Environmental Science Processes & Impacts

ROYAL SOCIETY OF CHEMISTRY

CRITICAL REVIEW

View Article Online
View Journal

Cite this: DOI: 10.1039/d2em00039c

Mercury pollution in China: implications on the implementation of the Minamata Convention†

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Mercury (Hg) is a toxic metal released into the environment through human activities and natural processes. Human activities have profoundly increased the amount of Hg in the atmosphere and altered its global cycling since the Industrial Revolution. Gaseous elemental Hg is the predominant form of Hg in the atmosphere, which can undergo long-range transport and atmospheric deposition into the aquatic systems. Hg deposition elevates the methylmercury (MeHg) level in fish through bioaccumulation and biomagnification, which poses a serious human health risk. Acute poisoning of MeHg can result in Minamata disease, while low-level long-term exposure in pregnant women can reduce the intelligence quotient of infants. After five sessions of intergovernmental negotiation, the Minamata Convention on mercury entered into force in August 2017 to protect human health and the environment from Hg pollution. Currently China contributes the largest quantity of Hg production, consumption, and emission globally. However, the status of Hg pollution in the environment in China and its associated health risk remains relatively unknown, which hinders the development of implementation plans of the Minamata Convention. In this paper, we provide a comprehensive review on the atmospheric release of Hg, distribution of air Hg concentration, human exposure to MeHg and health impacts caused by Hg pollution in China. Ongoing improvement of air pollution control measures is expected to further decrease anthropogenic Hg emissions in China. Air Hg concentrations in China are higher than the background values in the Northern Hemisphere, with spatial distribution largely influenced by anthropogenic emissions. Long-term observations of GEM in China show a decline in recent years. The net Hg transport outflow from China in 2013 is estimated to be 511 t year^{-1}, and ~60% of such outflow is caused by natural surface Hg emissions. Hg concentrations in fish and rice in China are relatively low and therefore the associated risks of human Hg exposure are low. Future research needs and recommendations for the implementation of the Minamata Convention are also discussed in this paper.

Received 30th January 2022
Accepted 6th April 2022

DOI: 10.1039/d2em00039c

rsc.li/espi

Environmental significance

Mercury is a global pollutant and human methylmercury exposure is a global environmental health issue. The Minamata Convention on mercury is aimed to protect human health and the environment, which entered into force in August 2017. China is one of the largest countries in terms of mercury production, consumption, and anthropogenic emission. However, the status of mercury pollution in China and its associated health risk remains relatively unknown. Here we summarized the current status of the mercury emission inventory from both anthropogenic activities and natural processes, the distribution of mercury in ambient air, and human exposure to methylmercury and health impacts in China. It is of great importance to fully understand the full picture of mercury pollution in China and to provide important implications on the implementation of the Minamata Convention in China.

Introduction

Since the tragedy of the Minamata disease outbreak in Japan in the 1950s, mercury (Hg) has been recognized as one of the most toxic heavy metals.[1] The toxicity of Hg depends on its chemical form with methylmercury (MeHg) being the most toxic. The major adverse effects of human MeHg exposure are neurological effects, but renal, cardiovascular, reproductive, and immune system effects were also reported.[1,2] Excessive exposure to MeHg through fish consumption was the main cause of Minamata disease, which led to the poisoning of more than

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† Electronic supplementary information (ESI) available. See https://doi.org/10.1039/d2em00039c

3000 people and a death toll of 1784.[2] Low-level, long-term MeHg exposure in pregnant women could reduce the intelligence quotient (IQ) of infants, which will cause significant economic loss to society.[3]

Human activities have profoundly altered the global cycling of Hg in the atmosphere. Hg is the only metal that occurs in the atmosphere mainly in the gaseous elemental mercury (GEM) form that has a long residence time (0.5–2 years) in the atmosphere, while other atmospheric forms including gaseous oxidized mercury (GOM) and particulate bound mercury (PBM) generally represent a minor fraction of the total atmospheric Hg and are readily deposited on the earth's surface because of their relatively short atmospheric residence time (*e.g.*, days to weeks).[4] Therefore, Hg can undergo long-range air transport after being released from anthropogenic sources such as coal combustion, artisanal and small-scale gold mining, metal smelting, cement production, and waste incineration, as well as natural processes such as volcanic eruption, geo-thermal activities, forest fires and emissions from soil and water surfaces.[5] It was first discovered in the 1980s that fish in pristine lakes in remote areas of North America and Europe contained elevated MeHg levels exceeding the World Health Organization (WHO) recommended food consumption limit of 1 mg kg^{-1} (wet weight).[6,7] Long range transport of Hg in the atmosphere followed by deposition of Hg in these aquatic systems caused an elevated MeHg level in fish in these lakes.[8] However, the temporal trends of Hg concentrations in aquatic biota often do not agree with atmospheric concentration trends, which may be caused by legacy Hg and changing biogeochemical processes.[9]

There is an international focus on the reduction of Hg emissions. The current mass loading of Hg in the atmosphere is about 3 times that in the pre-industrial era.[10] In February 2009, the Governing Council of the United Nations Environment Programme (UNEP) agreed on the need to develop a global legally binding instrument to mitigate the adverse impacts caused by Hg pollution. Five meetings organized by the intergovernmental negotiating committee (INC) were convened from 2011 to 2013. Upon the completion of negotiations in February 2013, the Minamata Convention on mercury was signed in October 2013 to protect human health and the environment from anthropogenic emissions and the release of Hg and Hg compounds. The convention formally entered into force in August 2017 and controlling the anthropogenic emission of Hg throughout its lifecycle has been a key obligation under the convention.

China currently has the largest national production, consumption and emission of Hg. It contributed 89% of the world's annual Hg production.[11] The Hg consumption increased from 803 ± 95 t in 2005 to 1272 ± 110 t in 2011 and decreased to 903 ± 115 t in 2014.[12] Polyvinyl chloride (PVC) production is the primary industrial Hg use, accounting for more than 60% of the global Hg demand. The anthropogenic Hg emission in China is ~25% of the global total.[13] The quantity of Hg use and emission poses a challenge in Hg pollution control in China under the Minamata Convention. Currently, the status of Hg pollution in the environment and the health risk of MeHg exposure in China remain unclear, hindering the development of the implementation plans of the Minamata Convention.

In this paper, we summarize the state of knowledge on Hg pollution in China, including Hg emissions from anthropogenic and natural sources, distribution of Hg concentration in ambient air, human exposure to Hg and its associated health risk. The goal is to provide science-based recommendations for guiding the implementation of the Minamata Convention in China.

Anthropogenic Hg emission in China

Anthropogenic Hg emissions refer to the mobilization and release of geologically sourced Hg into the environment by industrial activities.[14,15] Due to the rapidly growing economy and industrial activities, China is the largest source region of anthropogenic Hg emissions in the world.[15] Hg emissions from coal combustion in China were first investigated by Wang *et al.*[16] The source sector released 214 t of Hg to the atmosphere and 89 t of Hg to the environment as solid wastes in 1995. The atmospheric Hg emission inventory from all man-made sources in China in 1999 was first developed by Streets *et al.*,[17] which estimated total anthropogenic emissions of 536 ± 236 t. In the past two decades, the emission quantity and speciation distribution of Hg from anthropogenic sources in China have been greatly improved.

Previous Hg emission inventories in China estimated that anthropogenic sources released 356–1028 t $year^{-1}$ of Hg into the atmosphere and 643–1161 t $year^{-1}$ into land surfaces during the last two decades.[13,15,18–23] Huang *et al.*[18] estimated that Hg emissions into the environment in China increased from 1028 to 2152 t $year^{-1}$ from 2000 to 2012, yielding an average annual increase rate of ~6.3%. Liu *et al.*[21] estimated that the anthropogenic Hg emissions to the atmosphere varied from 375 to 574 t $year^{-1}$ during 2010–2017, with a continuous increase observed during 2010–2013 (from 375 to 574 t) and a slight decrease observed after 2013 (574 to 445 t $year^{-1}$). A positive-to-negative trend reversal was detected during 2011–2013. The uncertainties in these estimates ranged from −49% to 68% for atmospheric emissions and from −74% to 97% for land surface emissions.[13,15,20,23] Anthropogenic Hg emissions exhibit large long-term variation.[13,18,21–23] Due to the changes of Hg emission speciation driven by the application of air pollution control devices (APCDs), the fractions of gaseous elemental mercury (GEM, Hg^0) showed a continued decrease in recent years (*e.g.*, from 60% in 2000 to 51% in 2014). Modelling studies have shown that the change of the emission trends decreases both domestic Hg deposition and the outflows of Hg emission in China.[22,24]

Fig. 1A shows the global atmospheric Hg emissions from anthropogenic sources by the country in 2015. China contributes to 25.5% of global Hg emissions, although the cumulative atmospheric emissions of Hg in China are much smaller than in other industrial regions since the industrial era. For example, the total atmospheric Hg emissions in China from human activities during 1949–2012 were estimated to be 15 500 t,[25] which only accounts for 4.6% of the global cumulative

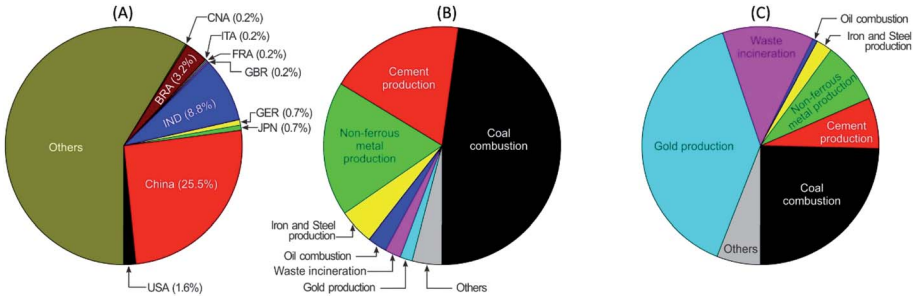

Fig. 1 (A) Anthropogenic atmospheric Hg emissions from the top 10 economies and other countries in the world in 2015;[5] (B) proportion of anthropogenic atmospheric Hg emissions in China by source sector in 2010;[13] (C) proportion of global anthropogenic atmospheric Hg by source sector in 2010.[27]

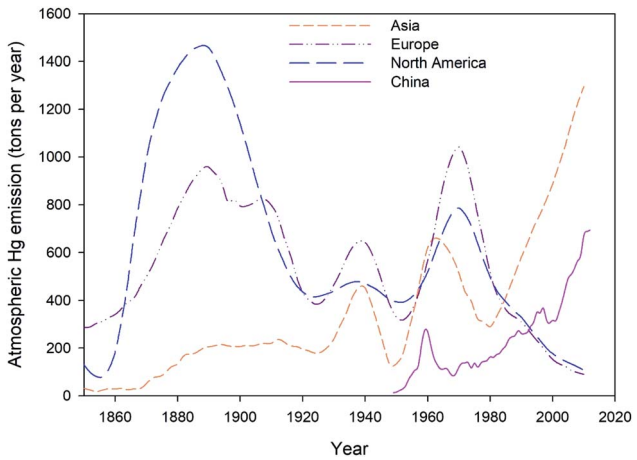

Fig. 2 Trends of anthropogenic air Hg emissions in China and different world regions.[25,26]

atmospheric Hg emissions (336 000 t) since the industrial era, and which is much lower than the contribution from North America (30%) and Europe (27%) (Fig. 2).[26] Recently, intensive air pollution control measures (*e.g.*, ultralow emission renovations of coal-fired power plants, elimination of small coal combustion boilers in industrial activities and domestic heating) have rapidly decreased (*e.g.*, 5.5% year^{-1}), and will continue to reduce anthropogenic Hg emissions in China.[21] This is anticipated to decrease atmospheric Hg concentrations and depositions noticeably in China, as well as anthropogenic Hg emissions globally. Coal combustion, cement production, nonferrous metal production, and waste incineration are important source sectors in China, which released approximately 253.8, 98.3, 97.4, and 11.6 tons of Hg to the atmosphere, respectively, in 2010, accounting for 47.8%, 18.5%, 18.3% and 2.2% of the total emissions in 2010, respectively (Fig. 1B).[13] The source profile of anthropogenic Hg emissions in China differs from those of other major source regions, where gold production, coal combustion, and waste incineration were the 3 largest sources (Fig. 1C). The anthropogenic emissions in China were concentrated in eastern, northern, and central China.[13,21,22] The top 5 largest source regions in 2017 were Hubei, Shandong, Hebei, Hunan, and Jiangsu provinces, accounting for approximately 37% of the total atmospheric Hg emissions in China.[21]

Liu *et al.*[28] showed that the monthly variation in Hg emission from coal fired power plants was highly correlated to industrial activities, with peak emission occurring in January and December and the lowest emission, in February. The former was caused by the increased year-end industrial activities and the latter was caused by reduced industrial activities during the holiday season in China. The diurnal variation of Hg emission from coal fired power plants exhibited higher emissions during daytime due to the higher energy demand.[28] Hg emission from industrial coal combustions showed a relatively consistent monthly level throughout the year, with a smaller emission quantity in February. The emission from coal combustion in residential and other sectors showed a strong seasonal variation, with peaks in the cold seasons. Such a seasonal pattern was more pronounced in regions that use coal heating in winter.[29] Overall, anthropogenic Hg emissions in China are an important source of atmospheric Hg at regional and global scales. Recent studies have significantly advanced our knowledge of anthropogenic Hg emissions in China. Future studies, however, are still needed to reduce the uncertainties of atmospheric Hg emission inventories and explore their long-term and seasonal trends, which would be helpful for better understanding regional and global atmospheric Hg cycling and the effectiveness of the implementation of the Minamata Convention.

Hg emission from natural surfaces in China

"Natural" emission includes the primary Hg^0 release from geogenic activities (*e.g.*, emission from volcanos, hot springs, *etc.*) and the re-emission of legacy Hg stored in terrestrial surfaces. Although the temporal and spatial features of geogenic Hg emission in China are largely undocumented,[30] its emission quantity is estimated to be smaller than the re-emission of legacy Hg stored in the terrestrial surfaces. Herein, the discussion mainly focuses on the re-emissions of legacy Hg (bidirectional and multi-phased air–surface exchange) involving the air–solution–particle interactions at air–surface interfaces.

The temporal and spatial patterns of the air–surface exchange of Hg^0 in China have been studied extensively.[31–37] The air–surface Hg^0 fluxes in China are statistically higher than those observed in Europe and North America, suggesting a strong anthropogenic influence in China.[22,36,38] Hg^0 re-emission fluxes from natural surfaces are highly diverse. The fluxes from natural Hg-enriched and anthropogenic polluted sites were 1–2 orders of magnitude higher than the fluxes from unpolluted terrestrial forest and grassland surfaces.[36,38] In addition to soil Hg content, other environmental parameters including solar irradiance and soil temperature have been qualitatively evaluated for their influence on Hg^0 re-emission statistically.[36] Shetty *et al.*[39] used the empirical functions of soil Hg, solar irradiation, and soil temperature to estimate a 462 t $year^{-1}$ Hg^0 re-emission from China's natural surfaces. However, there was a knowledge gap in the understanding of environmental processes driving air–surface Hg^0 exchange at that time. Moreover, the uncertainties in flux measurement techniques, insufficient temporal resolution and a long-term flux database for different surfaces limit the application of regression-based models.[36] Estimates from regression-based models are not globally applicable because of existing data gaps in site-specific characteristics, thus overlooking the heterogeneity of the air–surface interface.

Recently, research on the reduction kinetics of soil Hg enabled the building of a more physically robust air–surface Hg^0 flux scheme. Wang *et al.*[40] estimated the natural Hg^0 re-emission in China using an updated mechanistic model that incorporates the present understanding in Hg transformation in soil and on the cuticular surface.[41] They calculated 565.5 t $year^{-1}$ of emission from soils, 9.0 t $year^{-1}$ of emission from water bodies, and 100.4 t $year^{-1}$ uptake by vegetation in China. The mechanistic model results are validated against the predicted spatial pattern of forest foliage Hg^0 uptake and observed litterfall Hg deposition (Fig. 3A and B). The mechanistic approaches by Wang *et al.*[40] advance upon the estimate of Shetty *et al.*[39] in two ways. One is the confirmation of the role of vegetation as a Hg^0 sink. Vegetation is considered a major source in Shetty *et al.*,[39] accounting for 76% of the total re-emissions, since the earlier scheme treats Hg evasion like the evapotranspiration process that transports Hg from the root zone through vascular tissues in the foliage, followed by reduction and evapotranspiration into the atmosphere.[39,42] Recent experimental evidence using stable Hg isotopes excluded this pathway for cereal plants.[43,44] Vegetation foliage is denoted as a sink of Hg^0 in Wang *et al.*[40] (Fig. 3C), consistent with the observations in forest ecosystems.[34,45,46] The other advancement is the spatial quantification of soil Hg re-emissions with improved soil Hg data. Shetty *et al.*[39] estimated that the Hg^0 re-emissions mainly occurred on the east side of the "Heihe-Tengchong line," because of the higher soil Hg concentration in East China. In contrast, a larger natural Hg^0 re-emission in the western region of China (Fig. 3D) was found using the new mechanistic model.[40] The changed spatial distribution of natural Hg^0 re-emission also decreases the Hg emission outflow from China.[24] The 460–550 t $year^{-1}$ of natural Hg^0 re-emission in China is equivalent to the anthropogenic Hg emission (546 t $year^{-1}$ in 2013).[22]

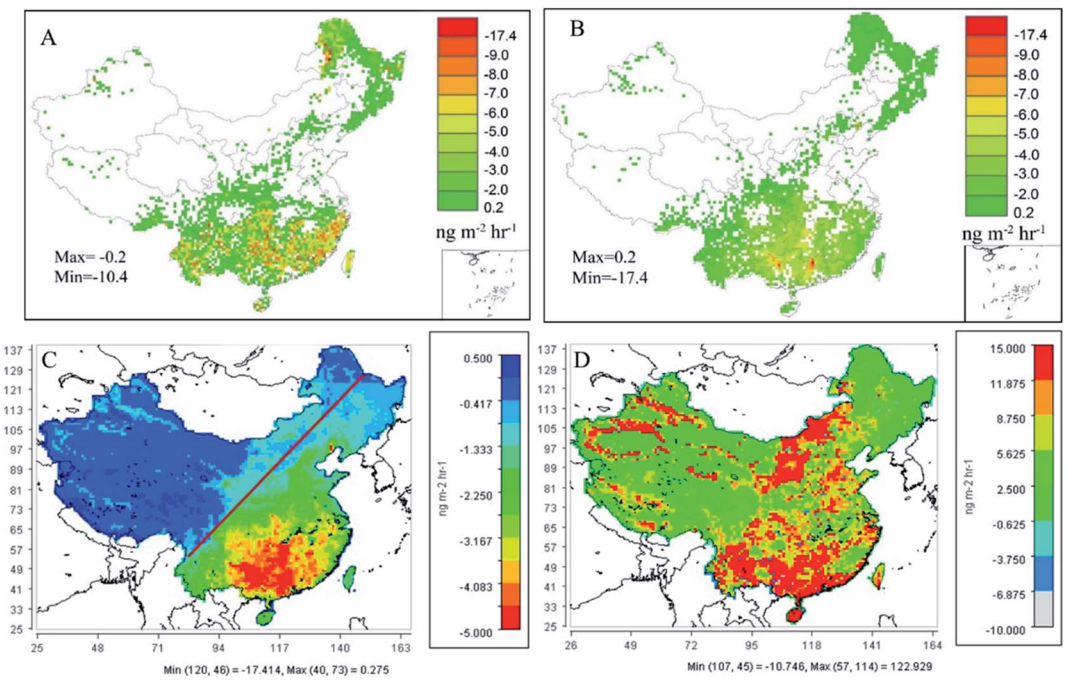

Fig. 3 (A) Measurement-based estimates of litterfall Hg deposition in forest ecosystems, (B) simulated results of annual Hg uptake by forest foliage; (C) simulated results of the mean annual air–foliage flux; and (D) simulated results of the mean annual air–soil flux. The redline refers to the well-known geodemographic demarcation line, the Heihe-Tengchong line. Data in (A) are from Wang *et al.*,[45] and data in (B)–(D) are from Wang *et al.*[40]

Legacy Hg stored in the surface soil layer of China is mainly from the accumulation of previous atmospheric Hg deposition (up to 60%). The natural archives (sediment and peat cores) in remote regions of China (Fig. S1†) have recorded continuously increasing atmospheric Hg deposition rates during the 1850s–1970s when there were few anthropogenic Hg emissions in China. The rapid increase of anthropogenic emission from China started from the 1980s due to economic reform and industrial development, while large quantities of anthropogenic Hg were released from the developed countries since the 1850s. The cumulative anthropogenic Hg emission from developed countries since the 1850s is 10–20 times higher than the modern-day anthropogenic Hg emissions in China.[47,48] This is consistent with the recorded atmospheric Hg deposition rates in natural archives of China, North America and Europe (Fig. S1 and S2†). The historical emissions had contributed to Hg re-emission from natural surfaces in China, specifically in West China where few point sources exist but with elevated natural Hg re-emissions.[22,40]

Hg concentration in air

The observational network of atmospheric Hg in China was built gradually since the 2000s. Average GEM, PBM and GOM concentrations and the Hg wet deposition flux at rural sites in China were 2.50 ± 0.93 ng m^{-3} (means: 1.33 to 5.07 ng m^{-3}, n = 20), 44.9 ± 48.7 pg m^{-3} (means: 2.3 to 180 pg m^{-3}, n = 12), 8.0 ± 5.2 pg m^{-3} (means: 0.8 to 21.4 pg m^{-3}, n = 12), and 9.9 ± 9.9 μg m^{-2} $year^{-1}$ (means: 2.0 to 32.3 μg m^{-2} $year^{-1}$, n = 13), respectively (Fig. 4, Tables S1 and S2†). Urban areas of China show a higher atmospheric Hg concentration and wet deposition fluxes as compared to rural areas, with a mean of 4.98 ± 1.98 ng m^{-3} (means: 2.57 to 9.72 ng m^{-3}, n = 14) for GEM, 175 ± 151 pg m^{-3} (means: 16.0 to 452 pg m^{-3}, n = 9) for PBM, 32.5 ± 29.4 pg m^{-3} (means: 5.3 to 61.0 pg m^{-3}, n = 7) for GOM, and 21.3 ± 17.5 μg m^{-2} $year^{-1}$ (means: 5.7 to 56.5 μg m^{-2} $year^{-1}$, n = 7) for wet deposition fluxes, respectively (Fig. 4). The concentrations and wet deposition fluxes were higher compared to those observed at the background sites in the Northern Hemisphere. For example, the means of GEM, PBM, and GOM concentrations, and the Hg wet deposition flux in ambient air in rural areas of China were 1.6, 8.5, 1.3, and 1.2 times higher than the means in North America and Europe (GEM = 1.51 ng m^{-3}, PBM = 5.3 pg m^{-3}, GOM = 6.2 pg m^{-3}, and wet deposition flux = 8.2 μg m^{-2} $year^{-1}$), respectively.[49–51] The contribution of domestic primary anthropogenic emissions to GEM, PBM, and GOM in China was estimated to be 41%, 71%, and 64%, much greater than those found in North

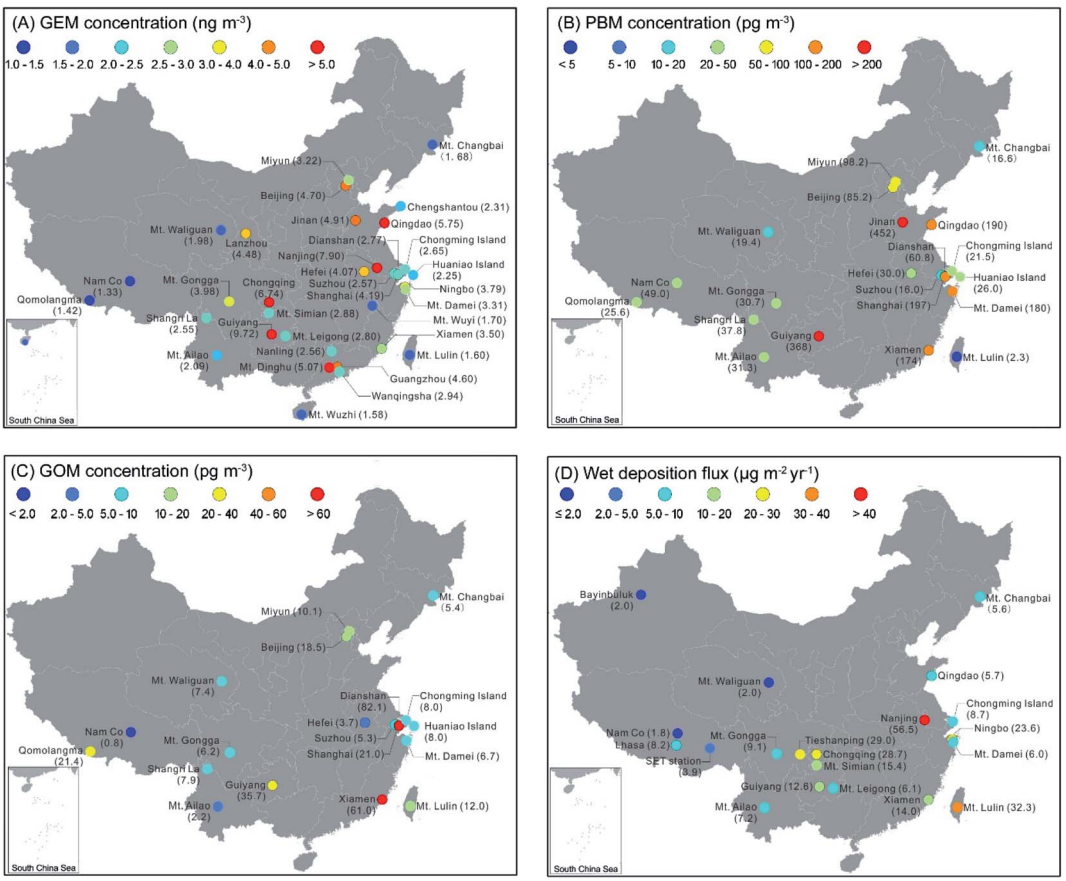

Fig. 4 Locations of the ground-based measurement sites and observed mean concentrations of (A) atmospheric gaseous elemental mercury (GEM), (B) particulate bound mercury (PBM), (C) gaseous oxidized mercury (GOM), and (D) the Hg wet deposition flux in China. Data are from the literature and references therein.[54–75]

America and Europe (5–7%, 41–66%, and 20–52% to GEM, PBM, and GOM[52]). The primary anthropogenic Hg emissions in China during 2010–2015 were 10 to 20 times higher than those in North America and Europe,[27] which explained the elevated atmospheric Hg concentrations and wet deposition fluxes in China.[27,53]

GEM concentrations in ambient air in the rural areas of China showed strong spatial variation (Fig. 4A), with the lowest at sites in the Tibetan Plateau (means = 1.33 to 1.98 ng m^{-3}, n = 3), followed by the sites in Taiwan and Hainan Island (means = 1.58 to 1.60 ng m^{-3}, n = 2), the site in Northeast China (mean = 1.68 ng m^{-3}, n = 1), and then the sites in East, Southwest, North, and Central South China (means = 1.70 to 5.07 ng m^{-3}, n = 14). The mean GEM concentrations in rural and urban sites are significantly correlated with the cumulative anthropogenic Hg emission (ΣHg emission) encountered by air masses in the source region (ANOVA, $R^2 = 0.43$, $p < 0.01$, Fig. 5A), suggesting that anthropogenic emission is an important factor controlling the spatial variation in the GEM concentration in China. No significant correlation is obtained between the concentrations of PBM (or GOM) and anthropogenic Hg emissions (R^2-PBM = 0.11, R^2-GOM = 0.10, $p > 0.05$ for both), indicating that the spatial variation in PBM and GOM is likely controlled by other factors such as the local emission (*e.g.*, biomass burning and dust related sources) and atmospheric transformation (*e.g.*, oxidation of GEM to GOM and gas-particle partitioning of GOM to PBM).[54–56,76–78] Hg wet deposition fluxes in China are significantly correlated with the concentrations of total Hg (THg) in precipitation (Fig. 5B), indicative of the influence of precipitation THg concentration. THg in precipitation is mainly from the scavenging of GOM and PBM in clouds and below clouds.[79] In China, PBM and GOM concentrations are highly elevated in urban areas, which explains the high Hg wet deposition fluxes in the urban areas of China.[57]

Hg emissions from land surfaces, biomass burning, and indoor Hg-containing products are also important sources of atmospheric Hg in China.[40,80,81] Previous studies in Nanjing and Lanzhou found that the ratios of GEM/CO were frequently higher in summer than in winter, suggesting that Hg emission from land surfaces plays an important role in atmospheric GEM concentrations in urban areas.[58,82,83] GEM emission from land surfaces, especially in urban areas, comes from the recycling of previously deposited anthropogenic Hg.[38] Other than urban areas where the atmospheric Hg is dominated by local sources,[83,84] long-range transport of Hg controlled by the monsoon climate is the dominant source of atmospheric Hg in rural areas of China. The identified transport routes are from the northern, eastern, and southern source regions to rural areas in northeastern, eastern, and southern China, and from the northwestern and southwestern source regions to rural areas in the Tibetan Plateau and southwestern China, respectively.[56,59–61,85,86] Transboundary transport of Hg emissions from South and Southeast Asia has been identified and shown to play an important role in atmospheric Hg pollution of the rural Tibetan Plateau and Southwest China.[60,62,63,77,87]

The long-term trend of GEM concentrations in China is shown in Fig. 6.[54,88,89] At Mt. Lulin, offshore of southeast China, the GEM concentration peaked in 2009, followed by a decline during 2009–2010, and then remained relatively

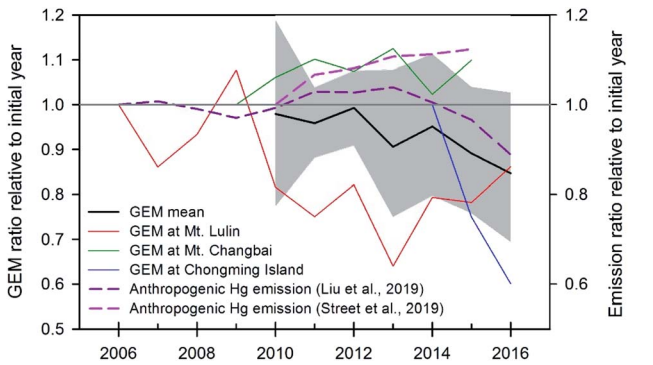

Fig. 6 Trend of measured atmospheric GEM concentrations at Mt. Lulin, Mt. Changbai, and Chongming Island and the estimated anthropogenic Hg emissions in China. Data are normalized by dividing the mean of each year by their corresponding value in the initial years. The shaded area indicates the one standard deviation (1sd) of the GEM concentration trend during 2010 and 2016. Data are from ref. 21, 27, 54, 88 and 89.

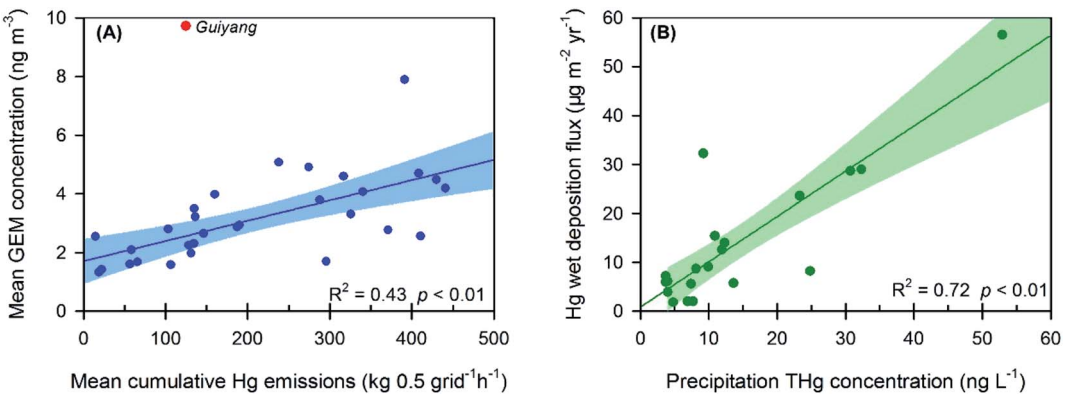

Fig. 5 Relationship (A) between the mean ground-based GEM concentration and cumulative anthropogenic Hg emissions encountered by the air mass during the last three days of atmospheric transport at the corresponding sites, and (B) between the Hg wet deposition flux and THg concentration in precipitation. Data are from Fig. 3.

constant from 2010 to 2016. The GEM concentration at Mt. Changbai, northeastern China, showed a continuous increase from 2009 to 2013 (+2.6% year^{-1}), followed by a decline in 2014. The GEM concentration at Chongming Island, Eastern China showed a 20% year^{-1} decline from 2014 to 2016. Overall, the GEM concentration mean remains relatively constant from 2010 to 2012 and shows a decline from 2013 to 2016 (Fig. 6). This is consistent with the trend of anthropogenic Hg emission in China,[21] but in contrast to the estimate by Streets *et al.*[27] (Fig. 6). The trend of the GEM concentration could be site-specific due to anthropogenic emission, transport patterns, and atmospheric transformation (*e.g.*, vegetation uptake activity and atmospheric oxidations) at local and regional scales.[21,88,90,91]

Mass balance of atmospheric Hg in China

Regional and global scale Hg modeling assessments have documented atmospheric Hg transport and outflow in China and the East Asian region. Most of the regional and global modeling assessments depicted that the Asian Hg outflow contributes to 15–25% of Hg deposition in North America and 10–15% in the European region,[92–98] except for one study reporting <5% of contribution in both regions.[99] It should be noted that the results of earlier modeling assessments are not directly comparable due to differences in emission inventories, Hg chemistry schemes and model configurations. Specifically, the natural emission in earlier studies did not specify the quantity and spatial distribution in global models.[92,94,95]

Wang *et al.*[24] applied CMAQ-Hg v5.1 with the updated Hg natural emission of Wang *et al.*[40] and anthropogenic Hg emissions in 2013 from Wu *et al.*[22] to reassess the Hg transport budget in China (Fig. 7). The natural and anthropogenic emissions (up to 1100 t year^{-1}) and chemical transport of Hg lead to an accumulation of 78 t year^{-1} of Hg in the atmosphere, 422 t year^{-1} deposition (49% by wet deposition, and ~2/3 of the deposition is caused by domestic emissions) in mainland China. The net Hg transport outflow from China is estimated to be 511 t year^{-1}, and nearly 60% of such an outflow is derived from natural surface Hg emission. This outflow contributes to about 10% of Hg deposition outside of East Asia, lower than the fraction estimated by earlier studies.[92–98] The decrease is based on the recently updated Hg emission quantity, as well as the speciation, and spatial and temporal distributions of the inventory data.

Most model results cannot fully explain the seasonal and spatial GOM patterns of field observations.[92–98] This is possibly caused by the measurement bias of GOM sampling using KCl denuders commonly used in ground-based monitoring of atmospheric Hg,[100,101] and the inadequate representation of atmospheric Hg chemistry implemented in the models.[102] The measured data of GOM are associated with large uncertainty because the measurement conducted by a Tekran system is strongly influenced by RH, the O_3 content and other factors.[100,101] In addition, the accuracy of emission inventories is critical for assessing the Hg budget, but available emission data in China do not provide the required resolution, speciation, and temporal and spatial patterns to constrain models.[22,24] More studies to better understand Hg chemistry during Hg cycling will improve the estimates of the Hg mass budget in China.

Health risks of Hg exposure in China

Human exposure to MeHg *via* fish consumption is a global public health concern.[103] In the 1980s, fish THg levels, even in remote regions in North Europe and North America, commonly exceeded the international standards set by the World Health Organization (WHO) and Food and Agricultural Organization (FAO) for the protection of human health (0.3–1.0 mg kg^{-1} and wet weight (ww)).[104,105] Fish MeHg contamination poses a particular challenge to global public health, because fish contains many beneficial nutrients.[103] Moreover, fishery is an important part of global agriculture.

In China, the THg concentration in fish is much lower than the national standards (1.0 mg kg^{-1} for carnivorous fish and

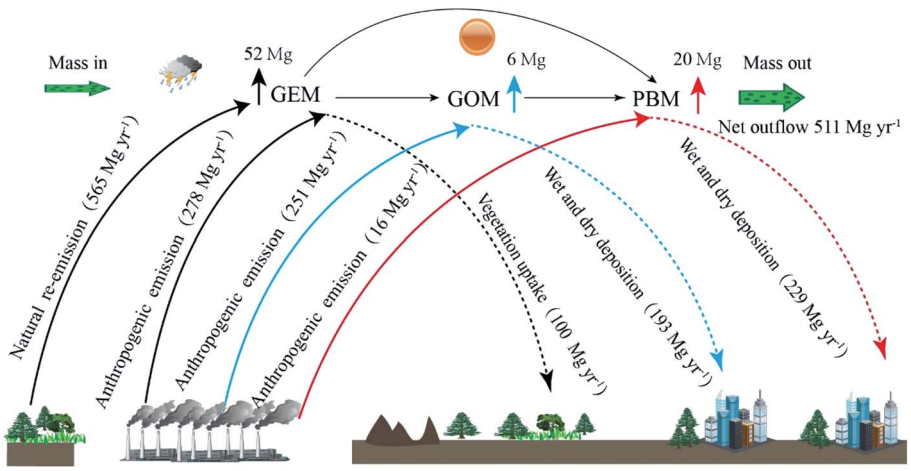

Fig. 7 Mass budgets of atmospheric Hg in mainland China. GEM: gaseous elemental mercury; GOM: gaseous oxidized mercury; PBM: particulate bound mercury. The data are obtained from Wang *et al.*[24]

0.5 mg kg^{-1} for others, ww). These are considered relatively safe levels.[106] Fish THg in China is within the average value range of 8.5–274.7 ng g^{-1} (ww) with the highest value of 1218 ng g^{-1} (ww).[107–112] A high THg concentration (100.5 ± 149.2 ng g^{-1}, median ± average deviation) was mainly observed in wild fish from the Tibetan Plateau, and the long lifespan and slow growth of the fish in low-productivity environments are likely the major biological factors.[112] Generally, fish THg increases with size, weight, age, trophic position and feeding habits.[106] Since fish in China is largely farmed, little Hg bio-accumulation occurs.[113] China is the largest country involved in fish farming with a production of 26.9 Mt in 2018, much greater than the fish catch of 10.4 Mt.[114] The low trophic level of farmed fish and overfishing in China significantly shorten the length of the food chain and reduce the fish Hg level. The effect of the fishing ban policy in the Yangtze River and coastal waters on fish MeHg levels remains to be observed when data become available.

Rice grown at Hg-polluted sites shows an elevated MeHg concentration. Rice consumption can be the primary source of human MeHg exposure in contaminated areas.[32,115,116] Hg in cereal is typically below 20 ng g^{-1} and is predominantly present in inorganic forms. Qiu *et al.*[117] reported that rice (*Oryza sativa* L.) grown in abandoned Hg mining areas contained MeHg levels of 1.61–174 ng g^{-1}, 10–100 times higher than other crops. Bio-accumulation factors (BAFs) for inorganic Hg (IHg) ranged from 0.00014 to 0.51 and from 0.71 to 50 for MeHg in rice grains collected from the Wanshan Hg mine.[118] BAFs for MeHg were on average >800 times higher than those for IHg in rice grains, indicating that rice grains are a strong bioaccumulator of MeHg, but not of IHg.[118]

Paddy field is the most prevalent type of land use in South and East Asia. The ephemeral paddy is an environment that favors Hg methylation.[119] Soil is the source of MeHg in rice plants. MeHg in soil is first absorbed by the roots and then translocated to the leaf and stalk.[120] During the ripening period, most of the MeHg stored in the leaf and stalk is transferred to the seed.[120] The majority of IHg in a rice grain is found in the hull and bran, while most of the MeHg is found in white rice.[121] Data from X-ray absorption near-edge spectroscopy (XANES) showed that MeHg in grains is primarily bound to cysteine and associated with proteins, which can be actively transported to the endosperm during seed ripening.[121] Based on stable isotope evidence, soil is the main source (80%) of Hg in rice grains.[122] The soil–water system is the original source of MeHg in rice plants and water, soil, and the atmosphere contributed to IHg in grains, leaves, stalks, and roots.[123]

Rice THg and MeHg levels in China are relatively low with a mean of 4.74 ng g^{-1} and 0.682 ng g^{-1}.[124] However, the THg and MeHg concentrations in rice from Hg contaminated areas (such as Hg mining areas) are significantly elevated.[116,117,125] Rice consumption constitutes 93.5–97.5% of the total MeHg intake among residents in the Wanshan Hg mining area, leading to consumption of rice, rather than fish consumption, being the main route of human MeHg exposure in inland areas of southern China.[115,116,125] Rice consumption can contribute to up to 95% of MeHg exposure in the Wanshan Hg mining area.[126] A toxicokinetic model of MeHg exposure using fish consumption underestimated the health risks of human MeHg exposure in China.[127] Fish contains micronutrients (*e.g.*, n-3 LCPUFA and selenium, the essential amino acids) that have protective effects on the intoxication of MeHg. Human MeHg exposure through rice consumption and disparities in n-3 LCPUFA may cause serious harmful effects at the same exposure level. For example, an increase of 1 μg g^{-1} in hair Hg in Wanshan children through rice consumption results in 1 point of IQ loss, much higher than 0.18 through fish consumption.[128]

Rice is the staple food for half of the world population and therefore MeHg accumulation in rice and its health risks are of great concern. Rice plants store 180 Mg of Hg in 2016 and are considered an active bioreactor in the global biogeochemical cycle of Hg.[129] Human hair is a convenient biomarker for evaluating health risks of human MeHg exposure because the THg levels in human hair are 250- to 300-fold higher than the values in blood.[130,131] The hair Hg levels corresponding to the provisional tolerable weekly intake (PTWI) and reference dose (RfD) are 2.3 and 1 μg g^{-1}, respectively. The hair THg concentration in the general population of China is relatively low, generally below 1 μg g^{-1} of the reference level.[91] However, the residents in Hg contaminated areas (chlor-alkali plants, Hg mining and small-scale artisanal gold mining areas) have elevated hair THg concentrations.[132–135] It is advisable to focus on Hg pollution control and remediation at Hg contaminated sites of China to reduce MeHg accumulation in rice. Populations in coastal areas of China with frequent marine fish consumption showed relatively higher hair THg concentrations.[107,111,135,136] Advisories on fish consumption are needed to reduce MeHg exposure in coastal areas.

Implications on the implementation of the Minamata Convention in China

Although developed countries have cumulatively contributed a majority of man-made release of Hg into the environment, China is the biggest emitter in the 21st century. Anthropogenic Hg released into the atmosphere in China peaked in 2012–2013 and decreased significantly thereafter (with an average of −32 t per year).[21,22] A 50% reduction of anthropogenic Hg emissions in East Asia would cause a significant decrease of Hg deposition in North America (17%) and Europe (10%).[137] Due to the low Hg content in fish in China, MeHg exposure through fish consumption is of smaller concern in China. Therefore, the primary beneficiaries of Hg pollution control in China are North America and Europe. In recent years, China has taken strong emission reduction measures that will lead to global benefits in reducing Hg deposition over time. China has developed plans to curb carbon dioxide emission and to achieve carbon neutrality before 2060. Attaining these goals relies on a dramatic transformation of China's energy use and emission reduction, especially on sustainable demand, decarbonizing electricity generation, electrifying end-use, fuel switching, and emission reduction approaches. It is anticipated that Hg emission from anthropogenic sources in China will continue to decline at an

expedited rate into the future. Based on our synthesis using available data, the following implications can be reached:

(1) Development of Hg emission inventories of high temporal and spatial resolution are needed for anthropogenic sources

Previous studies mostly focused on the spatial and long-term variations in anthropogenic Hg emission in China. To better understand the impacts of specific sources on Hg cycling in the local and regional atmospheric pool as well as to efficiently abate anthropogenic Hg emissions in China, there is a need to establish sector-based and facility-based anthropogenic Hg emission inventories. The establishment of specific emission inventories provides crucial information on the dominant sources and helps to make targeted strategies for mitigating the release of Hg into the atmosphere, water bodies, and soil. In addition, high-time resolution (*e.g.*, daily or monthly) inventories of speciated anthropogenic Hg emission from source sectors are needed for examining the effect of anthropogenic emission on the concentration changes of atmospheric Hg species. These high-resolution data allow adequate evaluation on the effectiveness of emission reduction.

(2) Improved estimates of natural Hg emission and a better understanding on the impact of climate change on Hg re-emission

Improving the data availability and quality of air–surface Hg^0 flux measurements, soil characteristics and the Hg content in remote regions covering diverse landscapes (*e.g.*, Tibet Plateau and tropical biome regions) will better constrain the natural emission quantity in China. A mechanistic understanding of Hg redox chemistry mediated by organic matter, microbes and solar radiation transfer in soil will enhance the capability of natural emission models. Since forests are the largest terrestrial carbon and atmospheric Hg sinks, rapid afforestation in China is anticipated to increase the terrestrial Hg sink, and possibly reduce the concentration of atmospheric Hg and its outflow in China. The 27% increase of forest cover during the last two decades created by China's ecological projects such as the "Grain for Green Program" should have increased the terrestrial Hg sink significantly, although a comprehensive assessment is yet to be performed. The reduction of anthropogenic Hg emission mandated by the Minamata Convention will continue to decrease the Hg concentration in the atmosphere. However, it may take decades to observe a distinct Hg decrease in biota due to complex legacy Hg re-cycling associated with climate change and changes in land use. The combined effect caused by the interplays of climate change, direct Hg emission reduction efforts, indirect benefits from other environmental programs and continued economic development in China on Hg recycling from the terrestrial system requires further investigation.

(3) The need for national Hg observational networks and a better understanding of the mass balance of atmospheric Hg in China

Observations of Hg levels in air, water, soil, and biomasses are important to assess environmental Hg pollution, its health risks, and the effectiveness of emission reduction measures. There are limited data sets documenting Hg levels in various environmental systems in China,[54,106,138] which should be integrated into coordinated observational networks to evaluate the response of environmental Hg levels to emission changes. Such networks must include sites in both polluted (*e.g.*, urban, industrial and Hg mining areas) and background areas, with measurements using automatic, manual and passive sampling techniques over a timeframe that allows adequate detection of a long-term trend. More technical development for the measurement of the atmospheric Hg species will improve the understanding of atmospheric Hg chemistry during Hg cycling. Finally, research and development of the national assimilation and prediction modelling systems of the atmospheric Hg mass balance will largely promote the estimation of the Hg mass budget in China by combining datasets of the high temporal and spatial time resolution of the anthropogenic and natural Hg emission inventories and the Hg observational networks.

(4) Prioritization for MeHg exposure through rice consumption at Hg contamination hotspots in China

The health risk of Hg pollution in China can originate from the consumption of fish, aquatic products and rice. Bioaccumulation of MeHg in rice grains at contaminated sites requires special attention to reduce the associated human health risks caused by rice consumption. Possible remediation and control measures should be examined at Hg contaminated sites. For instance, the paddy field at Hg contaminated sites can be changed to dry land, which can potentially eliminate methylation and MeHg exposure for local residents. Long-term cohort studies at Hg contaminated sites (*e.g.*, in the Wanshan Hg mining area) and animal studies should be conducted to set standards for maximum daily MeHg intake among the rice eating population. Further studies that identify and quantify the effects of MeHg exposures among residents, particularly in young children, should also be carried out to understand the impacts of long-term, low-level Hg exposure. In addition, the cost-benefit relationship of these mitigation measures should be investigated.

Author contributions

Feng Xinbin conceived and designed the manuscript. All authors drafted and revised the manuscript. All authors gave final approval for publication.

Conflicts of interest

There are no conflicts to declare.

Acknowledgements

This study was financially supported by the National Natural Science Foundation of China (41921004).

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