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# Temporal and spatial variation characteristics of atmospheric emissions of Cd, Cr, and Pb from coal in China

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ABSTRACT

Multiple-year inventory of atmospheric emissions of cadmium (Cd), chromium (Cr), and lead (Pb) from coal burning in China have been established for the period 1980–2008 by using best available emission factors and annual activity data which are specified by different sub-categories of combustion facilities, coal types, and air pollution control devices. Our results show that the total emissions of Cd, Cr, and Pb have rapidly increased from 31.14 t, 1019.07 t, and 2671.73 t in 1980 to 261.52 t, 8593.35 t, and 12 561.77 t in 2008, respectively. The industrial sector ranks as the leading source, contributing ~88.3%, ~86.7%, and ~81.8% of the total Cd, Cr, and Pb emissions, respectively. Remarkably uneven spatial allocation features are observed. The emissions are primarily concentrated in the provinces of the northern and eastern region of China owing to the dramatic difference in coal use by the industrial and power sectors. Monthly temporal emission profiles for different sectors are established by using indexes such as monthly thermal electricity generation, monthly gross industrial output values and monthly average ambient temperature. For the power plants, there are two peaks during cold and hot season while for the industrial sector, emissions are most substantial in the summer and autumn season. Further, uncertainties in the bottom-up inventories are quantified by Monte Carlo simulation, and the overall uncertainties are demonstrated as −16% to 45% for Cd, −13% to 20% for Cr, and −21% to 48% for Pb, respectively. To better understand the emissions of these metals and to adopt effective measures to prevent poisoning, more specific data collection and analysis are necessary.



## 1. Introduction

In recent years, the negative effects of atmospheric emissions of Cd, Cr, and Pb on ecosystems and public health have received attention throughout the world as poisoning accidents increased (Zhang and Wong, 2007; Lenz and Lens, 2009; Li and Zhang, 2010). A rising concentration of hazardous trace elements in aerosols of China's urban areas has been reported (Xiao et al., 2008; Cong et al., 2010; Fan et al., 2011).

In order to restrain increasing emissions and pollution of typical heavy metals (including Hg, As, Cd, Cr, and Pb) from fuel combustion and industrial process, the State Council of China's government has officially ratified a specific Comprehensive Prevention Plan for Heavy Metals Pollution (CPPHMP) for the 12th five-year-plan (2011–2015) in the early of 2011, which was proposed by the Ministry of Environmental Protection of China (MEP). By now, atmospheric emissions of some volatile trace elements such as Hg, As, and Se from coal burning have been established at different scales (Streets et al., 2005; Wang et al., 2006; Wu et al., 2006; Tian et al., 2010). However, detailed investigation of Cd, Cr, and Pb discharge in China is quite limited, and little is known about the historical trend of anthropogenic atmospheric emissions of these metals and their spatial characteristics. The main objectives of this study are to establish multiple-year bottom-up inventories of anthropogenic emissions of Cd, Cr, and Pb from coal burning in China for the period 1980–2008, as well as to identify the temporal and spatial variation characteristics in terms of economic sectors, coal types, and provinces.

## 2. Methodologies, data sources and key assumptions

In this study, atmospheric emissions of Cd, Cr, and Pb from coal burning are calculated by combining the provincial-level coal consumption data and detailed emission factors, which are specified by four different economic sectors (power plants, industry, residential use, and other use sectors). The algorithm of

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a bottom-up emission inventory can be expressed by the following equation:

$$E_T(t) = \sum_i \sum_j C_{i,j}(t)F_{i,j}(t)EF_{i,j}(t)\left(1 - P_{\mathrm{PM}(i,j)}(t)\right)\left(1 - P_{\mathrm{FGD}(i,j)}(t)\right) \quad (1)$$

where $E$ is the atmospheric emissions of Cd, Cr, or Pb; $C$ is the average content of Cd, Cr, or Pb in coal consumed in one province; $F$ is the amount of coal consumption; $EF$ is the fraction of Cd, Cr, or Pb in flue gas released from coal combustion facility; $P_{\mathrm{PM}}$ and $P_{\mathrm{FGD}}$ are the fraction of Cd, Cr, or Pb removed by the existing dust collectors (cyclones, wet scrubbers, electrostatic precipitators, fabric filters, etc.) and flue gas desulfurization (FGD) devices, respectively; $T$ is the national totals; $i$ is the province (autonomous region or municipality); $t$ is the calendar year; and $j$ is the emission source classified by economic sectors, combustion facilities, and the particulate matter (PM) and $SO_2$ control devices.

**Table 1**
Contents of Cd, Cr, and Pb in raw coals consumed in 2008 (unit: $\mu g\ g^{-1}$).

| Province | Cd | Cr | Pb | Province | Cd | Cr | Pb |
|---|---|---|---|---|---|---|---|
| Anhui | 0.20 | 29.84 | 14.76 | Jiangsu | 0.46 | 25.92 | 21.67 |
| Beijing | 0.67 | 25.47 | 25.21 | Jiangxi | 0.59 | 34.44 | 20.32 |
| Chongqing | 1.29 | 28.89 | 30.21 | Jilin | 0.14 | 18.35 | 26.50 |
| Fujian | 0.48 | 29.78 | 24.02 | Liaoning | 0.27 | 23.39 | 22.34 |
| Gansu | 0.14 | 23.16 | 9.39 | Ningxia | 0.69 | 27.12 | 21.56 |
| Guangdong | 0.63 | 32.65 | 24.23 | Qinghai | 0.05 | 27.98 | 9.88 |
| Guangxi | 0.76 | 55.38 | 30.41 | Shaanxi | 0.72 | 32.23 | 32.82 |
| Guizhou | 0.79 | 28.49 | 23.83 | Shandong | 0.52 | 21.93 | 20.56 |
| Hainan | 0.64 | 28.05 | 23.66 | Shanghai | 0.57 | 25.98 | 22.64 |
| Hebei | 0.56 | 26.55 | 26.71 | Shanxi | 0.75 | 21.82 | 26.14 |
| Heilongjiang | 0.14 | 15.60 | 22.70 | Sichuan | 1.80 | 33.12 | 28.62 |
| Henan | 0.55 | 24.84 | 17.37 | Tianjin | 0.68 | 25.05 | 25.35 |
| Hubei | 0.63 | 28.03 | 27.20 | Xinjiang | 0.23 | 13.07 | 7.41 |
| Hunan | 0.68 | 33.73 | 25.43 | Yunnan | 0.82 | 70.79 | 41.41 |
| Inner Mongolia | 0.16 | 14.75 | 26.34 | Zhejiang | 0.50 | 26.80 | 20.96 |
| | | | | China | 0.57 | 28.44 | 23.32 |

Note: 95% CI = 95% confidence interval.

### 2.1. Average content of Cd, Cr, and Pb in coal

The content of hazardous trace elements in coal can provide useful information on the pollution control during coal combustion and utilization from an environmental point of view (Dai et al., 2008; Vejahati et al., 2010). Even when present at only parts per million levels in coal, hazardous elements can result in tons of pollutants discharged into the environment (von Storch et al., 2003). Previous studies have indicated the modes of occurrence, contents and distributions of Cd, Cr, and Pb varied substantially among provinces, sources, and even the coal of the same seam (Bai, 2003; Song et al., 2007; Li et al., 2006; Lu et al., 1995; Zhou et al., 2010).

The geographical distribution of coal resources in China is extremely unbalanced; coal is abundant in the northern and western areas while rare in the southern and eastern areas in general. As a result, large quantities of mined raw coal have to be transported long-distances from production areas to demand areas leading to remarkable variation between the characteristics and content of trace elements in coal produced and consumed in a single province, which must be considered when developing an emission inventory.

In this paper, field test content data of Cd (616 samples), Cr (956 samples), and Pb (831 samples) for different Chinese raw coal are summarized from available published literature (Bai, 2003; Dai et al., 2006; Zhou et al., 2010). First, the average content of each of these elements in coal as produced on a provincial-level are determined by using a bootstrap simulation (Tian et al., 2011). Then, the weighted average content of Cd, Cr, and Pb in coal products as consumed are calculated by combining the annual coal flow matrix among 30 provinces in the Chinese mainland as described in our previous studies (Tian et al., 2010, 2011). The procedures and source samples for determining the average contents of Cd, Cr, and Pb in Chinese coal by provinces can be referred to in Tables S1–S4 in the supplementary materials for details. Table 1 illustrates the weighted average content of Cd, Cr, and Pb in coal as consumed by province. As can be seen, obvious variation of the element content in coal from different provinces will exert significant influence on the spatial distribution characteristics of trace element emissions from coal in China.

### 2.2. Coal consumption

In this study, coal combustion sources are divided into four large sectors (power plants, industrial sector, residential sector, and other sector) according to the operational patterns of combustion furnaces as well as their functions. Detailed coal consumption by sector and type of coal products are compiled from provincial-level data in the China Energy Statistical Yearbooks from 1980 to 2008 (DITS, 1992, 1998; NBS, 2001, 2004; NBS and NDRC, 2005–2009). Xizang Autonomous Region, Taiwan province, Hong Kong and Macau Special Administrative Region are not considered. Fig. 1 shows the historical trend in coal consumption by different sectors for the period of 1980–2008. A relatively high growth rate of coal consumption by power plants and industrial sector is found since the beginning of the 21st century. By the end of 2008, national total coal consumption had reached at 3459 million tons. Although the industrial sector still ranked as the leading consumer for coal use, its proportion has declined from 53.0% in 1980 to 48.1% in 2008. In contrast, the share of power plants has grown from 20.0% to approximately 45.8% of the total. Notably, the use of coal for residential sectors has decreased, which can be mainly ascribed to cleaner energy substitution such as LPG (Liquefied Petroleum Gas) and natural gas, especially in urban areas for abatement of PM and $SO_2$ discharge to improve the urban air quality.

### 2.3. Emission factors of Cd, Cr, and Pb

#### 2.3.1. Release rates from different coal-fired facilities

As mentioned above, the distribution and modes of occurrence of Cd, Cr, and Pb in different coals varies significantly; the occurrence of Cd, Cr, and Pb in the environment are Cd (II), Cr (VI), and Pb (II), respectively (Song et al., 2007). With rising furnace temperature during coal burning, these trace elements will become part of the flue gas, and react with surrounding gases. The release rates of Cd, Cr, and Pb depend on coal combustion technologies and operating conditions (Zhang et al., 2010), thus, it is necessary to give

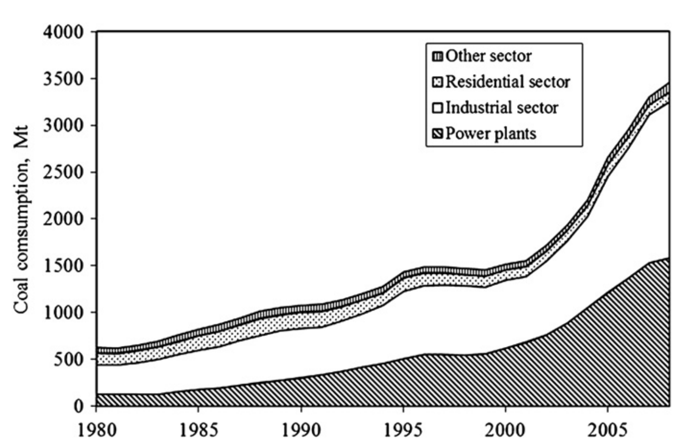


**Fig. 1.** Trend of coal consumption by sector in China, 1980–2008.

a detailed specification of the methods by which the coals are fed and burned. Here, coal combustion facilities are divided into four types: pulverized-coal boilers, fluidized-bed furnaces, stoker-fired boilers and coke furnaces. Presently, pulverized-coal boilers are predominant in coal-fired power plants in most of the provinces in China, representing over 90% of the total. The remaining share is divided between fluidized-bed furnaces and stoker-fired boilers which are mainly used in small coal-fired power plants. Unlike power plants, the stoker-fired boiler is the dominant boiler type used in the industrial sector (Tian et al., 2010). We adopt the arithmetic mean values (see Table 2) of release rates reported in references to calculate final emissions. Field test release rates of Cd, Cr, and Pb from different types of coal-fired facilities are summarized in Table S5 in the supplementary materials.

*2.3.2. Removal efficiencies of PM and $SO_2$ control devices*

After being emitted from a facility's coal-fired furnace, Cd, Cr, Pb and their compounds accompanied with fine fly ash in the flue gas can be partly removed to ash and desulfurization slurry by downstream PM and $SO_2$ control devices, such as electrostatic precipitators (ESPs), fabric filters (FFs), cyclones, wet scrubbers, as well as wet flue gas desulfurization (FGD) systems (Yi et al., 2008; Tian et al., 2010). The remaining flue gas will be emitted into the atmosphere through the stack. Field tests have demonstrated that the removal efficiency by different types of PM and $SO_2$ control devices varied substantially (Song et al., 2007; Dai et al., 2008; Zhang et al., 2010). In this study, we assume the arithmetic mean values of those reported in available references as the average removal efficiencies by different PM and $SO_2$ control devices, as shown in Table 2 and Table S6 in the supplementary materials.

*2.3.3. Emission factors of Cd, Cr and Pb for residential and other sectors*

Residential and other sectors are also important coal consumers in China. Because the coal-fired facilities applied in residential and other sectors are mainly poorly-controlled and uncontrolled small-scale stoves, we applied the emission factors of commonly-used stoves in the reported literature and the coal consumption to estimate atmospheric emissions of Cd, Cr, and Pb from residential and other sectors. The major combustion patterns for residential cooking and heating are traditional cook stoves and improved cook stoves, both of which are mostly fed with raw coal or briquettes and which are not equipped with any specific PM control devices. However, there is very little information about Cd, Cr, and Pb emissions from these sources. In this study, the emission factors of Cd, Cr, and Pb from residential use are assumed as: $2.77 \times 10^{-5}$ g kg$^{-1}$, $1.92 \times 10^{-5}$ g kg$^{-1}$, and $8.02 \times 10^{-3}$ g kg$^{-1}$ for improved cook stoves, respectively; and $1.07 \times 10^{-4}$ g kg$^{-1}$, $6.21 \times 10^{-4}$ g kg$^{-1}$, and $1.98 \times 10^{-2}$ g kg$^{-1}$for traditional cook stoves, respectively (Zhao et al., 1994; NPI, 1999). In addition, the emission factors for boilers used in other sectors are separately assumed at $3.76 \times 10^{-5}$ g kg$^{-1}$, $1.92 \times 10^{-5}$ g kg$^{-1}$, and $8.02 \times 10^{-3}$ g kg$^{-1}$ (US EPA, 2001), respectively.

**Table 2**
The release rate and removal efficiency of Cd, Cr, and Pb from coal-fired facilities and pollution control devices.

| Category | | Cd | Cr | Pb |
|---|---|---|---|---|
| Release rate (%) | Pulverized-coal boiler | 94.93 | 84.50 | 96.25 |
| | Stoker fired boiler | 42.53 | 26.74 | 40.10 |
| | Fluidized-bed furnace | 91.50 | 81.33 | 77.33 |
| | Coke furnace | 20.00 | 24.00 | 31.50 |
| Removal efficiency (%) | ESPs | 96.46 | 98.53 | 97.16 |
| | FFs | 97.63 | 95.13 | 99.00 |
| | Wet scrubber | 75.00 | 48.14 | 70.10 |
| | Cyclone | 22.91 | 30.04 | 12.10 |
| | Wet-FGD | 80.50 | 86.00 | 78.42 |
| | Coal washing | 32.21 | 57.99 | 36.30 |

## 3. Results and discussion

*3.1. Temporal trend of Cd, Cr, and Pb emissions by sector*

The historical trend and composition of Cd, Cr, and Pb emissions by sector are illustrated in Fig. 2. As can be seen, total emissions of Cd, Cr, and Pb have been increasing from 31.14t, 1019.07 t, and 2671.73 t in 1980 to 261.52 t, 8593.35 t, and 12 561.77 t in 2008, at an annual average growth rate of 8.0%, 7.9%, and 5.7%, respectively.

Industrial sector has been the leading source of Cd, Cr, and Pb emissions, increasing from 19.89 t, 788.58 t, and 965.47 t in 1980, to 230.88 t, 7454.26 t, and 10 271.45 t in 2008, accounting for ∼88.3%, ∼86.7%, and ∼81.8% of the respective totals. The contributions of power plants are ∼19.5% for Cd, ∼31.2% for Cr, and ∼16.4% for Pb, respectively. Among the four sectors, the emissions of Cd, Cr, and Pb from the industrial sector have increased fastest, at an annual growth rate of 9.2%, 8.4%, and 8.8%, respectively. The growth rate of emissions from power plants ranks the second, growing 7.8%, 6.5%, and 8.2%, respectively. However, emissions from industrial sector are still experiencing a rapid increase, mainly due to the growing coal use and less application of advanced PM and $SO_2$ control devices. Since the beginning of the 21st century, with the rapid expansion of the energy-intensive manufacturing industry, such as steel and cement production, coal consumption of industrial sector has maintained a relatively high growth rate. By the end of 2008, coal consumption of the industrial sector reached 1664.69 million tons, with an annual growth rate of ∼26.6% (since 2001). If the emission rates of the industrial sector are not better controlled, it will be difficult to reduce these three toxic metal emissions effectively, which will frustrate the implementation of the specific Comprehensive Prevention Planning for Heavy Metals Pollution for the 12th five-year-plan as mentioned above. The industrial sector is far from its maximum reduction potential.

Notably, negative growth was observed in emissions from the power sectors in 1997 and 1998, which can be mainly attributed to the decline in coal consumption during the period of the Asian financial crisis as referred to in the previous study (Hao et al., 2002). Emissions from the industrial sector have experienced two fluctuations, one between 1990 and 1991 and the other around 2000. The main reason is also due to decreased coal consumption by restructuring of the energy-intensive manufacturing industry (such as iron and steel, cement) during those periods.

Another highlighted feature is that the emissions of Cd, Cr, and Pb from the power plants have declined substantially since 2005 due to the co-benefit reduction effects by the existing and newly installed PM and $SO_2$ control devices in coal-fired power plants. By the end of 2008, the installed capacities of FGD in power plants reached about 379 GWe, accounting for nearly 66% of the total installed capacity of thermal power plants (SEPA, 2008). Due to widespread installation of advanced emission control devices such as ESPs, FFs, and FGD systems, final discharge rates of Cd, Cr, and Pb into the atmosphere per ton of coal used by power plants have been reduced significantly. The average emission factors of these three toxic elements has been reduced from 0.03 g t$^{-1}$ for Cd, 0.96 g t$^{-1}$ for Cr, and 1.00 g t$^{-1}$ for Pb in 2005 to 0.01 g t$^{-1}$, 0.69 g t$^{-1}$, and 0.84 g t$^{-1}$ in 2008, respectively. As mentioned in Table 2, the average removal efficiencies of Cd, Cr, and Pb by wet-FGD system reach about 80.5%, 86.0%, and 78.4%, respectively; efficiency is even more enhanced when wet-FGD is combined with other emission control devices. Accordingly, although the coal consumption by power plants has caught up with that of the industrial sector in recent years, the final discharge of hazardous elements into the environment has gradually decreased.

Because of the substitution by cleaner fuels like natural gas and LPG, especially in urban areas, the use of coal for residential sector

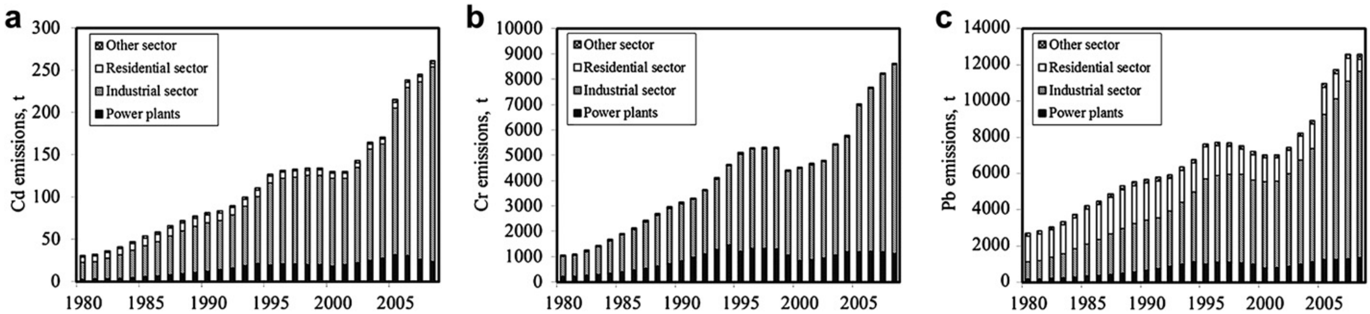


**Fig. 2.** Trend of Cd, Cr, and Pb emissions by sector, 1980–2008.

has been decreasing during the past decades, resulting in direct emissions of Cd, Cr, and Pb falling continuously. In 2008, the emissions of Cd, Cr, and Pb from residential coal use are estimated at 4.00 t, 10.69 t, and 663.86 t, respectively, even lower than those of 1980. For other sectors, coal consumption increased gradually during the recent past, which caused the emissions of these sectors to steadily grow. However, the harmful health effects of these metals should be considered in light of their indoor discharge and the long-time exposure of inhabitants.

According to the global inventory of trace elements in 1995, Cd, Cr, and Pb emissions in Asia by combustion of fuels in stationary sources are estimated at 237 t, 4282 t, and 4845 t in 1995, respectively (Pacyna and Pacyna, 2001). In comparison, our results of Cr and Pb emissions for the year 1995 are markedly higher. This is mainly owing to the large difference of the adopted average contents of Cr and Pb in coals. Our results show that the average content of Cd in Chinese coals is similar to the world average and American coals; while the contents of Cr and Pb are much higher than those of the world average and American coals. In addition, the emission factors used by Pacyna and Pacyna (2001) are suggested by the UN Economic Commission for European emission inventory, which do not take into account the relatively lower penetration of advanced PM and $SO_2$ control devices. Thus, our new emission inventory of Cd, Cr, and Pb for China can be useful to amend and supplement the global emission inventories.

### 3.2. Spatial variation characteristics of Cd, Cr, and Pb emissions

Since the introduction of 11th five-year-plan after 2005, atmospheric emissions of Cd, Cr, and Pb from coal in China have begun to grow at a more moderate pace in spite of the continuous rapid coal consumption increase. Emissions of Cd, Cr, and Pb by province in China for the year of 2008 are summarized in Table 3, and corresponding spatial emissions intensity can be seen in Fig. 3.

On the whole, emissions of Cd, Cr and Pb are highly concentrated in provinces of northern and eastern regions, such as Hebei, Shanxi, and Shandong. Heavy emissions from these areas are driven by dramatic coal consumption by industrial and power plants sectors. With the rapid application of advanced air pollution control technologies, especially the mandated installation of FGD for $SO_2$ abatement, emissions of Cd, Cr, and Pb in these provinces have declined even as power plant installation has increased owing to the co-benefit removal of trace elements by PM and FGD. Hence, comprehensive PM and $SO_2$ control policies issued by the central government and MEP have already obtained remarkable co-benefit achievements for abating atmospheric emissions of hazardous trace elements into the atmosphere. However, the shift and accumulation of hazardous trace elements from the atmosphere to the water and soil environment should be highlighted, since they may transformed to more toxic species and endanger the ecosystem and human health (Zhen et al., 2008; Lenz and Lens, 2009; You and Xu, 2010).

Provinces like Sichuan, Chongqing, Hubei, and Yunnan have relatively higher emissions mainly due to high contents of these three elements in raw coal produced and/or consumed in such areas. Beijing, Tianjin, and Shanghai share common features such as rapid economic development but have lower emissions due to lower coal consumption. These areas have little coal reserves so most of the energy demand is supplied by surrounding provinces; the share of clean energy (in terms of PM and $SO_2$ direct emissions) such as natural gas and electricity has been increased in order to reduce the emissions of PM and $SO_2$ to improve local urban air quality. However, this will increase the environmental pressure on energy-output regions. Provinces like Heilongjiang, Jilin, Xinjiang, and Gansu in the northeast and northwestern region emit far lower trace

**Table 3**
Provincial emissions of Cd, Cr, and Pb from coal burning in China (unit: t $a^{-1}$).

| Province | Cd | Cr | Pb | Province | Cd | Cr | Pb |
|---|---|---|---|---|---|---|---|
| Anhui | 3.66 | 308.29 | 273.50 | Jiangsu | 11.22 | 428.49 | 557.94 |
| Beijing | 2.17 | 58.11 | 112.94 | Jiangxi | 4.94 | 190.79 | 203.61 |
| Chongqing | 10.83 | 173.79 | 297.87 | Jilin | 1.96 | 201.70 | 381.54 |
| Fujian | 3.84 | 170.94 | 216.47 | Liaoning | 6.87 | 401.79 | 646.99 |
| Gansu | 1.06 | 86.57 | 112.48 | Ningxia | 2.86 | 84.33 | 104.11 |
| Guangdong | 8.11 | 306.31 | 340.57 | Qinghai | 0.17 | 31.80 | 31.21 |
| Guangxi | 5.87 | 287.11 | 256.93 | Shaanxi | 8.70 | 320.26 | 489.65 |
| Guizhou | 7.54 | 178.14 | 341.13 | Shandong | 24.14 | 685.25 | 901.99 |
| Hainan | 0.35 | 10.92 | 13.89 | Shanghai | 3.48 | 117.10 | 168.95 |
| Hebei | 24.62 | 752.29 | 1222.21 | Shanxi | 28.94 | 594.07 | 1156.77 |
| Heilongjiang | 1.99 | 175.31 | 356.12 | Sichuan | 31.46 | 385.24 | 634.94 |
| Henan | 17.22 | 579.09 | 724.93 | Tianjin | 3.29 | 79.89 | 136.22 |
| Hubei | 11.57 | 355.68 | 601.03 | Xinjiang | 2.21 | 87.97 | 106.14 |
| Hunan | 11.38 | 388.29 | 512.63 | Yunnan | 10.64 | 653.08 | 642.34 |
| Inner Mongolia | 4.07 | 241.44 | 732.49 | Zhejiang | 6.36 | 259.31 | 284.18 |
| | | | | China Total | 261.52 | 8593.35 | 12 561.77 |

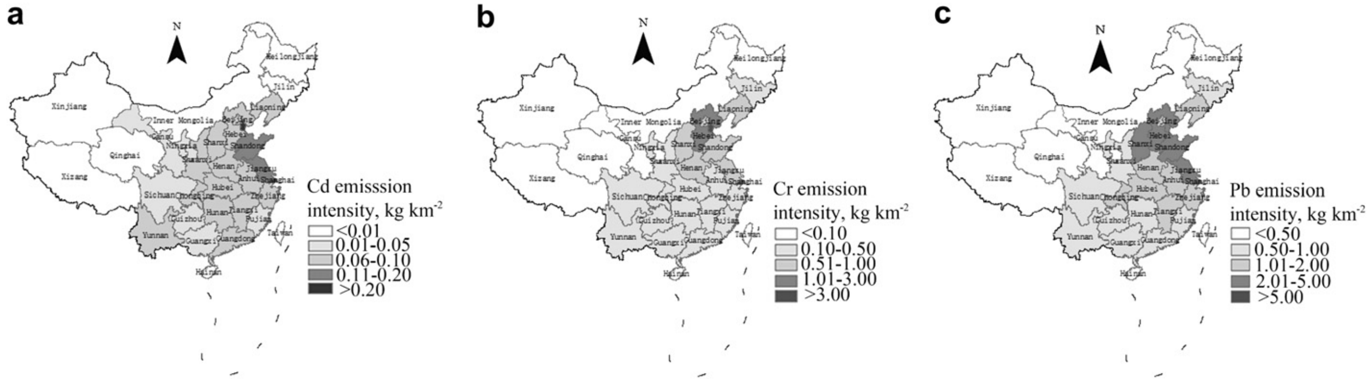


**Fig. 3.** Distribution of Cd, Cr, and Pb emission intensity by province in 2008.

elements because of less coal-fired power plant capacity, lower coal consumption by industrial sector, and fewer inhabitants. However, with the implementation of the Western Development Strategy in China, Transferring Electricity from West to East Program, and Rejuvenation Project of the Traditional Industrial Base in Northeastern China, many large coal-based construction projects are being planned or built in the northwestern and northeastern areas. The increasing coal output and consumption will result in the growth of Cd, Cr, and Pb emissions. More high-polluted manufacturing enterprises are moving to the western provinces under the motivating policies of the western local governments as well as the stress of increasing labor cost in developed eastern coastal areas. Although the policies can promote rapid economic growth in the west, they also have brought frequent Cd, Cr and Pb poisoning accidents to these places. Consequently, attention should be focused on the emissions of western regions due to the growing real and potential harmful emission impacts on the local environment and human health.

### 3.3. Monthly variation characteristics of Cd, Cr, and Pb emissions

In order to know about the temporal variation patterns of Cd, Cr, and Pb emissions throughout the year, and to explore possible effective ways to reduce emissions and prevent related poisoning accidents, monthly variation characteristics are investigated for different sectors by region. Normally, emissions of Cd, Cr, and Pb are closely correlated with the activity levels of a specific source category (Reis et al., 2009). Thus, we use monthly profiles of activity level as surrogates to approximately reflect the temporal variation of emissions for different source categories for the year 2008 (Fig. 4).

Monthly profiles of power plants for different regions are derived from the variation of coal-fired power electricity generation reported by the National Bureau of Statistics of China (see Fig. 4a). On the whole, power generation is closely related to the variation of ambient temperature in most regions with two peaks during cold (winter) and hot (summer) seasons; for provinces of the southwest China, peak values only arises in the cold season, power generation drops gradually until November which can be mainly explained by high proportion of hydropower generation in summer season.

The profiles of industrial sector are derived from monthly gross industrial output values reported by the National Bureau of Statistics (NBS) of China (see Fig. 4b). An apparent feature is that industrial sector has accomplished the lowest gross industrial output value in February, when the traditional Chinese New Year (Spring Festival) holiday normally occurs, and most manufacturing industries will reduce their production owing to declining demand of industrial products. The common characteristics for all regions is that the profile of industrial sector increase gradually from February to the end of the year, through a relative higher and steady period from May to September marked by small fluctuations. Hence, according to the seasonal variation of industrial activities, one may imagine that the emissions of Cd, Cr, and Pb from industrial sector are highly centralized in the summer and autumn season.

Compared with power plants and industrial sectors, the contribution of residential and other sectors to the total emissions of Cd, Cr, and Pb is comparatively small. However, it is interesting to consider their temporal variations. The coal consumption of residential and other sectors are mainly for cooking and supplying hot water and heat which will experience an obvious growth in cold seasons; therefore, the emission profiles from these two sectors mainly rely on ambient temperature change. Monthly average

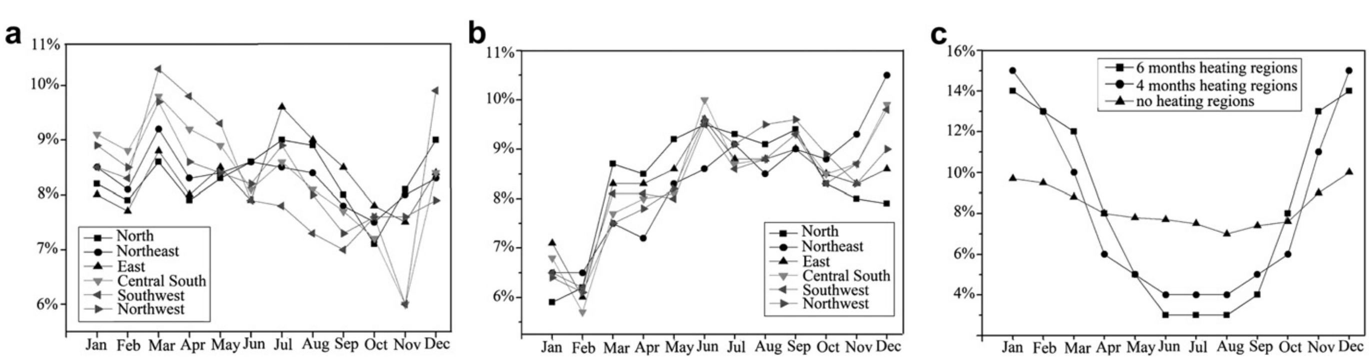


**Fig. 4.** Monthly temporal variation profiles of different emission sources.

**Table 4**
Uncertainties in the emissions of Cd, Cr, and Pb by sector in China in 2008.

| Sectors | Cd | Cr | Pb |
|---|---|---|---|
| Power plants | 23.42 (−7%, 32%) | 1098.31 (−9%, 10%) | 1327.90 (−6%, 15%) |
| Industrial sector | 230.88 (−21%, 254%) | 7454.26 (−15%, 54%) | 10 271.45 (−33%, 71%) |
| Residential sector | 4.00 (−72%, 70%) | 10.69 (−187%, 136%) | 663.86 (−98%, 111%) |
| Other sector | 3.22 (−108%, 92%) | 30.10 (−81%, 76%) | 298.56 (−75%, 73%) |
| Total | 261.52 (−16%, 45%) | 8593.35 (−13%, 20%) | 12 561.77 (−21%, 48%) |

ambient temperature change is considered to be the main factor influencing heat and hot water supply which is in reverse proportion to monthly coal consumption in different provinces. The temporal profiles for residential and other sectors are presented in Fig. 4c. The heating period in northern provinces is about 6 months (from October 15th to April 15th in the next year), like Xinjiang, Qinghai, Gansu, Inner Mongolia, Heilongjiang, Jilin, and Liaoning, and the peak values appear in January and December. For other northern provinces, of which the cold season is a little shorter, the heating period is usually 4 months (from November 15th to March 15th of the next year), such as Beijing, Hebei, Tianjin, Shanxi, Shaanxi, Shandong and Henan. However, for most provinces like Guangdong, Guizhou, Hunan and Sichuan in southern China, there are rarely any heating needs, or they only appear in some urban areas, so the profile of emissions of these provinces shows a relatively smooth monthly temporal variation.

### *3.4. Uncertainty analysis*

To better understand the uncertainties in our inventory, Monte Carlo simulation is used to quantify the uncertainty of Cd, Cr, and Pb emissions depending on available activity data and emission factors distribution (Zheng et al., 2009; Zhao et al., 2011). Emissions of Cd, Cr, and Pb from coal burning with uncertainty are shown in Table 4. The overall uncertainties in our inventories are estimated at −16% to 45% for Cd, −13% to 20% for Cr, and −21% to 48% for Pb emissions, respectively.

Emissions of coal-fired power plants are considered to have the least uncertainty (−7% to 32% for Cd, −9% to 10% to Cr, and −6% to 15% for Pb) among all of the four sectors. They have demonstrated relatively lower variations for the distribution of available activity data, release rates from different boiler types, as well as removal efficiencies of different control devices.

The industrial sector (−21% to 254%) is identified as the main contributor of uncertainties in Cd emissions. It can be mainly attributed to limited information on specific industrial manufacturing processes and facility parameters, and relatively poor resolution of coal burning technologies and emission control devices.

The residential sector is demonstrated to be the largest source of uncertainties in Cr and Pb emissions (−187% to 136% for Cr, and −98% to 111% for Pb). There have been few studies conducted on the real emission features from domestic residential sectors to date. In this study, emission factors of residential sector are mainly compiled by the average values of measurements from foreign and domestic studies (Zhao et al., 1994; NPI, 1999). It is inevitable that the determination of emission factors of some coal-fired furnace types will contain uncertainties.

Thus, more detailed investigation and field tests for all kinds of coal-fired facilities are very necessary for a better understanding of the emissions of these three hazardous trace elements.

## 4. Conclusions

By applying provincial-level coal use and detailed emission factors, which is categorized with different coals, boilers types, and equipped PM and $SO_2$ control devices configuration, bottom-up emission inventories of Cd, Cr, and Pb from coal in China have been developed for the period of 1980–2008. The national total atmospheric emissions of Cd, Cr, and Pb are estimated at about 261.52 t, 8593.35 t, and 12 561.77 t in 2008, at annual growth rates of 8.0%, 7.9%, and 5.7% since 1980, respectively. The industrial sector is considered the leading source of emissions, accounting for ∼88.3%, ∼86.7%, and ∼81.8% of the national total emissions, respectively.

As for the regional distribution, dramatic coal consumption by the industrial sector and power plants in the provinces of northern and eastern regions of China has brought the largest emissions of Cd, Cr, and Pb into the atmosphere. The heavy emissions of Sichuan, Chongqing, Hubei, and Yunnan province are caused by the high contents of these elements in coals. While the current emissions for the provinces of northeast and northwest regions are still not very large, they should be considered due to the potential development of high-polluting manufacturing enterprises.

To achieve a more reliable estimation of atmospheric emissions of Cd, Cr, and Pb from coal combustion in China, potential improvements include the better understanding and determination of elemental content in coals (bituminous coal, anthracite, and lignite), emission factors and more specific sub-categories of industrial sector.

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## Appendix. Supplementary data

Supplementary data associated with this article can be found in the online version, at doi:10.1016/j.atmosenv.2011.12.045.

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