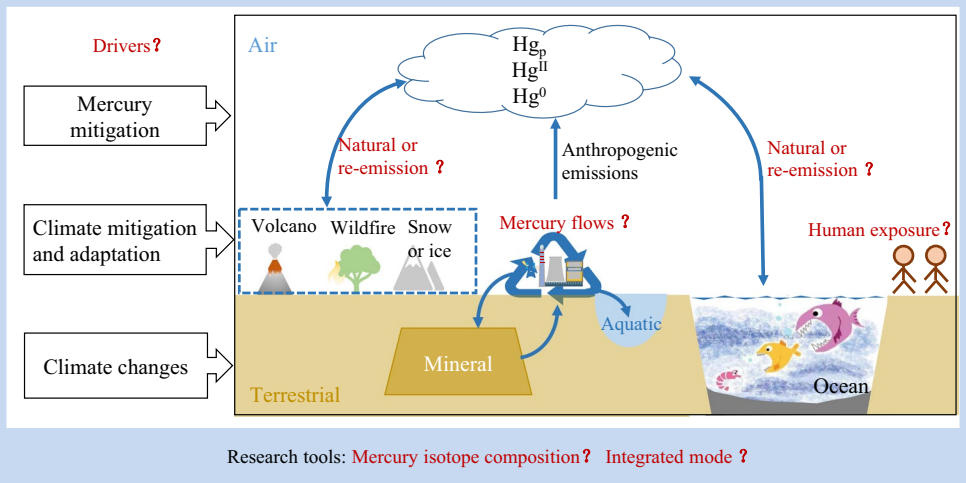
Graphical Abstract



Introduction

Mercury (Hg) is a natural element in the Earth's lithosphere, which can emit to the atmosphere through natural processes (e.g., weathering and volcanism) (UNEP 2019; Pirrone et al. 2010). Atmospheric Hg can transport on a global scale and deposit to surface terrestrial and marine ecosystems (UNEP 2008; Sikkema et al. 2011). Mercury in the surface environment is either re-emitted to the atmosphere, buried in stable long-lived reservoirs (e.g., soil and sediments), or accumulated in biota including humans (UNEP 2019). This natural Hg cycling has been greatly altered by human activities (e.g., coal combustion, gold and silver mining), which have liberated millions of tons of Hg from the lithosphere and augmented its flux between the lithosphere, the atmosphere, and surface ecosystems (Horowitz et al. 2014; Streets et al. 2011, 2019). Due to its characteristics of long-distance transport, persistence, bioaccumulation and toxicity, the overburden of Hg in the surface environment poses severe risks to ecosystem and human health globally (Douglas et al. 2012; Mergler 2021; So et al. 2021).

In response to the global risks, the Minamata Convention on Mercury was signed in 2013 and entered into force in 2017, aiming to protect humans and the environment from Hg pollution (UNEP 2019). However, the impact of climate and other global changes on the biogeochemical cycling of Hg is still unclear, which may doubt the ecosystem Hg recovery and health benefit from regulatory activities (Krabbenhoft and Sunderland 2013; Obrist et al. 2018; Wang et al. 2019). This makes it difficult to project when and to which extent the ecosystems will recover and how human health impact will be minimized. The Minamata Convention on Mercury calls for its effectiveness which will be evaluated for the first time before 2023 and periodically thereafter at intervals (UNEP 2013), yet the Convention itself does not spell out how this should be done. The negotiation process during the past three Conferences of Parties was fraught with conflict on the best indicators and methods to evaluate the effectiveness, as the concurrence of multiple synergistic and antagonistic driving factors challenges the attribution of ecosystem and human exposure change to regulatory activities.

Evidence is mounting that future climate change will likely be the main driving force of global Hg cycling, especially in sensitive regions such as the Arctic (Hansen et al. 2015; Stern et al. 2012). However, scientific information currently available is not sufficient to quantitatively answer how climate change affects the timing and extent of the ecosystem recovery due to the complexity of the system. For example, the increase of air temperature has the potential to liberate Hg from permafrost to the atmosphere and the aquatic ecosystem at a certain turning point, yet in regions experiencing glacier retreat global warming can accelerate the uptake of atmospheric Hg by vegetation (Wang et al. 2020a). Global ocean warming is also likely a major driving force of future methylmercury (MeHg) concentration change in marine predators (Schartup et al. 2019). Furthermore, actions to mitigate climate change such as the improvement of energy efficiency in power stations and replacement of fossil fuels by renewable sources are

expected to result in the reduction of Hg emissions as a co-benefit (Pacyna et al. 2016; Streets et al. 2009). Studies should therefore be undertaken to understand and model the relationships among those synergistic and antagonistic processes to allow the projection of changes in ecosystem Hg and human exposure in a changing climate. This is critically important for the development of mitigation and adaptation strategies that Hg-affected communities can take to reduce the risk while the ecosystem is toward recovery. New and rapidly developing scientific tools such as the Hg isotope fractionation measurement could potentially help address these challenges.

Future projection on global anthropogenic Hg emissions indicates up to 85% of reduction by 2035 under the best-case scenario, which would result in up to 50% and 35% reduction of Hg deposition in the Northern and Southern Hemisphere, respectively (Pacyna et al. 2016). However, model evaluation indicates that terrestrial and oceanic Hg reservoirs originating from legacy anthropogenic and natural Hg sources will continue to grow and emit more Hg under all but the most stringent emission control scenarios (Amos et al. 2013). In addition, the effectiveness of regulatory policies under the Minamata Convention on Mercury will be reduced with time when any of the countries delays or weakens their actions since more legacy Hg emissions will accumulate in the environment (Angot et al. 2018). Future Hg emission control trajectories are also highly uncertain, as they depend on both global socioeconomic development and technological innovation and penetration.

In this review, we focus on a critical analysis of major scientific knowledge gaps that are limiting our capability of projecting ecosystem recovery and health benefit under the Minamata Convention in the context of climate change. Our analysis is summarized in three sections: 1) Perspective of historical Hg change in the ecosystem; 2) Major factors/processes affecting ecosystem Hg recovery and human health; 3) Future assessment. It includes seven key scientific questions, which covered historical changes, natural emissions/re-emissions, ecosystem changes with atmospheric deposition, and ocean Hg cycle. The Hg stable isotope and integrated model are introduced as powerful tools to assess ecosystem recovery and health benefit, and future global social-economic pathways and mitigation options are also considered in the last question. These seven key scientific questions dealt with the most important aspects in global Hg biogeochemical cycle under the context of the Minamata Convention and climate change. Our goal is to assist the understanding of global biogeochemical cycling of Hg, inform policy-making on mitigation and community adaptation, and support the effectiveness evaluation and future improvement of the Minamata Convention on Mercury.

Perspective of Historical Hg Change in the Ecosystem

Q1 How have Hg concentrations in the atmosphere and biota, as well as human Hg exposure, changed over the past millennia? How do we determine "background" or "safe" concentrations and how does that affect our definition of "recovery"? What are the best indicators to evaluate the effectiveness of the Convention?

During the past decade, atmospheric Hg declines have been observed globally from both ground station- and ship-based measurements. Declines of Hg concentrations in Europe and North America were on the order of 1–2% per year (UNEP 2019). Inventory and modeling studies have attributed these trends to declined anthropogenic emissions, resulting from the phase-out of Hg from commercial products and the co-benefit from SO_2 and NO_x emission controls on coal-fired utilities (Zhang et al. 2016). Long-term Hg monitoring data in the Arctic have shown generally declining but weaker trends of Hg concentrations than the observations in the mid-latitudes, with an observed decrease rate of ~ 0.2% per year since 1994 (Wang et al. 2019). Complex long-term atmospheric Hg trends in some regions could be explained by changes in anthropogenic and natural Hg emission and atmospheric oxidants (Lyman et al. 2020). Chen et al. (2015) found an association between the weaker trends of atmospheric Hg concentrations in the Arctic and the change in temperature and sea ice content. Time-series data also become available in China in recent years. The observed atmospheric Hg trends generally agreed with the China's emissions trend (Liu et al. 2019). Mercury measurements on the Tibetan Plateau indicated stable atmospheric Hg concentrations during 2006–2009 and decreased trends during 2010–2015 (Tong et al. 2016). The GEM concentration means at Mt. Lulin, Mt. Changbai, and Chongming Island in China remains relatively constant from 2010 to 2012 and shows a decline from 2013 to 2016 (Feng et al. 2022). In contrast, no significant temporal trends were observed in the free tropospheric Hg which is a better indication of the global background level (Zhang et al. 2016). A few vertical atmospheric Hg profile measurements have also been reported. For example, Slemr et al. (2014) reported the aircraft-based investigation for the global upper troposphere/lower plumes stratosphere with elevated Hg from 2005 to 2013 (the ARCTAS program) and no significant trends were found. They found that many of the observed plumes were associated with biomass burning that were transported over long distances.

The atmospheric Hg deposition to aquatic ecosystems is often responsible for elevated MeHg levels in fish. It was reported that two-thirds of Hg in fish from US lakes originated from anthropogenic sources (Hammerschmidt and Fitzgerald 2006). However, the temporal trends of Hg concentrations in aquatic biota often did not agree with atmospheric trends, which might be caused by legacy Hg and changing biogeochemical processes (Wang et al. 2019). Hg concentrations in aquatic biota in North America, North Europe, and the Arctic have generally decreased since the 1970s. However, significant increases were also observed over the recent decades in North America and the Arctic. In the Great Lakes, fish Hg concentrations have presented a mixed temporal pattern since the 1970s (Blukacz-Richards et al. 2017): fish Hg declined in all species in the first few decades (up to 1995–2000), but that trend has been reversed in most species at some locations since 2005. When examining Hg data on 96,000 fish samples from 206 species in over 4200 lakes across western Canada and the USA, Eagles-Smith et al. (2016) found a significant and rapid decline in length-adjusted Hg concentrations in the 1970s, while no subsequent significant trend to 2012. The early declines in biotic Hg in most studies could be attributed to regional and local declines in atmospheric Hg concentrations and deposition, whereas the recent no-trends or trend reversals were a clear indication that other processes were also at play (Wang et al. 2019). In North Europe, Braaten et al. (2017) and Akerblom et al. (2014) found that the freshwater fish Hg concentrations (normalized to 1-kg pike) showed a consistent and significant decreasing trend during the past 50 years (1965–2015), which matched well with the declining atmospheric Hg trend over North Europe. In Arctic, some species in some locations had shown significant increases of Hg concentrations over recent decades, whereas others exhibited decreases or non-significant changes (Riget et al. 2011).

The fact that short-term trends in biota Hg do not always follow trends in atmospheric Hg should not discourage actions to reduce Hg emissions. Instead, implementation of the Minamata Convention on Mercury is necessary to achieve long-term results. Once an aquatic ecosystem has accumulated a sufficiently large quantity of Hg, biotic uptake of Hg would be controlled primarily by internal processes rather than new Hg inputs. Even if all the new Hg inputs are stopped, legacy Hg stored in the sediments and soils could be released under changing climatic and environmental conditions, methylated, and bioaccumulated and biomagnified in the food web. Therefore, it will take more time for biota Hg to decline, and in some cases, biotic Hg may even increase in the short term. Further attention is needed on the fate and effect of legacy Hg that is already stored in the environment, and on the factors and processes that affect the recovery of Hg in biota.

Data on human Hg exposure, especially long time-series data, are still severely lacking, with the exception of a few case studies such as the Faroe Islands and the Seychelle Islands (UNEP 2019). A review of national datasets from four countries showed a decline trend in blood Hg; urinary Hg also decreased, particularly in the US dataset where the recent Hg levels were approximately half of those a decade earlier (UNEP 2019). Sharma et al. (2021) reported significant declines in total Hg levels in three biological matrices (whole blood, cord blood, and breast milk) between 1966 and 2015 worldwide. A regional overview suggested the highest blood Hg levels in the populations in South America, followed by Africa and Asia, and with the lowest levels in Europe and North America.

A better understanding of historical atmospheric Hg levels and emission sources is critical for assessing both background Hg levels and future trends. Sediment and ice cores records showed that total Hg concentrations increased 2–7 folds after the Industrial Revolution compared with those in preindustrial times (Chellman et al. 2017). Sediment Hg in Lake Huguangyan, south China, showed significant increases since the 1980s, due to the increased Hg emissions from Asian countries. The ice core record in Wyoming, US, indicated a large Hg increase in the mid-1800s, which was attributed to mining activities during the Gold Rush, with a second large increase beginning in 1950 linked to industrialization (Chellman et al. 2017). Recent analysis of Hg isotopes of sedimentary archives indicated that the rise in industrial-use Hg is accompanied by an increase in δ^{202}Hg and Δ^{199}Hg values (Lepak et al. 2020).

Therefore, the definition of "background" or "safe" Hg concentrations in the environmental and biota matrixes is crucial. Even if anthropogenic Hg emissions are significantly reduced under the Minamata Convention on Mercury, Hg concentrations in the environmental and biota matrixes are unlikely to recover to the level of preindustrial times.

Ecosystem Hg recovery under the Minamata Convention on Mercury in a changing climate thus needs to be defined carefully with the ultimate goal to protect the wildlife and humans from adverse effects of Hg exposure. Decreased Hg loading to the environment will produce a response in fish MeHg, but the timeline and magnitude of the response will vary depending on ecosystem-specific variables and the form of the Hg loaded. Predicted benefits to wildlife would be primarily through improved hatching success and development of hatchlings to maturity, as Hg concentrations in prey fish decline (Scheuhammer et al. 2007). Evaluating the effectiveness of the Minamata Convention will require biomonitoring of multiple species that represent different trophic and ecological niches in multiple regions of the world. More national and global human

biomonitoring are needed to evaluate the effectiveness of the Minamata Convention.

Major Factors and Processes Affecting Ecosystem Hg Recovery and Human Health

Q2 How well do we know about natural emissions/releases or re-emissions/re-releases of mercury?

Natural Hg emissions include those from contemporary geogenic activities (e.g., emission from volcano, hot spring) and re-emissions of legacy natural Hg that was released in the past. Mercury emissions from contemporary geogenic sources were estimated to be 500 Mg yr^{-1} by the United Nations Global Mercury Assessment 2018 (Outridge et al. 2018). Edwards et al. (2021) reviewed volcanic Hg studies in the last fifty years. While Hg emissions from persistently degassing volcanos appeared to be constrained in a narrow range of 30–95 Mg yr^{-1}, great uncertainties existed with Hg releases from explosive eruptions; as a result, the best estimates of global volcanic Hg emissions still differed over an order of magnitude from 45 to 700 Mg yr^{-1} (Edwards et al. 2021). Even lesser known are Hg emissions from other geothermal–hydrothermal sources (e.g., hot spring), which are potentially large but poorly measured globally (Edwards et al. 2021). Long-range transportation of Hg after volcanic eruptions and chemical speciation transformation after the mixing of the plume and ambient air are still not well understood (Bagnato et al. 2007; Edwards et al. 2021; Babu et al. 2022).

Hg^0 re-emission fluxes from natural surfaces are highly diverse, and major data gaps exist for Africa, South Asia, Middle East, South America and Australia (Obrist et al. 2021; Sommar et al. 2016; Yu et al. 2018; Zhu et al. 2015a, b; Zhu et al. 2016). Spatiotemporal measurements in North America, Europe, and East Asia showed that the fluxes from natural Hg-enriched and anthropogenic polluted sites were 1–2 orders of magnitude higher than those from unpolluted terrestrial forest and grassland surfaces, particularly in Hg-enriched regions of East Asia (Agnan et al. 2016; Carpi and Lindberg 1998; Gustin et al. 2008; Lindberg et al. 2007; Zhu et al. 2016). Scaling up those filed measurements from limited geographical regions, the global air–surface exchange of Hg^0 in terrestrial ecosystems ranged from -513 to 1333 Mg yr^{-1}, and the largest uncertainty was observed from forested regions (-727 to 703 Mg yr^{-1}), due to insufficient spatiotemporal resolutions of data and a lack of standardization for measurement techniques and data comparability (Agnan et al. 2016; Zhu et al. 2016).

Wildfires can mobilize a substantially large amount of forest floor Hg that has been accumulated over decades or centuries rapidly into the air, dramatically increasing the atmospheric Hg loading (Friedli et al. 2009, 2001; Outridge et al. 2018; Wang et al. 2021). It was estimated that annual global wildfire Hg emissions ranged from 104 to 678 Mg (De Simone et al. 2015; Friedli et al. 2009; Huang et al. 2015; Kumar et al. 2018; Outridge et al. 2018), and more than 60% of which were from tropical regions (Friedli et al. 2009; Kumar et al. 2018; Shi et al. 2019; Wang et al. 2015). It was further estimated that over 75% of Hg emitted from wildfires were deposited into the ocean, and about 10% into the Arctic (De Simone et al. 2015; Kumar and Wu 2019). Wildfire-emitted Hg can enhance regional Hg deposition and increase ecological risk in Hg sensitive ecosystems (Garcia and Carignan 2005; Kelly et al. 2006; Vijayaraghavan et al. 2014). It is expected that climate change will lead to more frequent and severe wildfires and hence greater Hg emissions across the globe. Models projected an increase of 14–27% in wildfire Hg emissions by 2050 when compared with those in 2000 (Huang et al. 2015; Kumar et al. 2018; Shi et al. 2019; Turetsky et al. 2006).

Some of the Hg deposited onto snow and ice emits back to the atmosphere, the percentage of which largely depends on the speciation of Hg and the light conditions (Durnford and Dastoor 2011). The release of Hg from glaciers was studied only at a few glacial catchments. Glacier catchments in the Tibetan Plateau were found to have higher Hg yields than those from Arctic river basins (Zhang et al. 2019); even higher Hg loads were reported from the meltwater of the southwestern Greenland Ice Sheet (Hawkings et al. 2021). The Northern Hemisphere permafrost region represents a large store of Hg (Schuster et al. 2018, Lim et al. 2020; Schaefer et al. 2020). Thawing permafrost enhanced the THg and MeHg release in Arctic rivers (St Pierre et al. 2018), and has the potential to increase Hg concentrations in water and fish in the permafrost regions (Schaefer et al. 2020).

In summary, the Hg release from geogenic activities and the re-emission of legacy Hg stored in terrestrial systems and melting cryosphere impact the ecosystems across the globe. Specifically, the ongoing climate change will enhance the release of Hg from the cryosphere, which could offset, at least to some extent, the emission reduction efforts on anthropogenic emission sources. However, to fully address Question 2, the following critical questions remain to be answered: (1) How to estimate Hg re-emission fluxes from various natural surfaces? What are the important processes that determine Hg emissions and have we missed any major hot spots? (2) How to quantify the contribution of previously deposited Hg in Hg re-emission fluxes? How to determine the deposited Hg fate in terrestrial and marine ecosystems and their impact on biota Hg concentrations? and (3) How will the relevant climatic and environmental factors affect the Hg exchange among ecosystems, particularly in high-altitude and high-latitude cryosphere regions?

Q3 How are ecosystem Hg levels respond to changes in atmospheric deposition?

Atmospheric deposition is considered as the major driver for MeHg levels in the freshwater and marine ecosystems. This is confirmed by ecosystem-level studies (e.g., METAALICUS, Harris et al. 2007; Blanchfield et al. 2022) and by measurements of fish Hg levels across different spatial and temporal scales (Engstrom 2007). The METAALICUS study also suggests a quick recovery of MeHg concentrations in lake water and organisms when direct atmospheric depositions are decreased, although the degree of recovery varies at different trophic levels (Blanchfield et al. 2022). No direct observations are available for the marine environment, but Hg isotope data also confirm the atmospheric source of Hg in the surface ocean (Jiskra et al. 2021). This implies that reduction in human and wildlife MeHg exposure can be achieved by reducing anthropogenic emissions and subsequently atmospheric deposition, which is the scientific basis for the Minamata Convention (Krabbenhoft and Sunderland 2013). However, this does not mean synchronous variation trends of ecosystem Hg and atmospheric Hg deposition. Wang et al. (2019) reviewed long-term time-series data from ecosystems across the globe and found that fish Hg trends were often decoupled from atmospheric Hg trends, especially during the most recent decades, due to the influence of legacy Hg and biogeochemical and ecological processes that control its mobility, transformation (especially methylation), and biological uptake. Predicting future ecosystem Hg levels thus requires a comprehensive evaluation of the inventories and residence times of both "new" and legacy Hg, and environmental and ecosystem processes.

Future atmospheric Hg deposition trends are dependent on policy scenarios for Hg emission reduction. Pacyna et al. (2016) used two chemical transport models (GLEMOS, ECHMERIT) to evaluate future changes in Hg deposition in various geographic regions using three anthropogenic emissions scenarios of 2035, and projected that the 'new policy' and 'maximum feasible reduction' scenarios would result in 20–30% decrease in Hg deposition in all regions except South Asia. Lei et al. (2014) combined the effect of emissions changes and climate change through the CAM-Chem model using three emissions scenarios of 2050 (B1, A1B, A1FI), and projected a general increase in TGM around the globe under all the three scenarios due to an increasing use of fossil fuels. By integrating future changes in anthropogenic emissions, climate, and biogeochemical cycles into a three-dimensional atmosphere/ocean model and a two-dimensional land model, Zhang et al. (2021) projected future Hg deposition trends based on various policy scenarios. The results showed that the 'maximum feasible reduction' and 'new policy delayed' scenarios would reduce the atmospheric deposition in 2050 by 48% and 28%, respectively, compared to the 'current policy' scenario.

Q4 How does climate change affect Hg cycle in the ocean?

Globally humans are exposed to MeHg primarily via the consumption of seafood. Therefore, the ocean Hg cycle and its sensitivity to climate change are of particular importance to risk assessment of human health. The Arctic region is a hotspot for climate change. The rising temperature and increasing riverine input of dissolved organic carbon (DOC) have the potential to increase Hg methylation rate (Stern et al. 2012). Highly characteristic Hg isotope signatures reflect that Hg deposited to the cryosphere during spring can reemit back to the atmosphere contributing to the summer peak of atmospheric Hg concentrations (Araujo et al. 2022). The Hg export to the Arctic Ocean increases associated with enhancing permafrost thaw, glacier melt, and coastal erosion due to the warming surface air temperature (Chételat et al. 2022). Elemental Hg (Hg0) concentrations in the surface ocean might decline related to the enhancing air–sea exchange of Hg^0 and a lack of Hg^0 enrichment owing to contiguous ice (DiMento et al. 2019). Schartup et al. (2020), otherwise, found an increase of mono-methylmercury (MeHg) in sea ice during a regime shift from multi-year sea ice to seasonal ice indicating a higher MeHg exposure of sea ice-related biota (Schartup et al. 2020). However, the overall trends are the result of many competing processes and hard to discern at ecosystem levels. For instance, it was reported that the reduced sea ice cover in the Bering Sea was associated with decreased sea bird egg MeHg levels, indicating a dominant role of enhanced photodemethylation (Point et al. 2011). Lower Hg concentrations were observed in the predators with reduced sea ice-based foraging (McKinney et al. 2022).

Studies of Hg in the Southern Ocean are scarcer than their Arctic counterpart. There is a seasonal variation of mercury species in the polar sea ice environment, but the cause is still ambiguous including the influence of solar radiation, temperature, brine volume, and atmospheric deposition (Nerentorp Mastromonaco et al. 2016). Lower gaseous oxidized mercury (GOM) content has been recently found in the Antarctic sea ice region, which was attributed to the greater uptake by sea salt aerosols (Yue et al. 2021). Despite the limited study of the Hg cycle in the Southern Ocean, the effect of climate change on the Southern Ocean is probably distinct compared to that on the Arctic Ocean. Unlike its counterpart, sea ice extent in the Southern Ocean has been found a slowly increasing trend based on the satellite record that began in 1979 (Meehl et al. 2016). Therefore, the oceanic Hg cycle in the Southern Ocean affected by climate change should be remarkably different.

At a global scale, methylmercury can be biomagnified over 7–9 orders of magnitude in fish and marine mammals

compared to the seawater concentrations in the marine food webs. It was shown that nutrient supply was strongly linked to the bioavailability and trophic transfer of MeHg in marine ecosystems (Driscoll et al. 2012). Lee and Fisher (2016) reported an active uptake of MeHg by phytoplankton cells and found that the volume concentration factors (VCF) were inversely correlated with the radius of the cells. Schartup et al. (2018) carried out a quantitative study of the food web dynamics of MeHg involving different trophic levels of plankton (e.g., phytoplankton, copepod, and krill) and identified DOC, the growth dilution, and grazing rates as key factors in MeHg uptake in marine ecosystems. Their subsequent study incorporating long-term observation data and ecosystem modeling found seawater temperature and dietary shifts as important influencing factors (Schartup et al. 2019). When studying the MeHg bioaccumulation and food web dynamics in the global marine plankton ecosystem, Zhang et al. (2020) found a competing effect between the phytoplankton size and seawater MeHg concentrations on phytoplankton MeHg, and the biomagnification from phytoplankton to herbivorous zooplankton was determined by the zooplankton biomass. Wu et al. (2020, 2021) extended the bioaccumulation of MeHg in the plankton food webs to more trophic levels and found that trophic dilution occurs for food webs involving small zooplankton and that assimilation efficiency was a particularly sensitive factor for MeHg biomagnification. They also found a connection between the slope of trophic pyramid and MeHg biomagnification.

Although the links between climate change and the marine Hg cycle have become increasingly evident (e.g., Stern et al. 2012), the net effects remain unclear on the global scale, especially because of synergetic and antagonistic effects of multiple factors that operate simultaneously (Krabbenhoft and Sunderland 2013). The fact that future projections of many fundamental marine biogeochemical processes such as ocean warming, acidification, productivity, and export varying between models add another layer of uncertainty (Dutkiewicz et al. 2015). As a first step, Zhang et al. (2021) used a 3D ocean general circulation model to project Hg based on ocean biogeochemical parameters generated from an Earth System Model. Their results revealed large regional variabilities in a changing climate and highlighted the importance of plankton community structure. Future studies are clearly needed to better parameterize the linkage between the Hg cycle and ocean biogeochemistry in a changing climate.

Q5 Can Hg isotopic composition be used as a new tool for tracing Hg source, transport, and surface exchange in complex and changing environments? To trace the environmental recovery and human benefit in future, are there other methods we could use? How can these methods be integrated together to reduce the uncertainties?

Mercury isotopes can undergo mass-dependent fractionation (MDF), as well as odd- and even-mass independent fractionation (MIF) in the environment. The MDF of Hg isotopes (e.g., δ^{202}Hg signature) can be induced by many environmental processes, whereas significant MIF of odd-mass Hg isotopes (Δ^{199}Hg and Δ^{201}Hg signatures) is mainly associated with photochemical redox reactions (Bergquist and Blum 2007; Jiskra et al. 2012; Kritee et al. 2009; Rodriguez-Gonzalez et al. 2009; Wiederhold et al. 2010; Zheng et al. 2007; Zheng and Hintelmann 2009). Measurable even-Hg MIF anomalies (e.g., Δ^{200}Hg and Δ^{204}Hg signatures) are mainly observed in atmospheric samples, with GEM and oxidized Hg forms in precipitation, cloud water, air particles, and gaseous oxidized mercury (GOM) universally characterized by negative and positive Δ^{200}Hg values, respectively (Chen et al. 2012; Fu et al. 2021a, 2019b; Washburn et al. 2021). The mechanisms that induce even-MIF of Hg isotopes are related to atmospheric photochemical redox reactions at high altitudes (Chen et al. 2012; Fu et al. 2021a; Sun et al. 2016a, 2017).

Mercury stable isotopes are used to track Hg sources and transformation processes in environment. GEM, the dominant Hg form in the atmosphere, in remote regions worldwide is generally characterized by positive δ^{202}Hg values (mean of $0.52 \pm 0.38‰$ with site-specific mean values ranging from -0.21 to $1.19‰$) and negative Δ^{199}Hg (mean of $-0.19 \pm 0.06‰$ with site-specific mean values ranging from -0.29 to $-0.08‰$) and Δ^{200}Hg (mean of $-0.06 \pm 0.03‰$ with site-specific mean values ranging from -0.10 to $0.00‰$) values (Fig. 1; Demers et al. 2013; Fu et al. 2018, 2019a, 2016b; Kurz et al. 2020; Gratz et al. 2010; Nguyen et al. 2021; Enrico et al. 2016). These values are clearly distinguishable from those observed in urban areas worldwide (mean δ^{202}Hg = $-0.54‰$, mean Δ^{199}Hg = $-0.04‰$, mean Δ^{200}Hg = $0.00‰$) (Fig. 1; Demers et al. 2015; Fu et al. 2021b; Xu et al. 2017; Yu et al. 2016). The large variations in GEM isotopic compositions in the global atmosphere are thought to be caused by a combined effect of anthropogenic emissions, foliar uptake, and atmospheric oxidations (Fu et al. 2018, 2019a, 2016b). In addition, soil GEM emissions are generally characterized by negative and slightly positive δ^{202}Hg (means = -4.49 to $1.33‰$), negative to positive Δ^{199}Hg (means = -0.30 to $0.71‰$), and near-zero Δ^{200}Hg values, which is thought to have contributed significantly to the negative GEM δ^{202}Hg shift in the urban atmosphere (Fu et al. 2021b; Zhang et al. 2020; Zhu et al. 2022). On the other hand, observations of oxidized Hg forms in the atmosphere including GOM, particulate-bound Hg (PBM), precipitation, and cloud water Hg showed opposite Hg MIF isotopic signatures (e.g., overall positive Δ^{199}Hg (means: -0.12 to $0.83‰$) and Δ^{200}Hg (means: 0.02 to $0.14‰$) values) (Fu et al. 2021a, 2019b; Guo et al. 2021; Kwon et al. 2020; Qiu et al. 2021; Washburn et al. 2021). It is mainly

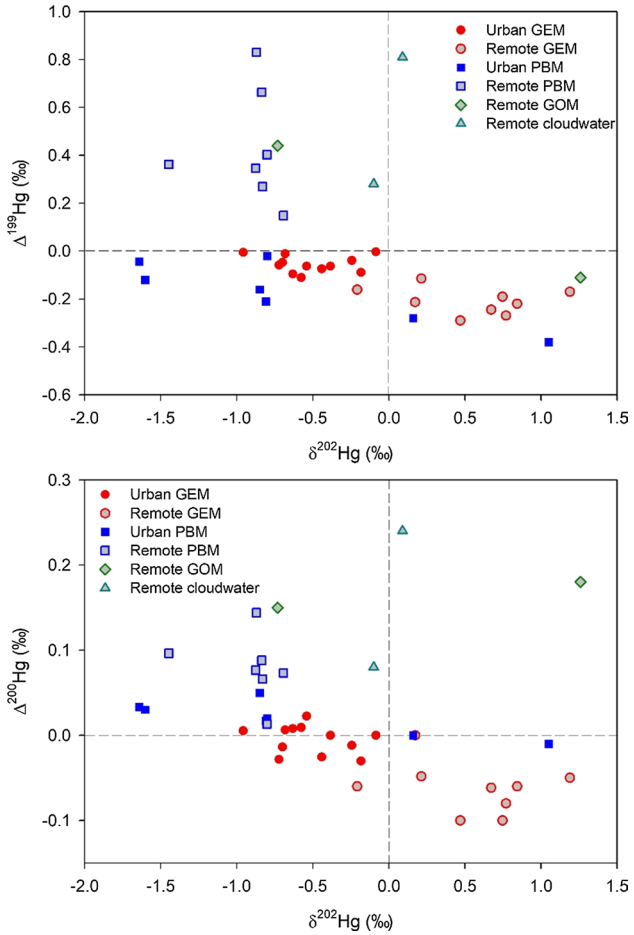


Fig. 1 Site-specific mean δ^{202}Hg, Δ^{199}Hg, and Δ^{200}Hg values of atmospheric gaseous elemental mercury (GEM), particulate-bound mercury (PBM), gaseous oxidized mercury (GOM), and cloud water at global sites

caused by atmospheric redox reactions in the gas phase and in aerosols and cloud droplets (Fu et al. 2021a; Huang et al. 2019).

With MIF signatures between GEM and oxidized Hg, it has been proposed that dry deposition of GEM constitutes a dominant fraction (50–92%) of Hg loading to forests, peat bogs, and Arctic tundra (Enrico et al. 2016; Obrist et al. 2017; Wang et al. 2020a, b). Jiskra et al. (2021) found that Hg in seawater was probably largely derived from the GEM dry deposition. It suggest that the GEM dry depositions to the Earth surface are likely underestimated (Enrico et al. 2016; Obrist et al. 2017; Wang et al. 2020a, b; Jiskra et al. 2021; Fu et al. 2021a). Either GEM is more readily removed from the atmosphere than previously thought, or our understanding of bi-directional exchange processes and rates of Hg(0) between the atmosphere and the Earth surfaces is incomplete (Holmes et al. 2010; Horowitz et al. 2017; Jiskra et al. 2021). Mercury stable isotopes have been also used to track re-emissions of Hg from organic soils and foliage (Jiskra et al. 2015; Yuan et al. 2019, 2021). Jiskra et al. (2015) calculated an approximately 28% loss of the previously deposited Hg in forest soils due to re-emissions in a Northern Sweden boreal forest. A recent study in a subtropical evergreen forest estimated that the re-emission flux of Hg from forest floors exceeded that of gross GEM depositions to forest soils, indicating that subtropical forest soils are an net sources of GEM (Yuan et al. 2021). Foliage is generally regarded as a net sink of GEM (Fu et al. 2016a); however, simultaneous re-emissions of Hg from foliage during foliar GEM uptake have estimated to be 30% on average to the GEM depositions to foliage (Yuan et al. 2019).

Lake sediments in remote areas provide records of historical variations in the atmospheric Hg deposition. Many previous studies have observed increased Hg accumulation in lake sediments since the industrialization, which is mostly likely attributed to the increasing anthropogenic atmospheric Hg emissions (Kang et al. 2016; Lepak et al. 2020; Outridge et al. 2007; Yin et al. 2016). Interestingly, the increase of sedimentary Hg accumulation in most regions is accompanied by a positive shift of δ^{202}Hg and Δ^{199}Hg in sediments, as those compiled by Lee et al. (2021). Currently, the mechanisms associated with the continuous increase of δ^{202}Hg and Δ^{199}Hg in sediments are not well clear. The increase of δ^{202}Hg was proposed to be partly caused by the δ^{202}Hg changing patterns of anthropogenic Hg emissions (Lee et al. 2021; Lepak et al. 2020), and the δ^{202}Hg values of which were estimated to increase since the industrialization because of the changes of anthropogenic source sectors (Sun et al. 2016b). The increase of Δ^{199}Hg was speculated to be related to increasing precipitation Hg deposition and accelerated photoreduction of Hg(II) in lake waters. Generally, the historical variations in the sedimentary Hg isotopic signatures should reflect isotopic signatures of atmospheric Hg deposition, especially under the condition that the lake is in a stead state and the input of Hg from the atmosphere is considerably large when compared with the Hg inventory in the lake (e.g., shallow lakes). Therefore, the sedimentary records should probably reflect similar changes in the isotopic compositions of atmospheric Hg deposition as those observed in lake sediments (Lepak et al. 2020). Numerous studies have defined the baseline Hg isotopic signatures in sediments before the industrialization (Lee et al. 2021). Hence, further studies to compare Hg isotope signatures between preindustrial and present-day sediments would help to evaluate the recovery of atmospheric Hg depositions and emissions.

In addition, Hg isotopes have been used to identify human Hg exposure sources and metabolic processes (Laffont et al. 2009, 2011; Li et al. 2014; Du et al. 2018; Sherman et al. 2013; Rothenberg et al. 2017). For example, a significant MDF of ~ 2‰ for δ^{202}Hg and no MIF between human hair

and fish diet have been reported (Laffont et al. 2009, 2011). A binary mixing model based on Δ^{199}Hg has been used to quantify human exposure from fish and rice consumption (Du et al. 2018). However, Hg isotope study on Hg biogeochemical cycle and human Hg exposure can be integrated together, which finally evaluate the ecosystem mercury recovery and health benefit.

Since Hg isotopes in the atmosphere and the Earth surfaces are controlled by many processes, a combination of Hg isotopes, Hg mass balance, and Hg cycling approaches would improve our understanding of Hg sources and transformation in the environment using Hg isotopes. Several studies have already accommodated the stable Hg isotopes in the global Hg chemistry and transport models (Sonke 2011; Sun et al. 2019; Song et al. 2022). However, the modeling results based on published Hg isotope fractionation enrichments factors (the standard model) seem to be inconsistent with the observations, and such a discrepancy is likely caused by our incomplete understanding of the fractionation of Hg isotopes in the environment and the transport and transformation of Hg in different Earth reservoirs (Sun et al. 2019). Therefore, further studies on isotope compositions of natural and anthropogenic Hg emissions and Hg species in various Hg reservoirs (e.g., atmosphere, water, soil, biota) as well as stable Hg isotope fractionation during environmental processes are needed, which will help to improve our understanding of Hg cycling in the environment and evaluate the effectiveness of Minamata Convention.

Data are adopted from the following: Das et al. 2016; Demers et al. 2013; Demers et al. 2015; Enrico et al. 2016; Fu et al. 2021b; Fu et al. 2018; Fu et al. 2019a; Fu et al. 2016b; Fu et al. 2019b; Gratz et al. 2010; Guo et al. 2021; Huang et al. 2016; Kurz et al. 2020; Nguyen et al. 2021; Qiu et al. 2021; Rolison et al. 2013; Washburn et al. 2021; Xu et al. 2017; Xu et al. 2019; Yu et al. 2016; Zheng et al. 2018.

Future Assessment

Q6 How far away are we from having a process-based integrated global Hg model? How well are these models constrained by observational data? How well do these models attribute the changes in ecosystem and human beings to the change of Hg emissions and release?

The construction of a state-of-the-art integrated global Hg model first needs a dynamic and online biogeochemical cycling simulation of Hg which couples multiple environmental media. A majority of three-dimensional (3D) models have been developed for the simulation of atmospheric Hg, including regional models (e.g., CAMx, CMAQ-Hg, STEM-Hg, WRF-Chem-Hg) and global models (e.g., GEOS-Chem-Hg, GLEMOS, GRAHM, ECHMERIT). Processes such as emission, transport, and deposition of atmospherically speciated Hg have been simulated with different horizontal and vertical resolutions in these models. Compared to atmospheric Hg, process-based models are limited for the simulation of Hg in soils and oceans. Smith-Downey et al. (2010) developed a mechanistic global model of soil Hg (i.e., GTMM) to simulate global distribution of soil Hg storage and emissions. Zhang et al. (2015) developed oceanic Hg simulation in the MIT general circulation model (MITgcm) to investigate the air–sea exchange of Hg and source contribution of rivers. Importantly, the lifetime of Hg is very different among different environmental media, ranging from several months in the atmosphere to millennia in the deep ocean and armored soil (Amos et al. 2013). The long lifetime of Hg in soils and oceans leads to significant legacy impacts of historical Hg activities on present Hg conditions (Amos et al. 2013), which may influence ecosystem Hg recovery.

The great differences in lifetime result in difficulties to couple existing models to achieve a dynamic simulation of biogeochemical Hg cycling among environmental media. Off-line boundary conditions are a common practice presently for the bi-directional exchange processes among environmental media (Horowitz et al. 2017; Shah et al. 2021). However, this practice restricts the evaluation of the legacy impacts of historical Hg activities on present Hg conditions and the subsequent assessment of ecosystem Hg recovery. Thus, a dynamic online biogeochemical cycling simulation of Hg which couples multiple environmental media is urgently needed. The modeling representation of biogeochemical Hg cycling in Earth system models (e.g., CESM) may be an alternative approach in future studies. The Earth system models couple the components of the atmosphere, land surface, ocean, and sea ice in a changing climate (Lehner et al. 2015; Wieder et al. 2021), and these components are to be coupled without resorting to any "flux adjustments." Meanwhile, long-term simulation can be conducted in these models accounting for the legacy impacts of trace elements, and the simulation of Earth's future climate states allows the assessment of ecosystem Hg recovery in a changing climate.

The construction of a fully coupled Hg simulation in Earth system models requires accurate representations of key processes at the interface among different environmental media, such as air–sea exchange, air–foliage exchange and air–soil exchange. Air–sea fluxes of GEM are observed by flux chambers (Floreani et al. 2019; Gardfeldt et al. 2001) and simulated by micrometeorological methods (Nightingale et al. 2000; Soerensen et al. 2010; Osterwalder et al. 2021), indicating low invasion of elemental Hg. However, high invasion of elemental Hg is confirmed by isotopic evidence in a recent study (Jiskra et al. 2021). The controversy indicates our knowledge gap on the air–sea exchange of Hg. Foliage-induced dry deposition of elemental Hg is simulated by the resistance-in-series model, accounting for

aerodynamic, quasilaminar, stomatal, in-canopy aerodynamic, ground, and cuticular resistances (Selin et al. 2008; Wesely 1989; Wright and Zhang 2015). However, higher stomatal resistances of elemental Hg (uptake of elemental Hg by foliage) have been confirmed by recent studies (Jiskra et al. 2018; Zhou et al. 2021), suggesting the improvement of the representation of stomatal resistances in the resistance-in-series model.

Besides the dynamic biogeochemical cycling simulation in Earth's system models, a state-of-the-art integrated model should also involve food web models and assessment models of human exposure risk. With the help of these models, the integrated model can attribute the changes in human exposure to the changes of Hg emissions and releases. Several studies have coupled atmospheric models to assessment models of human exposure risk to investigate the relationship (Chen et al. 2019; Giang and Selin 2016; Zhang et al. 2017). However, the lack of food web models in the studies led to ineluctable uncertainties of their results. Zhang et al. (2021) improved the model framework by introducing a 3D oceanic Hg simulation and a low-level food web along with coupled atmospheric and terrestrial simulation to investigate the impacts of future emission changes on human health. Modeling Hg cycling in comprehensive and systematic food webs has been still deficient, which hampers the accurate assessment of human exposure risk and the constriction of an integrated model. In addition, uncertainties still exist for Hg-related health impact assessments. Besides fetal IQ loss and adult myocardial infarction which possess dose–response relationships based on epidemiological surveys, other health problems such as metabolic syndrome (Zhang et al. 2020), hearing impairment (Hemmativaghef 2020), and immunotoxin impairments (Abu Zeid et al. 2021) are revealed to be associated with Hg exposure. However, these health impacts have still lacked dose–response relationships. The consideration of comprehensive health impacts is needed for the state-of-the-art integrated model.

A state-of-the-art integrated model should be constrained by various types of observations which are conducted in a long-term period. The types include atmosphere, soil, ocean, vegetation, sediment, biota, etc. In fact, Hg in various types of samples has been measured in numerous studies (UNEP 2019). However, the combination of the various types of observations and associated effectiveness has not been evaluated in an integrated model framework. Meanwhile, long-term observations are of much importance accounting for the legacy impacts of Hg in soils and oceans, which assists the integrated model to assess the ecosystem Hg recovery.

Q7 How do different social-economic pathways and mitigation options affect the ecosystem and human health?

There is no linear relationship between regulatory activities and changes of human exposure to Hg, as best demonstrated by case studies in the Arctic and elsewhere (UNEP 2019). One way to quantify the relationship between regulatory activities and changes of human Hg exposure is to integrate Earth model with social-economic system, which requires the full sequence information all the way through regulatory activities, emission projection, environmental transport and inter-compartmental migration, and mercury bioaccumulation, to human exposure and health (Fig. 2). A recently published analysis of Hg concentrations in blood and urine samples of pregnant women and children in the United States collected during 1999–2016 shows an increase in organic Hg (So et al. 2021), although Hg mitigation and adaption measures have been taken in the United States in the past decades and atmospheric Hg concentrations have

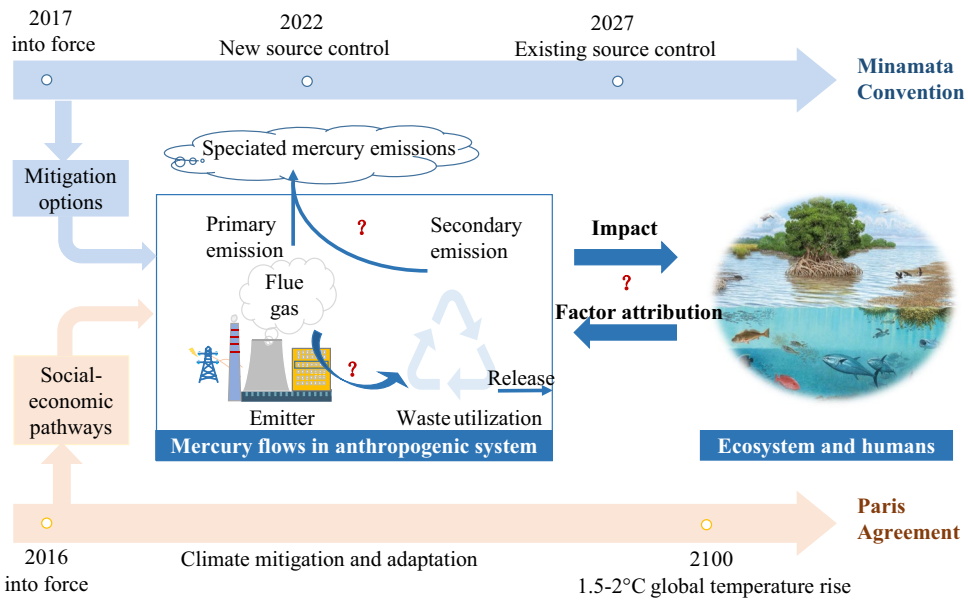


Fig. 2 Roadmap of regulatory activities and ecosystem recovery and human Hg exposure

kept decreasing (Zhang et al. 2016). Thus, we can expect that even substantial anthropogenic Hg emission reduction can result in low abatements or increase of Hg in ecosystem or human beings if the background Hg concentrations in atmosphere and global ocean are not stabilized (Wang et al. 2019). The scientific community has pointed out that aggressive regulatory activities should be taken as soon as possible to stabilize Hg cycle and to lower the adverse impact of climate change on human health (Amos et al. 2013; Angot et al. 2018; Krabbenhoft and Sunderland 2013; Wang et al. 2010, 2019).

However, future emission trajectories are highly uncertain and depend on changes both in the global social-economic pathways and in the mitigation options. National commitments under the Paris Agreement on climate change will also interact with the Minamata Convention on Mercury. Actions to reduce CO_2 emissions including the improvement of efficiency of energy production and usage and fossil fuels substitution with renewable sources are likely to have a positive impact on the reduction of anthropogenic Hg emissions (Pacyna et al. 2016; Streets et al. 2009; Wang et al. 2012). Several studies have projected global or regional atmospheric Hg emissions (Chakraborty et al. 2013; Mulvaney et al. 2020; Pacyna et al. 2016; Rafaj et al. 2013; Streets et al. 2009; Wu et al. 2018a, 2018b). Although the scenario assumptions on activity level and technology penetration vary across these studies, the differences in results are mainly caused by the assumptions on the degree of control technologies and the projected activity levels output from existing models such as the energy-related model. Therefore, the impact from the drivers of the social-economic pathways (e.g., detailed energy technologies, secondary metal production) on the Hg emissions is scarcely analyzed.

Moreover, the global social-economic pathways and mitigation options will also impact the relative emissions of different Hg species (Rafaj et al. 2013; Wu et al. 2018a), which is important for modeling the Hg behaviors in the environment. Future Hg speciation profiles are very sensitive to Hg sources, combustion and production conditions, and penetration technologies. In future, the detailed point source information collected by parties will aid to the determination of Hg speciation profiles. However, the uncertainty of new technologies on Hg behaviors in sources and their penetration situation will challenge the projection of speciated Hg emissions.

Recent advances on Hg flow studies indicate secondary anthropogenic Hg emissions and Hg releases to water and soil due to the use of wastes contaminated by Hg (Hui et al. 2017; Ouyang et al. 2021; Sundseth et al. 2012; Wu et al. 2015, 2016). Reducing the use of fossil fuel and promoting the waste safe disposal will promote the search of energy and resources from wastes. It is quite possible that Hg embodied in the historical waste streams will be activated and new sources will be embodied (Chakraborty et al. 2013). Besides, with the control of convention-related emission sources, the sectoral distribution of Hg emissions will be changed in future (Streets et al. 2009). Moreover, Hg control technology cannot eliminate Hg from the anthropogenic activities unless it is sealed or emitted/released to the environment. Therefore, it is easy for Hg to flow from one medium to another medium and the countermeasures sometimes conflict. Therefore, it is urgent to switch single medium Hg emission control to cross-media and cross-industries Hg flow management globally. In future, the impact of different social-economic pathways and mitigation options on global Hg flows requires future studies to provide whole-process control strategies.

Perspective and Recommendation

In this paper, we synthesized research progresses with respect to 7 scientific issues on ecosystem Hg recovery and health benefit under the Minamata Convention on Mercury in a changing climate. Declines of atmospheric Hg concentrations in Europe and North America can be clearly attributed to the decline of anthropogenic Hg emissions, whereas atmospheric Hg trends in China are more complex but generally agree with emission trends. The Hg concentrations in aquatic biota mostly decreased since the 1970s in North America, North Europe, and Arctic. However, significant increases were observed over recent decades in North America and the Arctic. Data on human exposure are severely lacking, with limited data showing significant declines from 1966 to 2015. It is difficult to set the "background" or "safe" Hg concentrations in environmental and biota matrixes. Biomonitoring in multiple species in different regions of the world is needed to evaluate the effectiveness of the Minamata Convention.

Large uncertainties exist with respect to the fluxes of natural emissions from geogenic activities and re-emissions of legacy Hg stored in the terrestrial surfaces. Glaciers, seasonal snow cover, permafrost, and sea/lake ice will continually liberate legacy Hg into the atmosphere and aquatic environments in a warming climate. The reduction in human and wildlife MeHg exposure can be ultimately achieved by reducing anthropogenic emissions; however, the former is expected to significantly lag behind the latter. Global biogeochemical cycling of Hg will be affected by rapid climate change, which remains a major uncertainty when evaluating the effectiveness of regulatory activities.

The combination of Hg isotopes, Hg mass balance, and Hg cycling approaches will improve the understanding of Hg sources and transformation in the environment. It is critical for identification of legacy Hg sources to decipher signals due to policy changes versus natural variations. The

construction of a state-of-the-art integrated Earth system model requires accurate understanding of key processes at the interface among different environmental media, which should also involve in food web model and assessment models of human exposure risk. To quantify the relationship between regulatory activities and changes of human Hg exposure, it is needed to integrate Earth system model with social-economic system, which requires the full sequence information of global biogeochemical cycle of Hg. However, future emission trajectories are highly uncertain and depend on changes both in the global social-economic pathways and in the mitigation options, such as the impacts from the Paris Agreement on climate change. The impact of different social-economic pathways and mitigation options on global Hg flows requires future studies to provide whole-process control strategies.

Systematic studies on global biogeochemical cycle of Hg (anthropogenic and natural emission, environmental transport and migration, Hg bioaccumulation, human exposure, and health effects) are urgently needed. Major factors/processes affecting ecosystem Hg recovery and human health such as natural re-emissions and climate changes should be better understood. A variety of new methods such as Hg stable isotopes, integrated Earth system models, and social-economic system should be coupled to attribute the changes in ecosystem recovery and human health benefits to the change of Hg emissions and releases and to evaluate the effectiveness of the Minamata Convention.

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Data availability The datasets generated during and/or analysed during the current study are available from the corresponding author on reasonable request.

Declarations

Conflict of interest On behalf of all authors, the corresponding author states that there is no conflict of interest.

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