China's retrofitting measures in coal-fired power plants bring significant mercury-related health benefits

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SUMMARY

China has implemented retrofitting measures in coal-fired power plants (CFPPs) to reduce air pollution through small unit shutdown (SUS), the installation of air pollution control devices (APCDs) and power generation efficiency (PGE) improvement. The reductions in highly toxic Hg emissions and their related health impacts by these measures remain poorly understood. Here, we evaluated the health benefits of reduced Hg emissions via retrofitting measures during China's 12th Five-Year Plan (2011-2015) by combining plant-level Hg emission inventories with the China Hg Risk Source-Tracking Model. We found that the measures reduced Hg emissions by 23.5 tons (approximately 1/5 of that from CFPPs in 2010), preventing 30484.77 total points of intelligence quotient decrements and 114 deaths between 2011-2015. These benefits were dominated by CFPP shutdowns and APCD installations, and nearly 50% of provincial health benefits were attributable to Hg reductions in other regions. We suggest that Hg control strategies should consider various factors, such as CFPP locations, population densities and trade-offs between reductions of total Hg (THg) and Hg^{2+}.

KEYWORDS: Hg emissions, coal-fired power plants, retrofitting measures, health benefits.

INTRODUCTION

Mercury (Hg) is a globally recognized contaminant posing great risks to both humankind and ecosystems.[1-3] Scientific evidence shows that organic Hg is the culprit of Minamata disease, one of the major environmental disasters in the 20th century.[4-6] Once emitted from emission sources (e.g. stacks of coal-fired power plants), Hg is involved in atmospheric transport and deposition along with the atmospheric circulation. After deposition, Hg can be converted into methylmercury (MeHg), and accumulated in terrestrial crops (e.g., rice) and aquatic food webs (e.g., marine fish). As a highly toxic substance, MeHg can be transmitted from mother to foetus, and then break through blood brain barrier and damage brain tissues of the foetus, resulting in the developmental disorders of the nervous system and intelligence quotient (IQ) decrements for the foetus.[7, 8] MeHg exposure can also diminish the cardiovascular protective effect of omega-3 polyunsaturated fatty acids, leading to the increase of cardiovascular disease risk toward adults, especially the elderly groups.[9, 10] In 2010, 7360 deaths from fatal heart attacks were related to the intake of MeHg in China.[3] Moreover, other symptoms have been reported among Hg exposure, such as forgetfulness, ataxia of motor function, constriction of the visual fields and damage to reproductive function.[9, 11-14] To protect our society from Hg-related hazards, 128 countries, including China, signed the *Minamata Convention on Mercury (Minamata Convention hereafter)*, which is a legally binding international treaty aiming at Hg control and officially came into effect in 2017.

Coal-fired power plants (CFPPs) are one of the largest anthropogenic sources of global atmospheric Hg emissions, and have been listed as a key sector for Hg emission reduction in the Minamata Convention. According to the latest Global Mercury Assessment 2018, the contribution of CFPPs to global anthropogenic Hg emissions decreased from 16% in 2010 to 14% in 2015, but the Hg emissions from CFPPs in East and Southeast Asia witnessed a growth of 19.2% during 2010-2015.[2] China, as the world's largest coal-fired power generator, emitted 73 tons Hg emissions from CFPPs in 2015, accounting for a quarter of total Hg emissions from global CFPPs.[15, 16] Moreover, a large number of CFPPs in China are located in densely populated areas, leading to an increase in Hg exposure risks. Thus, CFPPs are important for the elevation of MeHg exposure risk in many areas in China.[17] Given that, reducing Hg emissions from CFPPs has been put into China's political agenda.

In addition to Hg, CFPPs are major sources of emissions of CO_2 and pollutants such as $PM_{2.5}$, SO_2 and NO_x. The control of CO_2 and air pollution from CFPPs has been prioritized in China's national political agendas because China has been confronted with mounting international and domestic pressures to mitigate CO_2 emissions and improve the ambient air quality.[18] China's central and local governments have been retrofitting CFPPs in the context of the energy revolution. These retrofitting measures can be generally categorized into three types: 1) small unit shutdown (SUS) aimed at units with a capacity less than 300 MW,[19] 2) installation of efficient air pollutant control devices (APCDs) for reducing air pollutants including PM2.5, SO2 and NOx, 3) power generation efficiency improvement (PGE) via upgrading generation technologies. It has been verified that these retrofitting measures have made great contributions to the reduction of air pollutant and greenhouse gas emissions.[18, 20, 21] For instance, the installation of APCDs resulted in the reduction of emissions of SO_2, NO_x and $PM_{2.5}$ by 20.1 Tg, 3.9Tg and 2.15 Tg respectively during 2005-2025, and 111900 premature deaths have been avoided by the reduced $PM_{2.5}$ exposures.[18] In addition, these retrofitting measures also bring synergetic effects for reducing Hg emissions,[20, 22] an important pollutant with global concern. However, these synergetic effects on Hg reduction and its associated health benefits have not yet been assessed. According to the articles of the Minamata Convention, all the parties are required to regularly facilitate the evaluation of mitigation measure effectiveness and provide an assessment of the related impacts on human health. Thus, the Hg emission reductions and associated health co-benefits from CFPP retrofitting is vital for both fulfilling the international obligation and formulating refined mitigation policies.

Scholars have made initial attempts to investigate the impacts of CFPP retrofitting on Hg emissions. For instance, a recent study found that improving APCD's Hg removal efficiency substantially reduced THg emissions (over 40 t) from China's coal power sector during 2013-2017.[21, 23] However, these studies take all the CFPPs as a whole and cannot explicitly reflect the significant heterogeneity in Hg emissions from individual CFPPs (caused by their strikingly different Hg content in coal and APCD types). Although Liu et al.[24] compiled a high-resolution inventory on Hg from 1817 CFPPs in China based on the detailed APCD parameters, the plant-level retrofitting measures' effect on Hg emission changes remains poorly understood. Moreover, existing studies on Hg-related health impacts are at the national or provincial scale.[1, 3] As the real-world retrofitting actions occurred at the plant-level, a precise assessment comprehensively considering factors such as local population density and Hg deposition is thus in urgent need to reflect each CFPP's contribution to health benefits.

In order to track the route of Hg exposure and quantify the Hg-related health impacts from China's CFPP retrofitting campaign during the 12th Five Year Plan (FYP) period (2011-2015), in this study we combine plant-level Hg emission inventory model and the China Mercury Risk Source-Tracking Model (CMSTM) (details in Experimental Procedures). According to the specific parameters (e.g. coal consumption, application of APCDs, energy efficiency) of each power plant, the high-resolution Hg emission inventories are established to clarify the changes in emissions from CFPPs to the air. The CMSTM model consists of components such as atmospheric transport and deposition of Hg, changes in food MeHg, human intake of MeHg, and related health impacts. Each component tracks an important biogeochemical process between the emissions from stacks of CFPPs and human health. Based on the above models, we for the first time present a high-resolution map of both Hg reduction and related health benefits from the CFPP retrofitting measures. We then compare the benefits between different retrofitting measures, and make targeted suggestion to CFPP retrofitting in different areas by comprehensively considering factors such as CFPP locations and population densities. The findings of this study provide not only a scientific foundation for assessing the implementation of the Minamata Convention on Mercury in China's coal fired power sector but also useful information for more refined mitigation strategies.

Results

Emission reductions from CFPP retrofitting measures

The three CFPP retrofitting measures resulted in an overall emission reduction of 23.51 t (72.38% Hg^0, 26.82% Hg^{2+} and 0.80% Hg_P) during 2011-2015, equivalent to approximately 20% of the THg emitted by China's CFPPs in 2010. Hg emission reduction occurred in 29 out of the 34 provincial regions (Figure 1), among which Jiangsu, Inner Mongolia and Shandong are the top three provinces with greatest reductions (Figure 1A). Among the three retrofitting measures, over half of the national total emission reduction was attributed to the newly installed APCDs, while SUS and PGE had relatively lower contributions. The contributions of the three types of measures in different provinces are shown in Figure 1A. Meanwhile, China's top five state-owned power generation groups (Huaneng Group, Datang Corporation, Huadian Corporation, Guodian Corporation and State Power Investment Corporation), accounting for 46.29% of China's total CFPP capacity, contributed more than half of the emission reductions. The rest of the reductions were made by other large state-owned enterprises, local enterprises (owned by provincial or municipal governments) and private enterprises.

Among the three measures, the increasing APCD installation rate made the largest contribution to emission reductions. As illustrated in Figure 1B, 308 CFPPs in 24 provinces were equipped with new pollution control devices such as wet flue gas desulfurization (WFGD) and selective catalytic reduction (SCR) technology, which helped avoid 12.02 tons of atmospheric Hg emissions (81.69% Hg^0, 17.82% Hg^{2+} and 0.49% Hg_p, see Table S1 for more details). Notably, some of the newly installed APCDs reduced the THg emissions but also led the growth of Hg^{2+} or Hg_p emissions. For example, as SCR was deployed in many CFPPs, Hg^{2+} emissions in Anhui and Guizhou increased by 0.09 tons and 0.04 tons, respectively. This is because SCR can change the speciation profile of Hg emissions in flue gas by oxidizing Hg^0 to Hg^{2+}.[24, 25] SUS contributed 9.19 tons of Hg emission reductions nationwide (Table S1), only second to the reductions attributed to newly installed APCDs. The power generation capacity of the decommissioned CFPPs was equivalent to 5% of the national total in 2010, while the associated reduction in Hg emissions was equivalent to 7.75% of the total emissions from China's CFPPs in 2010, indicating that the decommissioned units were more Hg intensive than the national average level. Improving PGE via the recovery and utilization of waste heat is also an effective way to reduce Hg emissions. During the 12th FYP, the national average coal consumption rate (the amount of coal consumption per kilowatt-hour of electricity generated) of China's CFPPs decreased from 312 g/KWh to 297 g/KWh,[10,18, 19, 26] while PGE reduced atmospheric Hg emissions by 2.3 tons (data for each province are shown in Table S1).

Reduced atmospheric Hg deposition

The three retrofitting measures prevented 5.10 tons of atmospheric Hg deposition in China, of which 2.38 tons for CFPP shutdown, 2.17 tons for APCD installation and 0.55 tons for PGE. Figure 2 illustrates the spatial distributions of the reductions in atmospheric Hg deposition over China. Among all provincial regions, the Hg deposition reduction was the greatest in Inner Mongolia (0.38 t), followed by Shandong (0.36 t) and Hebei (0.35 t). It is interesting to note that there is a significant mismatch between the Hg emission reduction and deposition in many regions. Although the reduction in Hg emissions is higher in Jiangsu (0.26 t) than that in Inner Mongolia, the reduction in atmospheric Hg deposition over Inner Mongolia is 1.5 times greater than that over Jiangsu. The explanation for this phenomenon is that Inner Mongolia has a much larger territory than Jiangsu and therefore experienced a greater reduction in atmospheric Hg deposition from other regions. The air transport effect also played a vital role in the reduction of Hg depositions in each province. It was found that, for the provinces with the top ten largest Hg reductions, more than half of the reductions in Hg depositions resulted from decreases in the Hg transported from other provinces, especially from their neighbouring provinces (summarized in Table S2). For example, Hg deposition in Hebei decreased by 0.35 tons, but less than one-third of this decrease was derived from local CFPPs within Hebei's territory. The largest trans-regional contributors to deposition reductions in Hebei were Shandong (0.08 tons), Shanxi (0.04 tons) and Inner Mongolia (0.04 tons). Although there was no reduction in Hg emissions in Xizang, the province experienced a decline of 0.06 tons in Hg deposition due to the emission reduction measures of CFPPs in other provincial regions.

The health benefits from CFPP retrofitting

The total Hg reductions from CFPP retrofitting prevented 30484.77 total points of intelligence quotient (IQ) decrements and 114 deaths from fatal heart attacks (points and deaths hereafter) in total during 2011-2015 compared with 2010, equivalent to 9.09% and 9.26% of the total IQ decrements and deaths from fatal heart attacks caused by the Hg emissions from CFPPs in China in 2010.[3] As illustrated in Figure 3, the health benefits in each province had large spatial variability (see details in Table S3), and approximately 70% of the contributions (10720.97 points and 78 deaths) came from the top ten provincial regions with the greatest emission reductions.

Among the three emission reduction measures, the largest health benefits came from SUS, which contributed to the prevention of 15374.63 points of IQ decrements and 59 deaths, respectively. It is interesting to note that the SUS-associated reduction in Hg depositions was greater in Inner Mongolia than in Jiangsu, but the number of avoided deaths associated with SUS in Jiangsu was more than tenfold greater than that in Inner Mongolia, as Jiangsu has a much higher population density and MeHg concentration in consumed food[3]. Although reduced Hg emissions were more from the installation of APCDs than the SUS, the former contributed less to health benefits (11858.16 points and 43 deaths) than the latter. The health benefits associated with newly installed APCDs were undermined by devices such as SCR, which remove THg emissions from flue gas and change the Hg species by increasing the proportions of

Hg^{2+} and Hg_P that are easily deposited, thus adversely affecting human health[1]. Therefore, an undesirable situation occurred in Anhui Province: the health risks related to Hg pollution increased despite deceasing THg emissions. Compared with SUS and the installation of APCDs, PGE had much smaller contribution to health benefits (3251.99 points and 12 deaths). Regarding the health benefits gained by Hg reduction through efficiency improvement nationwide, only Zhejiang and Sichuan avoided more than 1 death.

Health benefits by power generation capacities

Most of the reductions in health impacts associated with PGE (avoided IQ decrements and fatal heart attacks: 2425.76 points and 9 deaths) and APCD installation (10702.69 points and 39 deaths) were attributed to large CFPPs with capacities over 300 MW, accounting for 74.36% and 89.80% of the total reductions, respectively. In particular, CFPPs with capacities over 1200 MW contributed more than half of the health benefits associated with the installation of APCDs. In contrast, the CFPPs with capacities under 100 MW accounted for only approximately 8% and 3% of the health benefits related to PGE improvement and the installation of APCDs, respectively. The CFPP shutdown campaign showed a completely different picture. As this campaign mainly targeted small units, almost two-thirds of the health benefits gained by CFPP decommission (9407.32 points and 37 deaths) were from CFPPs with capacities less than 300 MW.

Health benefits by power generation groups

The CFPPs from the five power generation groups were the leading contributors to the observed health benefits and were responsible for half and two-fifths of the health benefits associated with the newly installed APCDs and decommissioned CFPPs, respectively. Among the five groups, the Huaneng Group made the largest contribution (avoided IQ decrements and fatal heart attacks: 4732.85 points and 18 deaths) to the health benefits, especially for the health benefits of newly installed APCDs. Furthermore, an IQ decrement of 2610.71 and 10 deaths were avoided by SUS of the Guodian Corporation, ranking first among the five groups. The local CFPPs in different regions were the second largest contributor, responsible for approximately 30% of the health benefits of SUS and 20% of the health benefits of the other two retrofitting measures. Moreover, half of the decommissioned CFPPs with capacities less than 100 MW belonged to local enterprises, which have higher emission intensity and lower energy efficiency. The rest of the health benefits were attributed to individuals and private enterprises, captive power plants and other large state-owned enterprises (i.e., China Resources Group and State Grid Corporation of China). Notably, not all CFPP retrofitting measures resulted in health benefits; for instance, health impacts slightly increased because of the newly installed SCR device of a captive CFPP-Taigang stainless steel power plant.

Trans-regional effects for health benefits

As the sources of health benefits related to reductions in Hg deposition were clearly identified in the atmospheric transport model, the transregional effects on health benefits (namely, the source-receptor relationship of health benefits between provincial regions) were illustrated (Figure 4). The three provinces that received the largest cross-boundary health benefits were Zhejiang, Guangdong and Jiangsu, while Shandong, Jiangsu and Hebei served as the largest contributors to other provinces. For example, 5 out of the 10 deaths avoided in Jiangsu Province were attributable to CFPP retrofitting measures in other provincial regions, such as Shandong (14.44%), Hebei (5.74%) and Inner Mongolia (4.53%). The majority of deaths avoided in Shandong (55.05%) and Hebei (71.69%) were credited to the implementation of the three types of CFPP retrofitting efforts in other provinces. The retrofitting measures in Shandong prevented 13 deaths in other provinces, accounting for over 80% of the total contributions of Shandong, and the largest beneficiaries were Jiangsu (9.19%), Zhejiang (8.91%) and Shanghai (8.73%). Meanwhile, these three measures also exerted transregional effects on IQ decrements. More than half of the avoided IQ decrements in Hebei, Shandong and Jiangsu were associated with Hg reductions from CFPPs in other provincial regions (see details in Table S4). Health benefits were subject to not only Hg deposition but also the population density in each province. The amount of Hg deposited over Guangdong was comparable to that deposited over Guizhou, but the health benefits gained by the former were 5 times greater than those gained by Guizhou. The striking difference can be explained by the fact that the population in Guangdong was much larger than that in Guizhou. Furthermore, Sichuan is one of the most densely populated provinces, leading to greater health benefits in Sichuan than in less populated provinces under the same deposition reduction level. For the same reason, the health benefits gained by Inner Mongolia and Shaanxi were mismatched with their contributions to emission reductions (i.e., 2 deaths prevented locally versus 14 deaths prevented in other regions).

Uncertainty and sensitivity analysis

In this study, a Monte Carlo simulation with 10000 samplings was applied to evaluate the uncertainty ranges of the emission reductions and health impacts for each type of retrofitting measures.[3,20] We set P10 and P90 values of the statistical distributions as the lower and upper limits of the uncertainty ranges, respectively, as shown in Table S5. In summary, the uncertainty ranges of emission reductions, avoided IQ decrements and avoided deaths of fatal heart attacks were 13.35-36.67 tons, 5954.84-59032.78 points and 46-256 deaths, respectively. Moreover, a sensitivity analysis has been conducted to assess the sensitivity of the health impacts to various parameters such as population, the current incidence of fatal heart attacks and the probability of the causality of the associations (see details in Sensitivity Analysis of Health Benefits Section). We define the sensitivity coefficients as the change of the results caused by a marginal change in the parameters in the CMSTM. It is found that the sensitivities of avoided IQ decrements and deaths to changes in all parameters are similar. For example, if MeHg concentrations in food products which are induced by changes in the atmospheric Hg deposition decrease by 50%, the changing rates of the IQ decrements avoided by SUS, APCDs, and PGE are all -50%. However, the changing rates of the avoided deaths from fatal heart attacks related to SUS, APCDs, and PGE are -50.00335% (29.43), -50.00071% (6.23), and -50.00257% (21.54), respectively. The results show that the changing rates of $\Delta\mathrm{IQ}$ due to the change in food MeHg concentrations equal to the changing rate of food MeHg concentrations, while the changing rates of $\Delta\mathrm{CF}$ due to the change in food MeHg concentrations were very close to the changing rate of the food MeHg concentrations.

Discussion

For the first time, we performed a quantitative analysis of the health benefits of Hg reduction-related gained by unit-specific CFPP retrofitting measures during the 12th FYP in China. The results indicated that China has gained significant health benefits from Hg reductions by CFPP retrofitting, with 23.51 tons Hg emissions (17.14 tons of Hg^0, 6.35 tons of Hg^{2+} and 0.19 tons of Hg_P) avoided during this period. This figure equals to nearly one-fifth of the Hg released from China's CFPPs in 2010. The health benefits of the three different measures applied to CFPPs in China as a whole and in each provincial region were also quantified, respectively. The total reductions in Hg emissions prevented114 deaths from fatal heart attacks and 30484.77 points of IQ decrements, more than half of which were attributable to small-unit shutdown.

Guangdong, Jiangsu and Zhejiang provinces had the largest health benefits, accounting for nearly 30% of the national total. Therefore, this study provided a useful tool for assessing the impacts of plant-level retrofitting measures on air quality and human health. It improves the current framework for evaluating the benefits of retrofitting measures. In light of the tool, the prioritized CFPPs with large Hg emissions and health impacts can be identified, thus helping decision makers to develop a roadmap for Hg reduction from CFPPs. Moreover, the results provided valuable information for CFPPs to implement more targeted retrofitting measures that can maximize the health benefits associated with Hg mitigation, in the context of the Minamata Convention.

Plants decommissioning remains to be the priority

Although significant progress has been made in reducing Hg emissions via SUS, coal power industry still faces with great challenges given that a large number of pollutant intensive small units remain in operation. At the early stage of the 13th FYP (2016-2020), the total capacity of existing small units in China was 212.5 GW, equivalent to the total capacity of CFPPs in the USA. Since a large number of small units in China will come to the end of their service life before 2030, SUS will maintain to be the focus of pollution mitigation strategies for the next decade. From the view of provincial regions, the extant small units concentrated in several provincial regions such as Shandong (29.45 GW), Inner Mongolia (12.57 GW), Henan (10.61 GW), Jiangsu (10.23 GW), and Shanxi (9.62 GW), which together occupy approximately half of the national total. On one hand, small units, especially those with capacities under 300 MW, failed to meet the increasingly stringent environmental requirements even after ultra-low emission retrofit. On the other hand, the costs of efficiency improvements and the installation of APCDs are beyond the affordability of small units. Incentivized by the increasingly stringent environmental requirements (e.g., Emission Standard of Air Pollutants for Thermal Power Plants[27]), China will commit to promoting the early-retirement policy among small units with intensive pollution in the future.[28] However, if all the small units are decommissioned radically in a short time, some regions may encounter large power gap. For example, in Shandong Province (China's third largest provincial economy), small units with capacity less than 300MW still take over 35% of the province's total capacity. If all the small units are decommissioned immediately, Shandong will face a power shortfall of more than 100 million kilowatt hours.[29, 30] Thus, an appropriate timetable for provincial CFPP shutdown campaign in China is recommended, especially in regions where small units still play an important role. Meanwhile, the specific development plans for alternative energy are needed. For the provinces with abundant renewable energy resources, such as Inner Mongolia, Gansu and Qinghai, their local renewable energy could be further explored as a substitution for coal power. For provinces such as Shandong and Jiangsu, where coal power dominated and the potential of renewable energy is comparatively smaller, replacing pollutant-intensive small units by large ones with higher Hg removal efficiency could be a feasible policy for Hg emission control.

More efficient and targeted measures are required

The results demonstrate that health benefits associated with the reduced Hg due to PGE improvement and the installation of APCDs mainly occurred in large CFPPs with capacities over 300 MW. In addition, improving the PGE by upgrading the boiler requires substantial financial investments, which impedes the small units to pursue technology innovation and equipment upgrade. Furthermore, over half of the CFPPs are owned by the five major power generation groups in China. These five groups contributed approximately 50% of health benefits related to newly-built APCDs in this study. Therefore, the large CFPPs owned by the five state-owned power generation groups with sufficient funds for retrofitting should be prioritized when advocating PGE improvement and the application of efficient APCDs. According to the "Ultra-Low Emission and Energy Saving of Coal-fired Power Plant Plan",[28] the capacity of newly built CFPPs after 2014 must be above 600 MW, and energy efficient technologies such as ultra-supercritical boilers should be introduced. Thus, large CFPPs are predicted to take the lion's share of Hg reductions and their related health benefits brought by improving coal combustion efficiency and increasing APCDs application rate in the foreseeable future. Owing to the inherent characteristics of existing equipment, including steam pressure, temperature and combustion mode, the coal consumption rate may not be radically improved in a short time period, but the benefits of Hg emission reductions will emerge gradually over a long period[20]. Notably, phasing out recently built small units installed with adequate APCDs will waste money and resources, as the units have a long service life. Therefore, it is encouraged to transform these small units into cogeneration units (combine heat and power generation) to improve their overall energy utilization efficiency.

We also found that the application of new APCDs (e.g., SCR) in a few regions (e.g., Anhui) led to an undesired outcome that the amounts of Hg^{2+} and Hg_P emissions rose with decreasing THg emissions. Increases in Hg^{2+} and Hg_P cause increased human health impacts, as the possibility of the intake of MeHg increases. This undesirable situation suggests that when selecting Hg control devices, policy makers should prioritize the reduction of human health impacts rather than the reduction of THg. However, the latest pollutant emission standards for CFPPs set only a total emission reduction of 0.03 mg/m^3, while standards for Hg^{2+} and Hg_P have been lacking[27]. Thus, the emission limits of Hg^{2+} and Hg_P for CFPPs should also be included in environmental mandates in the future. Moreover, for those CFPPs with increased health impacts after the use of new control devices, measures that are able to reduce emissions of all Hg species should be implemented. For instance, if ESP is replaced by ESP-FF or ESP-WESP in the combination of SCR+WFGD+ESP, not only will increase the overall Hg removal efficiency by 25% but also effectively reduced the emissions of Hg^{2+} and Hg_P.[22] Furthermore, devices to meet the particular needs for Hg reduction would be good options such as activated carbon injection which can improve the removal efficiency to more than 90% without after-effect. In addition, there is much on-going research focusing on the removal technology of Hg emissions, including organic catalyst, oxidation mechanism, magnetosphere catalyst and so on.[31-35] In future work, we can follow the requirements of the Minamata Convention to conduct a comprehensive evaluation and enable stakeholders to identify the best available Hg removal devices that are not only able to reduce emissions but also are economically and technically feasible for any given CFPP.

Policy implications of trans-regional health impacts

Our analysis also reveals that mitigating Hg via retrofitting CFPPs produces significant mutual health benefits between different regions (see Figure 4). For example, owing to reductions in Hg emissions of CFPPs in Zhejiang, the number of Hg-related deaths outside the border of the provincial territory is 1.5 times higher than that inside the provincial territory. This is because Hg mitigation in one region not only reduces local Hg deposition but also decreases Hg deposition over neighbouring regions via atmospheric transport. Thus, it is crucial to promote joint efforts for CFPP retrofitting. As mentioned above, the cost for retrofitting is so high that some poor regions may not be able to afford the financial burden, which will hinder transitions to cleaner CFPPs. However, the emission reduction in underdeveloped provinces can also bring significant benefits in developed provinces. For instance, the reduced Hg emissions in Inner Mongolia avoided 7 deaths in total, while over one third of the health benefits occurred in Zhejiang, Fujian, Hebei and Shanghai. Following "Ecological Protection

Compensation Mechanism" policies, the regions which get the benefits from external emission reductions should compensate other regions where the reductions occurred.[36, 37] Consequently, the rich eastern regions can provide western coal-fired power bases with financial and technological supports to reduce the trans-boundary Hg deposition and the corresponding health losses.

Health benefits affected by population density

Furthermore, health benefits are associated not only with the total Hg reduction but also with the population density of each province. It has been verified that the same amount of pollutant emissions would cause greater overall health impacts in areas with higher population density.[3] For instance, the reduction in the Hg deposition of Sichuan is approximately the same as that of Gansu, but the number of avoided deaths in Sichuan is 5 times higher than that in Gansu, as the population in Sichuan is more than 2 times greater than that in Gansu. The results implied that population density should be taken into consideration when selecting sites for CFPPs or factories with large air pollutant emissions. Following the pace of Beijing that has already put a ban on newly-build CFPPs and eliminated all CFPPs by 2017, the densely urbanized Yangtze River Delta and Pearl River Delta should gradually close down outdated capacity and further prohibit the construction of new CFPPs, except for cogeneration projects. Moreover, China's western regions have sparse populations and rich coal resources, while the opposite is true in central and eastern China, so large coal power bases should be established in the western region to expand the scale of coal power transmissions from west to east. Therefore, when formulating the national air pollutant emission reduction strategy, the population density, resource endowments, installed capacities and power structures among different regions should be fully considered.

EXPERIMENTAL PROCEDURES

Resource Availability

Lead Contact

Further information and requests for resources and data should be directed to and will be fulfilled by the Lead Contact, Kuishuang Feng (fengkuishuang@gmail.com).

Materials Availability

This study did not generate new unique materials.

Data and Code Availability

The information on decommissioned CFPPs shown in Table S6 was derived from reports issued by the National Energy Administration.[38] Notably, this study only considered Hg emission reductions resulting from newly installed APCDs and PGE improvement during 2011-2014, as the latest available information on each unit's APCDs and the PGE was for 2014. In addition, the key parameters for plant-specific Hg emission estimation, including estimates of the amount of coal burned, the coal consumption rate and APCD application, were collected from the China Electricity Council and Ministry of Ecology and Environmental Protection (see details in Table S7 and Table S8).[39-41] The data are available for academic use at https://data.mendeley.com/datasets/c9rgdsxxvf/draft?a=55782c57-4d1c-41d6-930f-9b8c91dc67be.

Model description

Three types of measures for reductions in Hg emissions from CFPPs, i.e., SUS, the installation of new APCDs and PGE improvement, were considered in this study. To estimate the change of Hg emissions and related health impacts induced by retrofitting measures during the 12th FYP, this study selected the year 2010 as the base year to evaluate the emission reductions and the total related health benefits caused by retrofitting measures in CFPPs during the period concerned.

A plant-level inventory model for reduced Hg emissions

In this study, we compiled the inventory of reduced Hg emission from CFPPs as a substitute for the original one to serve as the first component in the CMSTM model.

The plant-level inventory covers Hg emissions avoided by three types of retrofitting measures implemented by 1119 CFPPs, namely, small unit shutdown aimed at units with a capacity less than 300 MW, installation of efficient air pollutant control devices and power generation efficiency improvement via upgrading generation technologies. The key parameters for each individual CFPP such as coal consumption, application of APCDs and energy efficiency are illustrated in Tables S6-S8. The reduced Hg emissions by each different retrofitting measures are elaborated as follows.

Emission reductions of small unit shutdown campaign. SUS generally refers to units with capacities less than 200 MW. Small units are much more pollution intensive than large units, as they lack APCDs and are installed with low-efficiency subcritical boils.[20, 24, 42] It was reported that the total capacity of decommissioned power plants reached

33.23 GW during the period of the 12th FYP, approximately three quarters of the capacity of Japan. Based on the accurate inventory of decommissioned CFPPs, the atmospheric Hg emissions of 234 CFPPs in 2010 was calculated as follows:[24, 43, 44]

$$E_{ij}^{1} = C_{ij} \times A_i \times (1 - Q_i \times \omega) \times R \times (1 - \eta_{ij}) \tag{1}$$

The notation E_{ij} (ton) is the Hg emissions of power plant *j* in region *i*; *C* (million tons) is the coal consumption of plant *j*; A_i (ton/million tons) is the average Hg content in consumed coal in region *i* (see details in Table S9); Q_i (%) is the percentage of washed coal in region *i*; ω (%) is the Hg removal efficiency for coal washing; *R* (%) is the release ratio of Hg in flue gas compared with the Hg concentration in feed coal (the value of R is usually about 99%[24,25]); and η_{ij} (%) represents the Hg removal efficiency of APCDs in power plant *j* in region *i* (see details in Table S10).

Emission reductions from newly installed APCDs. By the end of 2010, the CFPPs without desulfurization and denitration facilities accounted for 14% and 88% of the national total,[25] respectively. In recent years, the Chinese government issued a series of policies for end-of-pipe control facilities, such as the "Emission standard of air pollutants for thermal power plants"[27] and "Ultra-Low Emission and Energy Saving of Coal-fired Power Plant Plan"[28], aiming at the large-scale application of APCDs that are able to remove Hg from flue gas.[22, 45] The newly installed APCDs, including desulfurization and denitration facilities, doubled the Hg removal efficiency compared to boils only equipped with dust collectors. The calculation of the emission reductions by newly installed APCDs is expressed as follows:[22, 45]

$$E_{ij}^{2} = C_{ij}^{t2} \times A_i \times (1 - Q_i \times \omega) \times R \times ((1 - \eta_{ij}^{t1}) - (1 - \eta_{ij}^{t2})) \tag{2}$$

The notation E_{ij}^{1} (ton) is the reduction in emissions from newly installed APCDs; η_{ij} is the Hg removal efficiency of power plant *j* in region *I*; t1 and t2 represent 2010 and 2014 respectively.

Emission reductions by improving PGE. The coal consumption rate is the overall energy efficiency of power plants, implying that PGE improvements can reduce Hg emissions by reducing the coal consumption over one kilowatt hour of electric power. During the period concerned, a large-scale technical reform was launched for CFPPs, and the emission reduction of 571 CFPPs was calculated with the specific parameters of this study. The amount of coal saved via the decrease in the coal consumption rate was estimated by using the following equation:

$$E_{ij}^{3} = P_{ij}^{t2} \times (CCR_{ij}^{t1} - CCR_{ij}^{t2}) \times A_i \times (1 - Q_i \times \omega) \times R \times (1 - \eta_{ij}^{t2}) \qquad (3)$$

The notation P_{ij}^{t2} (kwh) is the electric energy production of power plant j in region i in t_2, and CCR_{ij} (ton/kwh) is the coal consumption rate of power plant j in region i.

The CMSTM model

The CMSTM model is used to evaluate the impacts of changing Hg emissions from CFPP on human health in China in this study. The model was first developed by Chen as el (2019).[3] Briefly, the model consists of five main components: the atmospheric Hg emission inventory, an atmospheric transport model, changes in food MeHg caused by atmospheric deposition, a human intake inventory of MeHg, and human health impacts due to MeHg intake. Two other components including Hg emissions from natural sources and foreign anthropogenic sources, and imports of food products from foreign countries serve as two external components that are quantified as constant contributions. We briefly describe the model as follows in this study, and more details about the model can be accessed when visiting the website.

First, Hg emission reductions in coal-fired power plants due to each of the three types of measures (i.e., SUS, the new installation of APCDs and PGE improvement) are grouped by provinces, volumes, and companies, respectively. Hg emission reductions in each group are treated as the atmospheric Hg emission inventory. We use the positions of the coal-fired power plants to spatialize the Hg emission reductions, which are used as the input of the atmospheric transport model.

Second, the chemical transport model of GEOS-Chem v9-02 is used to simulate atmospheric Hg transport and deposition over China. The chemical transport model was first introduced and described in details by Bey et al.[50] The model includes cycling of elemental Hg^0, divalent Hg^{II}, and particulate Hg^P between the atmosphere, land, and the mixed layer of the surface ocean.[46-49] A nested simulation at a horizontal resolution of 1/2°×2/3° over China was conducted in the global transport model.[3] A global simulation at a horizontal resolution of 4°×5° provided boundary conditions to the nested simulation. Both simulations were driven by GEOS-5 assimilated meteorological data from the NASA Global Modeling and Assimilation Office (GMAO). We conducted a 3-year simulation for 2008-2010 with 2008-2009 for spinup and 2010 for analysis. The use of the same simulated time interval for each emission scenario isolated the impacts of emissions that were not affected by meteorological factors. Through evaluation against a series of observations, the nested model improved its performance over China.

Third, we estimated changes in Hg concentrations in Hg-containing food products based on the simulated changes in Hg deposition. Hg concentrations in 10 categories of food products (Rice, wheat, beans, vegetables, pork, poultry, milk, eggs, marine fish, and freshwater fish) were estimated according to previous studies[51, 52]. We assumed that changes in MeHg concentrations in food products were proportional to changes in the atmospheric inputs of total Hg (THg) to environmental media, which was consistent with previous studies[53-55]. Changes in MeHg concentrations in food products are caused by changes in the atmospheric Hg deposition in a specific region.

Fourth, we compiled a MeHg intake inventory for the population of China, considering 1) MeHg concentrations in food products, 2) the interprovincial trade of food products, and 3) the evaluation of the estimated daily intake (EDI). MeHg concentrations in food products of different provinces were obtained from previous studies conducted by authoritative scientific research institutions in China and published in peer-reviewed journals. The multiregional input-output (MRIO) model was used to investigate the interprovincial trade of food products. The final demand data for the Farming, Forestry, Animal Husbandry and Fishery sector from the MRIO table are used for the simulation of food trade. The EDI of MeHg was calculated with the MeHg concentrations and the intake rate of each category of food product. The equation for the calculation of EDI is provided below:

$$EDI_j = \sum_{ik} \frac{SC_{ijk} \times I_{ij} \times C_{ik}}{W} \tag{4}$$

where SC_{ijk} is the source contribution of food i in province j driven by the supply of province k. I_{ij} indicates per-capita intake rate of food *i* by the population in province *j*. W represents average body weights of Chinese adult males and females. C_{ik} is the MeHg concentration in food *i* harvested in province *k*, and EDI_j represents the EDI of MeHg by the population in province *j*.

Finally, we evaluated the health impacts of Hg emission reductions in coal-fired power plants in China. Foetal IQ decrements and deaths from fatal heart attacks were evaluated with their dose-response relationships using dietary intake of MeHg, which were based on previous epidemiological studies[53, 56-59]. The assessment of the IQ effects uses the following equation:

$$\Delta IQ = \gamma\lambda\beta(\Delta EDI \times W) \tag{5}$$

The assessment of the deaths from fatal heart attacks uses the following equation:

$$\Delta CF = \sum_g N_g \times Cf_g \times \omega \times (1 - exp\,(-\varphi\lambda\beta(\Delta EDI \times W))) \tag{6}$$

where ΔIQ represents the changes in IQ points, and ΔEDI indicates the changes in EDI of MeHg. The coefficients β (μg Hg/L blood per μg Hg/day), λ (μg Hg/g hair per μg Hg/L blood), and γ (IQ points per μg Hg/g hair) represent the conversion factors of MeHg from intake to blood, blood to hair, and hair to IQ, respectively. ΔCF represents the changes in the deaths from fatal heart attacks due to changes in the intake of MeHg, N_g is the number of people aged ≥ 40 years of gender g, and Cf_g indicates the current incidence of fatal heart attacks among people aged ≥ 40 years of gender g. The coefficient φ (risk per μg Hg/g hair) represents the conversion factors of MeHg in the hair biomarker to fatal heart attack risks. The coefficient ω is introduced to represent the probability of the causality of the associations. W represents the average body weights. A detailed description of the dose-response relationships and their coefficients is provided by Chen at el (Section 5 Human Health Impacts).[3]

Sensitivity Analysis of Health Benefits.

To investigate the sensitivity of China Mercury Risk Source-Tracking Model (CMSTM), we define the sensitivity coefficients as the change of the results caused by a marginal change in each parameter of the CMSTM. The sensitivity coefficients are expressed as partial derivatives of the health risks with respect to each parameter.

In the CMSTM, the assessment of IQ (ΔIQ) and cardiovascular (ΔCF) effects caused by MeHg intake can be briefly expressed as below:

$$\Delta IQ = \gamma\lambda\beta(\Delta EDI \times W) \tag{7}$$

$$\Delta CF = \sum_g N_g \times Cf_g \times \omega \times (1 - exp\,(-\varphi\lambda\beta(\Delta EDI \times W))) \tag{8}$$

where γ represents IQ points per μg Hg g^{-1} hair; λ indicates μg Hg g^{-1} hair per μg Hg L^{-1} blood; β denotes μg Hg L^{-1} blood per μg Hg day^{-1}. Per represents the food trade structure, and *I* indicates the per-capita daily food intake. *C* denotes the total MeHg concentration in food products, while *R* represents the assumption that MeHg concentrations in food products respond proportionally to changes in deposited Hg. *P* is the number of people aged ≥40 years, while *CF* indicates the current incidence of fatal heart attacks among people aged ≥40 years. The coefficient φ represents the conversion factors of MeHg in the hair biomarker to fatal heart attack risks (risk per μg

Hg g^{-1} hair). The coefficient ω represents the probability of the causality of the associations. The full details of the equations are given in our previous study.[3]

Consequently, the sensitivity coefficients for parameters assessing IQ effects are given in the following equations.

$$\frac{\partial \Delta IQ}{\partial r} = \lambda\beta(Per \times I \times C \times R) \tag{9}$$

$$\frac{\partial \Delta IQ}{\partial \lambda} = \gamma\beta(Per \times I \times C \times R) \tag{10}$$

$$\frac{\partial \Delta IQ}{\partial \beta} = \gamma\lambda(Per \times I \times C \times R) \tag{11}$$

$$\frac{\partial \Delta IQ}{\partial Per} = \gamma\lambda\beta(I \times C \times R) \tag{12}$$

$$\frac{\partial \Delta IQ}{\partial I} = \gamma\lambda\beta(Per \times C \times R) \tag{13}$$

$$\frac{\partial \Delta IQ}{\partial C} = \gamma\lambda\beta(Per \times I \times R) \tag{14}$$

$$\frac{\partial \Delta IQ}{\partial R} = \gamma\lambda\beta(Per \times I \times C) \tag{15}$$

It can be observed that ΔIQ is in a linear correlation with all the parameters, which suggests 1) consistent sensitivity results of all the parameters to the IQ effect accounting for the equation, and 2) The changing rate of ΔIQ due to the change of each parameter equals to the changing rate of the parameter. Meanwhile, for the cardiovascular effects, the sensitivity coefficients for parameters are given in the following equations.

$$\frac{\partial \Delta CF}{\partial P} = Cf \times \omega \times \{1 - exp[-\varphi\lambda\beta(Per \times I \times C \times R)]\} \tag{16}$$

$$\frac{\partial \Delta CF}{\partial Cf} = P \times \omega \times \{1 - exp[-\varphi\lambda\beta(Per \times I \times C \times R)]\} \tag{17}$$

$$\frac{\partial \Delta CF}{\partial \omega} = P \times Cf \times \{1 - exp[-\varphi\lambda\beta(Per \times I \times C \times R)]\} \tag{18}$$

$$\frac{\partial \Delta CF}{\partial \varphi} = -\lambda\beta(Per \times I \times C \times R) \times P \times Cf \times \omega \times exp[-\varphi\lambda\beta(Per \times I \times C \times R)] \tag{19}$$

$$\frac{\partial \Delta CF}{\partial \lambda} = -\varphi\beta(Per \times I \times C \times R) \times P \times Cf \times \omega \times exp[-\varphi\lambda\beta(Per \times I \times C \times R)] \tag{20}$$

$$\frac{\partial \Delta CF}{\partial \beta} = -\varphi\lambda(Per \times I \times C \times R) \times P \times Cf \times \omega \times exp[-\varphi\lambda\beta(Per \times I \times C \times R)] \tag{21}$$

$$\frac{\partial \Delta CF}{\partial Per} = -\varphi\lambda\beta(I \times C \times R) \times P \times Cf \times \omega \times exp[-\varphi\lambda\beta(Per \times I \times C \times R)] \tag{22}$$

$$\frac{\partial \Delta CF}{\partial I} = -\varphi\lambda\beta(Per \times C \times R) \times P \times Cf \times \omega \times exp[-\varphi\lambda\beta(Per \times I \times C \times R)] \tag{23}$$

$$\frac{\partial \Delta CF}{\partial C} = -\varphi\lambda\beta(Per \times I \times R) \times P \times Cf \times \omega \times exp[-\varphi\lambda\beta(Per \times I \times C \times R)] \tag{24}$$

$$\frac{\partial \Delta CF}{\partial R} = -\varphi\lambda\beta(Per \times I \times C) \times P \times Cf \times \omega \times exp[-\varphi\lambda\beta(Per \times I \times C \times R)] \tag{25}$$

These equations show that ΔCF is in a linear correlation with P, *Cf*, and ω, but not with the other parameters, due to the part of $exp[-\varphi\lambda\beta(Per \times I \times C \times R)]$ in their partial derivatives. However, the calculation results of $exp[-\varphi\lambda\beta(Per \times I \times C \times R)]$ are very close to 1 in this study (within the range of [0.9995, 1] for each element in the exponential expression). This shows that the correlation between ΔCF and these parameters are very much close to linear. Consequently, the sensitivity results of all the parameters to the cardiovascular effects are also consistent accounting for the equation.

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AUTHOR CONTRIBUTIONS

J.L., S.L., L.C., and K.F. conceived the idea and supervised the entire project. J.L. and S.Z. evaluated the specific emission reduction of Hg from three retrofitting measures. J.L., led the construction of a plant-level inventory model for reduced Hg emissions. W.W., and J.M. collected the requisite data of each power plant. B.C., N.Z., and D.G. contributed to the planning and coordination of the project. L.C., led the construction of GEOS-Chem chemical transport model. L.C., S.L., J.Q., Y.L., and X.W. estimated health impacts through the Hg biogeochemical cycle. S.L., led the estimate of changes in Hg concentrations in Hg-containing food products. K.F., J.L., S.Z. and W.W. co-wrote the manuscript. K.F. led the drafting of this manuscript. All authors discussed the results and commented on the manuscript.

DECLARATION OF INTERESTS

The authors declare that they have no competing interests.

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Figures Legends

- **Figure1. Reduced Hg emissions by three retrofitting measures (unit: kg)**. (**A**) Reduced Hg by three retrofitting measures. (**B**) Reduced Hg by newly installed APCDs. (**C**) Reduced Hg by SUS. (**D**) Reduced Hg by PGE. (The pie charts in Figure 1A show the proportion of Hg reduced by different retrofitting measures. The gradations of colours in the map represent the amount of emission reduction in each province. The dots in Figures 1B-1D represent the emission reduction from each power plant, and indicates the specific location of power plants. The dotted background means on data or no emission reduced in this province.)
- **Figure 2. Spatial distributions of reduced Hg deposition over China and its surrounding areas** (The results of simulation by chemical transport model, which includes the cycling of Hg emissions between the atmosphere, land, and the mixed layer of the surface ocean. The gradation and density of the colour represent the amount of Hg deposition over China and its surrounding areas.)
- **Figure 3. The spatial distributions of health benefits over China** (The gradations of colours in

the map represent the amount of avoided deaths in each province. The histogram charts represent the proportion of deaths avoided by different retrofitting measures. The dotted background means on data or no health benefits in this province.)

- **Figure 4. Avoided deaths attributable to local and external Hg reduction** (Both the receptors and sources represent different provincial regions; x-axis shows the receptors of avoided deaths by Hg reduction in sources; y-axis shows the sources where their local Hg reduction contributed to avoided deaths in receptors; The grids indicate avoided deaths in receptors brought by Hg reductions in sources. Beijing (BJ); Tianjin (TJ); Hebei (HEB); Shanxi (SX); Inner Mongolia (IM); Liaoning (LN); Jilin (JL); Heilongjiang (HLJ); Shanghai (SH); Jiangsu (JS); Zhejiang (ZJ); Anhui (AH); Fujian (FJ); Jiangxi (JX); Shandong (SD); HEN (Henan); Hubei (HB); Hunan (HN); Guangdong (GD); Guangxi (GX); Hainan (HAN); Chongqing (CQ); Sichuan (SC); Guizhou (GZ); Yunnan (YN); Xizang (XZ); Shaanxi (SHX); Gansu (GS); Qinghai (QH); Ningxia (NX); Xinjiang (XJ).)