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Viewpoint

# Developing Methylmercury-Targeted Strategies to Safeguard Rice Consumers

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**ABSTRACT:** Mitigating mercury (Hg) risk in the rice-paddy system is crucial for safeguarding food safety and human health, as rice is a main source of human exposure to neurotoxic methylmercury (MeHg). Current mitigation strategies predominantly focus on reducing the availability of inorganic Hg (IHg) for Hg methylation, achieved primarily through Hg emission control and *in situ* Hg immobilization. While these IHg-targeted approaches have effectively reduced MeHg bioaccumulation and subsequent human exposure, their efficacy is largely undermined by Hg transformations and fluctuating environmental conditions due to the complex and protracted pathway linking IHg from environmental sources to MeHg at the point of human exposure. In light of recent advancements in MeHg-related transformations, we emphasize the development of MeHg-targeted strategies to improve the overall efficiency of Hg risk management in rice-paddy systems. MeHg-targeted strategies include microbial regulation to diminish net MeHg production, facilitating MeHg demethylation in soils, and promoting the *in vivo* MeHg degradation within rice plants. Although these approaches are still in their nascent stages, they hold significant promise due to their potential high mitigation efficacy and reduced uncertainties, owing to the shorter pathway between MeHg production and human exposure. Integrating IHg- and MeHg-targeted strategies offers a comprehensive and synergistic approach, paving the way for more effective mitigation of human exposure to MeHg in rice-paddy systems.

## INTRODUCTION

Methylmercury (MeHg), renowned as the pollutant causing the Minamata disease, has been a global concern for decades due to its potent neurotoxicity. Despite constituting a minor proportion in natural environments, typically less than 2% of the total Hg (THg) in soils and sediments,[1] MeHg poses the greatest threat to humans compared to other Hg species. In natural environments, MeHg is predominantly produced through the microbial methylation of divalent inorganic Hg (IHg), which primarily originates from parent materials and atmospheric deposition. Upon emission from both natural and anthropogenic sources, Hg undergoes long-distance atmospheric transport in the form of gaseous elemental Hg ($Hg^0$). The oxidation of $Hg^0$ to IHg facilitates Hg deposition onto surface soils and waters, where it becomes rapidly immobilized through the aging process. Only a small fraction of legacy Hg remains bioavailable for microbial methylation, after which MeHg bioaccumulates and magnifies along food chains, ultimately reaching humans (Figure 1). These interconnected processes underpin the global mercury cycle and its flux to human diets.

The ongoing mitigation strategies focus on reducing the IHg availability for methylation, as IHg serves as the precursor of MeHg formation. This goal is primarily pursued through Hg emission control and *in situ* IHg immobilization, with the former being a regional or global policy while the latter being a site-specific approach. The Minamata Convention, ratified by 151 parties currently, aims to phase out the use of mercury and has significantly contributed to reducing anthropogenic Hg emissions. For instance, China, the largest emitter of anthropogenic Hg, has achieved a reduction in annual Hg emissions from approximately 510 t in 2010 to about 360 t in 2020, making a 30% decrease within a decade.[2] As a result, the atmospheric Hg levels and Hg depositions to the earth's surface have also been observed to decline.[3,4] These efforts to curb Hg emissions hold promise for a decreasing trend in MeHg bioaccumulation.[5] In addition to Hg emission control, *in situ* Hg immobilization is another prevalent approach to mitigate Hg risks. The principle of this approach is amending specific agents to reduce Hg mobility and its subsequent uptake and methylation by microorganisms. Among various immobilization agents, biochar[6] and selenium (Se)[7] have been widely reported to mitigate MeHg bioaccumulation. For example, the application of Se at a dose of 6 mg/kg was reported to decrease MeHg accumulation in rice grains by 55% in a pot experiment.[8]

## CHALLENGES OF TRADITIONAL MITIGATION APPROACHES

The effectiveness of our efforts to mitigate Hg risks by reducing anthropogenic emissions has been lower than anticipated. It is estimated that the changes in human Hg

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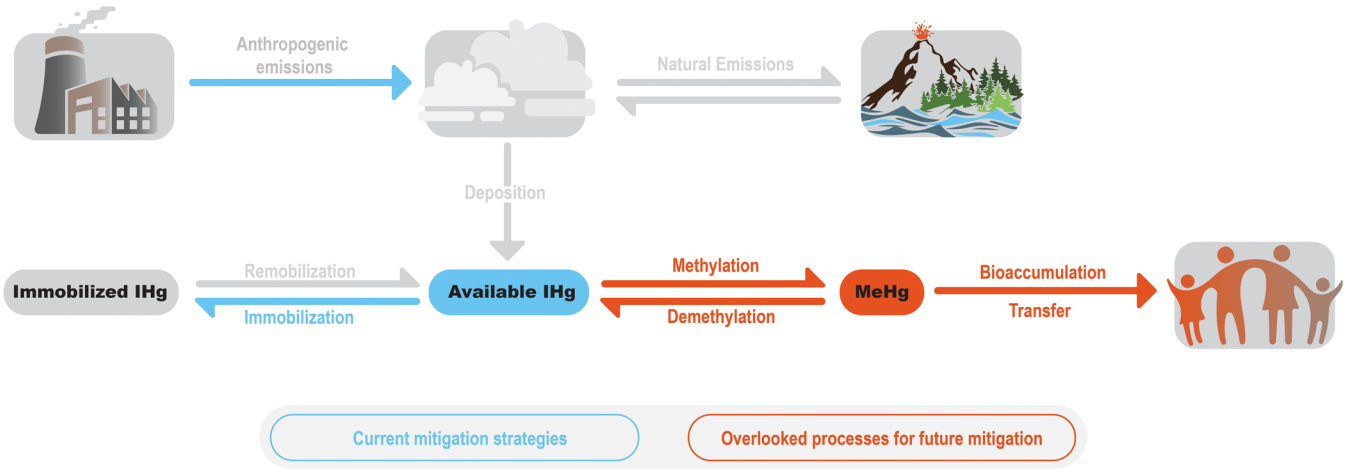


**Figure 1.** How Hg makes its way from environments to humans.

exposure represent only 1/3−1/2 of the reductions in Hg emissions.[9] Such limited efficacy and sensitivity can be partially attributed to the complex and protracted pathway between Hg emissions and human exposure to MeHg (Figure 1), which also results in substantial uncertainties in predicting changes in MeHg accumulation following the implementation of Hg emission control measures. The long-distance transport of emitted Hg in the atmosphere, diverse Hg transformation processes in natural environments, and prolonged MeHg accumulation in organisms collectively contribute to this delayed and attenuated response.[5] Additionally, while newly deposited Hg exhibits higher bioavailability for methylation compared to aged Hg, it undergoes rapid aging upon entering soils, reducing its potential for Hg methylation and subsequent bioaccumulation. Further complicating these dynamics is the remobilization of legacy Hg under changing environmental conditions, which can contribute to Hg methylation (Figure 1).[10] This remobilized Hg undermines the progress made by IHg-targeted measures and highlights the need for more comprehensive strategies to address Hg risks effectively.

Worse still, the effectiveness of Hg emission control is anticipated to decline in the forthcoming years. This decline is primarily attributed to the substantial pool of natural Hg emissions, which constitute approximately half of the annual global Hg emissions. Along with the fulfillment of the Convention, the proportion of anthropogenic Hg emissions would keep going down. Consequently, even a moderate reduction in anthropogenic Hg emission will have a limited impact on atmospheric Hg levels and subsequent Hg deposition in the future. Furthermore, the significant decreases in anthropogenic Hg emissions have been primarily achieved through the rapid installation of air pollution control devices (ACPDs), which typically exhibit removal efficiencies lower than 90%.[2] This suggests that future Hg emission control will increasingly rely on elevating the Hg removal efficiency rather than solely deploying additional ACPDs. However, the dedicated Hg removal technologies, which offer removal rates exceeding 97%, are costly, about 2 to 5 times higher than the ACPDs.[11] These limitations in Hg removal devices and technologies suggest that Hg emission control may encounter significant challenges in the future.

Similarly, the immobilization of Hg using various agents has its own limitations. Commonly used immobilization agents, namely biochar or activated carbon, have been frequently reported to enhance Hg methylation by stimulating the growth of the microbial Hg methylators.[12,13] For example, applying biochar at a dose of 1% has been shown to elevate MeHg concentration in soils by 100%.[12] While the addition of biochar could temporarily inhibit the phytoavailability of MeHg, this fraction can persist in soils and become phytoavailable again upon soil drainage. This poses a particular concern in crop rotation systems where maize or wheat are planted following rice harvest.[14] In addition to biochar, selenium (Se) is a natural antagonist factor of Hg as it can bind Hg to form inert HgSe compounds.[15,16] High doses of Se, ranging from 3 to 500 mg/kg, have effectively reduced MeHg production.[7,15,17] However, such high Se doses may pose Se risks due to the narrow range of Se-rich and Se-contaminated soils (0.4−3 mg/kg). As a result, there are currently no effective yet environment-friendly agents for Hg mitigation, particularly for nonmining impacted paddy fields. Moreover, the immobilized Hg could be readily remobilized in changing environments. Regular farming activities, such as straw return and fertilization, have been reported to mobilize Hg and increase net MeHg production.[10,18] This suggests that immobilizing Hg to inhibit Hg methylation and the following MeHg bioaccumulation may not be an effective strategy in environments that are heavily impacted by human activities. Therefore, there remains a pressing need to develop effective strategies to mitigate Hg risks.

## EXPENDING MITIGATION FOCUS BEYOND IHG TO INCLUDE MEHG

Here, we advocate expanding our focus on risk mitigation beyond IHg to include MeHg in the future. Historically, our understanding of Hg risks and subsequent mitigation strategies have predominantly concentrated on IHg, which has led to the development of IHg-targeted approaches, such as Hg emission control and *in situ* Hg immobilization. However, the scope of potential strategies extends well beyond these conventional methods. As MeHg is the primary form of Hg absorbed by humans, the pathway linking MeHg to human exposure is considerably shorter than that of IHg (Figure 1). The shorter pathway suggests high efficiency and reduced uncertainty in predicting changes in human exposure risks when targeting MeHg concentrations for regulation.

The major obstacle to developing MeHg-targeted mitigation strategies is our insufficient understanding of Hg biogeochem-


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ical cycling, particularly aspects related to MeHg. Significant knowledge gaps persist regarding the dominant microorganisms that control Hg methylation, the mechanisms and drivers of MeHg demethylation, and the potential transformations of MeHg following bioaccumulation. Specifically, although sulfate-reducing bacteria (SRB), iron-reducing bacteria (FeRB), and methanogens have been identified as the key groups harboring Hg methylation gene *hgcA*, it is not clear which group is mainly responsible for the production of MeHg in paddy soils. Furthermore, these three groups are also implicated in MeHg demethylation,[19] yet how these microorganisms contribute to these two opposite processes and, thus, the net production of MeHg remains unknown. In addition to microbial drivers, abiotic factors may play a previously overlooked role in MeHg degradation. For instance, minerals containing Cu, Fe, or Mn have been reported to be involved in MeHg degradation in soils and sediments,[20−22] yet their contribution to net MeHg accumulation in natural environments is still elusive. Meanwhile, the existence of other abiotic demethylation pathways in soils has yet to be explored. Furthermore, for a long period, MeHg was deemed as nondegradable once entering food chains, despite some pioneer studies revealing that Se-containing protein could mediate MeHg degradation in fish.

## RECENT ADVANCEMENTS FACILITATING THE DEVELOPMENT OF MEHG-TARGETED STRATEGIES

Recently, some advancements in MeHg transformations have been achieved, expanding our understanding of how MeHg makes its way from the environment to our dining table, especially in the rice-paddy systems. These advancements include the identification of the dominant microorganisms governing net MeHg production in paddy soils and the discovery of a reactive oxygen species (ROS)-mediated MeHg demethylation pathway in rice plants, as introduced below.

Recent research has revealed that *hgcA*-containing *Geobacteraceae*, namely FeRB, are mainly responsible for net MeHg production in paddy soils. This finding stemmed from a national-scale study encompassing 67 paddy soils across a latitudinal span of 3,600 km. The dominant role of *hgcA*-containing *Geobacteraceae* was established based on 3 pieces of evidence: their highest relative abundance on a national scale (50%), the strongest correlation between their relative abundance and Hg methylation potential (MeHg%), and a significant correlation between iron reduction and MeHg concentration.[23] Although the mechanisms underlying FeRB dominance in net MeHg production remain elusive, it is plausible that FeRB are involved in regulating the availability of IHg, which is the limiting factor of Hg methylation in paddy soils.[10] Additionally, FeRB may play a role in MeHg degradation, acting as both microbial methylators and abiotic driving factors (discussed below). Nonetheless, the identification of key microorganisms controlling net MeHg production suggests the possibility of microbial regulation to mitigate Hg risks. Indeed, soil Se has been found to play a critical role in shaping the spatial distribution pattern of *hgcA*-containing *Geobacteraceae*, as soil Se concentrations are significantly negatively correlated to the relative abundance of *hgcA*-containing *Geobacteraceae*. Notably, soil amendment with Se at a dose of 0.4 mg/kg, comparable to the background soil Se concentration of 0.39 mg/kg, did inhibit Fe reduction and MeHg production. The reductions in soil MeHg concentration and Fe reduction were significantly correlated in soils with high abundances of *hgcA*-containing *Geobacteraceae*.[23] This suggests that using Se to inhibit *Geobacteraceae* activities and thus MeHg production might be a promising approach to mitigate Hg risks.

Additionally, an efficient pathway of MeHg demethylation has been reported in crop plants, notably rice plants.[24−26] This previously unrecognized demethylation pathway is mediated by ROS generated *in vivo* with the aid of thiols, indicating a light- and microbe-independent pathway of MeHg degradation. This process is estimated to have reduced MeHg accumulation in rice grains by 72−80%. Organic pollutants are generally known to be degradable and have been reported to be degraded in organisms, while the degradation of organic MeHg within organisms has rarely been considered. As a result, our understanding of MeHg behaviors in the postabsorption period has stagnated for decades. The discovery of MeHg degradation after absorption offers insights and potential possibilities for facilitating this process for risk mitigation. In fact, the presence of soil-dissolved organic matter (DOM) was reported to elevate the demethylation ratios in rice plants.[24] This promotion might be attributed to better plant growth, as evidenced by the higher biomass of rice plants exposed to soil DOM-containing solutions compared to those without. Enhanced growth might signify a higher concentration of ROS, elevated thiol concentrations, accelerated MeHg uptake by rice roots, or faster MeHg transportation within rice plants. Although the precise mechanisms by which DOM promote *in vivo* MeHg degradation remain unclear, this finding indicates that environmental factors can influence the MeHg demethylation ratio. Therefore, manipulating these influencing factors to favor *in vivo* MeHg degradation could represent a viable approach to reducing MeHg accumulation in rice grains.

Apart from the aforementioned findings in Hg biogeochemistry in the rice-paddy system, the recent groundbreaking studies demonstrating the efficient production of ROS in soils/sediments suggest the potential for abiotic MeHg demethylation. These works elucidate that in soils/sediments with fluctuating redox potentials, reduced iron species could generate hydroxyl radicals via Fenton and Fenton-like reactions.[27−30] The generation of ROS in soil differs fundamentally from that under solar radiation, although light may be involved in ROS generation indirectly, for instance, by influencing root oxygen loss,[30] which provides oxygen or hydrogen peroxide ($H_2O_2$) for Fenton and Fenton-like reactions. In addition, microorganisms may also indirectly participate in ROS generation. Particularly, FeRB reduce Fe oxides to form active Fe species for Fenton reactions. These ROS generated in soils/sediments can participate in metal(loid) transformations and pollutant degradation.[30,31] Therefore, although direct evidence for ROS-mediated MeHg degradation in soils/sediments has not been reported, it is very plausible that ROS in soils/sediments contribute to MeHg degradation in soils. This is mainly because ROS, particularly hydroxyl radicals, are highly oxidative and efficient in degrading MeHg. Further research is required to investigate the existence of ROS-mediated MeHg degradation and to assess the relative contribution to net MeHg production.


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## POSSIBLE MEHG-TARGETED STRATEGIES FOR FURTHER RISK MITIGATION OF HG

Therefore, we suggest three potential avenues for developing MeHg-targeted mitigating approaches, i.e., regulating microorganisms to curb net MeHg production, enhancing the abiotic demethylation pathway to diminish MeHg concentration, and facilitating MeHg degradation *in vivo* to reduce net MeHg bioaccumulation. These approaches aim to cut MeHg down from links between MeHg formation in environments and its transfer along food chains. In fact, microbial regulation has widely been used in agriculture to control pests, promote plant growth, etc.[32] Meanwhile, facilitating the degradation of pollutants is not new for mitigation risks. Unfortunately, such approaches have seldom been considered for MeHg, limiting an efficient mitigation of human exposure to Hg.

We also highlight the importance of a systematic evaluation regarding mitigation efficacy and ecological effects before field application. First, similar to other mitigation measures, the efficacy of approaches might be impacted by a series of environmental factors, such as farming activities. In addition, the long-term efficacy should also be considered. For instance, whether soil amendment with Se could efficiently inhibit FeRB activities during the whole period of rice growth is critical to evaluating the mitigation of MeHg accumulation. Second, a healthy microbiome is critical for sustainable agriculture.[33] Whether MeHg-targeted approaches, such as microbial regulation, would adversely impact the functions of microorganisms in the paddy soils remains unexplored. This is particularly important, as rice paddies are a main source of greenhouse gases, including $CO_2$ and $CH_4$.[34] Meanwhile, MeHg-targeted approaches should also not negatively influence food quality and quantity, which are critical for achieving Sustainable Development Goals (SDGs) 2 and 3.

It is noteworthy that developing MeHg-targeted strategies does not constitute a replacement of the ongoing global strategy; rather, it entails the concurrent application of both to enhance mitigation effectiveness. The fundamental approach to Hg risk mitigation continues to be the IHg-targeted mitigation strategy, particularly the reduction of anthropogenic Hg emissions. This is mainly because, on the one hand, Hg emission control is indispensable for sustaining low atmospheric Hg concentrations and mitigating the subsequent Hg deposition in the future. Without effective Hg emission control, atmospheric Hg concentration would rise, leading to increased Hg deposition and elevated MeHg production. On the other hand, microorganisms and plants/algae are more sensitive to IHg than MeHg. Controlling low IHg concentrations is particularly critical in aquatic systems, where atmospheric Hg deposition is a main Hg source, as the elevated IHg concentrations might threaten the growths of microbes and algae, both of which are vital drivers in maintaining the functions of aquatic ecosystems. Therefore, the integration of MeHg-targeted approaches, particularly in Hg-risk sensitive areas such as paddy soils, into the global IHg-targeted strategies offers a comprehensive and synergistic approach, paving the way for more effective mitigation of Hg risk globally.

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### Notes

The authors declare no competing financial interest.

### Biography

Huan Zhong is a professor at the School of Environment, Nanjing University. His main research interests are in the biogeochemical cycling of mercury, with a focus on novel pathways of mercury transformation in the environment and in organisms. Prof. Zhong's lab, in collaboration with multidisciplinary scientists, reports the light- and microbe-independent pathway of methylmercury demethylation at the entry of food chains, i.e., primary producers.

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