Journal of Cleaner Production 114 (2016) 343–351

Contents lists available at ScienceDirect

Journal of Cleaner Production

journal homepage: www.elsevier.com/locate/jclepro

Potentials of whole process control of heavy metals emissions from coal-fired power plants in China

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ARTICLE INFO

Article history:
Received 22 August 2014
Received in revised form
27 April 2015
Accepted 1 May 2015
Available online 21 May 2015

Keywords:
Heavy metals
Atmospheric emissions
Coal-fired power plants (CFPPs)
Whole process control
Co-benefit removal effects
China

ABSTRACT

Recently, more and more poisoning accidents associated with toxic heavy metals have been reported throughout China, coal-fired power plants (CFPPs) sector is regarded as one of the most important source categories of anthropogenic atmospheric releases of heavy metals due to tremendous annual coal consumption (about 1785.3 Mt in 2012). In this paper, with the concept of whole process control, the co-benefit or synergistic removal efficiencies of different control measures used in CFPPs of China are evaluated, the combination of coal washing before burning plus post combustion cleaning of selective catalytic reduction (SCR) + electrostatic precipitator/fabric filters (ESP/FFs) + wet flue gas desulfurization (WFGD) configuration is identified to be the best available control technology for heavy metals abatement of CFPPs at present and in the near future. However, the widely application of special mercury control (SMC) technologies in Chinese CFPPs in future is greatly needed. Furthermore, three energy scenarios and three control scenarios were assumed to forecast the future trend of heavy metals emissions. Under the same control scenario, the change of the energy saving and energy structure will give rise to about 24.1% and 24.6% of abatement potential for heavy metals in 2020 and 2030, respectively. Whereas, under the current energy consumption pattern and air pollution control policies, the installation of SCR + SMC + ESP/FFs + WFGD will result in about 21.0–44.1% and 36.3–67.5% of reduction for heavy metals emissions in 2020 and 2030, respectively. Finally, integrated control suggestions are proposed to minimize the final toxic heavy metals discharges.

1. Introduction

Normally, Heavy metal (HM) is a general collective term which applies to the group of metals and metalloids with an atomic density greater than 4 g/cm^3. In recent years, more and more poisoning accidents associated with various heavy metals pollution have been reported in China, such as arsenic (As) poisoning in Guizhou and children with excessive blood lead (Pb) levels in Qingyuan city of Guangdong province (Chen et al., 2012; Xiao et al., 2011). In February 2011, to cope with the serious heavy metals pollution situation, the State Council of China officially approved the 12th Five-Year Plan for comprehensive prevention and control of heavy metals pollution (MEP, 2011).

China is one of the few countries in the world whose energy mix is highly dependent on coal, and coal consumption constitutes approximately 70% of the country's total primary energy consumption (Tian et al., 2013). According to Chinese statistics (NBSC, 2013), coal-fired power plants (CFPPs) generated over 80% of the annual total electricity (3925.5 TWh) and consumed over 50% of the total coal consumption (1785.3 million metric tons (Mt)). Therefore, although heavy metals present in only trace levels in the coal, the huge use in coal consumption by electricity generation has resulted in significant emissions of heavy metals to the atmosphere and accumulatively deposited into surface water and soil environment.

Studies on emission characteristics of selective heavy metals from CFPPs have been conducted (Ito et al., 2006). Tian et al. (2014) developed an integrated emission inventory of eight heavy metals from CFPPs of China for the year 2010. Wang et al. (2012) estimated Hg emissions from China's power sector in 2008 and discussed the

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http://dx.doi.org/10.1016/j.jclepro.2015.05.008
0959-6526/

mitigation potential of Hg emissions from CFPPs for the target year of 2020 and 2030 under three control scenarios.

Since 2006, the wide application of electrostatic precipitator (ESP)/ fabric filters (FFs) and flue gas desulfurization (FGD) as well as the increasing installation of selective catalytic reduction (SCR)/selective non catalytic reduction (SNCR) has remarkably changed the conventional air pollutants emission status of CFPPs in China. Nevertheless, there are few studies on systematically evaluating the mitigation potential of typical heavy metals emissions from CFPPs within the whole process control concept which starting from the feed coal to the end tail cleaning on accounting of the current status and future trends of CFPPs in China till now. Little is known about the integrated countermeasures for diminishing the final stack discharges of heavy metals from CFPPs of China based on the present situation.

In this paper, we will first discuss the control potentials of typical heavy metals emissions by conducting various energy and emission control scenario analyses with a new whole process control concept. Then, integrated control suggestions for minimizing diminish the final stack discharges of heavy metals from CFPPs of China are proposed.

2. Methodology

The calculation of heavy metals emissions from CFPPs is based on combining coal consumption data with specific emission factors grouped by different boiler patterns and various air pollutant control device (APCD) configurations, which can be expressed by the following equation (Tian et al., 2014, 2010).

$$E_{Total} = \sum_i E_i = \sum_i \sum_k \sum_m \sum_n A_{i,k} \times C_i \times R_m \times (1-\eta_{PM_n}) \times (1-\eta_{SO_2}) \times (1-\eta_{NO_X}) \times (1-\eta_{Hg}) \quad (1)$$

where, E is the annual atmospheric emissions of Hg, As, Se, Pb, Cd, Cr, Ni or Sb (tons/yr) from CFPPs in China; A is the amount of coal burned (tons/yr); C is the provincial average concentration of one toxic heavy metal in feed coal (ug/g); R is the average release ratio of one toxic heavy metal in flue gas compared with the element concentration in feed coal from pulverized-coal (PC) boilers, circulating fluidized-bed (CFB) boilers or stoker fired (SF) boilers (%); η_{PM}, η_{SO_2} and η_{NOx} represent the averaged fraction of one heavy metal co-benefit removed from flue gas by the conventional $PM/SO_2/NO_X$ emission control devices (%), respectively, while η_{Hg} represents the added Hg removal efficiency by the special mercury control (SMC) technologies (activated carbon injection (ACI), bromide injection into the furnace (BIF), oxidation catalysts, low-temperature mercury capture, the thief carbon process, etc.) (%) if any; i, k, m and n denote province (municipality, autonomous region), power plant, boiler type and PM emission control technology type.

As can be seen from Eq. (1), if we want to reduce the final emissions of heavy metals from CFPPs, the possible and feasible options include decreasing the volume of coal consumption, lowering the heavy metals concentration in feed coal through coal cleaning and/or adoption of high-quality raw coal with lower contents of heavy metals and/or coal transformation to liquid or gas fuels, adjusting the boiler furnace pattern with lower release rate, as well as adopting advanced $PM/SO_2/NO_X/Hg$ control technologies with high co-benefit or specific removal efficiency.

2.1. *Average concentration of each heavy metal in feed coal by province*

Although the content of heavy metal in per kg coal is really affected by the moisture level, the mass concentration of heavy metal in coal (μg/g) is still used in this study. This is mainly due to wide application of coal consumption on as received basis and the limited information about per energy content for heavy metal in coal (μg/J) in Chinese domestic field test results and related statistics.

Coals of China can be classified into three main coal ranks: anthracite, bituminous coal and lignite. The qualities of coals that are mined from different regions vary substantially and are largely determined by the distinction of coal-forming plants and coal-forming geological environments. According to the coal samples mined from three main coal-fields of China (Duan et al., 2011; Li et al., 2014; Zhu et al., 2015), the moisture, ash content, net calorific value and sulfur content on an air dried basis in above three coals are about 2.1–32.4%, 15.8–25.6%, 17.4–26.1 MJ/kg and 0.6–2.1%, respectively (please see Supplementary Data Table S1 for more details). On the nationwide statistical levels, Tian et al. (2013) concluded that the concentration of Hg, Se, Pb, and Cr in Chinese coals generally decreased in the order: anthracite > bituminous coal > lignite. While Ni, As, and Sb are more enriched in lignite than in bituminous coal and anthracite. Improvement of coal quality before coal utilization may help to minimize environmental pollution. Therein, coal washing is an alternative choice.

The original field test data sources, computational methodologies and geographical distinctions of average concentrations of various heavy metals in the coal as produced and consumed have been discussed in detail previously (NBSC, 2013; Tian et al., 2013, 2012a, 2012b, 2010). Thus, here we mainly focus on the determination of co-benefit removal rates of heavy metals in coal washing process. The provincial average concentrations of 8 hazardous heavy metals in coal as produced and consumed are listed in Supplementary Data Table S2–S3 due to paper length limitation.

Previous studies have proved that most of heavy metals show a strong affinity to inorganic minerals (pyrite, illite, and kaolinite) and the most likely forms of occurrence are association with sulfur in pyrite (Finkelman, 1994; Song et al., 2006). Luttrell et al. (2000) and Song et al. (2006) indicated that sulfur in pyrite could be removed effectively during physical coal cleaning and the removal efficiency could reach up to about 50.0%. Consequently, not only ash and SO_2 discharge, but also the heavy metals concentration in feed coal, which associated with the ash and inorganic minerals can be effectively reduced by coal washing process before coal burning. Meanwhile, the heating value of cleaned coal is much higher than raw coal (You and Xu, 2010). The final concentration of heavy metals in the cleaned coal can be calculated based on the following equation:

$$C_{cc} = \frac{C_{rc} A_{rc} (1-F)}{P_{cc}} \quad (2)$$

where, C_{cc} is the averaged concentration of one element in the cleaned coal; A_{rc} and C_{rc} are the amount and averaged concentration of one element of raw coal input in the production of cleaned coal, respectively; F is the fraction of one element removed by the coal washing process; P_{cc} is the amount of cleaned coal as produced.

As discussed above, the removability of various heavy metals during physical coal cleaning is mainly determined by the modes of occurrence of heavy metals in coals as well as the wash ability of inorganic minerals. The average removal efficiencies of Hg, As, Se, Pb, Cd, Cr, Ni and Sb during coal washing process have ever been discussed in detail in our previous studies. Therein, by collecting and compiling the relevant field test data reported in published literature, the arithmetic average removal efficiencies of Hg, As, Se, Pb, Cd, Cr, Ni and Sb during coal washing process were presumed to be about 50.0%, 54.0%, 30.0%, 36.3%, 32.2%, 58.0%, 58.5% and 35.7%, respectively (Tian et al., 2013, 2012a, 2012b, 2011, 2010). Other parameter values of A_{rc}, C_{rc} and P_{cc} which used in Eq. (2) are provided in the Supplementary Data Table S3–S4. Finally, the provincial average concentrations of 8 heavy metals in cleaned coal are calculated and listed in Supplementary Data Table S5.

According to the outputs of feed coals and provincial average concentrations of heavy metals in feed coals, the six regional weighted-average concentrations of heavy metals in feed coals are calculated and illustrated in Table 1. Here, provincial average concentration of arsenic in washed coal is used as an example to display the co-benefit removal rate of heavy metals during coal washing process. As can been seen from Table 1, because of distinct concentrations of heavy metals in raw coal mined from different regions and uneven outputs of cleaned coal throughout the country, the calculated weighted-average concentration of arsenic in cleaned coal from N, NE, E, CS, SW and NW regions have decreased by about 49.0%, 44.5%, 44.8%, 56.0%, 42.3% and 41.5%, compared to those in coal as consumed, respectively (NBSC, 2013; Tian et al., 2013). As a result, if totally burned with cleaned coal by all the power plants throughout the country, the emissions of Hg, As, Se, Pb, Cd, Cr, Ni and Sb from burning of cleaned coal will be 71.8 t, 181.5 t, 404.4 t, 546.7 t, 11.7 t, 243.8 t, 218.1 t and 56.7 t, respectively, approximately 39.5%, 45.9%, 12.0%, 22.5%, 12.3%, 51.7%, 51.2% and 31.1% lower than those from directly burning equal amount of raw coal as consumed in 2010 (Tian et al., 2014).

2.2. Emission factors of heavy metals from CFPPs

Normally, various heavy metals in coal can be classified into three groups according to their volatility during high temperature coal combustion process: (1) high volatile elements, such as Hg, Se, etc.; (2) semi-volatile elements like As, Pb, Cd, Cr, Ni, Sb, etc.; (3) low or no volatile elements like Mn, Rb, etc. (Xu et al., 2004). When coal is burned in boiler furnaces, part or even most of these heavy metals are volatilized and released into flue gas with gaseous status or condensed and adhered with fine particles depending on the volatility performance. Notably, the volatility of certain heavy metals depended on the calcium content in the ash (Kaakinen and Jorden, 1975). Once in the vapor phase during the coal combustion process, the heavy metals (e.g., As, Se, Sb, Pb, Cd, etc.) may react with compounds in the ash (e.g., calcium oxide and silicates) and are removed from the vapor phase (Bool and Helble, 1995). The coal ashes consist mainly of SiO_2 and Al_2O_3 (generally greater than 60% of the total ash). The CaO content in Chinese coal ashes generally decreases in the order lignite < anthracite < bituminous coal (Li, 2014; Liu, 2005; Zhu, 2015). When the flue gas pass through downstream conventional air pollution control devices, such as SCR, ESP/FFs and FGD, part of these heavy metals with gaseous forms or adhered with fine particles can be synergistically removed from the flue gas. In this study, a literature review on the heavy metals released rates of coal-fired boilers and the co-benefit removal efficiencies of various APCDs for heavy metals was conducted thoroughly (more details can be seen Supplementary Data Table S6–S7). Here, the arithmetical average release rate and co-benefit removal efficiency are adopted to evaluate the mitigation potential of heavy metals from CFPPs (see Fig. 1).

Until now, the comprehensive and quantitative studies on phase distinctions of various HMs (except Hg) emissions in China are quite limited. Therefore, only estimated emission of Hg speciation from CFPPs would be discussed in this study. Generally, Hg in combustion flue gas existing in primary three forms, namely, elemental mercury (Hg^0), gaseous oxidized mercury (Hg^{2+}), and particle-bound mercury (Hg^P). On the basis of field tests of Hg speciation conducted outside and inside China, the average proportion of Hg^0, Hg^{2+} and Hg^P in total Hg emissions for the boilers installed with different APCD configurations were determined to be 4.4–81.0%, 18.6–86.0% and 0.4–9.6%, respectively (see Supplementary Data Table S8 for more details) (Bustard et al., 2004; Tian et al., 2014).

2.3. Scenario projection on heavy metals emissions for 2020 and 2030

On accounting of the current status of CFPPs and future demand of energy-saving and pollution reduction, three different energy scenarios, namely reference energy scenario, improved energy scenario and alternative renewable energy scenario, were projected for the target years 2020 and 2030. The reference energy scenario is presumed that related legislation and annual energy consumption trends will remain unchanged in the future. The alternative renewable energy scenario is characterized by the effective implementation of renewable and new energy-saving policies. The improved energy scenario is regarded as a moderate plot in which energy consumption falls in between those of above two scenarios. Wang et al. (2012) indicated that the development of electricity consumption per capita (ECPC) in the periods of 2010 to 2020 and 2020 to 2030 for China will be similar to the periods of the 1970s to

Table 1
The regional weighted-average concentrations of 8 hazardous heavy metals in coal as produced, coal as consumed and cleaned coal of China, μg/g (NBSC, 2013; Tian et al., 2013, 2012a, 2012b, 2010).

Categories	Regions[a]	Hg	As	Se	Pb	Cd	Cr	Ni	Sb
coal as produced	N	0.19	4.83	2.43	26.64	0.40	18.18	11.01	0.88
	NE	0.20	6.02	1.62	22.92	0.14	20.86	16.19	0.85
	E	0.29	4.73	5.43	16.48	0.28	26.66	21.16	0.50
	CS	0.18	4.72	4.74	20.64	0.55	30.66	12.70	0.81
	SW	0.35	6.79	3.10	30.31	1.12	40.78	22.15	3.24
	NW	0.19	3.68	2.66	24.41	0.61	25.28	16.00	2.02
coal as consumed	N	0.19	4.77	2.33	25.91	0.38	18.27	10.96	0.84
	NE	0.20	5.80	1.48	23.98	0.14	17.94	12.55	0.79
	E	0.23	4.46	3.73	22.89	0.39	23.18	15.83	1.02
	CS	0.17	4.46	3.89	23.19	0.60	27.19	14.40	1.31
	SW	0.35	6.72	3.09	30.60	1.12	40.89	22.04	3.14
	NW	0.18	3.55	2.15	17.08	0.51	20.05	13.98	1.36
cleaned coal	N	0.11	2.43	2.17	19.07	0.36	9.28	5.78	0.67
	NE	0.12	3.22	1.20	20.42	0.12	9.37	6.22	0.67
	E	0.15	2.46	3.35	15.92	0.27	11.22	8.04	0.56
	CS	0.12	1.96	3.83	15.93	0.50	13.29	6.63	0.57
	SW	0.22	3.88	2.96	24.85	1.04	20.67	11.77	2.59
	NW	0.12	2.07	2.84	14.65	0.67	9.40	6.82	0.87

[a] N, NE, E, E, CS, SW and NW represented the Northern Region, the Northeastern Region, the Eastern Region, the Central and Southern Region, the Southwestern Region, and the Northwestern Region, respectively (more details about six administration districts of China can be seen in Supplementary Data Table S2 and Fig. S1).

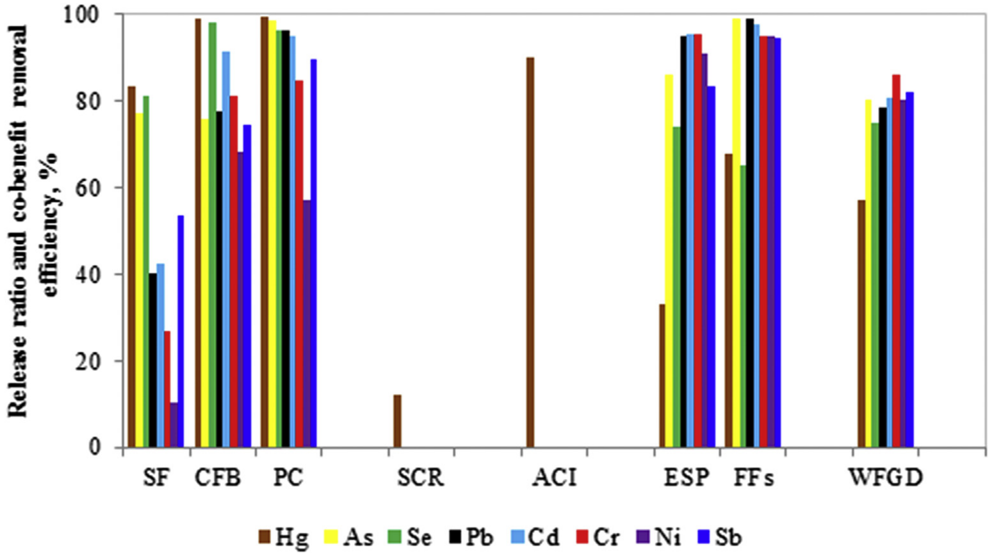

Fig. 1. Average release ratio and co-benefit removal efficiency of 8 heavy metals from CFPPs.

2000 and 2000 to 2010 for developed countries, respectively. On basis of the statistics from the International Energy Agency (IEA) and the above assumptions, we assumed the level of Chinese ECPC for 2020 and 2030 will probably be at about 4900–5500 kWh and 5100-5700 kWh, respectively (IEA, 2013). Then, combined with the parameters on the future trend of development on population, thermal power proportion and the standard coal consumption rate (CEC, 2013a; ERINDRC, 2008; NPDSRG, 2007; Wang et al., 2012), we projected that about 1.28–1.78 billion tons of coal equivalent (tce) will be fired by power plants at the three scenarios (see details about the key parameters in Supplementary Data Table S9).

Besides, in order to evaluate the mitigation potential of the application share of different APCD configurations for heavy metals in 2020 and 2030, three pollution control scenarios, namely, business-as-usual (BAU) scenario, best-available-control-technology (BACT) scenario, and intensive scenario with advanced high-efficiency control-technology and SMC technology (HECT) scenario, were considered. Based on the current status of $PM/SO_2/NO_X/Hg$ control devices applications in CFPPs (MEP, 2014a, 2014b) and the national planning of air pollution control for the 12th Five-Year Plan (CPGPRC, 2012; NBSC, 2011), as well as the future emission reduction goals (CEC, 2013a), different APCDs application rates and foreseen emission limits of air pollutants (dust, SO_2 and NO_X) for thermal power plants are assumed for the three pollution control scenarios (see more details in Supplementary Data Table S10–S11). In addition, the current legislative limits of conventional flue gas emissions for thermal power plants from China, EU and U.S. are included in Table S11. Due to paper length limitation, the specific instructions for the formation of three different energy scenarios (projections of the coal consumption by the CFPPs of China) and three different control scenarios (projections of the APCDs applications for the CFPPs of China) are put in Supplementary Data Section S1–S2.

3. Results and discussion

3.1. Best available APCDs configuration for heavy metals emissions control of CFPPs

The heavy metals co-benefit removal efficiencies of the current and future APCD combinations, also shown in Table 2, are calculated by using the arithmetic mean of removal efficiency of separately APCD listed in Fig. 1. Currently, most CFPPs in China have similar APCDs configurations, consisting of ESP and wet flue gas desulfurization (WFGD). But the combination of ESP and WFGD is not at all the best available technology for most heavy metals compared with the synergetic removal efficiencies of other APCD configurations. As can be seen from Table 2, the best available APCDs configurations to control Hg and other elements are SCR + SMC + ESP + WFGD and SCR + FF + WFGD in the foreseeable future, respectively.

3.2. Future trends and mitigation potential of heavy metals emissions from CFPPs in China

Previous studies have demonstrated that CFPPs, the important Hg, As, Se, Pb, Cd, Cr, Ni and Sb producer, have reached the maximum output in 2006 during 2000–2010. The national total emissions of Hg, As, Se, Pb, Cd, Cr, Ni, and Sb were estimated at about 118.5 t, 335.5 t, 459.4 t, 705.5 t, 13.3 t, 505.0 t, 446.4 t, and 82.3 t in 2010, respectively (Tian et al., 2014).

The positive or negative growth rates of 8 heavy metals emissions in three control scenarios for the target years of 2020 and 2030, compared to those in 2010, are summarized in Table 3 and Table 4, respectively. As can be seen from Table 3, under the improved energy scenario, the discharges of Hg, As, Se, Cd, Cr, Ni and Sb from CFPPs will increase by 7.1–36.8% for BAU scenarios from 2010 to 2020. However, the emissions of 8 heavy metals for HECT scenarios in 2020 will decrease by 0.2–36.8%, compared to those in 2010. This is mainly because of increasing application rate of SCR, FFs and WFGD and relatively higher co-benefit removal efficiency of ESP/FFs plus WFGD configuration. Specially, the emissions of Se and Sb from CFPPs for BACT scenarios in 2020 still represent positive growth, compared to those in 2010. For Sb emission with positive growth, this is mainly because of the relatively high application rate of ESP and low co-benefit removal efficiency of Sb through ESP (see Table 2). Whereas, with respect to Se emission with positive growth, Cheng et al. (2009) indicated that Se is readily volatilized during coal combustion due to its low boiling point (217 °C) compared to the other low or no volatile elements. A full-scale study on partitioning of heavy metals demonstrated that about 57.5% of Se was present in the gas phase in the stack gas of coal-fired power plant equipped with SCR + ESP + WFGD (Ito et al., 2006). Moreover, high volatile elements (e.g. Hg, Se) are inclined to

Table 2
Average 8 heavy metals co-benefit removal efficiencies of different APCD combinations.

APCD combination	Co-benefit removal efficiency (%)[b]							
	Hg	As	Se	Pb	Cd	Cr	Ni	Sb
ESP	33.17	86.20	73.78	97.16	96.46	98.53	93.52	83.50
ESP + WFGD	71.41	97.29	93.41	99.39	99.31	99.79	98.70	97.05
SCR + ESP + WFGD	74.82	97.29	93.41	99.39	99.31	99.79	98.70	97.05
SNCR + ESP + WFGD	71.41	97.29	93.41	99.39	99.31	99.79	98.70	97.05
SCR + SMC + ESP + WFGD	97.48	97.29	93.41	99.39	99.31	99.79	98.70	97.05
FF	67.92	99.00	65.00	99.00	97.63	95.13	94.83	94.30
FF + WFGD	86.28	99.80	91.20	99.78	99.54	99.32	98.97	98.98
SCR + FF + WFGD	87.91	99.80	91.20	99.78	99.54	99.32	98.97	98.98
Coal washing	50.00	54.00	30.00	36.30	32.21	57.99	58.49	35.67

[b] See more detail in the separate Supplementary Data.

Table 3
The positive or negative growth rates of 8 heavy metals emissions in 2020, compared to those in 2010.

Category	2020-reference[c]			2020-improved[c]			2020-renewable[c]		
	BAU	BACT	HECT	BAU	BACT	HECT	BAU	BACT	HECT
Hg	33.4%	13.7%	−4.4%	16.9%	−0.3%	−16.2%	1.3%	−13.7%	−27.4%
As	32.7%	3.7%	−22.4%	16.3%	−9.1%	−32.0%	0.8%	−21.3%	−41.1%
Se	47.4%	30.3%	13.9%	29.2%	14.2%	−0.2%	11.9%	−1.1%	−13.5%
Pb	13.9%	−7.8%	−27.9%	−0.1%	−19.2%	−36.8%	−13.5%	−30.0%	−45.2%
Cd	22.2%	0.9%	−19.2%	7.1%	−11.6%	−29.2%	−7.2%	−23.4%	−38.7%
Cr	20.7%	−4.2%	−27.0%	5.8%	−16.1%	−36.0%	−8.3%	−27.3%	−44.5%
Ni	31.6%	7.1%	−15.6%	15.3%	−6.1%	−26.0%	−0.1%	−18.7%	−35.9%
Sb	56.1%	25.4%	−3.0%	36.8%	10.0%	−15.0%	18.5%	−4.8%	−26.4%

[c] "−" represents negative growth.

Table 4
The positive or negative growth rates of 8 heavy metals emissions in 2030, compared to those in 2010.

Category	2030-reference[d]			2030-improved[d]			2030-renewable[d]		
	BAU	BACT	HECT	BAU	BACT	HECT	BAU	BACT	HECT
Hg	−7.5%	−44.1%	−66.3%	−19.2%	−51.2%	−70.6%	−30.3%	−57.9%	−74.6%
As	−19.4%	−47.4%	−54.8%	−29.6%	−54.0%	−60.5%	−39.3%	−60.3%	−65.9%
Se	14.0%	0.2%	−1.4%	−0.4%	−12.5%	−13.8%	−14.0%	−24.5%	−25.7%
Pb	−27.1%	−48.9%	−54.4%	−36.3%	−55.3%	−60.2%	−45.0%	−61.5%	−65.6%
Cd	−19.1%	−39.7%	−43.5%	−29.3%	−47.4%	−50.7%	−39.0%	−54.6%	−57.4%
Cr	−23.5%	−44.5%	−48.0%	−33.2%	−51.5%	−54.5%	−42.4%	−58.2%	−60.8%
Ni	−11.1%	−33.6%	−38.4%	−22.4%	−42.0%	−46.1%	−33.0%	−49.9%	−53.5%
Sb	−2.5%	−32.3%	−38.5%	−14.8%	−40.9%	−46.2%	−26.5%	−49.0%	−53.6%

[d] "−" represents negative growth.

condense on the submicron fly ash particles being captured by ESP with lower efficiency compared with other six heavy metals (see Fig. 1) (Tang et al., 2012).

From Table 4, we can see that the discharges of each heavy metals (except Se) under all of scenarios for 2030 turn out to be much lower than those for 2010, because of the gradually accelerated application of advanced APCDs and the increasing usage rate of renewable energy in CFPPs. Under the reference energy scenario for 2030, the Se discharges for BAU and BACT scenarios will still increase by 14.0% and 0.2% from 2010 to 2030 due to the relatively lower co-benefit removal efficiency, respectively. The declined share of Se emissions in power sector, caused by increasing ESP, FFs and WFGD installation, will be offset by the added coal combustion and this also indicates the further control of Se emission from power generation sector just using traditional APCDs in the future might be insufficient.

The specific emissions of Hg speciation are listed in Supplementary Data Table S12. Hg^{2+} is water-soluble and can be effectively removed by the downstream WFGD scrubber. Therefore, the proportion of Hg^0 in the total Hg emission reaches up to about 74.3–74.8% in 2020 due to the continuous increase of WFGD application from 2010 to 2020 (see Supplementary Data Table S12). Specially, compared with Hg^{2+} and Hg^P, Hg^0 is much easier to transport long distances even causing trans-boundary concerns due to relatively higher residence time in the atmosphere. However, Hg^0 (the primary existing form of Hg in the combustion zone) can be effectively absorbed or oxidized by the SMC technologies. Therefore, the proportion of Hg^0 in the discharged total Hg has decreased from 73.1% in 2010 to 36.3–67.8% in 2030 (see Supplementary Data Table S12).

The heavy metals emissions in the renewable energy scenarios are 24.1% and 24.6% lower than those in the reference energy scenarios in 2020 and 2030, respectively. This reveals the reduction potential of heavy metals emissions as a result of application of renewable energy in power sector. If the application rates of all of the APCD configurations remain the same as 2010, the Hg, As, Se, Pb, Cd, Cr, Ni and Sb emissions from CFPPs in the improved energy scenario for 2020 will be as high as 171.3 t, 484.8 t, 663.9 t, 1019.5 t, 19.3 t. 729.9 t, 645.2 t and 119.0 t, respectively. However, the estimates of the improved BACT scenario are actually 21.0–44.1% lower than these values because of widespread application of the combination of SCR + ESP/FFs + WFGD configuration. The heavy metals emissions from CFPPs in 2030 would be only 4.9% lower than those of 2020 if the application rate of control technologies is kept the

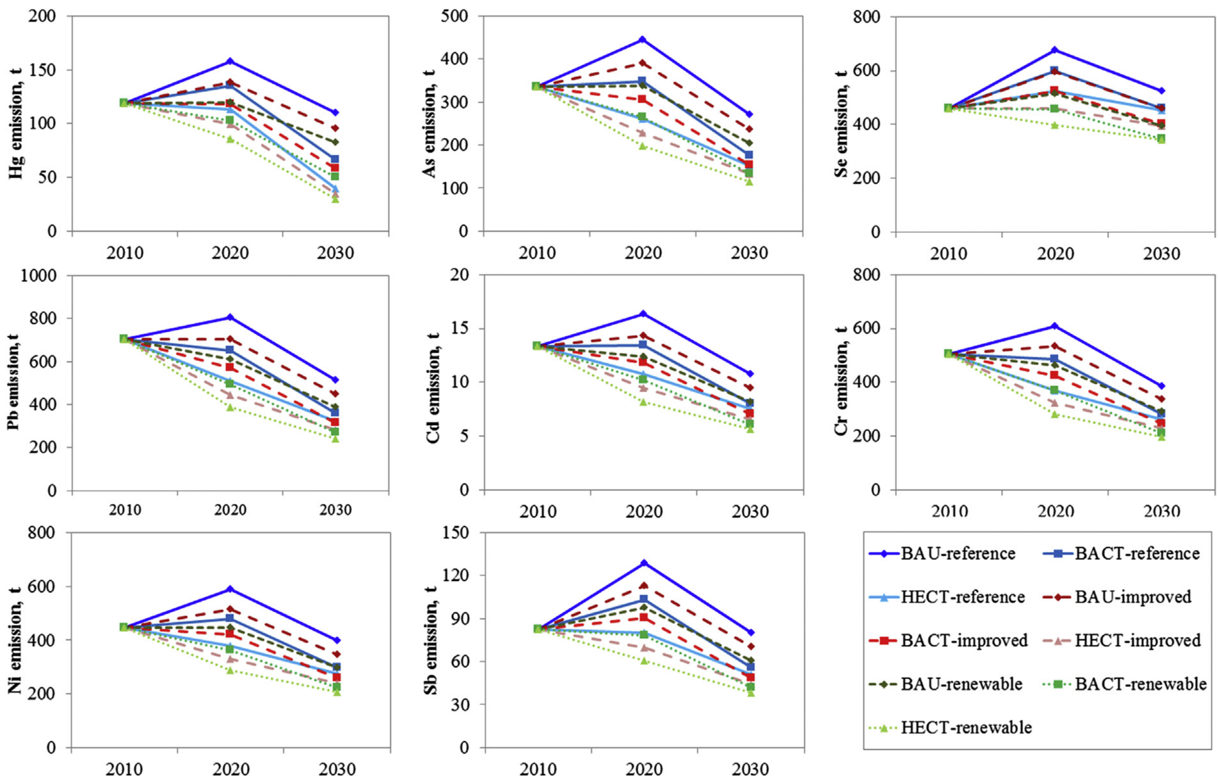

Fig. 2. Projected Hg, As, Se, Pb, Cd, Cr, Ni and Sb emissions from Chinese coal-fired power plants (CFPPs) in different scenarios.

same, which are about 57.1–207.8% higher than the actually estimated emissions of heavy metals under the improved scenario for 2030.

As can be seen from Fig. 2, under the alternative renewable HECT scenario, the significant potential reductions of Hg, As, Se, Pb, Cd, Cr, Ni and Sb can achieve 32.5 t, 137.9 t, 62.1 t, 319.1 t, 5.2 t, 225.0 t, 160.2 t and 21.7 t in 2020, compared to those in 2010, respectively. With the decreasing proportion of thermal power (see Table S9) and increasing application rates of advanced APCDs (see Table S10), the largest mitigation potential of Hg, As, Se, Pb, Cd, Cr, Ni and Sb emissions from CFPPs can reach 88.4 t, 221.1 t, 117.9 t, 463.1 t, 7.7 t, 307.0 t, 239.0 t and 44.1 t under the alternative renewable HECT scenario for 2030, respectively. On the basis of above analysis, we can conclude that only to increase application rate of advanced APCDs (e.g., SMC, SCR, FFs, WFGD, etc.) and renewable energy jointly can the discharges of heavy metals from CFPPs be well-controlled.

Mitigation potential of Hg emissions from CFPPs in China was ever projected by Wang et al. (2012), they estimated the Hg emissions from CFPPs for 2020 and 2030 as about 71.4–137.0 t and 28.0–104.2 t, respectively. In this study, under the three energy scenarios and three control scenarios, the total Hg emissions from CFPPs in China for the target years of 2020 and 2030 are projected at about 86.0–158.1 t and 30.1–109.6 t, respectively, which are well consistent with the results by Wang et al. (2012).

3.3. Proposals on heavy metals emission control strategies

3.3.1. Implement coal consumption cap in key areas

With the extensive growth of industry and the economy as well as the rapid urbanization process in China, huge electricity demand from various industrial activities and domestic use of residents would request increasing amounts of fossil fuel consumption.

In response to the severe $PM_{2.5}$ and haze pollution in the eastern China, coal consumption cap in key areas has been proposed in *Air Pollution Prevention and Control Actions Plan* issued by State Council of China in 2013 (CPGPRC, 2013). With implementation of coal consumption cap policy, the negative growth of coal consumption resulting in electricity deficit in the eastern provinces can be supplemented by increasing imported electricity from Western Region using long-range Super-High Voltage Electricity Transportation, natural gas supply, coal transformation application and non-fossil energy usage (e.g., wind, solar, etc.).

3.3.2. Stop mining and burning of coal with high heavy metals concentrations

It should be noted that the electricity deficit of eastern China is become larger and larger in recent years, while coal is predominantly deposited in western and northern China and the two areas account for 80.0% of the country's total coal reserves. Moreover, generating electricity from CFPPs by burning locally mined coal and transporting electricity from west to east is regarded as a landmark project within the national program for the West China Development. Therefore, huge power shortage in developed regions, unbalanced energy distribution and backward economic development mode relying on local coal mining and burning in certain provinces, are the main obstacles which restrict the local government to ban mining and burning such coals with high concentrations of heavy metals totally. Because of relatively low coal production and high concentrations of heavy metals in coal as produced, this measure can be carried out by Zhejiang and Guangxi. In order to do this, the Chinese government should encourage power plants to burn low heavy metals coal with price comparable to high heavy metals coal and shut down small coal mines and provide a social welfare network for the displaced workers.

Cheng (2014) indicated that sulfur concentration of coal have significant positive correlation coefficients with heavy metals such as Se, Pb, As, Ni, Sb, etc. Therefore, in response to *12th Five-Year Plan for comprehensive prevention and control of heavy metals pollution* to reduce heavy metals emissions, stopping utilizing high-sulfur coal in provinces is a another optionally effective measure.

3.3.3. Promote coal washing before combustion

According to the statistical energy data, the total production of cleaned coal has increased to 647.5 Mt in 2012, while only 34.5 Mt was burned directly by CFPPs throughout the country, accounting for only 5.3% of the total cleaned coal production and 1.7% of total power coal consumption (NBSC, 2013). Dramatically environmental and economic benefit will be gained by improving the consumption of cleaned coal in power sector, especially for provinces such as Jiangsu, Shandong, Guangdong, which ranking among the top three provinces with the highest thermal power generation and large amounts of coal input from other coal-export provinces. In addition, the raw coal production from Inner Mongolia, Shanxi and Shaanxi are predominant in China, accounting for about 66.5% of the total raw coal production. Nevertheless, only about 11.2%, 31.7% and 10.9% of raw coal are washed before burning in these three provinces, respectively. The main reason is that coal-mining companies are not willing to wash raw coals and supply cleaned coals in case of added capital investment and possible secondary water and soil pollution risks. Thus, Chinese central government should adjust the coal price system and raise financial supplement intensity for coal-mining enterprises from main provinces in thermal power generation and/or raw coal production to increase the utilization rate of cleaned coal in power sector.

3.3.4. Increase the application rate of advanced APCD configurations

Generally speaking, submicron particle capture efficiency in ESP is significantly less efficient than that in FFs (Nelson et al., 2010). Moreover, for most of heavy metals, significant enrichment is observed in the finer particle sizes. Therefore, the widespread application of FFs and ESP plus FFs in newly built or retrofitted coal-fired boilers is significantly for strengthening abatement of heavy metals emissions.

Previous studies have demonstrated that WFGD system installed by CFPPs play also the role of second-stage dust removal with the total filtration efficiency reaching 80.0% (Konieczyński and Zajusz-Zubek, 2011). By the end of 2012, the installed capacity of coal-fired boilers without FGD is still as high as 113.3 GWe, accounting for about 13.8% of the total installed capacity (CEC, 2013b; MEP, 2014a). In the present condition of energy-saving and pollution reduction, it should be a valid strategy to reduce the discharges of heavy metals by further increasing the application rate of WFGD in coal fired power plants during the 12th Five-Year Plan period.

By the year of 2012, none coal-fired power plant in Chongqing and Qinghai has employed SCR device (MEP, 2014b). Moreover, the total installed capacity of coal-fired boilers equipped with SCR systems in Shandong, Inner Mongolia, Hebei and Liaoning provinces is only 21.5 GWe, accounting for only about 10.8% of the total installed capacity of coal-fired boiler of these four provinces. Nevertheless, the electricicty generated by these four provinces is as high as 953.6 TWh, accounting for about 24.3% of the national total electric energy production (CEC, 2013b; MEP, 2014b). Thus, the provinces with high installed capacity and electricity generation should be regarded as key areas in *Emission Standard of Air Pollutants for Thermal Power Plants* (GB13223-2011) (MEP, 2012), and performed more stringent air pollutants (e.g., NO_X, Hg & its compounds, etc.) emission limits.

Currently, the combination of ESP plus WFGD is the most common APCD configurations used in CFPPs of China. But, on the basis of technical feasibility and cost-benefit for CFPPs, the combination of SCR + ESP/FFs + WFGD configuration may be the best available control technology to co-benefit reduce Hg and other heavy metals emissions for China (can be seen in Table 2), especially for above mentioned provinces with huge coal use and thermal power generation.

3.3.5. Promote SMC technologies application

The contamination of Hg is a global problem that no one country can solve alone. In response to curb the adverse impacts of Hg pollution on environment and human health, Minamata Convention on Mercury was agreed through consultation by 139 countries in October 2013. With the signature of global mercury convention, China will be confronted with great challenges to abate Hg emissions from CFPPs.

Presently, SMC technologies can generally be divided into two categories: add halogens into the furnace to oxidize Hg^0 into Hg^{2+} and inject powdered activated carbon into the flue gas upstream of the particle control equipment to absorb Hg^0 (the sorbent with Hg attached along with the fly ash can be valid captured by particle control device) (Liu et al., 2012; Qu et al., 2009). By 2015, it is anticipated that about 148 GWe of installed capacity of coal-fired boilers equipped ACI systems will be achieved in the power industry of United States in response to *Mercury and Air Toxics Standards* (MATS) (U.S. EPA, 2011). However, there are only a few pilot-tests or demonstration projects initiated in Chinese power sector presently.

In addition to above two SMC technologies, beginning in 2007, the Department of Energy (DOE) of United States and others have developed and tested other technologies (e.g., oxidation catalysts, low-temperature mercury capture, the thief carbon process, etc.) to achieve reductions of 90 percent or greater Hg at low costs (U.S. Gao, 2009). Especially for the thief carbon process (the extracted carbon from the boiler is injected into exhaust gas to act as sorbent for mercury capture), the removal efficiency of Hg was regarded as to be comparable to those achieved by commercially available sorbents. Nevertheless, the price of thief carbon sorbents is only less than 10 percent of the typical cost of sorbents (U.S. Gao, 2009). Therefore, those SMC technologies with high Hg removal efficiency and low cost may be the commercially available alternative for mercury removal from boilers' exhaust gas in the future.

3.3.6. Strengthen energy conservation and boost electricity generation using renewable energy

As we all know, widespread application of advanced APCD configurations may reduce but not totally eliminate heavy metals release from CFPPs. By replacing coal burning with other clean energy and renewable energy sources for electricity generation in medium and long-term future, such as hydropower, wind power, solar power and nuclear power and so on, the emissions of heavy metals may be further eliminated. By the end of 2012, the application rate of hydropower, wind power and solar power is only about 17.2%, 2.1% and 2.0% in power sector throughout the country, respectively. In response to global climate change, Chinese government committed to reduce the amount of carbon dioxide emitted per unit of gross domestic product by 40–45% from 2005 levels by 2020 on United Nations Framework Convention on Climate Change in 2009. Specially, the strategies of energy-related carbon emission reduction also apply equally to heavy metal reduction. Therefore, the development of clean and renewable energy utilization technologies has a broad prospective with the increasing of energy crisis and environmental pollution by electric power sector in China. In addition, it is necessary for electric power sector to strongly develop supercritical CFB technology because of high fuel-flexible and low release rate of SO_2, NO_X as well as toxic heavy metals.

3.3.7. Other strategies to control heavy metals contamination

In order to take further steps to control heavy metals contaminations, it is still insufficient to reduce heavy metals emissions from CFPPs by mean of the pollution control technologies and strategies mentioned above. Especially, the implementation of more strict emission standards have been regarded as one of the most effective tools in controlling air pollutants emissions. However, except for Hg & its compounds, other heavy metals & their compounds are still not regulated with specific emission ceilings in GB13223-2011. Consequently, the emission ceilings for other heavy metals & their compounds should be determined and regulated in the series of emission standards for thermal power plants in the near future. Besides, the execution of heavy metals emission reduction objective also requires the restraint of legislation and regulation, the support of economy, and the guidance of policy. Moreover, the awareness of public participation in environmental decision making is also an effective way in solving pollution and improving environmental quality.

4. Conclusions

This study evaluated the performances of different control measures on diminishing 8 types of heavy metals emissions, discussed the control potentials of these toxic elements emissions by conducting various scenario analyses with a whole process control concept, and proposed integrated control suggestions for diminishing the final discharges of heavy metals from CFPPs of China.

Presently, coal washing is an effective way to reduce the initial input and final discharges of heavy metals. The emissions of heavy metals from cleaned coal fired by power plant throughout the country would be 12.0–51.7% lower than those from directly combusting equal amount of raw coal as consumed by power plants. The best available APCDs configurations to control Hg and other elements are SCR + SMC + ESP + WFGD and SCR + FF + WFGD in the foreseeable future, respectively.

The significant mitigation potential of heavy metals emissions from 2010 to 2020 is primarily due to the increasing application rates of SCR, FFs and WFGD. Furthermore, with the continuous electricity growth and increasing demand for coal consumption, it might be difficult for thermal power plant to reduce Hg and Se emissions only by use of traditional APCDs in the future. More mitigation can be attained by further enhancement of clean and renewable energy and SMC applications. Under the alternative renewable HECT scenario for 2020 and 2030, the heavy metals emission can be reduced by 13.5–45.2% and 25.7–74.6% from the emission level of 2010, respectively.

Finally, integrated control suggestions have been proposed to minimize the final heavy metals emissions on accounting of the current status of CFPPs and future demand of energy-saving and pollution reduction.

Acknowledgments

This work is jointly funded by the National Natural Science Foundation of China (21177012 and 21377012), the Special Program on Public Welfare of the Ministry of Environmental Protection (201409022) and the Open fund of State Environmental Protection Key Laboratory of Sources and Control of Air Pollution Complex (No.SCAPC201305).

Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.jclepro.2015.05.008.

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