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Trend and characteristics of atmospheric emissions of Hg, As, and Se from coal combustion in China, 1980–2007

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Abstract. Emissions of hazardous trace elements in China are of great concern because of their negative impacts on local air quality as well as on regional environmental health and ecosystem risks. In this paper, the atmospheric emissions of mercury (Hg), arsenic (As), and selenium (Se) from coal combustion in China for the period 1980–2007 are estimated on the basis of coal consumption data and emission factors, which are specified by different categories of combustion facilities, coal types, and the equipped air pollution control devices configuration (Dust collectors, FGD, etc.). Specifically, multi-year emission inventories of Hg, As, and Se from 30 provinces and 4 economic sectors (thermal power, industry, residential use, and others) are evaluated and analyzed in detail. Furthermore, the gridded distribution of provincial-based Hg, As, and Se emissions in 2005 at a resolution of $1^\circ \times 1^\circ$ is also plotted. It shows that the calculated national total atmospheric emissions of Hg, As, and Se from coal combustion have rapidly increased from 73.59 t, 635.57 t, and 639.69 t in 1980 to 305.95 t, 2205.50 t, and 2352.97 t in 2007, at an annually averaged growth rate of 5.4%, 4.7%, and 4.9%, respectively. The industrial sector is the largest source for Hg, As, and Se, accounting for about 50.8%, 61.2%, and 56.2% of the national totals, respectively. The share of power plants is 43.3% for mercury, 24.9% for arsenic, and 33.4% for selenium, respectively. Also, it shows remarkably different regional contribution characteristics of these 3 types of trace elements, the top 5 provinces with the heaviest mercury emissions in 2007 are Shandong (34.40 t), Henan (33.63 t), Shanxi (21.14 t), Guizhou (19.48 t), and Hebei (19.35 t); the top 5 provinces with the heaviest arsenic emissions in 2007 are Shandong (219.24 t), Hunan (213.20 t), Jilin (141.21 t), Hebei (138.54 t), and Inner Mongolia (127.49 t); while the top 5 provinces with the heaviest selenium emissions in 2007 are Shandong (289.11 t), Henan (241.45 t), Jiangsu (175.44 t), Anhui (168.89 t), and Hubei (163.96 t). Between 2000 and 2007, provinces always rank at the top five largest Hg, As, and Se emission sources are: Shandong, Hebei, Shanxi, Henan, and Jiangsu, most of which are located in the east and are traditional industry-based or economically energy intensive areas in China. Notably, Hg, As, and Se emissions from coal combustion in China begin to grow at a more moderate pace since 2005. Emissions from coal-fired power plants sector began to decrease though the coal use had been increasing steadily, which can be mainly attributed to the increasing use of wet flue gas desulfurization (WFGD) in power plants, thus the further research and control orientations of importance for these hazardous trace elements should be the industrial sector.

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1 Introduction

Atmospheric emissions of hazardous trace elements such as mercury (Hg), arsenic (As), and selenium (Se) from coal combustion is one of the main sources of anthropogenic discharge and pollution. The negative effects of Hg, As, and Se on the environment and public health are well-documented in literature (Duker et al., 2005; Zhang and Wong, 2007; Lenz et al., 2009), and have received a wide range of attention throughout the world.

China is one of the few countries in the world whose coal consumption constitutes more than 75% of the country's total energy sources (You et al., 2009). With the rapid development of economy, coal consumption has been increasing considerably. Though it is estimated that coal constitution will drop to about 54% in 2020 (NDRC, 1998), the total amount of raw coal as consumed is expected to rise to as much as 2.6 billion tonnes till then (You et al., 2009).

In fact, the rising Hg, As, and Se emissions with the growth of coal combustion have led to serious environmental pollution and economic losses in China. Elevated atmospheric Hg concentration in urban areas has been reported in several recent studies. Many densely populated cities, such as Beijing, Changchun, Guiyang and Chongqing, tend to have higher concentrations of atmospheric particulate Hg and total gaseous Hg, especially in the cold season (heating period) (Wang et al., 1996a; Fang et al., 2001; Liu et al., 2002; Feng et al., 2004; Jiang et al., 2006). In Sichuan and Guizhou provinces, high atmospheric mercury deposition, both total and wet, due to high emissions from coal combustion is found during fall and winter months (Fang et al., 2009). It is reported that the average gaseous elemental Hg concentration in China is higher than 6 ng/m^3 while the global background range is 1–3 ng/m^3 (Zhang and Wong, 2007; Tayban et al., 2005). Atmospheric Hg contamination in China should receive great attention right now.

China is one of the most serious As poisoning countries in the world. Cases of endemic As poisoning caused by coal-burning have been found in Xinjiang, Inner Mongolia, Sichuan and Guizhou provinces, particularly in southwest China, since the early 1970s. Guizhou province is classified as the heaviest coal-burning As poisoning district in China. 265 people died of As poisoning in Xingren county in Guizhou from 1976 to 2003 and more than 3000 As poisoning cases have been reported until now (Tian et al., 2008).

Selenium is an essential trace element for human beings and animals. However, high concentration of Se may be toxic for organism and results in hair and nail loss as well as nervous system disorders in humans (Zhu et al., 2008). There are two notable seleniferous regions in China: the Ankang region in Shaanxi Province and En-shi Prefecture in southwest Hubei Province owing to the exposure and burning of high selenium content hard coal. During the past few decades, 477 cases of human Se poisoning and more than 10 000 cases of swine Se poisoning have been reported in Enshi (Mao et al., 1990). Currently, cases of Se poisoning among swine and goats are still being reported in some high-Se villages, and several human cases of Se poisoning, while sporadic, are reported again in recent years (Zhu et al., 2008).

In recent years, there is a widespread concern over the control and reduction of atmospheric Hg emissions in China. Emission of Hg from coal combustion has been estimated for the whole country and some particular regions. Feng and Hong (1996) estimated that 296 tons of Hg were released in China by coal combustion sources in 1994. Wang et al. (2000) estimated the total Hg emitted to the atmosphere from coal combustion as 213.8 tons in 1995 and over 2400 tons of Hg had been emitted from 1978 to 1995 through coal combustion with an average increasing rate of 4.8% per year. Jiang et al. (2005) estimated 219.5 or 161.6 tons of Hg (based on different coal mercury contents) were released into the atmosphere from coal combustion in 2000. Streets et al. (2005) estimated 202.4 tons in 1999, of which 51% came from industrial use, 33.6% from power plants, 9.8% from residential use, and 5.6% from other uses. Wu et al. (2006) estimated the total Hg emissions from coal combustion in China increased from 202.4 tons to 256.7 tons with an annual growth rate of 3.0% during 1995–2003.

However, the comprehensive and detailed studies on As and Se emissions in China are quite limited. Wang et al. (2008a) estimated that 442.31 tons of As were released from coal combustion in 1997, 76.9 tons of which were from coal-fired power plants. Until now, there have been few published estimates to date of Se emissions in China and we preliminarily estimated the total Se emitted from coal combustion in China in 2005 in our previous study (Tian et al., 2008, 2009).

In this paper, the historical trend and characteristics of anthropogenic atmospheric emissions of Hg, As, and Se from coal combustion during the period of 1980–2007 are evaluated. Specially, temporal atmospheric emission inventories of Hg, As, and Se are developed and discussed in detail by economic sectors, coal types, provinces, and regions between 1995 and 2007.

2 Methodology

The emissions of Hg, As, and Se from coal combustion are calculated using provincial-level coal consumption data and detailed emission factors. The basic formulas can be expressed as follows:

$$E_{i,j} = C_{i,j} F_{i,j} EF_{i,j} (1-P_{\mathrm{DC}(i,j)})(1-P_{\mathrm{FGD}(i,j)}), \tag{1}$$

$$E_i = \sum E_{i,j}, \tag{2}$$

$$E_T = \sum E_i. \tag{3}$$

where E is the emissions of atmospheric Hg, As, or Se; C is the averaged Hg, As, or Se content of coal as consumed in one province; F is the amount of coal consumption; EF is the fraction of Hg, As, or Se released from coal combustion; P_{DC} and P_{FGD} are the fraction of Hg, As, or Se removed by the existing dust collectors and flue gas desulfurization (FGD) devices, respectively; T is the national totals; i is the province (autonomous region or municipality); and j is the emission source classified by economic sectors, combustion facilities, and the equipped PM and SO_2 control devices.

2.1 Averaged content of Hg, As, and Se in raw coal as produced

China is a huge country with 34 provinces and regions, and the content of Hg, As, and Se in coal mined from different places varies substantially due to the coal-forming plants and the coal-forming geological environments. Table 1 shows the analyzed results of Hg, As, and Se content in raw coal as produced from 30 provinces (autonomous regions, and municipalities) on the Chinese mainland; Hong Kong Special Administrative Region, Macau Special Administrative Region, and Taiwan province are not included tentatively. Xizang is also not considered because the amount of coal produced and consumed in this area is very small.

The provincial values of averaged Hg content adopted in our estimation as shown in Table 1 are summarized from both USGS data and domestic literature data (Wang et al., 2000; Zhuang et al., 1999; Chen et al., 2006; Feng et al., 2002; Huang and Yang, 2002; Ren et al., 2006; Zheng et al., 2007). We use the mean values of Hg content of coals for each province reported in literatures above. For provinces with no other coal samples, such as Gansu, Ningxia, Qinghai, etc., we use only USGS data. Among these 30 provinces, the lowest mean concentration of Hg is 0.025 mg/kg in Xinjiang province while the highest concentration is 0.368 mg/kg in Guizhou province. As a result, the national production-weighted averaged Hg content in coal as produced in China is estimated at about 0.185 mg/kg in 2005.

The distribution characteristics of As content of Chinese coals have been investigated by some researchers and institutions (Chen et al., 1989, 2002; Wang, 2000; Wang, 2005, 2008b; Ren et al., 2006). Chen et al. (1989) reported the range of As content of coals from different provinces. Wang (2000) compiled As concentration of 1018 coal samples collected by geological survey bureaus from all over the country. Wang (2005) evaluated the range and the arithmetic mean values of As content in coals of five coal basins as well as each province based on the tested results of 297 coal samples. Based on the literatures above, we determined the averaged As content of raw coal of each province. Consequently, the national production-weighted averaged As content in coal as produced in China is estimated at about 4.853 mg/kg in 2005.

Compared with Hg and As, there are relatively fewer works targeted on Se content of coals mined from different provinces in China (Chen et al., 1989; Chen and Tang, 2002; Bai, 2003; Zhang et al., 2007). Bai (2003) collected 1018 coal samples and indicated that Se content of most Chinese coal was between 0 and 52.9 µg/g with an averaged value of 4.01 µg/g. In this study, we use the mean value of Se content of coals for each province compiled from literatures above. However, limited by a lack of access to more specific data, the values for Beijing, Gansu, Guangdong, Guangxi, Ningxia and Qinghai are assumed to be equal to the average Se content of the corresponding coal-cumulating areas listed in Bai (2003). Consequently, the national production-weighted averaged Se content of coal as produced in China is estimated at 3.248 mg/kg in 2005.

No significant difference in trace elements content in coal samples among different years has been reported. Therefore, we assume that the averaged content of Hg, As, and Se of raw coal as produced did not change during the period of this study.

2.2 Averaged content of Hg, As, and Se in raw coal and coal products as consumed

The geographical distribution of coal resources in China is extremely unbalanced, less in the southern and eastern areas while abundant in the northern and western areas in general. As a result, large quantities of coal produced have to be transported long-distance from production areas to consumption areas, leading to remarkable variation between the trace elements content in coal as produced and consumed in one province.

According to the statistical data retrieved from China Energy Statistical Yearbooks (1997–2008) and China Coal Industry Yearbooks (2005–2008), annual coal flow matrixes among 30 provinces are established to quantify in-province coal use and inter-province coal flows from 2004 to 2007. From 1995 to 2003, only 1996 and 1999 coal flow matrixes are set up due to statistical data restrictions. It has been proven that the inter-province coal supply patterns were relatively steady for the majority of provinces between 1996 and 1999 (Streets et al., 2005). Therefore, we apply the 1996 flow matrix to 1995, 1997 and 1998, and the 1999 matrix to the other years (2000 to 2003). For the years before 1995, we use the 1996 flow matrix instead due to lack of inter-province coal transportation data.

Table 1 presents the calculated results of the weighted averaged content of Hg, As, and Se in raw coal as consumed in China by province in 2005. As can be seen, for province of which coal is obtained mainly from within-province supply, such as Guizhou, Shaanxi and Shanxi, there is little change in Hg, As, and Se content between raw coal as produced and raw coal as consumed. In provinces with large amounts of coal export (Shanxi, Shaanxi, Inner Mongolia, etc.), Hg, As, and Se content of raw coal is relatively lower, leading to the weighted averaged element content of raw coal consumed in most provinces lower than that produced. Consequently, the national consumption-weighted averaged Hg, As, and Se content of raw coal as consumed in China is much lower than that of raw coal as produced.

Presently, coal washing before combustion is an effective way to reduce ash and SO_2 emission. It can reduce sulfur pyrites content by 40% (You et al., 2009). This way, not only ash and SO_2 emission, but also trace elements concentration are reduced and the heating value of cleaned coal is increased.

In China, coal briquettes and coke are produced from both raw coal and the cleaned coal. The final content of Hg, As, and Se in the cleaned coal, briquettes and coke are calculated based on the following equation:

$$C_{\mathrm{cc}/b/c,i} = \frac{(C_{\mathrm{rc},i} M_{\mathrm{rc},i} + C_{\mathrm{cc},i} M_{\mathrm{cc},i})(1-F)}{P_{\mathrm{cc}/b/c,i}}. \quad (4)$$

Table 1. Results of averaged content of Hg, As, and Se in raw coal as produced and consumed by province in China, 2005 (mg kg^{-1}).

Province	Raw coal production (Mt)	Raw coal consumption (Mt)	Hg_P	Hg_C	As_P	As_C	Se_P	Se_C
Anhui	86.35	56.54	0.299	0.282	2.902	3.037	8.001	7.291
Beijing	9.45	26.01	0.340	0.154	3.280	2.947	0.600	2.365
Chongqing	34.00	25.63	0.135[a]	0.136	5.620[a]	5.625	3.290[a]	3.293
Fujian	20.00	43.76	0.070	0.103	9.890	5.591	4.300	3.235
Gansu	36.20	32.29	0.050	0.058	4.250	4.005	0.360	0.691
Guangdong	4.77	94.66	0.050	0.125	7.330	4.179	5.320	2.412
Guangxi	7.00	31.30	0.330	0.296	17.205	8.397	5.320	3.558
Guizhou	106.15	68.00	0.368	0.367	6.719	6.714	3.815	3.812
Hainan	0	3.38	0[b]	0.086	0[b]	1.635	0[b]	1.080
Hebei	86.39	141.68	0.133	0.144	4.087	3.524	1.840	2.396
Heilongjiang	97.37	54.89	0.118	0.141	3.240	4.050	0.550	0.692
Henan	187.61	131.04	0.273	0.261	2.765	3.063	4.738	4.617
Hubei	7.78	70.01	0.160	0.203	5.155	3.857	9.710	5.389
Hunan	69.04	66.88	0.105	0.158	11.950	8.718	3.200	3.487
Inner Mongolia	256.08	100.61	0.198	0.167	6.035	4.822	0.945	0.934
Jiangsu	28.18	150.46	0.179	0.169	2.433	2.811	7.825	4.163
Jiangxi	25.65	30.31	0.215	0.217	7.860	6.218	11.700	8.007
Jilin	27.15	60.79	0.271	0.211	12.745	8.043	4.220	2.122
Liaoning	66.41	91.94	0.171	0.168	5.764	5.179	0.868	1.323
Ningxia	26.60	23.36	0.200	0.200	3.955	3.955	5.300	5.300
Qinghai	5.55	6.72	0.040	0.051	2.660	3.428	0.360	0.555
Shaanxi	152.46	40.90	0.141	0.141	3.415	3.388	3.380	3.350
Shandong	140.30	215.39	0.212	0.194	5.055	4.543	3.912	3.707
Shanghai	0	41.01	0[c]	0.154	0[c]	2.943	0[c]	2.916
Shanxi	554.26	93.45	0.168	0.167	3.633	3.591	3.484	3.435
Sichuan	79.05	57.20	0.135	0.145	5.620	5.706	3.290	3.292
Tianjin	0	32.80	0[d]	0.138	0[d]	2.944	0[d]	2.541
Xinjiang	39.42	34.11	0.025	0.026	2.236	2.249	0.198	0.236
Yunnan	64.62	42.33	0.133	0.141	8.910	8.768	1.333	1.456
Zhejiang	0.44	94.73	0.350	0.154	11.000	3.185	12.100	3.199
Arithmetic average			0.180	0.165	6.138	4.571	4.073	3.028
Weighted average	–	–	0.185	0.178	4.853	4.478	3.248	3.200

$_P$ stands for raw coal as produced and $_C$ stands for raw coal as consumed.
[a] The value is assumed to be equal to that for Sichuan province due to lack of samples.
[b,c,d] No raw coal produced in Hainan, Shanghai, and Tianjin, thus the values of Hg, As, and Se content are assumed to be zero.

where $C_{cc/b/c,i}$ is the averaged content of Hg, As, or Se in the cleaned coal, briquettes or coke; $M_{rc,i}$ and $C_{rc,i}$ are the amount and averaged Hg, As, or Se content of raw coal input in the production; $M_{cc,i}$ and $C_{cc,i}$ are the amount and averaged content of cleaned coal input in the production of briquettes or coke; F is the fraction of Hg, As, or Se removed by the coal washing process, briquette production or coke making process; $P_{cc/b/c,i}$ is the amount of cleaned coal, briquettes or coke as produced; and i is the province (autonomous region or municipality).

The removability of Hg, As and Se during physical coal cleaning is mainly determined by the modes of occurrence of Hg, As, and Se in coal as well as the washability of inorganic minerals. Elements associated with inorganic phases offer a potential for removal by physical coal cleaning procedures, whereas those with affinity to organic constituents will not be reduced effectively (Finkelman et al., 1999; Wang et al., 2003, 2008a; Bai et al., 2003; Wang, 2007). The modes of occurrence of Hg, As, and Se vary significantly in different coals and definitive determination of the forms of occurrence of these elements is very difficult. On the whole, Hg and As show a strong affinity to inorganic materials and the most likely forms of occurrence are association with sulfides, especially with pyrite (Finkelman, 1994; Feng et al., 2002; Guo et al., 2004; Song et al., 2006; Wang et al., 2008a).

The removal effect of Hg and As is approximately similar to the sulfur in pyrite and it is indicated that sulfur in pyrite can be removed effectively during physical coal cleaning and the removal efficiency can reach up to 50% (Song et al., 2006; Luttrell et al., 2000). As a result, a large proportion

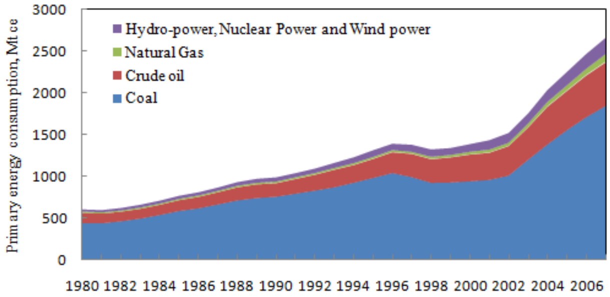

Fig. 1. Trend of primary energy consumption and its structure in China, 1980–2007.

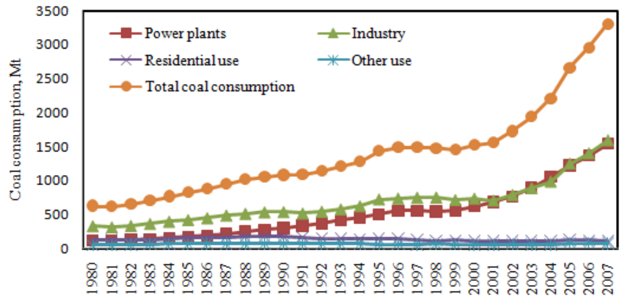

Fig. 2. Trend of coal consumption by different sectors in China, 1980–2007.

of Hg and As associated with pyrite can be washed away simultaneously. The reported Hg removal efficiency of coal cleaning varies from about 30% to about 75% and almost all the physical coal cleaning procedures tested on Chinese coals have a Hg removal efficiency larger than 50% (Akers and Dospoy, 1994; Luttrell et al., 2000; Wang et al., 2003; Song et al., 2006; Wang, 2007). The As removal efficiency of coal cleaning can vary from about 40% to about 70% (Akers and Dospoy, 1994; DeVito et al., 1994; Finkelman, 1994; Luttrell et al., 2000; Wang et al., 2003; Wu et al., 2005; Song et al., 2006; Wang, 2007) and most estimates are between 50% and 60%. Thus, we assume the average removal efficiency of Hg and As to be 50% and 54% respectively independent of mercury and arsenic content in raw coal.

Unlike Hg and As, Se exists in many different forms and the main form of occurrence is quite different from coal to coal. Analysis of chemical properties shows that Se is not only a sulfophilic element which tends to associate with sulfides, but also a biophile element which is much easier to accumulate in organic matters. Moreover, Se may also exist in exchangeable state and residual fractions. Previous studies have confirmed that Se shows a high organic affinity in coal and in most cases exists in organic phases (Finkelman, 1994; Liu et al., 2003; Zhu et al., 2003; Wang, 2007). As a result, the removal efficiency of Se is relatively low and mostly ranges from 20% to 41.9% (DeVito et al., 1994; Finkelman, 1994; Wang et al., 2003; Song et al., 2006; Wang, 2007). In this study, we assume an average Se removal efficiency of 30%.

According to the temperature levels, the coke making process can be divided into four stages: primary coal carbonization (400°), semi-coke formation (600°), secondary coal carbonization (850°) and coke formation (1000°) (Zajusz-Zubek et al., 2003). When it comes to the final stage, high volatility will be found, especially for Hg. The mean volatility is found for Se and the lowest for As. Experimental coking suggested that a coke kept nearly 10%–15% of the total Hg (Wang et al., 2000; Hong et al., 2002; Zajusz-Zubek et al., 2003; Huang et al., 2004), 60%–65% of the total Se and 70% of the total As (Zajusz-Zubek et al., 2003; Huang et al., 2004); while the rest escaped into volatile matter. Thus, we assume that 10% of the Hg, 60% of the Se and 70% of the As remains after the coking process.

2.3 Coal consumption

Coal consumption data by sectors and types of coal products is provincial-level data compiled from China Energy Statistical Yearbooks from 1980 to 2007 (DITS, 1992, 1998; NBS, 2001, 2004; NBS and NDRC, 2005∼2009). Taiwan province, Hong Kong and Macau Special Administrative Region are not included. In 1980, the total energy consumption in China was only 602.75 million metric tons of standard coal equivalent (Mtce). With the continuously rapid growth of economy, China's consumption of primary energy increased steadily and quickly (except for a few years in late 1990s) as can be seen in Fig. 1, especially from the beginning of the 21th century. By the end of 2007, the total energy consumption in China had reached 2655.83 Mtce with coal consumption accounting for about 70 percent of the totals. Although the use of cleaner high-quality energy such as hydro-power, natural gas, wind power and nuclear power has been introduced and enlarged to optimize the energy structure in China, the coal-dominated energy structure will remain unchanged in a relatively long time.

The industry sector is the leading sector in total coal consumption while the power sector comes next as can be seen in Fig. 2. A relatively high growth rate in coal consumption of power plants and industry sector can be found since the beginning of the 21st century. By the end of 2007, the amounts of coal consumed in the industry sector and power plants were 1584.38 and 1532.38 million tons, representing 48.0% and 46.4% of the totals, respectively.

2.4 Emission factors of Hg, As, and Se

The release rates of Hg, As, and Se depend greatly on combustion technology and operation conditions, it is necessary to give a detailed specification of the ways in which coal is burned in China. In this study, all the coal combustion sources are divided into 4 categories: power plants (POW),

Table 2. The release rates of Hg, As, and Se from coal combustion.

	Release Rate (%)			
Boiler type	Hg	As	Se	Literature cited
pulverized-coal boiler	99.50			Zhou et al. (2008)
pulverized-coal boiler	99.53			Jiang et al. (2007)
pulverized-coal boiler	99.10			Zhu et al. (2002)
pulverized-coal boiler	99.20			Meij et al. (2002)
pulverized-coal boiler	99.13			Lee et al. (2006)
pulverized-coal boiler	99.77			Zhang et al. (2008)
pulverized-coal boiler	99.73	98.90	93.97	Otero-Rey et al. (2003)
pulverized-coal boiler		99.47		Guo et al. (2004)
pulverized-coal boiler			99.70	Andren and Klein. (1975)
pulverized-coal boiler		97.00	95.00	A'lvarez-Ayuso et al. (2006)
In this study				
stoker fired boiler	83.15	77.18	80.95	Wang et al. (1996b)
fluidized-bed furnace	98.92	75.60	98.05	Demir et al. (2001)
coke furnace	85.00	30.00	40.00	Zajusz-Zubek et al. (2003)
pulverized-coal boiler	99.42	98.46	96.22	Averaged value

industrial use (including construction) (IND), residential use (RES), and other uses (OTH) including storage, post and telecommunications, wholesale and retail trades, catering industry, transportation, and so on. Then, the sources of power sector and industry sector are further classified into different sub-groups based on types of combustion facilities, the equipped particulate control devices, as well as flue gas desulfurization (FGD) systems. By the end of 2007, the amounts of power plants installed with SCR devices to reduce NO_x emission were relatively small (about 19.6 GWe) and some of them were believed not to fully operate most of the time, thus the influence of SCR devices on Hg, As, and Se emissions is not considered tentatively in this study.

Coal combustion facilities are divided into 4 types: pulverized-coal boiler, fluidized-bed furnace, stoker fired boiler and coke furnace. Presently, pulverized-coal boilers take a large proportion in coal-fired power plants in most of the provinces in China, representing over 90% of the totals. The remaining share is represented by fluidized-bed furnaces as well as stoker fired boilers which are mainly used in small coal-fired power plants. Different from power sector, stoker fired boiler is the dominant boiler type used in the industry sector. The release rates of Hg, As, and Se from different combustion boilers are compiled and listed in Table 2.

Besides, the conventional air pollution control devices (APCD) that are utilized on coal-fired utility boilers or industry boilers for reducing NO_x, SO_2 and PM, will affect Hg, As, and Se speciation and are effective in reducing the final Hg, As, and Se emissions.

Presently, all coal-fired power plants in China have adopted particulate control devices, such as cyclone, wet scrubber, fabric filters (FFs), as well as electrostatic precipitators (ESP). ESP is now the most widely used particulate abatement measure in major electric power plants and central heating plants in China due to its high efficiency and relatively lower operation expenses. The share of ESP has reached more than 95% and is as high as almost 100% in provinces such as Guizhou, Guangxi and Yunnan. Fabric filters are more effective in reducing fine particles and gaseous Hg, As, and Se emissions, and have been applied gradually since 2001. By the end of 2007, about 2% of the total installed capacity heat units have been equipped with FFs. Wet scrubbers and cyclones are the two main types of PM control device used on industrial boilers. More and more ESP and FFs have also been introduced into the industrial sector in recent years for complying with more stringent emission standards.

SO_2 post-combustion control technologies are mainly used in power plants and fall into three classifications: wet, semi-dry, and dry systems. In China, the total capacity of coal-fired power plants equipped with wet-FGD was only 1 GWe by the end of 1995 (You et al., 2009). Since 2000, the installed capacity of power plants with FGD increased rapidly and had grown up to 265.6 GWe, representing almost 50% of the total capacity by the end of 2007. Moreover, of all the coal-fired units performed FGD, more than 92.3% used wet scrubbers.

In this study, we compile and apply the time-varying provincial-level technology data (Wang, 1999, 2010; Jiang et al., 2005; Zhang, 2005; Streets et al., 2005; NDRC, 2008) for our emission inventory calculations. In recent years, there has been much research on the removal efficiency of Hg from the flue gas by the existing PM and SO_2 control devices. We adopt the average values of the reported efficiency in literature (Table 3). In the case of relatively fewer sampling data, the removal efficiencies of As and Se by FF and wet scrubber are taken from just one study, as well as the removal efficiency of Se by cyclone.

Residential use is also an important coal-consuming sector in China. The major combustion types for residential cooking and heating are traditional cookstoves and improved cookstoves, both of which are without any PM control device. However, there is very little information about Hg, As, and Se emissions through residential use. Here, we choose to use the emission factors for coal/briquette combustion provided by NPI (1999), and the emission factors of Hg, As, and Se through residential use are assumed at 6.5×10^{-5} g/kg, 9.5×10^{-5} g/kg, and 6.5×10^{-4} g/kg, respectively.

3 Results and discussion

3.1 Emissions by sectors

As can be seen in Fig. 3, with the continuously rapid growth of economic activity and coal consumption, the total emissions of Hg, As, and Se in China have been growing steadily during the past years, increasing from 73.59 t, 635.57 t, and 639.69 t in 1980 to about 305.95 t, 2205.50 t, and 2352.97 t in 2007, at an annually averaged growth rate of 5.4%, 4.7%, and 4.9%, respectively.

Among all of the coal consuming sectors, emissions of Hg, As, and Se from power plants have been increasing fastestly, at an annual growth rate of 7.7%, 10.9%, and 9.2%, and reaching at 132.36 t, 550.08 t, and 786.83 t in 2007, respectively. Although the total emissions of Hg, As, and Se from the industrial sector did not increase as fast as those from the power sector (nearly 5%), industrial coal combustion was still the largest single sector for Hg, As, and Se emissions among all the coal consumption sectors, increasing from 40.8 t, 382.67 t, and 364.81 t in 1980, to 155.46 t, 1348.70 t, and 1322.78 t in 2007, respectively. Power sector and industrial sector combined had accounted for almost 90 percent of the total emissions. Coal consumption for residential and other uses decreased during the period due to fuel substitution with more cleaner fuels such as natural gas and electricity, especially in urban areas, resulting in the emissions of Hg, As, and Se dropped down. In 2007, the total emissions of Hg, As, and Se from residential coal use were estimated at 6.96 t, 10.18 t, and 69.64 t, respectively, even lower than those of 1980.

As can be seen from Fig. 4, the emissions of Hg, As, and Se from power plants and industrial sector in China have experienced continuous and rapid growth since 1980, except for two fluctuations around 1991 and 1999. In recent years, the increase rates of total Hg, As, and Se emissions have slowed down quite remarkably. The main reason is that the emissions of Hg, As, and Se from power sector declined substantially since 2005 thanks to the co-benifit reduction effects of the existing and newly installed APCD in coal-fired power plants, such as ESP, FFs and FGD. However, the emissions from coal combustion in industrial sector still experienced a rapid increase owing to the lower penetration of advanced PM and SO_2 control devices.

In view of the large proportion of power sector and industrial sector in the emissions of Hg, As, and Se, the relationship between the increase rates of coal consumption and Hg, As, and Se emissions of the two sectors are discussed.

Table 3. Removal efficiencies of Hg, As, and Se from coal-fired power plants.

	Removal efficiency (%)			
Control device	Hg	As	Se	Literature cited
ESP	30.2			Zhu et al. (2002)
ESP	28.9	96.1	49.1	Helble (2000)
ESP	27			Pavlish et al. (2003)
ESP	33			Yokoyama (1999)
ESP	49.6	98.3	82.4	Meij and Henk (2007)
ESP	24			Chu and Porcella (1995)
ESP	42			Afonso et al. (2001)
ESP	36			Yang et al. (2007)
ESP	29			Srivastava et al. (2006)
ESP	32	95		Brekke et al. (1995)
ESP		87.5		Radian Corporation (1989)
ESP		51.8		Guo et al. (2004)
ESP			71.3	Xu et al. (2005)
ESP		88.5	92.3	Ondov et al. (1979)
FF	58			Pavlish et al. (2003)
FF	28.5			Chu and Porcella (1995)
FF	82			Afonso et al. (2001)
FF	90			Yang et al. (2007)
FF	89			Srivastava et al. (2006)
FF	60	99	65	Brekke et al. (1995)
wet scrubber	4.3			Chu and Porcella (1995)
wet scrubber	26			Afonso et al. (2001)
wet scrubber		96.3	85	Ondov et al. (1979)
cyclone	12	35	40	Huang et al. (2004)
cyclone	0			Chu and Porcella (1995)
cyclone		51		Radian Corporation (1989)
WFGD	70			Renninger et al. (2004)
WFGD	75			Díaz-Somoano et al. (2007)
WFGD	60	97	98	A'lvarez-Ayuso et al. (2006)
WFGD	50.2	75	65.6	Meij and Henk (2007)
WFGD	30.9			Chu and Porcella (1995)
WFGD		51.5		Radian Corporation (1989)
WFGD		98	61	Brekke et al. (1995)
In this study				
ESP	33.17	86.20	73.78	Averaged value
FF	67.92	99	65	Averaged value
wet scrubber	15.15	96.30	85	Averaged value
cyclone	6	43	40	Averaged value
WFGD	57.22	80.38	74.87	Averaged value

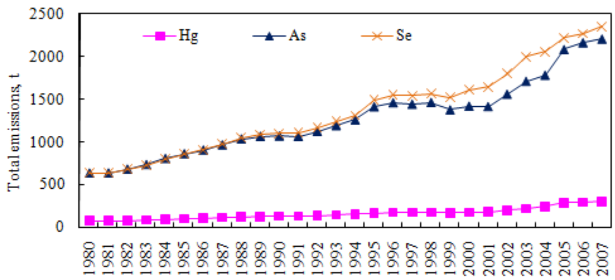

Fig. 3. Trend of the total emissions of Hg, As, and Se from coal in China, 1980–2007.

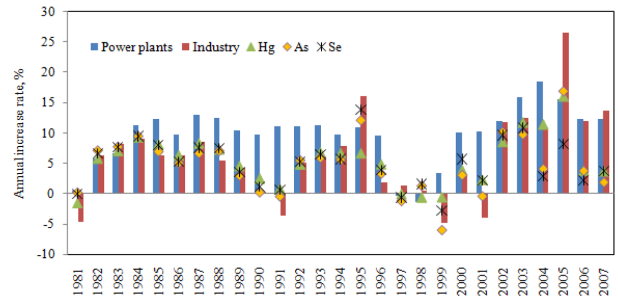

Fig. 5. Annual increase rate of coal consumption and Hg, As, and Se emissions, 1980–2007.

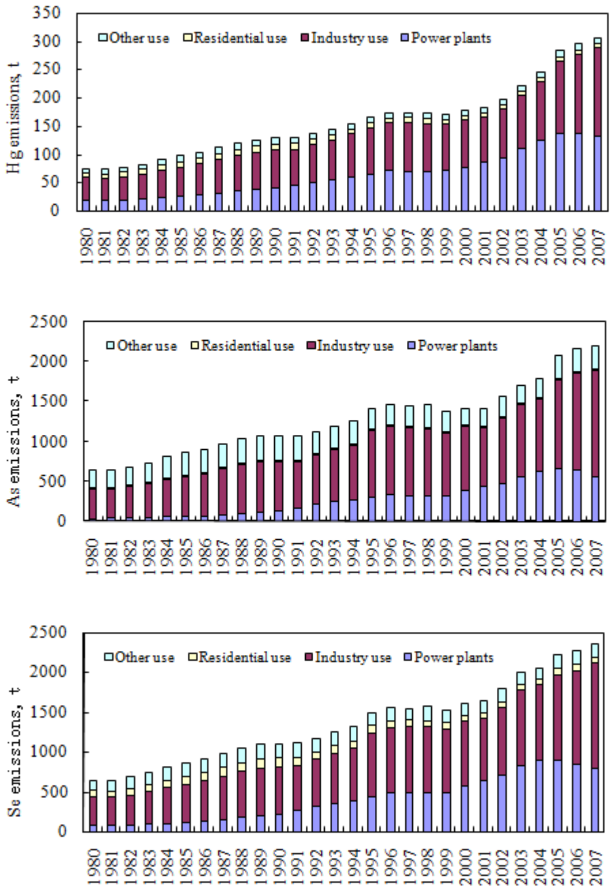

Fig. 4. Trend of Hg, As, and Se emissions by sectors in China, 1980–2007.

As can be seen from Fig. 5, the coal consumption in power sector and industrial sector presented a rapid growth tendency in most of the years over the past decades. The increase rate of coal used for power generation was greater than that for industrial use except for a few years. In 1997 and 1998, negative growth was found in coal consumption of power sector over the last year, which can be mainly attributed to the Asian financial crisis as referred to in our previous study (Hao et al., 2002). As far as the industrial sector is concerned, there were two large fluctuations, one was between 1990 and 1991, another was around 2000. Since the beginning of the 21th century, with the rapid expansion of energy intensive manufacturing industry, such as steel and cement production, the coal consumption in power sector and industrial sector both has maintained a relatively rapid growth rate. By the end of 2005, the coal consumption of the industrial sector had reached 1244.66 million tons, increased by 261.36 million tons over the previous year.

It can be concluded that the emissions of Hg, As, and Se from these two sectors are well associated with the coal consumption changes in each sector. However, the annual increase rates of the total Hg, As, and Se emissions are much more similar to that of industrial coal use compared to power sector. The decrease/increase in the total Hg, As, and Se emissions is basically consistent with the variation trend of the industrial coal use. For example, the coal consumption in power sector increased by 63.58 million tons in 2001 while the industrial coal use decreased by 29.26 million tons. As a result, the total Hg and Se emissions increased slightly while As emission decreased over the previous year. Total emissions of Hg, As and Se are sensitive to the coal consumption changes in the industrial sector. The main reason is that the proportion of industrial coal use is very large, and the net emission rate is relatively higher than those of power plants. Although the coal consumption of power sector has caught up with that of industrial sector in recent years, the Hg, As, and Se emissions released by per ton coal use in power sector are much lower due to the widely installation of advanced pollution control devices such as ESP, FFs, and Wet Limestone-Gym FGD.

Further, we can find in Fig. 5 that the growth rates of coal used in the power plants and industry in 2004 are close to those in 2003, and the growth rates of Hg are comparable between the two years. However, the growth rates of As and Se significantly decrease in 2004. This can be explained by the combined effects of the difference in elements content in coal as consumed deduced from different inter-provinces coal flow matrix between 2003 and 2004 as mentioned before, the difference in allocation pattern of coal used in the power and industry among 30 provinces, as well as the difference in reduction efficiency of different dust collectors and FGD between Hg and the other two elements (see Table 3).

Since the beginning of the 21th century, especially the introduction of 11th five-year-plan after 2005, Hg, As, and Se emissions from coal combustion in China have begun to grow at a more moderate pace in spite of the continuous rapid coal consumption. This can be attributed to two main reasons: (1) the implementations of various APCD, especially the request for widely installation and operation of FGD to reduce SO_2 emissions in coal-fired power plants (SEPA, 2007); (2) close-down and suspension of small-scale thermal power plants (NDRC, 2008). By the end of 2007, the installed capacity with FGD had reached 265.6 GW, of which more than 92.3% used wet scrubber systems (see Fig. 6). As mentioned above, the removal efficiency of Hg, As, and Se by WFGD may reach 57%, 80%, and 75%, respectively, and can be even higher when used in combination with other emission control devices. In 2007, small thermal power plants with a capacity of 14.38 MW have been shut down in China (NDRC, 2008). With the implementation of a series of comprehensive regulations and policies, the proportion of the installed capacity featured with heavy energy consumption, heavy pollution and low efficiency dropped dramatically. As a result, the net emissions from power sector began to decrease. The increase rates of the total Hg, As, and Se emissions are much lower than that of coal use, and the further research and control orientations of importance should be put on industrial coal use.

3.2 National and regional Hg, As, and Se emissions

The total emissions of Hg, As, and Se by provinces in China for 2000, 2005 and 2007 are summarized in Table 4. Some provinces show much higher emission growth, e.g., Hebei, Inner Mongolia, Shandong; however, some other provinces show declined Hg, As, and Se emissions during this period, e.g., Beijing, Shanghai, Ningxia.

Hg, As, and Se emissions from coal combustion are strongly affected by specific source-related trends in each province which can mainly be divided into 4 types: (1) the large increase in Hg, As, and Se emissions for Hebei province, is primarily attributed to greatly increased coal consumption in the industrial sector; (2) the increase in annual Hg, As, and Se emissions in Shandong, Inner Mongolia, Jiangsu, Henan and Shanxi province, is mainly driven by the continuously rapid increase in coal used for power generation; (3) emission reduction in Beijing is primarily due to the dramatic reduction in industrial coal use as well as the fast penetration of advanced air pollution control technologies, such as ESP and FGD, in the power sector; and (4) emission reduction in Ningxia and Guangdong province in spite of the increase in coal use can be attributed to the combined effects of implements of various APCD.

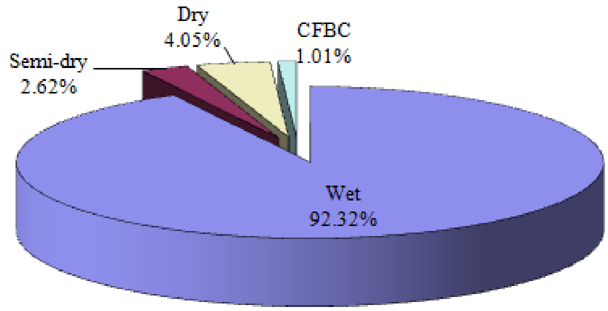

Fig. 6. Constitution of FGD installation in power plants in China, 2007.

Between 2000 and 2007, provinces always ranked in the top five largest Hg, As, and Se emissions are: Shandong, Hebei, Shanxi, Henan, and Jiangsu, most of which are located in the east and are traditional industry-based or economically intensive areas in China. Meanwhile, some provinces are also demonstrated as high emission districts with one or more elements emissions higher than other provinces, e.g. As of Hunan, As of Jilin, and Se of Anhui. This is mainly owing to the high element content in the raw coal produced and/or consumed in these areas as can be seen in Table 1.

In general, the provincial emission inventories of Hg, As, and Se coincide well with their industrialized levels and population. All provinces emitting large amounts of Hg, As, and Se are relatively developed economically and/or densely populated, which causes much higher coal consumption in industrial sector or power generation.

In order to investigate the regional distribution characteristics of Hg, As, and Se emissions, we divide China into six large regions, namely the Northern Region, the Northeastern Region, the Eastern Region, the Central and Southern Region, the Southwestern Region, and the Northwestern Region. Hg and Se emissions in the Eastern Region contribute the largest portion, about 92.44 t of Hg and 919.97 t of Se in 2007, a share of approximately 30.2% and 39.1%, respectively, of the national totals. As emission in this district comes second, about 541.60 t, representing 24.6% of the totals. The next three largest contributors of Hg and Se are the Central and Southern Region, the Northern Region and the Southwestern Region. For As, the largest contributor is the Central and Southern Region, a little different from Hg and Se. This is mainly due to the high As content of coal consumed in Central and Southern provinces. Emissions in the Northwestern Region are much smaller compared to other regions which can be attributed to the lower element content in raw coal both as produced and consumed.

It can be seen that the emissions of Hg, As, and Se from coal combustion in China are mainly concentrated in the Eastern Central and Southern areas, which cover about 45%

Table 4. Provincial inventories of Hg, As, and Se emissions in China (t/a).

Province/region	2000			2005			2007		
	Hg	As	Se	Hg	As	Se	Hg	As	Se
Northern Region	38.59	254.30	241.18	59.63	379.76	386.49	62.90	389.50	393.21
Beijing	3.79	24.82	24.42	3.00	22.85	24.67	2.52	20.35	22.62
Tianjin	2.24	16.59	19.96	3.62	23.62	29.59	3.22	23.42	27.76
Hebei	11.46	88.08	82.70	18.71	149.75	159.39	19.35	138.54	160.37
Shanxi	13.70	58.41	90.19	20.92	79.87	136.40	21.14	79.70	142.60
Inner Mongolia	7.39	66.40	23.91	13.39	103.67	36.43	16.68	127.49	39.86
Northeastern Region	21.34	216.94	95.00	29.76	263.29	107.80	32.89	293.26	124.77
Liaoning	10.10	83.56	39.73	13.43	95.95	47.01	14.96	110.08	54.35
Jilin	6.90	104.06	46.36	9.60	121.91	47.02	10.99	141.21	56.86
Heilongjiang	4.34	29.33	8.91	6.73	45.43	13.77	6.93	41.96	13.55
Eastern Region	54.12	326.26	646.50	88.97	527.21	876.72	92.44	541.60	919.97
Shanghai	5.09	27.30	45.07	5.43	29.71	42.34	4.94	27.40	37.98
Jiangsu	11.91	51.16	156.35	17.54	82.00	184.38	17.00	76.69	175.44
Zhejiang	9.26	65.18	112.08	10.34	59.08	88.56	10.78	59.94	91.30
Anhui	12.09	36.32	140.72	14.55	40.41	158.95	15.15	43.20	168.89
Fujian	1.65	34.84	27.05	3.10	51.22	43.10	3.46	62.46	49.11
Jiangxi	3.58	32.82	72.73	6.17	42.97	88.75	6.71	52.67	108.15
Shandong	10.56	78.64	92.50	31.84	221.82	270.62	34.40	219.24	289.11
Central and Southern	35.03	347.95	398.54	61.89	472.77	526.11	71.27	549.51	613.82
Henan	14.66	43.33	108.33	26.54	79.70	189.66	33.63	95.02	241.45
Hubei	7.54	64.14	134.83	10.80	70.58	128.04	13.56	84.75	163.96
Hunan	2.80	82.78	36.48	9.07	187.42	102.11	9.22	213.20	104.89
Guangdong	4.70	73.01	79.90	8.13	71.89	65.68	7.18	72.01	57.17
Guangxi	5.20	83.73	37.85	7.13	62.02	39.49	7.45	83.08	44.77
Hainan	0.12	0.96	1.15	0.21	1.17	1.13	0.24	1.46	1.58
Southwestern Region	19.32	203.87	147.01	32.86	339.71	221.02	35.42	334.51	224.59
Chongqing	2.50	45.36	35.08	2.64	45.00	36.11	2.77	41.37	34.46
Sichuan	4.00	46.38	42.93	6.64	72.59	67.29	6.69	72.76	68.45
Guizhou	10.45	68.57	56.83	17.51	107.45	88.72	19.48	109.60	91.53
Yunnan	2.37	43.56	12.17	6.07	114.68	28.90	6.48	110.79	30.15
Xizang									
Northwestern Region	8.65	70.17	81.21	10.99	100.66	104.55	11.03	97.12	76.60
Shaanxi	2.50	14.68	23.69	4.57	31.88	49.13	5.85	34.54	50.17
Gansu	1.16	19.14	8.11	1.59	28.09	9.68	1.52	28.22	6.77
Qinghai	0.21	3.56	1.55	0.28	5.33	1.69	0.26	5.02	1.28
Ningxia	4.00	19.50	42.31	3.62	18.40	38.76	2.07	8.83	12.57
Xinjiang	0.78	13.28	5.54	0.93	16.97	5.29	1.32	20.50	5.81
China	177.05	1419.49	1609.45	284.09	2083.41	2222.69	305.95	2205.50	2352.97

of national territory and account for over 54% of national Hg, 50% of national As, and 65% of national Se emissions. The Southwestern and Northwestern Regions, covering nearly 55% of the national territory, emit far less Hg, As, and Se, with a combined share of less than 15%, 20%, and 13%, respectively. This is mainly due to their less installed capacities of coal-fired power plants and low consumption of coal by industrial sector. With the implementation of western development strategies and programs to transfer electricity from west to east in China, several large energy-based construction projects have begun since 2000 in Guizhou, Yunnan, Shaanxi provinces, and Ningxia autonomous region, which are all located in the southwestern and northwestern areas. The increased coal use in the western power plants will result in increased Hg, As, and Se emissions, and much attention should be paid to limit their potential negative effects on the atmospheric environment and human health.

3.3 Mapping $1^\circ \times 1^\circ$ gridded distribution of Hg, As, and Se emissions

Figure 7 shows the distribution of the total Hg, As, and Se emissions from coal combustion in China in 2005 at a resolution of $1^\circ \times 1^\circ$ from all sources combined. Power plants are treated as point sources and their emissions are precisely located at their latitude/longitude coordinates. Normally, most grids are composed of part or whole of several counties. The emissions from industrial sector are firstly divided into each county with the proportion of industrial GDP in one province, and then allocated to each grid according to the share of each county-area in one grid. Whereas, the emissions from residential and other use sectors are firstly divided into each county with the proportion of populations in one province, and then allocated to each grid with the share of each county-area in one grid.

As can be seen from Fig. 7, the emissions of hazardous trace elements from coal combustion in China distribute very unevenly due to the remarkable difference in economic and energy consumption structure, degree of development, density of population, as well as regional area of each province. Hg, As, and Se emissions from coal combustion are mainly concentrated around the populated and industrial centers of China – the coastal provinces in the eastern and the northern areas. Besides, several provinces such as Guizhou, Hunan, Yunnan, stand out in coal-related Hg, As, and Se emissions due to the high Hg, As, and Se content of raw coal mined in these provinces. Owing to the different distributions of Hg, As, and Se content of raw coal in different districts, the distribution of the total Hg, As, and Se emissions is much different from each other.

3.4 Comparison with other inventories

Until now, the comprehensive and detailed studies on As and Se emissions in China are quite limited. Therefore, only Hg emission estimates are compared with other studies.

As shown in Fig. 8, the trend of Hg emissions in our study agrees well with other studies while the values for the same year calculated are lower somewhat. This can be attributed to the difference in the averaged provincial content of Hg in raw coal. Feng and Hong (1996) estimated a relatively high emission of Hg on the basis that the national averaged Hg content in China was 0.30 mg/kg. The national averaged Hg content adopted by Wu et al. (2006) and Streets et al. (2005) was 0.19 mg/kg. Jiang et al. (2005) estimated the Hg emission based on two different coal Hg contents which were 0.15 mg/kg and 0.20 mg/kg. Furthermore, the national averaged Hg content was 0.22 mg/kg in Wang's study (2000). In our study, according to these previous studies and some other new published test results as mentioned before, we determine the national averaged Hg content in China to be 0.18 mg/kg, a little lower than those of previous studies.

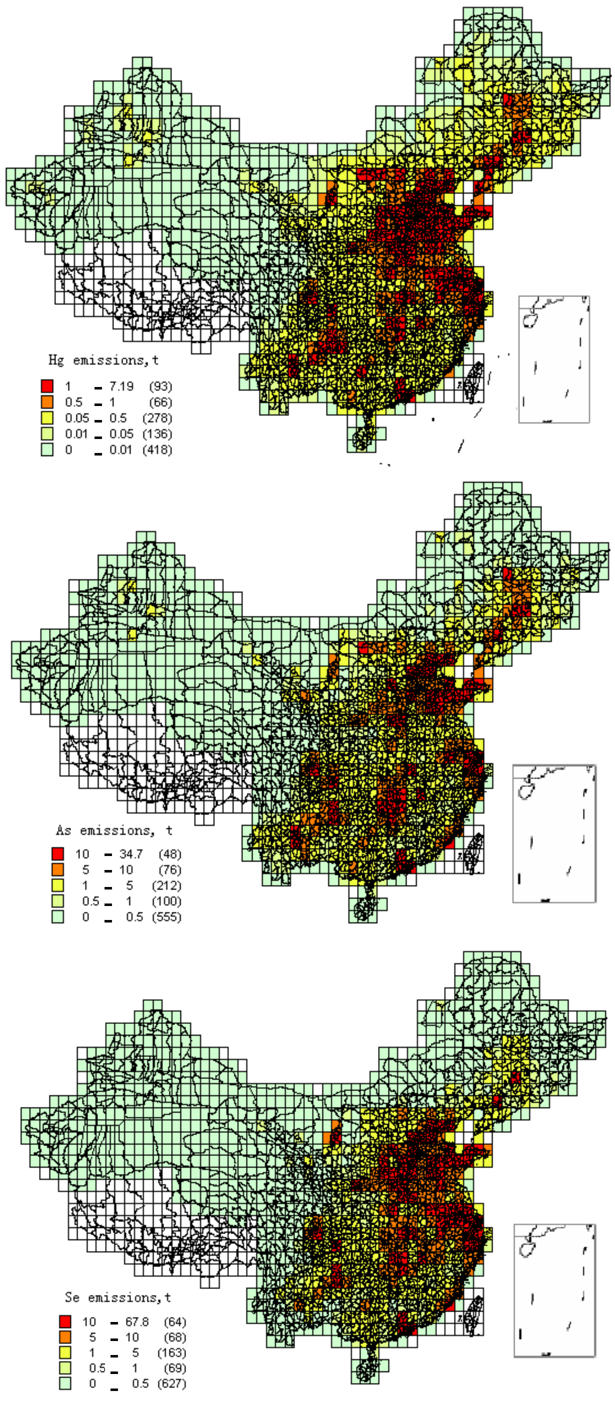

Fig. 7. Gridded total emissions of Hg, As, and Se from coal in China for the year 2005, $1^\circ \times 1^\circ$ (unit: $t\,yr^{-1}$ per grid cell).

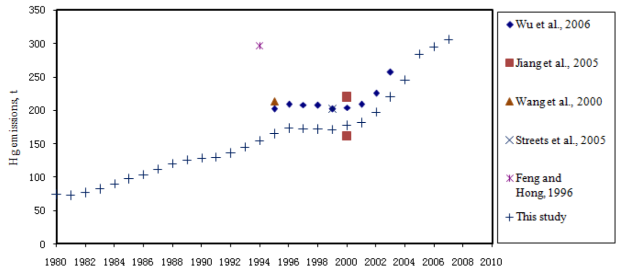

Fig. 8. Comparison of Hg emission estimates from coal in China.

4 Analysis of uncertainties

As mentioned above, uncertainties remain in the estimation of Hg, As, and Se emissions from coal combustion in China, which can be usually ascribed to three aspects: (1) the uncertainty in the averaged element content of raw coal, (2) the uncertainty in combustion release rates and emission factors, and (3) the uncertainty in degree of Hg, As, and Se removal by the existing air pollution control devices such as ESP/FFs and FGD. The Hg, As, and Se removal rates by various control devices such as wet FGD highly depend on the speciation of these elements in the flue gas (elemental vs. oxidized forms). Oxidized metals can be easily removed in WFGD compared to elemental forms. Coal properties such as Cl content affect the elements speciation in the flue gas, and thus the removal rate in WFGD. By now, actual measurements of Hg, As, and Se emission rates and species profiles from Chinese combustion facilities are still very limited, as well as the capture performance of Hg, As, and Se in Chinese emission control devices, especially for As and Se. Recent results indicate that the removal efficiencies of ESP+WFGD also vary with different coal types (Wang et al., 2010). There are even large discrepancies in estimates of the typical Hg, As, and Se content of coal in many provinces. However, the new results presented here improve understanding of the emissions of these hazardous trace elements from coal combustion and could be helpful for further control of Hg, As, Se emissions from coal in China. The remaining uncertainties of our inventories can be reduced in the future by additional field testing data for all kinds of coal and combustion facilities in China.

5 Conclusions

The calculated national total atmospheric emissions of Hg, As, and Se from coal combustion have rapidly increased from 73.59 t, 635.57 t, and 639.69 t in 1980 to 305.95 t, 2205.50 t, and 2352.97 t in 2007, at an annually averaged growth rate of 5.4%, 4.7%, and 4.9%, respectively.

The industrial sector is the largest source for Hg, As, and Se emissions, accounting for about 50.8%, 61.2%, and 56.2% of the national totals in 2007, respectively. Power plants sector ranks the second, with a share of 43.3% for mercury, 24.9% for arsenic, and 33.4% for selenium, respectively.

Emissions of Hg, As, and Se from coal combustion in China have begun to grow at a more moderate pace since 2005. Meanwhile, emissions from coal-fired power plants began to decrease which mainly thanks to the wide spread installation of WFGD in both new and in-use coal-fired power plants.

Our results show different regional distribution characteristics for these 3 trace elements, the top 5 provinces with the heaviest mercury emissions in 2007 are Shandong (34.40 t), Henan (33.63 t), Shanxi (21.14 t), Guizhou (19.48 t) and Hebei (19.35 t); the top 5 provinces with the heaviest arsenic emissions in 2007 are Shandong (219.24 t), Hunan (213.20 t), Jilin (141.21 t), Hebei (138.54 t) and Inner Mongolia (127.49 t); while the top 5 provinces with the heaviest selenium emissions in 2007 are Shandong (289.11 t), Henan (241.45 t), Jiangsu (175.44 t), Anhui (168.89 t) and Hubei (163.96 t).

In general, atmospheric emissions of Hg, As, and Se caused by coal combustion are mainly concentrated in the more populated and industrialized areas of China, i.e., the Eastern Central and Southeastern areas.

For a better reliable estimation of Hg, As, and Se emissions from coal combustion in China, long-term field testing and continuously monitoring for all kinds of coal combustion facilities in China are necessary.

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