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Mercury distribution and emission reduction potentials of Chinese coal-fired industrial boilers

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Abstract
Coal-fired industrial boilers (CFIBs) are an important emission source of mercury. In this study, the mercury flow of the CFIBs in China, including mercury input and output (atmospheric mercury and mercury-containing wastes), was assessed. Besides, the mercury output inventory of wastes (ashes and desulfurization products) from the Chinese CFIBs in 2015 was established for the first time based on the established activity database and updated mercury removal efficiencies. The total mercury inputs of Chinese CFIBs in 2015 were estimated to be 148.4 t, of which the boilers with a capacity of 10 t/h or below were the dominant source, with a total contribution of 41.6%. The atmospheric mercury emissions of CFIBs were 80.7 t, while the total mercury entering wastes including ashes and desulfurization products were 67.77 t. Among them, the mercury in ashes accounted for 80.8% of the mercury in wastes in the country. Provinces with high atmospheric mercury emission intensity (> 3000 g/km^2 and > 100 mg/cap) were primarily concentrated in North China, Northeast China, and Northwest China with high heating demands in winter. Significant reductions of mercury emissions were achieved under four scenarios from 2015 to 2030, where the mercury emissions decreased by 75.4–84.6%. However, the reduction potential of mercury emissions narrowed from 2025 to 2030, with an average annual decline of 3.3–6.0%. Besides, based on the energy policies and environmental control measures, three mercury emission reduction paths were proposed for the stages of 2015–2025 and 2025–2030. The uncertainties of mercury inputs, atmospheric mercury emissions, and mercury in wastes of the CFIBs in China in 2015 were estimated at the ranges of − 18.2%– + 20.8%, − 25.1%– + 29.7%, and − 40.7%– + 48.8%, respectively.

Keywords Coal-fired industrial boiler (CFIB) · Mercury · Emission inventory · Mercury-containing wastes · Scenario projection · Emission reduction potential

Introduction

Mercury as a kind of toxic substance has adverse effects on the ecological environment and human health due to its persistence, long-distance migration, high bio-enrichment, and high biotoxicity. In recent years, mercury has become an important global toxic and harmful environmental pollutant (Li et al. 2020; Zhang et al. 2016; Zhao et al. 2017). Human activities have raised the total concentration of mercury in the atmosphere by about 450% above the natural levels (UNEP 2019). In October 2013, 91 countries and governments, including China, signed *the Minamata Convention on Mercury*, in which coal-fired industrial boilers (CFIBs) were identified as one of the five key sources of atmospheric mercury emissions listed in Annex D of the Convention (UNEP 2013). Specifically, the Convention proposed establishing and maintaining an inventory of mercury emissions and release from relevant sources within five years and further formulating a national plan and proposing various measures to control emissions.

Mercury introduced into the combustion system of CFIBs by coal feeds can exit the system through a limited number

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of pathways (Xu et al. 2003). Mercury in coal enters the flue gas through various physic-chemical processes during coal combustion (Tian et al. 2015). The flue gas mercury is partially released into the atmosphere after passing through the air pollution control devices (APCDs), and the rest is dispersed in some wastes, such as fly ash, slag, and gypsum (Tang et al. 2012). Mercury in the aforementioned mercury-containing wastes can be released into the environment again under certain conditions, which has great environmental risks (Hao et al. 2017; Pudasainee et al. 2017). Therefore, the mercury emissions in flue gas and mercury-containing wastes are of great concern for their toxicological and environmental effects.

There are a large number of CFIBs in China, most of which are small in capacity and distributed in densely populated areas. In 2014, the mercury emission limit value (50 μg/m^3) was added for the first time to the *Emission Standard of Air pollutants for Boiler* (GB 13271–2014) in China (MEE 2014). Even though the proportion of coal in China's energy consumption structure is trusted to decline with the implementation of various policies about carbon emission peak and neutrality (CSC 2022), the coal-dominated energy consumption structure will maintain for a certain period of time. Therefore, reducing the atmospheric mercury emissions from the CFIBs remains a huge challenge in China.

At present, the mercury in CFIBs is mainly controlled by the existing APCDs for conventional air pollutants, such as dust removal, desulfurization, and denitration devices (Wu et al. 2016). Mercury emissions of CFIBs have been studied, and basically, the compilation methods of mercury emission inventory and emission prediction have been established (Gao et al. 2021; Wang et al. 2021). Tian et al. (2015) and Cheng et al. (2015) systematically established the atmospheric mercury emissions inventories from anthropogenic sources in mainland China during 1949–2012 and 2000–2020, respectively; however, the mercury removal efficiency of different APCDs of CFIBs was completely adopted the data of coal-fired power plants (CFPPs). Gao et al. (2021) firstly established a highly resolved county-based atmospheric mercury emission inventory for the CFIBs in China, but their research on the potential reductions of mercury emission was only based on the consideration of the *Three-Year Action Plan to Win the Blue-Sky Defence*. Qi et al. (2014) predicted a mercury emission reduction of 100.6–130.8 t in 2020 based on the scenario of setting the atmospheric mercury emission limit of CFIBs but did not take the mercury reduction potential of technological upgrading into account. With the implementation of a series of comprehensive energy policies and emission control measures for CFIBs, the control efficiency of conventional pollutants of CFIBs has been improved, which further affected the control effect of flue gas mercury. However, only limited individual studies have been carried out on the mercury in the wastes of CFIBs (Tang et al. 2012; Tong et al. 2020; Zheng et al. 2018), and the mercury inventory of CFIBs in wastes has not been established. Therefore, there is insufficient data support for China to formulate action plans for mercury emission reduction and fulfill its obligations under the Convention.

This study comprehensively considered the mercury flow and integrated the calculation methods of mercury input and output of the CFIBs. Given the date of entry into force of the Convention and the date of the implementation of GB 13271–2014 in China, this study took 2015 as an example and for the first time established the mercury input inventory and output inventory including atmospheric mercury emissions and mercury in the wastes (ashes/desulfurization products), based on the established activity database and the updated factors of mercury content in consumed coal and the mercury removal efficiency. In addition, scenario projections have been conducted to investigate the mercury emission trend from 2025 to 2030 by comprehensively considering the energy policies and environmental control measures, and to further quantify the reduction potential of atmospheric mercury emissions.

Material and methods

Overall framework of the methods

In this study, a deterministic bottom-up emission factor method was established based on the established activity database and the updated factors of mercury content in consumed coal and the mercury removal efficiency to assess the mercury inputs and outputs (atmospheric mercury emissions and mercury in the wastes) of CFIBs in China. The mercury inputs of the CFIBs were related to the coal consumption and the mercury content in consumed coal, and the calculation method was shown in Eq. (1). The atmospheric mercury emissions of the CFIBs were calculated using the emission factor as shown in Eqs. (2) and (3). The mercury in the wastes (ashes and desulfurization products) was calculated with the consideration of mercury removal efficiency of the APCDs of CFIBs, as shown in Eqs. (4) ~ (6).

$$I = \sum_i [A_i \times M_i] \tag{1}$$

$$E_e = \sum_i \sum_j \sum_k [A_{i,j,k} \times ef_{i,j,k}] \tag{2}$$

$$ef_{i,j,k} = M_i \times R_j \times \prod_s (1 - \eta_{s,k}) \tag{3}$$

$$E_r = E_a + E_d \tag{4}$$

$$E_a = \sum_i \sum_j \left[A_{i,j} \times (1 - R_j) \times M_i + \sum_k \left(A_{i,j} \times M_i \times R_j \times \eta_{PM,k} \right) \right] \quad (5)$$

$$E_d = \sum_i \sum_k \left[\left(I_{i,k} - E_{a,i,k} \right) \times \eta_{SO_2,k} \right] \quad (6)$$

where I, E_e, E_r are the mercury inputs, atmospheric mercury emissions, and the mercury in wastes, t; E_a and E_d are the mercury in ashes (fly ash and bottom ash) and desulfurization products, t; A is the coal consumption of CFIBs, Mt; M is the mercury content in consumed coal, mg/kg; ef is the emission factor, kg/t; R is the mercury release ratio; η is the mercury removal efficiency of the APCDs; i, j, k, and s represent the province, boiler type, type of APCDs and pollutants (PM/SO_2/NOx), respectively. The impact of coal washing was neglectable in this study as the coal washing rate of CFIBs in China was less than 4% according to our previous study (Gao et al. 2021).

Besides, in view of the relationship between atmospheric mercury emissions of CFIBs and geographical location, economic development level, and population, the emission intensity per unit area and per capita in the province were adopted in this study to investigate the spatial distribution characteristics of mercury emissions of the CFIBs in China, as shown in Eqs. (7) ~ (8).

$$IA_j = E_{e,j} \times 10^6 / S_j \quad (7)$$

$$IP_j = E_{e,j} \times 10^5 / P_j \quad (8)$$

where IA and IP are the emission intensity per unit area and per capita, g/km^2 and mg/cap; S is the area, km^2; P is the population size, 10,000 people. Previous studies mainly used the territorial area to calculate the pollutant emission intensity (Liu et al. 2018; Tong et al. 2021). To characterize the atmospheric mercury emission intensity per unit area of the CFIBs in China more accurately, the urban land-use scale index from the *China Statistical Yearbook 2016* was used in this study (NBS 2016).

Activity rates

Based on the environmental statistics from the Chinese Ministry of Ecology and Environment (MEE) on national key enterprises, an activity database of CFIBs at the provincial scale was established in this study, including coal consumption, boiler capacity, boiler types, and installed APCDs. In 2015, there were about 460,000 CFIBs with a gross capacity of about 1.4 million t/h in mainland China. The provincial composition of the capacities of CFIBs in 2015 is illustrated in SI Fig. S1. The CFIBs in China were mainly small-scale boilers (capacity ≤ 10 t/h), accounting for 46% of the total number of CFIBs. Large-scale boilers (capacity > 65 t/h) were primarily distributed in northern heating regions and coal-producing areas, which were related to the heating supply and economic development demands (Gao et al. 2021).

As the CFIBs are applied in various industrial production processes as well as the domestic heating supply in China, coal consumption by the CFIBs is not counted in any specific sector in China's energy statistics. Here, the coal consumption of CFIBs in different provinces in 2015 was calculated by an industry-category method established in this study. The data were obtained from the energy balance table and the terminal energy consumption data in the *China Energy Statistical Yearbook 2016* (DESNBS 2016). The industry-category method was expressed as follows:

$$A_j = A_{H,j} + A_{I,j} - A_{R,j} - A_{C,j} \quad (9)$$

where A_H is the coal consumption of the heating production sector, Mt; A_I is the coal consumption of the industry sector, Mt; A_R is the coal consumption used as raw materials, Mt; A_C is the coal consumption of the cement and steel industry sector, Mt.

The total coal consumption in China in 2015 was about 4,000 Mt, of which about 711 Mt was consumed by the CFIBs (as shown in SI Table S1). The spatial distribution of CFIBs in China is presented in Fig. 1. The coal consumption of CFIBs was mainly distributed in the northern regions such as Northeast China, North China, and the developed coastal regions. Shandong, Inner Mongolia, Liaoning, Shanxi, and Hebei were the top five provinces, accounting for about 35% of the total coal consumption of CFIBs. Grate furnace (GF), circulating fluidized bed (CFB), and pulverized coal (PC) boilers were the three major types of CFIBs in mainland China, where GF was the dominant type and accounted for more than 69% of the number of boilers (as shown in SI Table S2).

Mercury content in consumed coal

The coal resources in China are unevenly distributed, with about 67% of the national coal production coming from Inner Mongolia, Shanxi, and Shaanxi. Tian et al. (2013) summarized the average mercury content in raw coal from 30 provinces and other coal exporting countries using mathematical-statistical methods. Considering the complexity of coal transportation among different provinces, our previous study (Gao et al. 2021) established the coal transportation matrix of each province based on raw material production and energy consumption (DESNBS 2016; NBS 2016; SAMSS 2016), and then obtained the mercury content in consumed coal in different provinces in combination with the mercury content in raw coal from Tian et al. (2013). The mercury contents in raw coal of different provinces in China in 2015 are illustrated in SI Table S3. The average mercury

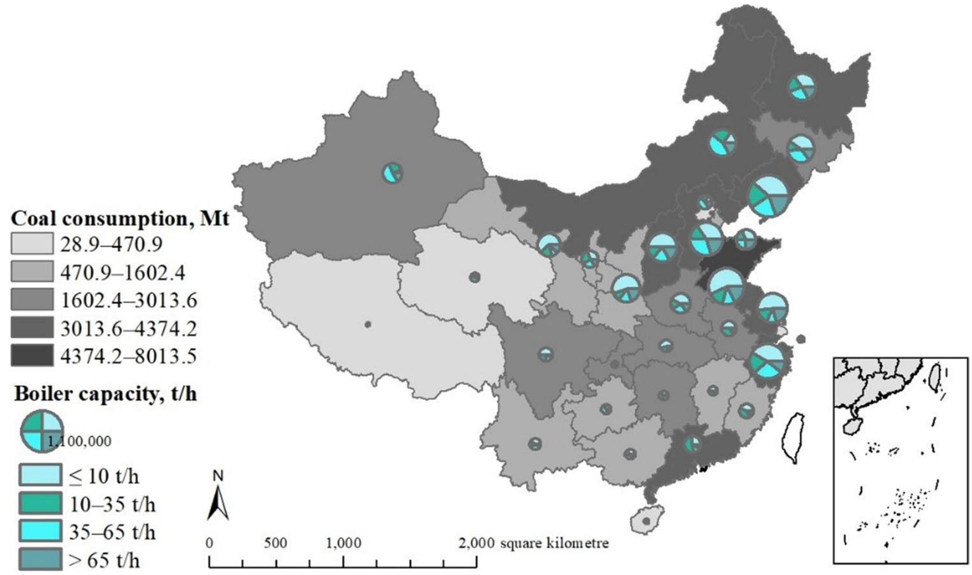


Fig. 1 Provincial distribution of coal consumption and boiler capacity of the CFIBs in China in 2015

content in consumed coal was 0.23 mg/kg in China. The mercury content in consumed coal was relatively high in southwestern China, where the highest mercury content was 0.39 mg/kg in Guizhou.

Mercury release ratio and removal efficiency of APCDs

During the coal combustion process in the boiler, mercury was released from the coal and then redistributed into different output materials along with flue gas flow (Tong et al. 2020; Zhao et al. 2019). The studies of Huang et al. (2004), Reddy et al. (2005), and Kim et al. (2010) have shown that the behavior of mercury release from coal was affected by factors such as the combustion conditions, flue gas conditions, and so on, which was closely related to the type of boilers. No latest research results show that the mercury release ratios of different boiler types have changed, so this study adopted a series of measured results from previous studies on the mercury release ratio of different boiler types, as presented in SI Table S4. (Gao et al. 2021; Huang et al. 2004; Meij et al. 2002; Tong et al. 2020; Wang et al. 2010; Yue et al. 2020; Zhang et al. 2008; Zhang et al. 2015). The mercury release ratio of PC boilers was higher than that of GF and CFB boilers because of the higher combustion temperature (Wei et al. 2017; Wang et al. 2010).

The mercury emission control of CFIBs was mainly carried out by the existing dust removal, desulfurization, and denitration devices (Gao et al. 2021). In our previous study (Tong et al. 2020), the atmospheric mercury emission concentration could be as low as $0.11-0.60$ μg/Nm^3, which was lower than the emission limit (50 μg/Nm^3) in GB 13271–2014. The APCDs of CFIBs were many and various, and their synergistic efficiencies for mercury removal were different. According to the environmental statistics of national key enterprises whose pollutant emissions accounted for 80% of the total pollutant emissions in the region, the composition of dust removal and desulfurization devices of the CFIBs in China in 2015 were presented in SI Fig. S2 and SI Fig. S3. All CFIBs were equipped with dust removal devices, the main three types of which were fabric filter (FF), wet scrubber (WS), and cyclone (CYC). The FF generally showed a higher mercury removal efficiency because of its more stable performance (Tong et al. 2020). Nearly all Hg^P could be simultaneously captured by the electrostatic precipitator (ESP) or FF, resulting in about an $18-30$% reduction in the flue gas mercury concentration (Wang et al. 2010). The wet flue gas desulfurization (WFGD) was the most widely used desulphurization technology for CFIBs, and $67-98$% of Hg^{2+} could be absorbed in wet flue gas and enter the desulphurized products such as gypsum and wastewater (Hao et al. 2017; Liu et al. 2018; Wang et al. 2010; Zhang et al. 2008). In 2015, the installation rate of WFGD for CFIBs in China was not high, where the proportion of small-scale CFIBs installed with WFGD was 8.7%. As for the denitration devices, selective catalytic reduction (SCR) could promote the oxidation of Hg^0 to Hg^{2+}, especially for the coal with high Cl content (Zhang et al. 2012). However, only 6.3% of CFIBs were installed with the denitration devices, because the NOx emission limits in the national standard GB 13271–2014 were only implemented on July 1, 2014. With the stricter emission standards of air pollutants and the improvement of environmental management requirements of CFIBs, advanced APCDs such as FF, FF-ESP, WFGD, and wet electrostatic precipitator (WESP) have been applied to the CFIBs, and the synergistic mercury removal efficiency could be as high as $80.97-98.86$% (Tong et al. 2020). Therefore, this study updated the mercury removal efficiencies of different APCDs for CFIBs in China combined with typical field test data (Li et al. 2017; Pudasainee et al. 2012; Pudasainee et al. 2017; Tang et al. 2012; Tong et al. 2020; Wang et al. 2010; Zhang et al. 2012; Zhang et al. 2015; Zhang et al. 2017), as presented in Fig. 2.

Scenario projections

In recent years, CFIBs have become a key target for air pollution control, subject to a series of energy policies and environmental control requirements. In this study, two energy consumption scenarios and two environmental control scenarios for CFIBs in 2025 and 2030 were developed based on the current policies and future development demands. Two energy consumption scenarios were developed for the coal consumption of CFIBs according to *China's Low Carbon Development Pathways by 2050* (NDRCC 2009) and the *China Energy Outlook 2030* (CERS 2016): the energy reference (REF) scenario assumed that future energy consumption trends would remain unchanged, with the coal consumption of CFIBs falling to 566 Mt and 560 Mt by 2025 and 2030, respectively; and the energy alternative (ALT) scenario assumed that a series of new energy-saving policies would be introduced and gradually implemented, with the coal consumption of CFIBs falling to 482 Mt and 415 Mt by 2025 and 2030, respectively (as shown in SI Fig. S4). Additionally, two environmental control scenarios were designed: the business-as-usual (BAU) scenario assumed that the emission control measures of CFIBs would be maintained and all goals in the *Three-Year Action Plan to Win the Blue-Sky Defence* could be achieved; and the available-control-technology (ACT) scenario assumed that stricter emission standards and policies would be implemented. In the ACT scenario, advanced APCDs would be developed and applied, and the ultra-low emission retrofitting would be carried out to the boilers with a capacity above 35 t/h (as shown in SI Table S6). Therefore, a total of four scenarios (REF-BAU, REF-ACT, ALT-BAU, and ALT-ACT) were designed based on the two sets of scenarios. The mercury emissions of CFIBs in China in 2025 and 2030 were estimated under four designed scenarios. Besides, based on the development of energy consumption and environmental control scenarios, three mercury emission control paths were categorized and proposed: (1) energy substitution with natural gas, electricity, biomass, and other clean energy; (2) phasing out boilers with the capacity less than 35 t/h or 10 t/h; (3) upgrading the APCDs of boilers, including the ultra-low emission retrofitting for boilers with a capacity of 35 t/h or above. The mercury emission reduction potentials were further investigated for these three paths (energy substitution, phaseout of boilers, and upgrading of APCDs).

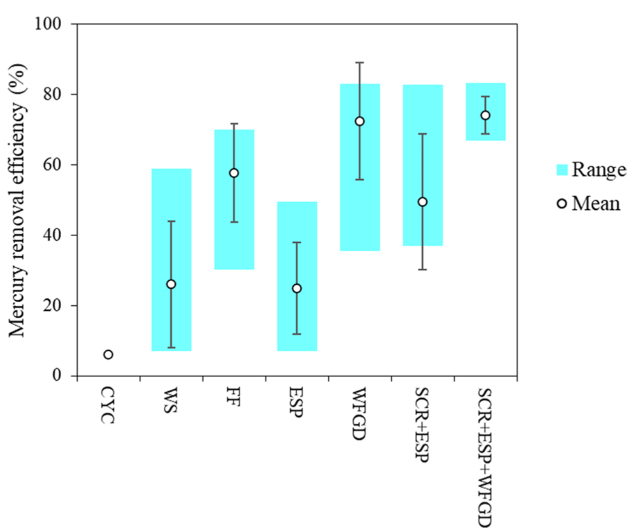


Fig. 2 Mercury removal efficiency of air pollution control technologies for the CFIBs

Results and discussion

Mercury inputs of the CFIBs in China

The total annual mercury inputs of CFIBs in 2015 were estimated to be 148.4 t. The distribution of mercury inputs of the CFIBs in China in 2015 is shown in Fig. 3, where the contributions of different boiler capacity ranges are given, too. Boilers with the capacity of 10 t/h or below were the leading input source, contributing 41.6% of the national total mercury inputs, followed by the boilers of (35, 65] t/h and (10, 35] t/h which contributed 22.5% and 21.4%, respectively. There were a huge number of boilers with a capacity of 10 t/h or below, whose coal consumption accounted for 40.5% of the total coal consumption of CFIBs, thus resulting in a large contribution to the mercury inputs. Obviously, the contributions of different boiler capacity ranged varied across provinces. Except for Beijing, Inner Mongolia, Shanghai, Guangdong, Guizhou, Qinghai, and Xinjiang, small-scale boilers (capacity $\leq$ 10 t/h) were the primary source of mercury input in other provinces, with a contribution rate ranging from 31.1 – 70.6%.

The provincial mercury inputs of the CFIBs in China in 2015 are presented in SI Table S7. The spatial distribution of mercury inputs was basically consistent with that of the CFIBs. The CFIBs in China were mainly used for various industrial production and residential or commercial heating supply, and their mercury inputs were closely related to factors like the geographical location, economic activity level, population scale, and so on. The mercury inputs of CFIBs were mainly concentrated in North China and Northeast China, where the heating demands were huge during

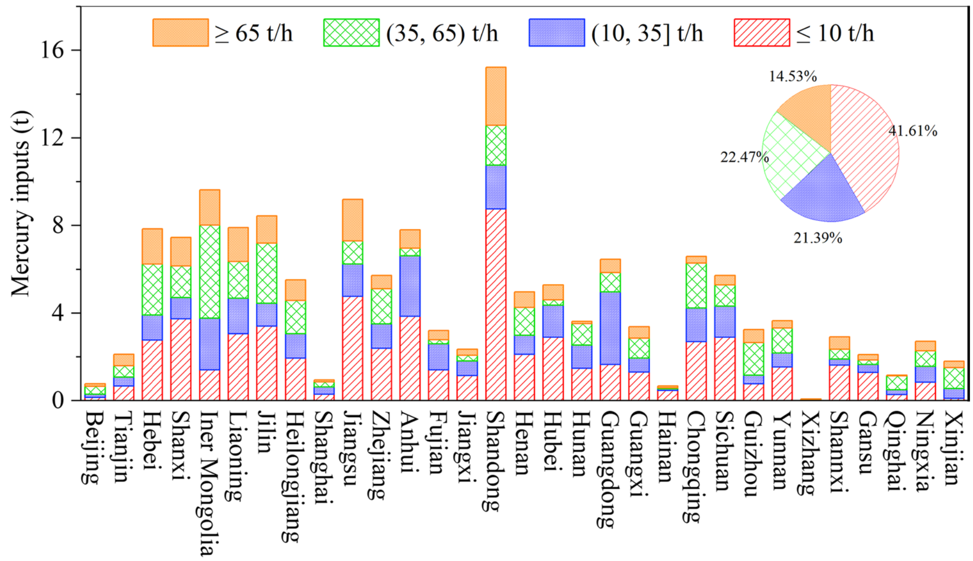


Fig. 3 Distribution of mercury inputs of the CFIBs in China in 2015

the heating season, and the economically developed East China, accounting for 18.7%, 14.7%, and 29.9% of the total mercury inputs of CFIBs in China, respectively. Shandong, Inner Mongolia, Jiangsu, Jilin, and Liaoning were the top five provinces in mercury inputs, with a total contribution of 33.9% to the national mercury inputs of CFIBs. However, although the coal consumption of CFIBs in Anhui and Chongqing accounted only for 3.0% and 3.1% of the total national coal consumption, respectively, their mercury inputs accounted for 5.2% and 4.4%, respectively, mainly due to the high mercury content in consumed coal. The mercury content in consumed coal in Anhui and Chongqing was 0.36 mg/kg and 0.30 mg/kg, respectively, which was much higher than the national average of 0.23 mg/kg. On the contrary, the coal consumption of CFIBs in Xinjiang accounted for 4.2% of the national coal consumption, but its contribution rate to the mercury input was only 1.2% due to the low mercury content in consumed coal (only 0.06 mg/kg).

Mercury outputs of the CFIBs in China

Mercury emissions of boilers with different capacity ranges

The total atmospheric mercury emissions of CFIBs in 2015 were estimated at 80.7 t, including 59.2 t of Hg^0, 19.9 t of Hg^{2+}, and 1.4 t of Hg^P according to the measured data shown in SI Table S5. The *Global Mercury Assessment 2018* assessed the atmospheric mercury emissions of CFIBs in China at 25 t in 2015 (UNEP 2019), which was underestimated due to the underestimation of coal consumption and inaccurate statistics on the APCDs installation ratio and mercury removal efficiency. The mercury emission limit for new or restructured low-rank coal-fired units in the USA is 0.04 lb/GWh, equivalent to 45.2 mg/t coal (EPA 2013; Sebor 2014). The average mercury emission per unit mass of coal in mainland China was 122.1 mg/t, which was 2.7 times the limit for low-rank coal-fired units in the USA. In comparison with CFPPs, the air pollutants removal efficiency of the APCDs of CFIBs was outdated, and the operation and management levels were low (Yue 2019). As a low-stack source of atmospheric mercury emissions, CFIBs have a great influence on the urban atmospheric environment.

The contribution of the atmospheric mercury emissions from the CFIBs in China with different boiler capacities in different regions is illustrated in Fig. 4. Small-scale boilers (capacity ≤ 10 t/h) were the main source of atmospheric mercury emissions, with a total contribution of 52.0%. The composition of APCDs of the CFIBs in China with different boiler capacities is illustrated in SI Fig. S2 and SI Fig. S3. Notably, the contribution of small-scale boilers to the atmospheric mercury emissions was 9.9% higher than that to the mercury inputs, which was mainly attributed to the low installation rate of various APCDs. The synergistic mercury removal efficiency of the dust removal devices and the desulfurization devices for the large-scale boilers (capacity ≥ 65 t/h) was about 47.3% and 26.1%, respectively, which was 31.7% and 19.9% higher than that for the small-scale boilers. Large-scale boilers were generally equipped with efficient APCDs like ESP-FF, and their pollutant emission control efficiency and management level were better than those of the small-scale boilers (Song et al. 2021).

Spatial characteristics of atmospheric mercury emissions

The spatial distribution of atmospheric mercury emissions of the CFIBs was consistent with the mercury inputs. As illustrated in Fig. 5(a), the atmospheric mercury

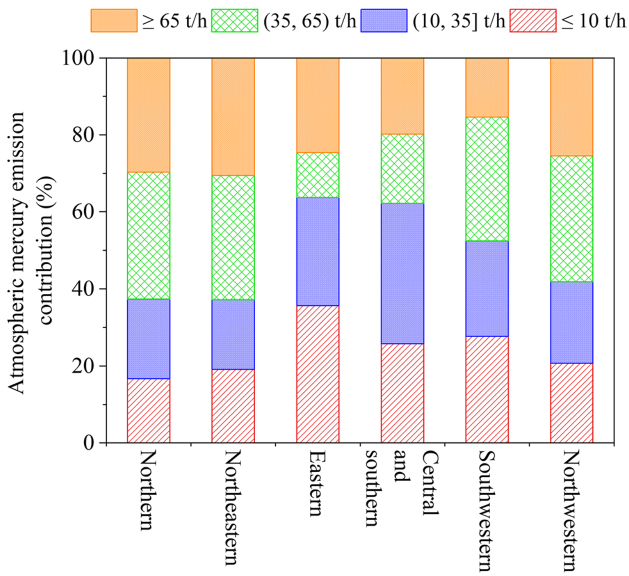


Fig. 4 Contribution of the atmospheric mercury emissions of the CFIBs in China with different boiler capacities in different regions

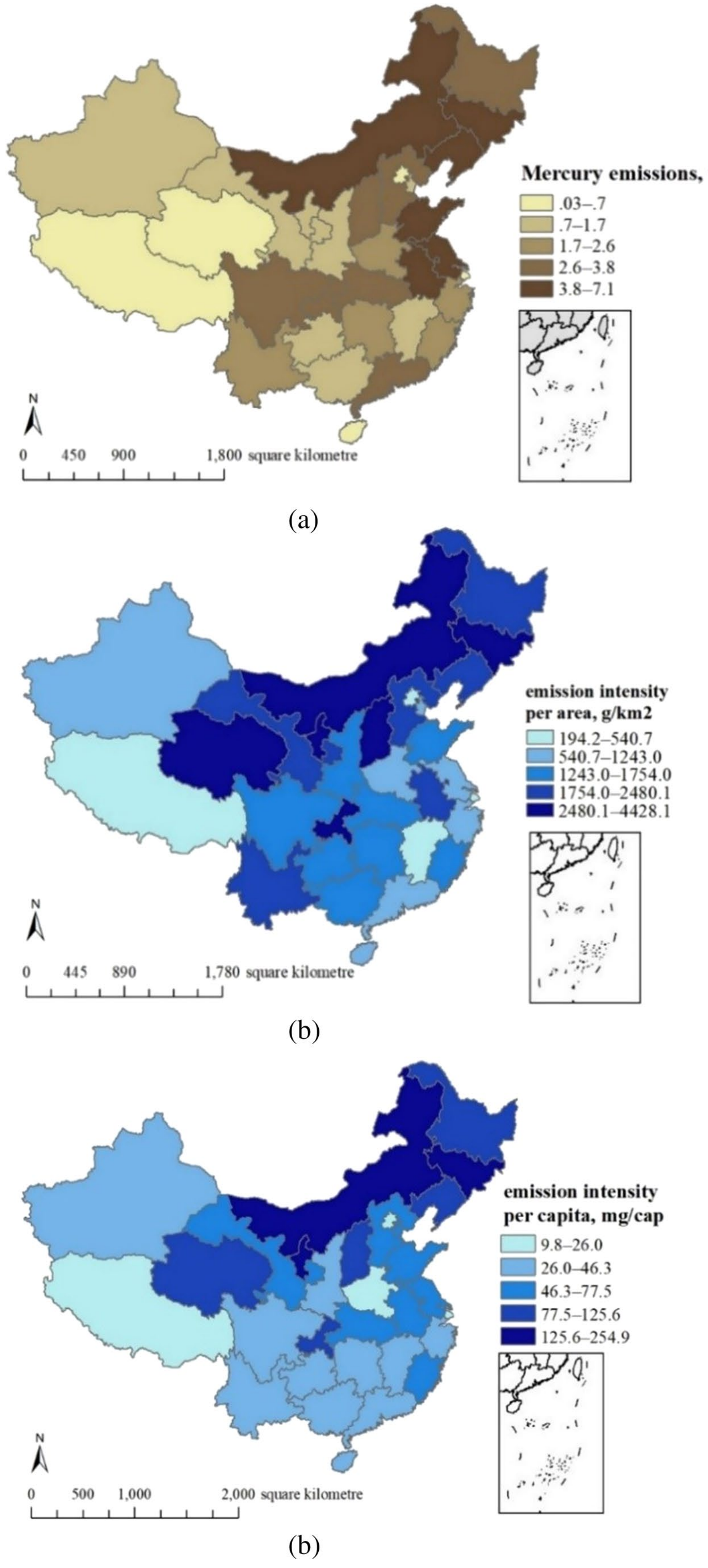
emissions of the CFIBs mainly came from the northern and northwestern provinces with high heating demands and the economically developed eastern coastal provinces. Among them, Shandong, Jilin, Jiangsu, Liaoning, and Inner Mongolia were the major contributors, and the total atmospheric mercury emissions from these five provinces accounted for 34.2% of the total atmospheric mercury emissions of the CFIBs in China. Shandong had the largest coal consumption (11.3%) and the highest contribution rate of atmospheric mercury emissions (8.8%). The Beijing-Tianjin-Hebei (BTH) region, the Yangtze River Delta (YRD) region, and the Fen-Wei Plains (FWP) region, where the economic activity level and pollution emissions were highly concentrated, were the key regions for air pollution control in China. The atmospheric mercury emissions in the provinces where these key regions are located accounted for 65.0% of the national total emissions. Likewise, the atmospheric mercury emissions in different provinces were also affected by the application of APCDs in the CFIBs. The coal consumption of the CFIBs in Heilongjiang and Jilin was comparable, but the atmospheric mercury emissions of Jilin were 2.1 t more than that of Heilongjiang. That was because the mercury content in consumed coal of Jilin was higher than that of Heilongjiang. Meanwhile, the installation rate of FFs for CFIBs in Heilongjiang was 10.4% higher than that in Jilin.

The atmospheric mercury emission intensity per unit area and per capita of the CFIBs in China was 1525 g/km^2 and 58.9 mg/cap, respectively. The spatial distribution of mercury emission intensity is presented in Fig. 5(b) and Fig. 5(c). As shown, provinces with high emission intensity (> 3000 g/km^2 and > 100 mg/cap) were mainly located in North China, Northeast China, and Northwest China, that is, the areas with high heating demands in winter. Although the atmospheric mercury emissions in some provinces were relatively low, their emission intensity was high, such as Qinghai and Ningxia. The main reason was that Northwest China had a small population but a high standard of urban construction land per capita, leading to the "double high" atmospheric mercury emission intensity (both per unit area and per capita). On the contrary, even if Shandong and Jiangsu ranked in the top five in terms of atmospheric mercury emissions, their mercury emission intensity was "double low" due to their high level of economic development, large urban construction area, and large population.

Mercury in wastes of the CFIBs

In addition to the atmospheric mercury emissions, part of the mercury in the flue gas enters the fly ash, bottom ash, and the desulfurization products (wastewater, gypsum) along with the particulate captured by the dust removal devices and SO_2 removed by the desulfurization devices. As shown in SI Table S7, the total mercury in the wastes including ashes and desulfurization products of CFIBs in China in 2015 were estimated at 67.7 t, of which 54.7 t was to ashes and 13.0 t was to the desulfurization products, accounting for 36.9% and 8.7% of the total mercury output, respectively. Boilers with a capacity above 35 t/h were the dominant source of the mercury in wastes, which was basically opposite to the atmospheric mercury emissions, and their mercury in ashes and the desulfurization products accounted for 49.6% and 52.0% of the total mercury in wastes, respectively. The mercury in different wastes was related to the mercury input and removal efficiency. The total mercury inputs of CFIBs in Jilin and Liaoning accounted for 11.0% of the national mercury inputs, which was higher than their proportion in the national amount of mercury in wastes. The mercury removal efficiency of dust removal devices and desulphurization devices of small-scale boilers were 11.5% and 12.6% in Jilin, and 0.8% and 1.8% in Liaoning, respectively, lower than the national average mercury removal efficiency of dust removal (16.4%) and desulphurization (6.2%) devices. With the upgrading of conventional air pollutants (PM/SO_2/NOx), control technologies, and the implementation of ultra-low emission retrofitting of CFIBs, the mercury contents in ashes and desulphurization products would further increase, and the environmental risks brought by the secondary mercury emissions in the reutilization of ash and desulfurization gypsum cannot be ignored.

Fig. 5 Spatial distribution of **a** the atmospheric mercury emissions, **b** the emission intensity per unit area, and **c** the emission intensity per capita



Scenario analysis for 2025 and 2030

Mercury emissions in 2025 and 2030

In this study, the atmospheric mercury emissions were the primary mercury output flow, accounting for 54.4%. Taking the further tightening of the air pollutants emission control of the CFIBs into consideration, this study investigated the mercury emission reduction potential of the CFIBs under four scenarios focusing on the atmospheric mercury emissions and the results are illustrated in Fig. 6. Apparently, mercury emissions were significantly reduced from 2015 to 2030 under the implementation of energy policies and environmental control measures. By 2025 and 2030, the atmospheric mercury emissions decreased by 70.9–79.1% and 75.4–84.6%, respectively, compared with 2015 under the four scenarios. As presented in Fig. 6(a), with the strictest energy policies and environmental control measures in the ALT-ACT scenario, the atmospheric mercury emissions of the CFIBs in China achieved the maximum emission reductions, decreasing to 16.9 t and 12.4 t in 2025 and 2030, respectively. The atmospheric mercury emissions of the CFIBs could be effectively reduced if the existing energy policies and environmental control measures were strictly implemented in the future.

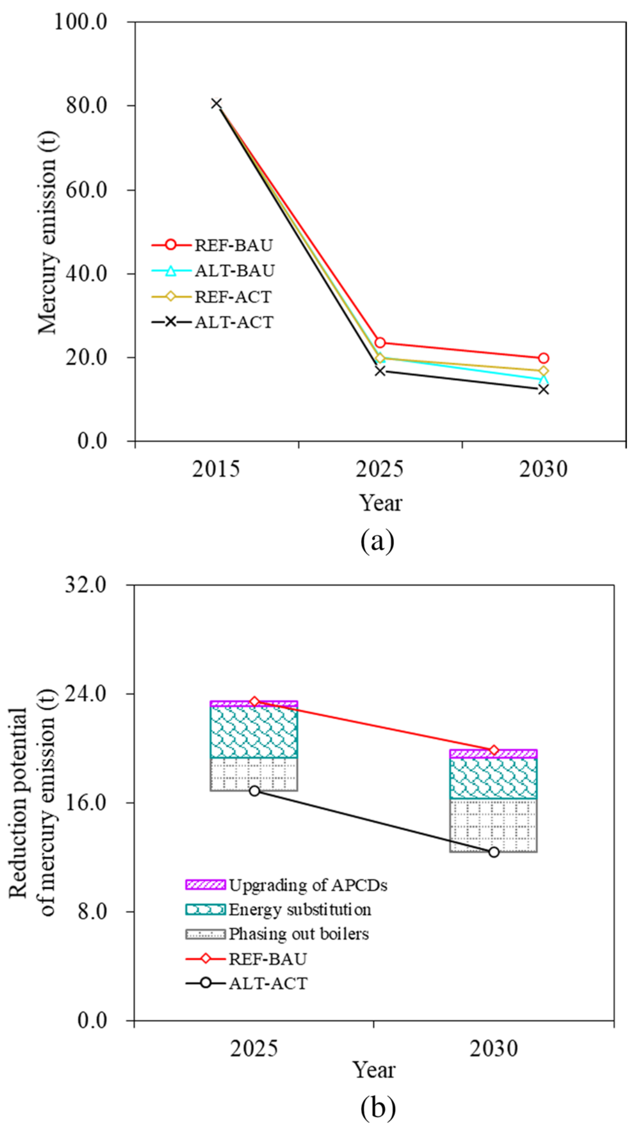


Fig. 6 Scenario projections of **a** the mercury emissions in 2025 and 2030, **b** the mercury emission reductions in 2025 and 2030

Overall, the reduction potential of the atmospheric mercury emissions from 2015 to 2025 was great, with the average annual reduction rate ranging from 21.9 to 26.9% under the four scenarios. However, the reduction potential of the atmospheric mercury emissions narrowed from 2025 to 2030, and the reduction rate slowed down with an average annual decline of 3.3–6.0%. The State Council successively issued the *Air Pollution Prevention and Control Action Plan* in September 2013 and the *Three-Year Action Plan to Win the Blue-Sky Defense* in June 2018 to control the emissions of conventional air pollutants (particle matter, SO_2, and NOx) of the CFIBs in China. The Chinese government set the air pollution control targets for CFIBs for 2017 and 2020. Thus during 2015–2025, huge changes have taken place in the boiler scale and air pollution control technologies of the CFIBs. The elimination of small and medium-scale (capacity ≤ 35 t/h) CFIBs accelerated, which promoted the application of high-efficiency dust removal and desulphurization devices for the CFIBs. Atmospheric mercury emission control of the CFIBs in China was mainly achieved through the synergistic control of the conventional air pollutant control devices. Therefore, the improvement of the synergistic mercury removal efficiency resulted in a significant decrease in the atmospheric mercury emissions of CFIBs. As a result, the atmospheric mercury emissions of the CFIBs were significantly reduced from 2015 to 2025, which was attributed to the implementation of various air pollution control measures for the CFIBs in China.

Reduction paths for mercury emissions

The reduction of the atmospheric mercury emissions of CFIBs from 2015 to 2025 mainly came from the phaseout of small-scale boilers and upgrading of the APCDs, with the contribution rates of 37.3–39.0% and 33.5–36.8%, respectively, as shown in Fig. 6(b). From 2025 to 2030, the contribution of APCDs upgrading to the emission reduction decreased to 32.1–35.1% due to the widespread application of high-efficiency APCDs during 2015–2025. In general, the source control measures (phaseout of the small-scale boilers and energy substitution) have much greater emission reduction potential than the end control measures (upgrading of APCDs). Based on the REF-BAU scenario, the atmospheric mercury emissions of CFIBs further reduced by 6.6 t and 7.5 t in 2025 and 2030, respectively, and the maximum emission reduction under the ALT-ACT scenario can be achieved. Under the circumstances, the contribution rates of the phaseout of small-scale boilers, energy substitution, and upgrading of APCDs were 39.4–52.7%, 33.2–57.5%, and 7.8–27.4%, respectively.

Based on the scenario projections of mercury emissions of the CFIBs in 2025 and 2030, this study proposed the feasible emission reduction paths for the CFIBs in China in the stage of 2015–2025 and 2025–2030. Three emission reduction paths were put forward: (1) phasing out boilers with a capacity below 35 t/h, and the atmospheric mercury emissions could be reduced by 21.7–24.8 t for 2015–2025 and 2.0–4.1 t for 2025–2030; (2) upgrading the APCDs including the ultra-low emission retrofitting for boilers with a capacity of 35 t/h or above, and the atmospheric mercury emissions could be reduced by 20.0–22.6 t for 2015–2025 and 0.3–1.3 t for 2025–2030; (3) energy substitution including the substitution of coal by clean energy, and the atmospheric mercury emissions could be reduced by 13.8–17.6 t for 2015–2025 and 0.5–1.3 t for 2025–2030.

Uncertainty

Uncertainties of the inventory of mercury inputs, atmospheric mercury emissions, and mercury in wastes of the CFIBs in China included the uncertainties caused by activity levels and emission factors, as well as other factors that affect the inventory results. The Monte Carlo simulation was used for the uncertainty analysis (Liu et al. 2018; Zhang et al. 2015; Tong et al. 2021). The probability distribution characteristics of all input parameters are provided in SI Table S8 (Gao et al. 2021; Liu et al. 2018; Streets et al. 2005; Tian et al. 2015; Tong et al. 2021). The uncertainties of mercury inputs, atmospheric mercury emissions, and mercury in wastes of the CFIBs in China in 2015 were estimated at the ranges of − 18.2 to + 20.8%, − 25.1 to + 29.7%, and − 40.7 to + 48.8%, respectively. The uncertainties of the inventory mainly came from the mercury content in consumed coal and the mercury removal efficiency of the pollution control technologies. The available test data is relatively limited, and the measured data are not representative enough. Moreover, the influence of mercury removal efficiency on the coal quality has not been considered at present. Comparatively, Streets et al. (2005) calculated the uncertainty of mercury emissions in China in 1999 to be − 60 to + 60% for industrial coal use. Tian et al. (2015) estimated the uncertainty of atmospheric mercury emissions of the CFIBs in China in 2010 at the range of − 44.6 to + 60.7%. Notably, the uncertainty of atmospheric mercury emissions of the CFIBs in China in this study was lower due to the more detailed mercury content in consumed coal and the updated mercury control efficiency of the APCDs. The uncertainties of mercury inputs, atmospheric mercury emissions, and mercury in wastes of the CFIBs in China could be further reduced by using refined point source activity data.

Conclusions

In this study, the mercury inputs and outputs including the atmospheric mercury emissions and mercury in wastes of the CFIBs in China were assessed by a deterministic bottom-up emission factor method. The inventory of mercury in wastes of the CFIBs in China in 2015 was firstly obtained. Scenario projections have been conducted to investigate the mercury emission reduction potential in 2025 and 2030 by considering the energy policies and environmental management measures comprehensively.

Based on the established activity database and the updated mercury content in consumed coal and mercury removal efficiency, the national atmospheric mercury emissions of the CFIBs in China in 2015 were estimated at 80.7 t, including 59.2 t of Hg^0, 19.9 t of Hg^{2+}, and 1.4 t of Hg^P. Boilers with a capacity of 10 t/h or below were the dominant mercury input source, which was mainly attributed to the low installation rate of various APCDs. Provinces with high mercury emission intensity (> 3000 g/km^2 and > 100 mg/cap) were mainly concentrated in North China, Northeast China, and Northwest China where the heating demand in winter is high. Intensive mercury emissions were mainly concentrated in Shandong, Jilin, Jiangsu, Liaoning, and Inner Mongolia, while the total mercury emissions of these five provinces accounted for 34.2% of the total national mercury emissions. The total mercury in wastes of the CFIBs in China in 2015 was estimated to be 67.7 t, including 54.7 t in the ashes and 13.0 t in the desulfurization products which accounted for 36.9% and 8.7% of the national mercury outputs, respectively. Boilers with a capacity above 35 t/h were the dominant source of the mercury in wastes, which was basically opposite to the atmospheric mercury emissions. From 2015 to 2030, the atmospheric mercury emissions of the CFIBs in China were significantly reduced, and by 2025 and 2030, the atmospheric mercury emissions under the four scenarios decreased by 70.9–79.1% and 75.4–84.6%, respectively, compared with 2015. Finally, three emission reduction paths were put forward based on the energy policies and environmental control measurements.

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Author contribution All authors contributed to the study conception and design. Yali Tong: Conceptualization, Methodology, Data curation, Formal analysis, Writing—original draft; Kun Wang: Conceptualization, Writing—review and editing, Resources; Jiajia Gao:

Conceptualization, Software, Writing—review and editing; Tao Yue: Writing—review & editing; Penglai Zuo: Investigation, Writing—review and editing; Chenlong Wang: Investigation, Methodology; Li Tong: Data curation, Methodology; Xiaoxi Zhang: Formal analysis, Validation; Yun Zhang: Investigation, Visualization; Quanming Liang: Investigation, Methodology; Jieyu Liu: Investigation, Visualization. All authors read and approved the final manuscript.

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Data availability The raw or partial processed data required to reproduce these findings are not publicly available at this time as the data also forms part of an ongoing study, but are available from the corresponding author on reasonable request.

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