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Atmospheric Mercury Outflow from China and Interprovincial Trade

Long Chen,[†,‡] Sai Liang,[§] Yanxu Zhang,[∥] Maodian Liu,[⊥] Jing Meng,[#] Haoran Zhang,[⊥] Xi Tang,[†,‡] Yumeng Li,[§] Yindong Tong,[∇] Wei Zhang,[○] Xuejun Wang,[*,⊥] and Jiong Shu[*,†,‡]

[†]Key Laboratory of Geographic Information Science (Ministry of Education), East China Normal University, Shanghai 200241, China

[‡]School of Geographic Sciences, East China Normal University, Shanghai 200241, China

[§]State Key Joint Laboratory of Environment Simulation and Pollution Control, School of Environment, Beijing Normal University, Beijing 100875, China

[∥]School of Atmospheric Sciences, Nanjing University, Nanjing, Jiangsu 210023, China

[⊥]Ministry of Education Laboratory of Earth Surface Process, College of Urban and Environmental Sciences, Peking University, Beijing 100871, China

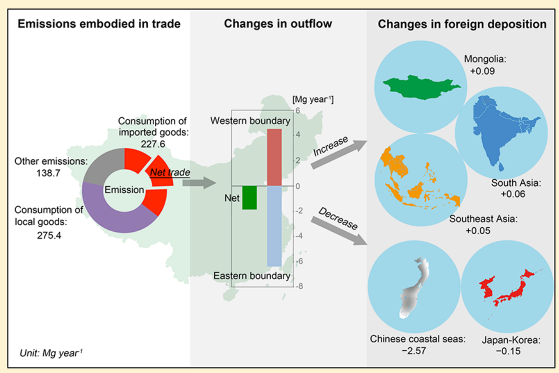
[#]Department of Politics and International Studies, University of Cambridge, Cambridge CB3 9DT, U.K.

[∇]School of Environmental Science and Engineering, Tianjin University, Tianjin 300072, China

[○]School of Environment and Natural Resources, Renmin University of China, Beijing 100872, China

Supporting Information

ABSTRACT: Mercury (Hg) is characterized by its ability to migrate between continents and its adverse effects on human health, arousing great concern around the world. The transboundary transport of large anthropogenic Hg emissions from China has attracted particular attention, especially from neighboring countries. Here, we combine an atmospheric transport model, a mass budget analysis, and a multiregional input−output model to simulate the atmospheric Hg outflow from China and investigate the impacts of Chinese interprovincial trade on the outflow. The results show outflows of 423.0 Mg of anthropogenic Hg, consisting of 65.9% of the total Chinese anthropogenic emissions, from China in 2010. Chinese interprovincial trade promotes the transfer of atmospheric outflow from the eastern terrestrial boundary (−6.4 Mg year^{-1}) to the western terrestrial boundary (+4.5 Mg year^{-1}) and a net decrease in the atmospheric outflow for the whole boundary, reducing the chance of risks to foreign countries derived from transboundary Hg pollution from China. These impacts of interprovincial trade will be amplified due to the expected strengthened interprovincial trade in the future. The synergistic promotional effects of interprovincial trade versus Hg controls should be considered to reduce the transboundary Hg pollution from China.



1. INTRODUCTION

Mercury (Hg) is a global toxic pollutant that is characterized by long distance atmospheric transport, which contributes to its ability to migrate across the oceans between continents.[1,2] Owing to dry and wet scavenging, Hg can be deposited in terrestrial and aquatic ecosystems. Methylmercury (MeHg) can be formed via the methylation and bioaccumulation of Hg in food webs after deposition, adversely affecting human health, for example, by causing neurocognitive deficits in children and cardiovascular problems in adults.[3−7] Several human health disasters have already occurred owing to MeHg exposure (e.g., Minamata disease),[8] promoting the launch of the Minamata Convention on Mercury.[9] The convention focuses on the transboundary Hg pollution and controls around the world.

China is the largest Hg emitter in the world, releasing approximately 33% of the global total anthropogenic emissions to the air each year.[1,10,11] The transboundary transport of large Hg emissions outside China has aroused great concerns from all countries of the world, especially neighboring countries. For instance, combining monitoring data and meteorological data, scholars suggested that high Hg concentrations observed at Mt. Fuji and Cape Hedo in Japan might be related to the transboundary transport of Hg from China.[12−15] These concerns require studies focusing on atmospheric Hg outflows from China and the subsequent impacts on neighboring

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regions. Previous studies have illustrated that anthropogenic emissions from East Asia significantly contribute to Hg deposition over the rest of the world. For instance, 70−75% of the total emissions from East Asia were transported outside the region, and the maximum occurred in spring and early summer, contributing 20−30% of the total atmospheric deposition over remote regions.[16,17] Anthropogenic emissions from East Asia were the primary sources for atmospheric Hg deposition over global oceans,[2,18] especially the Arctic.[19,20] However, atmospheric Hg outflows from China and the associated impacts on neighboring regions have not been investigated.

China is a vast country with substantial disparities in socioeconomic development across provinces, such as in the consumption of resources and energy, population growth, and lifestyles, resulting in frequent and substantial interprovincial trade. Interprovincial trade separates production activities and final consumption and subsequently induces embodied air, water, and soil pollution.[21−24] The atmospheric Hg emissions embodied in interprovincial trade have been well-documented in China, resulting in a comprehensive virtual atmospheric Hg emission network among Chinese provinces.[25−27] The network shows that a large amount of Hg emitted from inland provinces is caused by the final consumption in coastal provinces.[25,26] In addition to Hg emissions, previous studies also found that 32% of atmospheric deposition over China was embodied in interregional trade and that deposition was considerably redistributed by this trade.[28] Considerable impacts of interprovincial trade on atmospheric Hg emissions and deposition within China have been verified by previous studies, but impacts of interprovincial trade on Hg transport outside China are poorly understood.

In this study, we combine an atmospheric transport model, a mass budget analysis, and a multiregional input−output model to simulate the atmospheric outflow of anthropogenic Hg emitted from human activities in China and subsequent deposition over neighboring seas and lands and investigate the impacts of Chinese interprovincial trade on Hg outflow and deposition. Accordingly, suggestions on controlling trans-boundary Hg pollution from China are proposed. The findings in our study are relevant to efforts on international collaboration to reduce transboundary Hg pollution.

2. MATERIALS AND METHODS

2.1. Study Area and Associated Anthropogenic Emissions. The study domain is located from 70°E to 150°E and 11°S to 55°N, which represents the East Asian domain. This area covers China and other parts of East Asia, such as Japan, the Republic of Korea, India, Indonesia, Vietnam, Thailand, and Mongolia [Supporting Information (SI),Figure S1]. To evaluate the impacts of interprovincial trade on foreign deposition, we choose several representative neighboring seas and lands that border on terrestrial China or that are located offshore and classify these regions into five groups according to their locations, namely, the Chinese coastal seas, Japan−Korea, Southeast Asia, South Asia, and Mongolia (SI, Data set S1; Figure S1). To evaluate the impacts of interprovincial trade on atmospheric outflow, we divide the whole geographic boundary of China into the eastern terrestrial boundary and western terrestrial boundary. The eastern terrestrial boundary includes all the coastlines and national boundaries located in the northeast provinces (SI, Figure S1).

Human activities, such as coal combustion, nonferrous metal smelting, and cement production, emit large quantities of Hg into the atmosphere each year. In this study, anthropogenic emissions from China from the producer perspective (i.e., production-based emissions) are referenced from our previous work,[28] which compiled a Chinese emission inventory in 2010 by multiplying energy usage and product yields by the respective emission factors. The anthropogenic emissions are distributed as point and nonpoint sources in terms of industrial productivity and gridded population density. Seasonal variations have not been evaluated for the emissions due to data unavailability. The production-based emissions for each province and each economic sector are given in the SI (Data set S4 and S5, respectively). Anthropogenic emissions from other parts of Asia are referenced from the AMAP/UNEP (Arctic Monitoring and Assessment Programme/United Nations Environment Programme) global anthropogenic emission inventory in 2010.[1]

2.2. Evaluation of Trade-Induced Emissions. The calculation of the interprovincial trade-induced Hg emissions is based on an environmentally extended multiregional input−output (EE-MRIO) analysis of the interactions between different economic sectors (SI, Data set S2) and provinces in China multiplied by sector-specific emission intensities. The MRIO table of China in 2010 developed by Liu et al.[29] is used in this study to evaluate interprovincial trade-embodied Hg emissions among Chinese provinces. A brief introduction of the MRIO approach is shown as follows:

$$\mathbf{X} = (\mathbf{I} - \mathbf{A})^{-1}\mathbf{Y} \tag{1}$$

where $\mathbf{X}$ is a vector of the total monetary output for different sectors, $\mathbf{Y}$ is a vector of final demand for different sectors, including final consumption ($\mathbf{F}$) (i.e., urban household consumption, rural household consumption, government consumption, and investment) and international export ($\mathbf{E}$). $\mathbf{I}$ represents the unsity matrix, and $\mathbf{A}$ denotes the direct requirement coefficient matrix. Element a_{ij} in matrix $\mathbf{A}$ is defined as the intermediate input from sector i to the production of a unit output for sector j. $(\mathbf{I} - \mathbf{A})^{-1}$ is the *Leontief inverse matrix*, which is the foundation of the MRIO approach.

Multiplying by the direct emission intensity vector (ψ) for eq 1, we calculate sector- and province-specific consumption-based emissions with the corresponding final consumption $\mathbf{F}$. We define ψ_0 as a vector with zero for a given sector or province but the direct emission intensity for other sectors or provinces. Through multiplying by the vector, we calculate emissions embodied in imports (EEI) for the given sector or province with its corresponding final consumption $\mathbf{F}$ (eq 2). The hats over ψ_0 and $\mathbf{F}$ mean diagonalizing vectors of ψ_0 and $\mathbf{F}$. The correspondence relationships between direct emission sources and sectors of the Chinese MRIO table are shown in the SI (Data set S3).

$$\mathrm{EEI} = \hat{\psi}_0(\mathbf{I} - \mathbf{A})^{-1}\hat{\mathbf{F}} \tag{2}$$

To evaluate the impacts of interprovincial trade, we set up a hypothetical scenario with an absence of interprovincial trade. We assume the trade partners could produce the same goods that are originally involved in interprovincial trade locally, and then EEI of a given province are assumed to be relocated from its trade partners to the province. Similar to previous studies,[30,31] the assumed relocation of EEI reveals the same

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technologies when producing the same goods between trade partners and is used to evaluate the difference between existence and absence of interprovincial trade. The results of trade-induced emissions are shown in the SI (Data sets S4 and S5). Meanwhile, the net emission flows induced by interprovincial trade are shown in Figure S2 (SI).

2.3. Simulation of Atmospheric Hg Outflow and Deposition. The GEOS-Chem chemical transport model (version 9-02; http://geos-chem.org) is used to simulate atmospheric Hg deposition over the study area and atmospheric Hg outflow from China. The model is a 3-D atmosphere model that is integrated to a 2-D surface slab ocean and a 2-D soil reservoir for the Hg cycle.[32−34] Three Hg species, namely, elemental Hg (Hg^0), divalent Hg (Hg^{II}), and particulate Hg (Hg^P), are tracked in the model. Hg^0 can be oxidized to Hg^{II} by Br atoms, while Hg^{II} can be reduced to Hg^0 under light in cloud droplets.[33] Meanwhile, the balance of gas−particle partitioning is maintained between Hg^{II} and Hg^P.[35] Dry deposition and wet scavenging of atmospheric Hg in the model follow the resistance-in-series scheme from Wesely[36] and the scheme from Liu et al.,[37] respectively. The model is driven by the assimilated meteorological fields from the Goddard Earth Observing System (GEOS-5) conducted by the NASA Global Modeling and Assimilation Office (GMAO).

Using the method presented in our previous work,[28] a nested model can be conducted over East Asia at a native horizontal resolution of $^1/_2° \times ^2/_3°$ and 47 vertical levels from the surface to 0.01 hPa. Before performing the nested simulation, a global 4° × 5° resolution simulation is conducted first for lateral boundary conditions of the nested simulation. The global simulation is driven by emissions combining the Chinese emission inventory in China and the AMAP/UNEP global anthropogenic emission inventory outside China.[1] The nested model's performance has been evaluated against a series of observations in our previous work. In this study, we run the nested model with two emission scenarios representing the existence and absence of interprovincial trade, respectively, in 2010 under an initial spin-up of the last three months in 2009. The lateral boundary conditions are provided by global simulations during 2008−2010. The outputs are archived monthly and are used to illustrate seasonal variations in atmospheric outflow and deposition.

2.4. Calculation of Regional Hg Mass Budget. The atmospheric Hg outflow from China is estimated by calculating the Hg mass budget for China using the GEOS-Chem simulation results for each modeling month. We use a schematic for the regional mass budget calculation proposed by Lin et al.[16] In general, the change in atmospheric Hg burden within a region over a simulation period can be influenced by the Hg mass entering and leaving the region via atmospheric transport, atmospheric Hg emissions, and atmospheric Hg deposition via dry and wet scavenging. Meanwhile, the net change in atmospheric Hg burden within a region also equals the difference between the atmospheric Hg burden at the beginning and at the end of the simulation period. The schematic can be expressed by the following equations

$$\mathrm{FB} - \mathrm{IB} = \mathrm{IM} - \mathrm{OM} + E - D \quad (3)$$

$$\mathrm{OF} = \mathrm{OM} - \mathrm{IM} = E - D - \mathrm{FB} + \mathrm{IB} \quad (4)$$

where FB is the atmospheric burden within the region at the end of the simulation period and IB is the atmospheric burden within the region at the beginning of the simulation period. IM, OM, E, and D represent the Hg mass that enters the region, the Hg mass that leaves the region, the atmospheric emissions in the region, and the atmospheric deposition in the region over the simulation period, respectively. OF represents the net atmospheric outflow from the region via atmospheric transport, which can be defined as the difference between the Hg mass leaving the region and entering the region. All the terms are in Mg per period. In this study, we evaluate all the terms driven by Chinese anthropogenic emissions to illustrate the impacts of human activities in terrestrial China on neighboring seas and lands.

3. RESULTS AND DISCUSSION

3.1. Atmospheric Hg Outflow from China. In 2010, human activities in China totally release 641.7 Mg of Hg to the air, 218.7 Mg of which is deposited in terrestrial China and 423.0 Mg of which is transported outside terrestrial China (Figure 1). The atmospheric outflow consists of 65.9% of the total Chinese anthropogenic emissions, and the large contribution indicates that China serves as both an important Hg emitter and an important Hg exporter globally. For the whole year, the atmospheric burden over China driven by Chinese anthropogenic emissions remains nearly constant.

Moreover, seasonal variations are observed for atmospheric Hg deposition and outflow for China (Figure 1b). In summer months, large near-source deposition occurs in China due to the large amount of rain, which increases atmospheric deposition over terrestrial China and subsequently reduces atmospheric outflow from terrestrial China. The largest deposition (24.2 Mg $month^{-1}$) and smallest outflow (27.7 Mg $month^{-1}$) occur in August. In winter months, in contrast, the atmospheric deposition over terrestrial China decreases and atmospheric outflow increases subsequently due to the lower amount of rain and limited scavenging in winter in China. The smallest deposition (13.1 Mg $month^{-1}$) and largest outflow (41.4 Mg $month^{-1}$) values occur in February. The seasonal difference reveals that human activities in China contribute more to domestic Hg pollution in summer months and neighboring Hg pollution in winter months.

3.2. Impacts of Interprovincial Trade on the Atmospheric Outflow. Human activities in China released a total of 641.7 Mg of Hg to the air in 2010, 503.0 Mg of which is related to the final consumption of the Chinese population (SI, Data set S4). The remaining mass is related to noneconomic activities (i.e., residential coal consumption and the use of Hg-added products) and foreign consumption. For a specific province in China, the final consumption includes consumption of local goods and imported goods, inducing local emissions (i.e., on-site emissions embodied in the own consumption of the province) and emissions in other provinces that are involved in interprovincial trade (i.e., EEI), respectively. For the whole nation, the imports of goods and services induce 227.6 Mg $year^{-1}$ of Hg emissions. Meanwhile, among the EEI, 85.6 Mg $year^{-1}$ of Hg is embodied in net interprovincial trade, which represents the net transfer from embodied Hg importers to embodied Hg exporters within China (SI Data set S4).

Figure 2 illustrates the impacts of interprovincial trade on the net atmospheric Hg outflow from the whole boundary of China. Due to Chinese interprovincial trade, approximately 1.9 Mg $year^{-1}$ of net Hg would have been transported outside China, but it is deposited in terrestrial China (1.6 Mg $year^{-1}$) and stored in the atmosphere (0.3 Mg $year^{-1}$). Most of the embodied Hg importers are developed provinces in China

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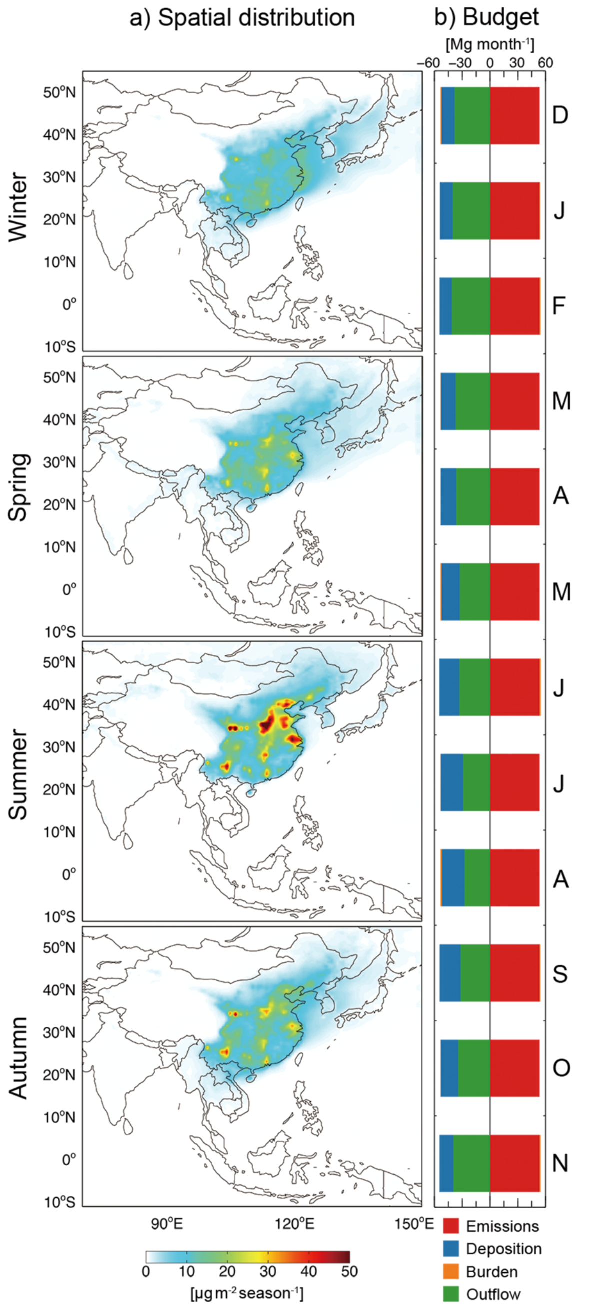

Figure 1. Spatial distribution of atmospheric Hg deposition over East Asia driven by Chinese anthropogenic emissions (a) and the Hg mass budget for China (b). The season "winter" includes December (D), January (J), and February (F). The season "spring" includes March (M), April (A), and May (M). The season "summer" includes June (J), July (J), and August (A). The season "autumn" includes September (S), October (O), and November (N). The mass budget includes atmospheric emissions, atmospheric deposition, atmospheric burden, and atmospheric outflow.

located on the southeast coast, while most of the embodied Hg exporters are developing provinces in China located in inland regions. As a result of interprovincial trade, emissions flow from the southeast coast to inland regions (SI, Figure S2), which was also reported by previous studies.[26,28] These emissions are easily deposited in terrestrial ecosystems and stored in the atmosphere over terrestrial China due to the greater distance from the terrestrial boundaries.

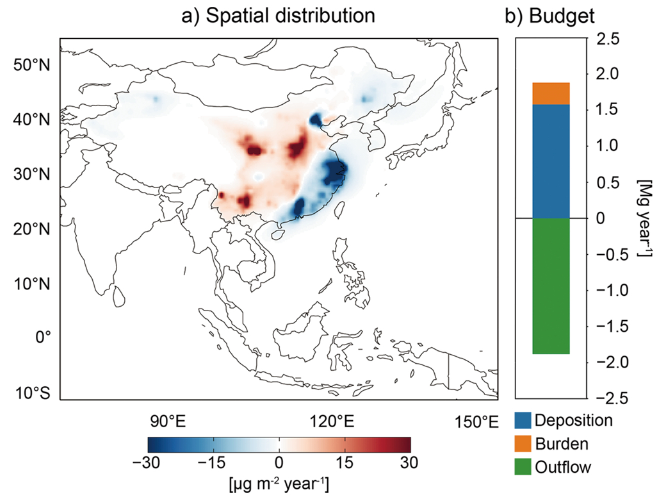

Figure 2. Changes in the spatial distribution of atmospheric Hg deposition over East Asia (a) and changes in the Hg mass budget for China (b) as driven by Chinese interprovincial trade.

Compared to the small net change for the whole boundary, interprovincial trade contributes more to the change in spillover channels. The emission flows from the southeast coast to inland regions during interprovincial trade contribute to the decrease in spillover from the eastern terrestrial boundary and the increase from the western terrestrial boundary (Figure 3). The decrease from the eastern terrestrial boundary is estimated to be −6.4 Mg year^{-1}, consisting of 7.5% of the emissions embodied in net interprovincial trade, while the increase from the western terrestrial boundary is estimated to be +4.5 Mg year^{-1}. The decrease in spillover from the eastern terrestrial boundary contributes to the decrease in atmospheric deposition over the seas and lands leaving the eastern terrestrial boundary, such as the Chinese coastal seas, the Sea of Japan, Japan−Korea, and some regions in Southeast Asia (Figure 3). However, the increase in spillover from the western terrestrial boundary contributes to the increase in atmospheric deposition over the lands outside the western terrestrial boundary, such as Mongolia, South Asia, and some regions in Southeast Asia (Figure 3).

The transfer of atmospheric outflow from the eastern terrestrial boundary to the western terrestrial boundary is a highly significant change in transboundary Hg pollution from China. As we know, human MeHg exposure stems almost entirely from the consumption of seafood, such as fish and shellfish harvested from marine regions.[38−40] Thus, marine regions are the critical receptors of Hg posing health risks to human beings.[3,41,42] Accordingly, the decrease in atmospheric outflow from the eastern terrestrial boundary and the subsequent decrease in atmospheric deposition over the seas outside the eastern terrestrial boundary may reduce the Hg in seafood harvested from the seas, which can reduce the MeHg exposure risks for humans who rely heavily on marine-based diets, such as the populations of Japan and the Republic of Korea. In general, through the transfer of atmospheric outflow between different boundaries, interprovincial trade reduces the chance of risks to foreign countries derived from transboundary Hg pollution from China.

Moreover, the transfer of atmospheric outflow from the eastern terrestrial boundary to the western terrestrial boundary is characterized by significant seasonal differences (Figure 3). The seasonal differences are attributed to the prevailing wind

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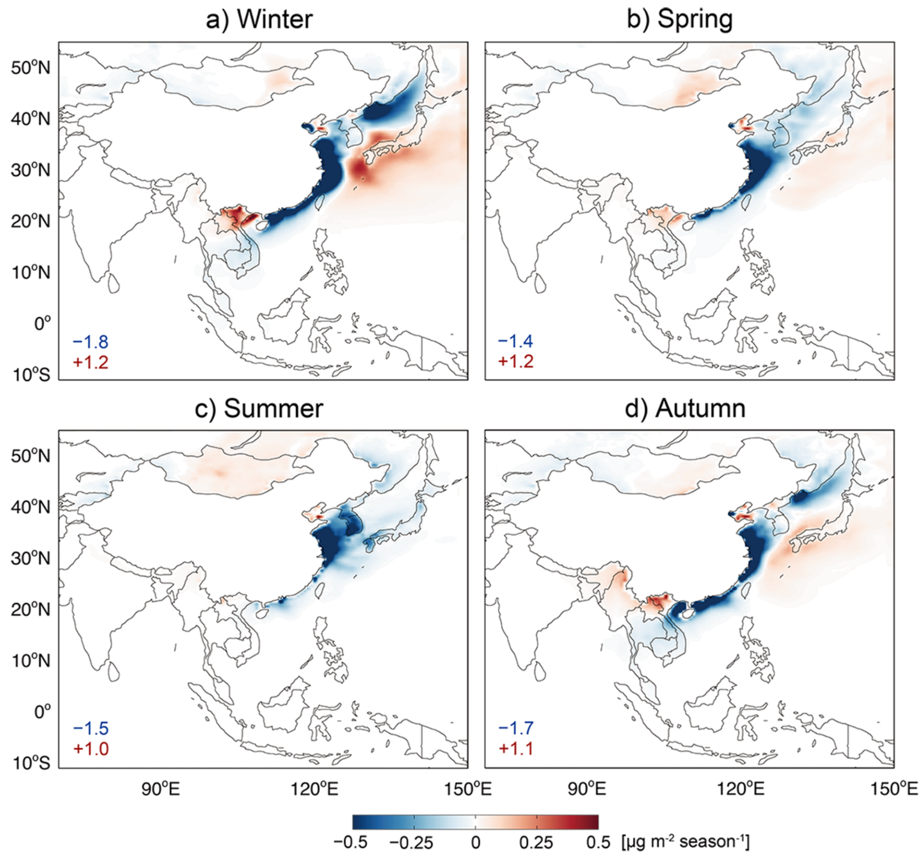

Figure 3. Changes in the spatial distribution of atmospheric Hg deposition over East Asia except for terrestrial China as driven by Chinese interprovincial trade. The definitions of the seasons are the same as those for Figure 1. The number in blue located in the lower left corner of each panel indicates the sum of decreasing global atmospheric deposition driven by Chinese interprovincial trade (unit: Mg $season^{-1}$), and the number in red indicates the sum of increasing global atmospheric deposition driven by Chinese interprovincial trade (unit: Mg $season^{-1}$).

directions of the East Asian Monsoon (SI, Figure S3). In the autumn and winter months, seasonal winds from Siberia to the Northwest Pacific bring atmospheric Hg from the southeast coast of China to the downwind seas outside the eastern terrestrial boundary, and the decrease in deposition occurs over the seas due to exported emissions outside the southeast coast of China (Figure S3a,d). Meanwhile, the seasonal winds also bring atmospheric Hg from southwest China to downwind regions in South Asia, and the increase in deposition occurs over these regions due to imported emissions into southwest China (SI, Data set S4, Figure S2). An increase in deposition is observed over the sea and land near southern Japan during interprovincial trade, which is attributed to both imported emissions into the North China Plain (e.g., Hebei, Shandong) (SI, Data set S4) and the seasonal winds of the East Asian Monsoon. Meanwhile, wind shear over the region may contribute to greater deposition and amplify the increase (Figure S3a,d).

In the spring and summer months, the prevailing wind direction is from the Northwest Pacific to Siberia, the opposite direction of the wind in the autumn and winter months (Figure S3b,c). The seasonal winds bring atmospheric Hg from inland China (e.g., Henan, Gansu, Shaanxi, and Inner Mongolia) to downwind regions outside the western terrestrial boundary, such as Mongolia. An increase in deposition occurs over these downwind regions due to the imported emissions in inland China during interprovincial trade (SI, Data set S4, Figure S2). Due to the seasonal winds, the decrease in atmospheric outflow from the eastern terrestrial boundary of China in the spring and summer months is weaker than that in autumn and winter months. This phenomenon indicates that interprovincial trade reduces the Hg risks to foreign countries mainly in the autumn and winter months. Finally, a decrease in deposition is found along the line of the Yellow Sea, the Korean Peninsula, the Sea of Japan, and northern Japan all the year round. The westerlies of the Northern Hemisphere throughout the year may contribute to this phenomenon, resulting in these regions as the most important regions benefiting from Chinese interprovincial trade.

3.3. Projected Impacts of Interprovincial Trade.

A large potential impact of Chinese interprovincial trade on the changes of atmospheric Hg outflow can be inferred. At present, the relocation of emissions embodied in the net interprovincial trade (85.6 Mg $year^{-1}$) results in the transfer of atmospheric outflow from the eastern terrestrial boundary to the western terrestrial boundary and a net decrease in the atmospheric outflow from the whole boundary. Subsequently, atmospheric deposition of Chinese anthropogenic Hg decreases over neighboring seas and lands outside the eastern terrestrial boundary, for example, −10.5% for Chinese coastal seas and −2.2% for Japan−Korea, and increases over neighboring lands outside the western terrestrial boundary (Figure 4). These changes in outflow and deposition can be amplified with an increase in net interprovincial trade.

In addition to the emissions embodied in net interprovincial trade, the trade induces another 142.0 Mg $year^{-1}$ of Hg

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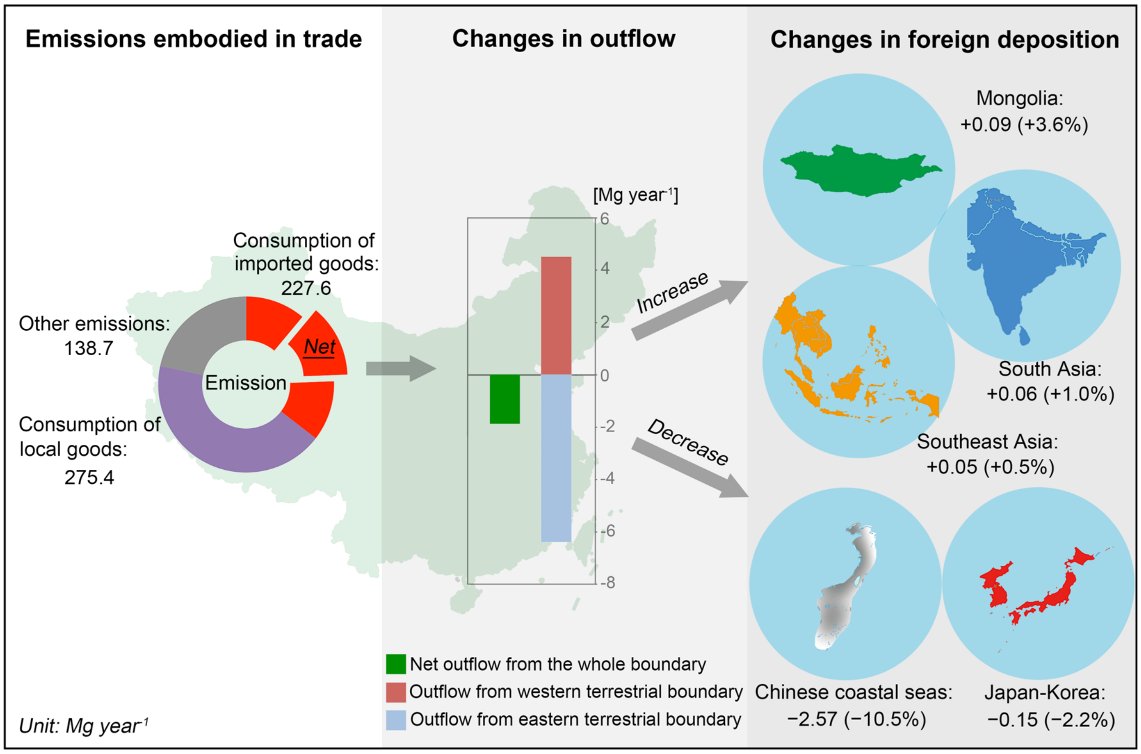

Figure 4. Changes in the atmospheric Hg budget for China and the atmospheric Hg deposition over neighboring seas and lands driven by trade-induced emissions of China. The total national anthropogenic emissions consist of emissions from each province, including the emissions embodied in the consumption of local goods, consumption of imported goods, and other emissions (e.g., residential coal consumption and the use of Hg-added products). The "net" emissions represent the emissions embodied in net interprovincial trade. The unit of all the numbers except numbers in parentheses is Mg year^{-1}. The numbers in parentheses represent the proportion of the change induced by interprovincial trade to the total deposition of Chinese anthropogenic Hg over each neighboring region. The definition of each neighboring region is described in the SI (Data set S1), and the Chinese coastal seas include the Bohai Sea, Yellow Sea, East China Sea, and South China Sea.

emissions that is offset due to trade balance (Figure 4). Meanwhile, the consumption of local goods by the national population also induces 275.4 Mg year^{-1} of Hg emissions that are not involved in interprovincial trade (SI, Data set S4; Figure 4). If the interprovincial trade is strengthened in the future, the original trade balance would be broken and the consumption of local goods would be transferred to the consumption of imported goods. Then, the emissions embodied in net interprovincial trade would become larger, which would further promote the transfer of atmospheric outflow from the eastern terrestrial boundary to the western terrestrial boundary and a net decrease in the atmospheric outflow from the whole boundary given the premise of constant total anthropogenic emissions. This change will further reduce the MeHg exposure risks to humans who live in the regions outside the eastern terrestrial boundary of China and those who rely heavily on the marine-based diets. For instance, Japan−Korea totally receives 6.3 Mg year^{-1} Hg from Chinese anthropogenic emissions. If the decrease responded proportionally to the increase of net interprovincial trade from 85.6 to 503.0 Mg year^{-1}, we would expect a decrease of Chinese anthropogenic Hg from 6.3 to 5.5 Mg year^{-1} deposited in Japan−Korea. That is, the maximum for transboundary impacts of China on Japan−Korea can be reduced by 11%, which is equivalent to the emission ratios of some critical emission sectors in China (e.g., coal-fired power plants). In general, a further reduction in transboundary Hg pollution from China is projected with the increase in net interprovincial trade.

3.4. Policy Implications of Transboundary Hg Controls. Policies on Chinese anthropogenic sources would contribute considerably to the mitigation of Hg-related health risks in Chinese neighboring regions in consideration of the considerable atmospheric Hg outflow. The mitigation would vary between seasons, with more mitigation occurring in the winter months due to the larger atmospheric Hg outflow in those months. Chinese interprovincial trade promotes the transfer of atmospheric outflow from the eastern terrestrial boundary to the western terrestrial boundary and a net decrease in the atmospheric outflow from the whole boundary and an increase in the atmospheric deposition over terrestrial China. The decrease in deposition over neighboring regions outside the eastern terrestrial boundary is more remarkable in the autumn and winter months. Rapid industrialization and urbanization have substantially increased the consumption of goods and services and associated interprovincial trade in the past decades in China.[25] This change reveals that a considerable portion of Hg from Chinese human activities has not been transported outside the eastern terrestrial boundary and has been deposited in terrestrial China or transported outside the western terrestrial boundary due to increasing interprovincial trade in the past decades. Chinese interprovincial trade, to some extent, has offset the adverse effects of increasing anthropogenic Hg emissions from China on neighboring seas and lands outside the eastern terrestrial boundary of China.

Since the 11th Five-Year Plan, the Chinese government has committed to build and develop urban agglomerations, which

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calls for more domestic consumption and domestic imports. More domestic consumption and imports promotes more emissions embodied in interprovincial trade, which will flow from coastal developed urban agglomerations to inland developing regions in the future. The projected increase in interprovincial trade is expected to promote the transfer of atmospheric outflow from the eastern terrestrial boundary to the western terrestrial boundary and the further reduction in transboundary Hg pollution from China. Alongside with the Hg controls implemented in China, interprovincial trade has a synergistic promotional effect on the reduction in atmospheric outflow, especially for the reduction in atmospheric deposition over regions outside the eastern terrestrial boundary of China. Thus, the combination of Hg controls and interprovincial trade could be considered to reduce the transboundary Hg pollution from China.

3.5. Uncertainties and Recommendations. Our model results are subject to uncertainties from various sources, including the compilation of production-based emissions, the calculation of trade-induced emissions, and the simulation of the atmospheric chemical transport model. Since the production-based anthropogenic emissions from China are referenced from our previous work,[28] an overall uncertainty of [−25.0%, 29.0%] from that study is used in this study. These uncertainties are derived from knowledge gaps on Hg concentrations in fuel/raw materials, Hg removal efficiencies, and activity rates. The calculation of trade-induced emissions includes an additional uncertainty from the MRIO analyses, which was estimated to be 13.0% according to previous studies.[22,43,44] These uncertainties are derived from knowledge gaps in economic statistics, sectoral mapping, and data harmonization.[45,46] The simulation of the atmospheric chemical transport model is subject to errors in emission inputs and the model representation of tropospheric physical and chemical processes. However, performing Monte Carlo and other sensitivity simulations that require high computational costs is computationally prohibitive. Instead, we use the normalized root-mean-square deviation (NRMSD) between the simulated and observed results of atmospheric deposition to represent the uncertainties of the model in terms of the method presented by Zhang et al.[44] On the basis of the method and the model evaluations in our previous work,[28] the NRMSD is estimated to be 23.1% for atmospheric Hg simulation over the model domain in 2010. In summary, based on the aggregation of the uncertainties above, we estimate an overall uncertainty of [−34.1%, 37.1%] for atmospheric deposition and outflow in section 3.1 and [−36.4%, 39.3%] for changes in atmospheric deposition and outflow in section 3.2.

The MRIO database used in this study is derived from the work of Liu et al.,[29] while many other MRIO databases have been developed for China, such as those developed by Shi and Zhang,[47] Zhang and Qi,[48] and Wang et al.[49,50] Especially, Wang et al.[49,50] have recently developed a more resolution-detailed and time-detailed MRIO database for China, which also comes with accompanying standard deviation tables. The use of the database from Liu et al.[29] makes it easy to compare our results to those of most previous studies that used the database, and it will be critical and possible to compare and harmonize the results based on the other databases in future studies. Meanwhile, to evaluate the impacts of interprovincial trade on atmospheric deposition over neighboring regions, we choose several representative neighboring seas and lands but do not include all neighboring regions of China in this study. Through additional measurements and models of the marine Hg cycle and human dietary habits, the evaluation of human MeHg exposure risks from the consumption of seafood harvested from marine regions outside the eastern terrestrial boundary of China would assist further analysis of this subject in the future. Moreover, transboundary Hg pollution has interactional impacts between China and neighboring countries. We investigate the impacts of Chinese emissions on neighboring countries from the perspective of interprovincial trade in this study. The impacts of emissions from neighboring countries on China need to be studied from the perspective of international trade in the future.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acs.est.8b03951.

Additional information on study domain and boundaries for regional definitions (Figure S1), net emission flows induced by interprovincial trade in 2010 (Figure S2), seasonal distribution of wind speed and wind direction over the East Asian domain in 2010 (Figure S3) (DOCX)

Additional information on study domain and boundaries for regional definitions (Data set S1), sector classification of the Chinese MRIO table and allocation of emissions in this study (Data sets S2 and S3), provincial production-based and trade-induced emissions and associated sector-specified data in 2010 (Data sets S4 and S5) (XLSX)

■ AUTHOR INFORMATION

Corresponding Authors

*J.S. phone: +86-21-54341198; e-mail: jshu@geo.ecnu.edu.cn.
*X.W. phone: +86-10-62759190; e-mail: xjwang@urban.pku.edu.cn.

ORCID

Long Chen: 0000-0001-9574-7307
Sai Liang: 0000-0002-6306-5800
Jing Meng: 0000-0001-8708-0485
Xuejun Wang: 0000-0001-9990-1391

Notes

The authors declare no competing financial interest.

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