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Quantitative assessment of atmospheric emissions of toxic heavy metals from anthropogenic sources in China: historical trend, spatial distribution, uncertainties, and control policies

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Section

Section S1. Mathematical description of bootstrap simulation

Bootstrap is a numerical technique originally developed for the purpose of estimating confidence intervals for statistics. This method can provide solutions of confidence intervals in situations where exact analytical solutions may be unavailable and in which approximate analytical solutions are inadequate. Confidence intervals for a statistic are inferred from its sampling distribution. For example, the 2.5th and 97.5th percentiles of sampling distribution enclose a 95% confidence interval. The brief mathematical description of bootstrap simulation is as follows:

A random sample $X=(x_1, x_2, ..., x_n)$ of size n is observed from a completely unspecified probability distribution F. The sampling distribution $R\ (X, F)$ is the function of X and F. Assume $\theta=\theta(F)$ is a parameter of F, F_n is the empirical distribution function of X, $\hat{\theta} = \hat{\theta}(F_n)$ is the estimator of θ, and the estimation error can be expressed as:

$$\mathrm{R}(X, F) = \hat{\theta}(F_n) - \theta(F) \triangleq T_n \qquad (1)$$

The basic steps of computing the distribution R (X, F) by bootstrap simulation are summarized as follows:

(1) The value of observed samples $X=(x_1, x_2, ..., x_n)$ are finite overall samples (called original samples), $x_i \sim F(x)$, $i=1, 2, ..., n$. The empirical distribution function of original samples is shown as:

$$F_n = \begin{cases} 0 & x < x_{(1)} \\ k/n & x_{(k)} \le x < x_{(k+1)} \\ 1 & x \ge x_{(n)} \end{cases} \qquad (2)$$

where, $x_{(1)} \le x_{(2)} \le ... \le x_{(n)}$ is the statistics of $x_1, x_2, \ldots, x_n$ sorted in ascending order.

(2) Monte Carlo simulation is used to randomly simulate N groups of samples $X^*_{(j)} = (x^*_1, x^*_2, ..., x^*_n)$, $j=1, 2, ..., N$ (a very large number) from F_n, and these regeneration samples called bootstrap samples. The generation method of empirical distribution function by Monte Carlo simulation can be expressed as: (a) generate a random integer η with independence and uniformity between 0 and $M\ (M >> n)$ by computer; (b) let $i=\eta$ % n, and i is the remainder of n divide η; (c) find the sample x_i as the regeneration sample x^* in observed samples, and x^* is the needed random sample.

(3) Calculate the statistics of bootstrap samples:

$$R^*(X^*, F_n) = \hat{\theta}(F_n^*) - \hat{\theta}(F_n) \rightarrow R_n \qquad (3)$$

where, F_n^* is the empirical distribution function of bootstrap samples. As small samples can't derive $\theta(F)$, $\hat{\theta}(F_n)$ is used to approximate it.

(4) Use the distribution of R_n (under given situation) to simulate the distribution of T_n, say:

$\theta(F) \approx \hat{\theta} - R_n$, which can receive *N* numbers of *θ(F)*. Then, the distribution and eigenvalue of unknown parameter *θ* can be obtained.

Section S2. Removal efficiencies of 12 HMs through coal cleaning and coke process

Some studies have reported that coal cleaning is an effective and feasible way to reduce atmospheric emissions of heavy metals before coal burning (Luttrell et al., 2000; Wang et al., 2006). By the year of 2012, only about 20.9% of total raw coal production is washed before burning, and is primarily used for coke making in iron and steel industry (NBS, 2013a). In view of the operation characteristics and the application situation of coal cleaning processes in China, we assume the average removal efficiency of Hg, As, Se, Pb, Cd, Cr, Ni, Sb, Mn, Co, Cu and Zn to be 50.0%, 54.0%, 30.0%, 36.3%, 32.2%, 58.0%, 58.5%, 35.7%, 68.2%, 39.3%, 31.8% and 48.6% (Quick and Irons, 2002; Bai, 2003; Wang et al., 2003a; Yao et al., 2012), respectively.

Due to excessive temperature range (400 °-1000 °) in coke process (Zajusz-Zubek and Konieczyński, 2003), high emission will be found, especially for volatile substance. According to analyze the data of heavy metals discharge in coke process as described in other studies (Helble et al., 1996; Guo et al., 2002; Guo et al, 2003; Yi et al., 2007; Konieczyński et al., 2012), we presume that 10.0% of Hg, 70.0% of As, 60.0% of Se, 68.5% of Pb, 80.0% of Cd, 76.0% of Cr, 90.3% of Ni, 30.0% of Sb, 92.4% of Mn, 92.9% of Co, 80.0% of Cu and 73.9% of Zn remains after the coking process.

Section S3. Temporal variation trends of HM emissions from other primary anthropogenic sources

1 HM emissions from liquid fuels combustion

Although liquid fuels only take up about 8.9% of the total primary energy production and account for nearly 18.8% of total energy consumption in 2012, the liquid fuels consumption is also one of major contributors for atmospheric Ni emissions due to the relatively high content of Ni in fuel oil (Tian et al., 2012b). Furthermore, with the rapid growth of vehicle/plane populations and transport turnover (including passenger and cargo turnover), the consumptions of gasoline, diesel oil and kerosene in China have reached 116.0, 184.1 and 22.0 Mt in 2012, respectively.

Because of the large use of leaded gasoline in China before 2001, none can afford to neglect the accumulated emissions of Pb from gasoline consumption by vehicles during 1949 to 2012, although the leaded gasoline production has been forbidden since 2001.

In this study, we estimate that the discharge of Ni from liquid fuels combustion have increased from 12.8 t in 1949 to 604.5 t in 2012. Therein, fuel oil combustion contributes over 82.1% of the total liquid fuels consumption category in 2012. Notably, the total Ni emission from liquid fuels consumption category has increased slightly (less than 2% annually) since 1980 despite of the rapid growth of distillate oils (gasoline, diesel oil, and kerosene), which is mainly because of the lower Ni content in distillate oils and relatively constant supply of fuel oil in China in the past three decades (NBS, 2013b; Wang et al., 2003b; Tian et al., 2012b).

In terms of Pb emission from gasoline combustion category, the reduced lead content of gasoline is the primary reason for the sharp decrease in total Pb emissions in 1991 and 2001 (Li et al., 2012), as with national total Pb emission. For the first sharp emission decline, the total emissions has decreased by 36.8% from 12 832.2 t in 1990 to 8107.5 t in 1991. For the other sharp decline, the total emissions have decreased by 98.1% from 12 866.7 t in 2000 to 248.3 t in 2001. However, the Pb emissions from this category have continued to increase in the following years due to the gradually increase of gasoline consumption with the rapid growth of urban vehicle populations (please see Fig. S5).

2 HM emissions from brake and tyre wear

During the period of 1949 to 2012, the amount of civilian vehicles has increased from 0.1 million units to 109.3 million units. Furthermore, the passenger turnover of highways and freight turnover of highways have increased continuously to 1846.8 billion passenger-kilometer and 5953.5 billion ton-kilometer, respectively (NBS, 2013b). As a result, the total Pb, Cr, Sb, Mn, Cu and Zn emissions from brake and tyre wear have increased remarkably to 333.5, 124.0, 530.1, 133.8, 2720.1 and 954.7 t in 2012, respectively. Especially during 2000 to 2012, the annual growth rate of these HM emissions from brake and tyre wear is up to about 17.5%, which is closely related to the rapid growth of civilian vehicle population (see Fig. S5). For other HMs (As, Se, Cd, Ni and Co), the extraordinarily low emissions from brake and tyre wear category are estimated due to trace level of these elements in brake linings.

Table

Table S1 Summary of heavy metal species and the associated emission sources categories

Sector		Category	Hg	As	Se	Pb	Cd	Cr	Ni	Sb	Mn	Co	Cu	Zn
Coal combustion	Power plant	Raw coal	●	●	●	●	●	●	●	●	●	●	●	●
		Cleaned coal	●	●	●	●	●	●	●	●	●	●	●	●
		Briquette	●	●	●	●	●	●	●	●	●	●	●	●
		Coke	●	●	●	●	●	●	●	●	●	●	●	●
	Industrial sector	Raw coal	●	●	●	●	●	●	●	●	●	●	●	●
		Cleaned coal	●	●	●	●	●	●	●	●	●	●	●	●
		Briquette	●	●	●	●	●	●	●	●	●	●	●	●
		Coke	●	●	●	●	●	●	●	●	●	●	●	●
	Residential sector	Raw coal	●	●	●	●	●	●	●	●	●	●	●	●
		Cleaned coal	●	●	●	●	●	●	●	●	●	●	●	●
		Briquette	●	●	●	●	●	●	●	●	●	●	●	●
		Coke	●	●	●	●	●	●	●	●	●	●	●	●
	Other sector	Raw coal	●	●	●	●	●	●	●	●	●	●	●	●
		Cleaned coal	●	●	●	●	●	●	●	●	●	●	●	●
		Briquette	●	●	●	●	●	●	●	●	●	●	●	●
		Coke	●	●	●	●	●	●	●	●	●	●	●	●
Non-coal combustion	Biomass burning	Straw	●	●	●	●	●	●	●	●	●	●	●	●
		Wood	●	●	●	●	●	●	●	●	●	●	●	●
	Liquid fuel combustion	Crude oil	●	●	●	●	●	●	●		●	●	●	●
		Gasoline	●	●	●	●	●	●	●		●	●	●	●
		Diesel for stationary sources	●	●	●	●	●	●	●		●	●	●	●

	Diesel for transportation	●	●	●	●	●	●	●		●	●	●	●
	Fuel oil	●	●	●	●	●	●	●		●	●	●	●
	Kerosene for stationary sources	●	●	●	●	●	●	●		●	●	●	●
	Kerosene for transportation			●	●	●	●	●		●	●	●	●
Nonferrous smelting	Primary copper	●	●	●	●	●	●	●	●	●		●	●
	Secondary copper	●	●	●	●	●	●	●	●			●	●
	Primary lead	●	●	●	●	●	●	●	●			●	●
	Secondary lead	●	●		●	●	●					●	●
	Primary zinc	●	●	●	●	●	●	●	●			●	●
	Secondary zinc	●	●		●	●	●						●
	Primary aluminum					●	●	●					●
	Secondary aluminum	●	●		●	●	●	●		●		●	●
	Nickel							●					
	Antimony								●				
	Gold (large scale)	●											
	Mercury mining	●											
Non-metallic minerals manufacturing	Cement	●	●	●	●	●	●	●		●		●	●
	Glass	●	●	●	●	●	●	●				●	●
	Brick	●	●	●	●	●	●	●	●	●	●		
Ferrous smelting	Pig iron	●	●	●	●	●	●	●	●	●		●	●
	Steel	●	●	●	●	●	●	●	●	●		●	●
Municipal solid waste (MSW) incineration	Municipal solid waste	●	●	●	●	●	●	●	●	●		●	●
Brake and Tyre wear (B&TW)	Brake pad		●	●	●	●	●	●	●	●	●	●	●
	Tyre		●	●	●	●	●	●	●	●	●	●	●

Table S2 The emission source classification by coal combustion sector

Economic sector	Fuel type	Boiler type	PM control device	SO_2 control device	NO_X control device
Coal-fired power plant	raw coal	pulverized-coal boiler	ESP	WFGD	SCR
	raw coal	pulverized-coal boiler	ESP	WFGD	
	raw coal	pulverized-coal boiler	ESP		
	raw coal	pulverized-coal boiler	FF	WFGD	SCR
	raw coal	pulverized-coal boiler	FF	WFGD	
	raw coal	pulverized-coal boiler	FF		
	raw coal	pulverized-coal boiler	wet scrubber	WFGD	
	raw coal	pulverized-coal boiler	wet scrubber		
	raw coal	pulverized-coal boiler	cyclone	WFGD	
	raw coal	pulverized-coal boiler	cyclone		
	raw coal	fluidized-bed furnace	ESP	WFGD	SCR
	raw coal	fluidized-bed furnace	ESP	WFGD	
	raw coal	fluidized-bed furnace	ESP		
	raw coal	fluidized-bed furnace	FF	WFGD	SCR
	raw coal	fluidized-bed furnace	FF	WFGD	
	raw coal	fluidized-bed furnace	FF		
	raw coal	fluidized-bed furnace	wet scrubber	WFGD	
	raw coal	fluidized-bed furnace	wet scrubber		
	raw coal	fluidized-bed furnace	cyclone		
	raw coal	stoker fired boiler	ESP	WFGD	
	raw coal	stoker fired boiler	ESP		
	raw coal	stoker fired boiler	FF		
	raw coal	stoker fired boiler	wet scrubber		

	raw coal	stoker fired boiler	cyclone		
	cleaned coal	pulverized-coal boiler	ESP	WFGD	SCR
	cleaned coal	pulverized-coal boiler	ESP	WFGD	
	cleaned coal	pulverized-coal boiler	ESP		
	cleaned coal	pulverized-coal boiler	FF	WFGD	SCR
	cleaned coal	pulverized-coal boiler	FF	WFGD	
	cleaned coal	pulverized-coal boiler	FF		
	cleaned coal	pulverized-coal boiler	wet scrubber	WFGD	
	cleaned coal	pulverized-coal boiler	wet scrubber		
	cleaned coal	pulverized-coal boiler	cyclone	WFGD	
	cleaned coal	pulverized-coal boiler	cyclone		
	cleaned coal	fluidized-bed furnace	ESP	WFGD	SCR
	cleaned coal	fluidized-bed furnace	ESP	WFGD	
	cleaned coal	fluidized-bed furnace	ESP		
	cleaned coal	fluidized-bed furnace	FF	WFGD	SCR
	cleaned coal	fluidized-bed furnace	FF	WFGD	
	cleaned coal	fluidized-bed furnace	FF		
	cleaned coal	fluidized-bed furnace	wet scrubber		
	cleaned coal	fluidized-bed furnace	cyclone		
	cleaned coal	stoker fired boiler	ESP	WFGD	
	cleaned coal	stoker fired boiler	ESP		
	cleaned coal	stoker fired boiler	wet scrubber		
	cleaned coal	stoker fired boiler	cyclone		
	briquette	pulverized-coal boiler	ESP	WFGD	
	coke	pulverized-coal boiler	ESP	WFGD	

	raw coal	stoker fired boiler	ESP
	raw coal	stoker fired boiler	FF
	raw coal	stoker fired boiler	wet scrubber
	raw coal	stoker fired boiler	cyclone
	raw coal	stoker fired boiler	
	raw coal	fluidized-bed furnace	wet scrubber
	raw coal	coke furnace	FF
	raw coal	coke furnace	wet scrubber
	raw coal	coke furnace	
	cleaned coal	stoker fired boiler	ESP
	cleaned coal	stoker fired boiler	FF
	cleaned coal	stoker fired boiler	wet scrubber
Coal-fired industrial boiler	cleaned coal	stoker fired boiler	cyclone
	cleaned coal	stoker fired boiler	
	cleaned coal	fluidized-bed furnace	wet scrubber
	cleaned coal	coke furnace	FF
	cleaned coal	coke furnace	wet scrubber
	cleaned coal	coke furnace	
	briquette	stoker fired boiler	wet scrubber
	briquette	stoker fired boiler	cyclone
	briquette	stoker fired boiler	
	coke	stoker fired boiler	wet scrubber
	coke	stoker fired boiler	cyclone
	coke	stoker fired boiler	
Coal-fired residential	raw coal	stove	

sector	cleaned coal	stove	
	briquette	stove	
	coke	stove	
Coal-fired other sectors	raw coal	stoker fired boiler	wet scrubber
	raw coal	stoker fired boiler	cyclone
	raw coal	stoker fired boiler	
	cleaned coal	stoker fired boiler	
	briquette	stoker fired boiler	
	coke	stoker fired boiler	

Table S3 Statistical parameters of bootstrap mean contents of Hg, As, Se, Pb, Cd, Cr, Ni and Sb in produced coal by provinces (Tian et al., 2013)

Provinces	Hg	As	Se	Pb	Cd	Cr	Ni	Sb
Anhui	0.43	2.89	7.54	13.24	0.11	31.25	19.57	0.25
Beijing								
Chongqing	0.31	5.66	3.69	30.44	1.22	28.44	20.9	1.71
Fujian	0.07	9.93	1.22	25.53	0.31	30.48	16.42	0.38
Gansu	0.27	4.14	0.51	8.35	0.08	23.7	19.3	0.7
Guangdong	0.07	8.3	0.6	24.4	0.25	74	24.9	
Guangxi	0.33	16.94	5.03	29.94	0.41	116.41	22.48	5.55
Guizhou	0.39	6.68	3.82	23.81	0.79	28.47	22.87	6.01
Hainan								
Hebei	0.15	4.88	2.31	29.3	0.23	32.52	14.61	0.41
Heilongjiang	0.12	3.42	0.9	22.15	0.13	15.48	10.49	0.79
Henan	0.2	2.2	4.86	16.78	0.54	24.94	11.84	0.37
Hubei	0.2	5.3	8.76	47.39	0.36	40.52	18.61	1.17
Hunan	0.12	10.59	3.72	26.29	0.64	37.03	13.25	1.54
Inner Mongolia	0.22	5.77	1.1	26.67	0.1	13.02	6.35	0.7
Jiangsu	0.69	2.74	6.11	20.98	0.06	19.82	15.48	0.55
Jiangxi	0.16	7.41	8.39	19.33	0.56	39.75	22.66	1.83
Jilin	0.4	11.57	4.06	29	0.15	23.09	15.34	1.02
Liaoning	0.17	5.51	0.85	19.68	0.16	26.24	24.13	0.81
Ningxia	0.22	3.65	4.27	14.05	1.1	10.63	10.95	0.27
Qinghai	0.25	2.68	0.3	10.72	0.03	30.82	12.2	0.91
Shaanxi	0.21	3.87	3.43	35.17	0.75	32.73	18.86	2.95
Shandong	0.18	5.23	3.66	16.64	0.39	20.62	23.77	0.47
Shanghai								
Shanxi	0.17	3.84	3.85	26.23	0.75	21.57	15.41	1.13
Sichuan	0.29	5.38	3.31	28.29	1.95	33	19.28	1.7
Tianjin								
Xinjiang	0.06	2.97	0.24	2.68	0.12	7.83	8.26	0.67
Yunnan	0.36	8.82	1.48	42.54	0.8	73.62	24.32	0.97
Zhejiang	0.65	12.04	12.02	17.25	0.47	24.2	9.95	0.73

Table S4 Mn Content of Raw Coal as Mined in China, by Province

Provinces[a]	Number of samples	Minimum (μg/g)	Maximum (μg/g)	Arithmetic mean (μg/g)	Literature cited
Anhui	47	0.80	76.30	27.69	(Tang et al., 2002; Wu, 2006; Li et al., 2011)
Beijing[b]	/	/	/	45.80	/
Chongqing	20	5.23	291.00	66.65	(Zhao et al., 2002; Zhuang et al., 2003)

Fujian	7	30.00	459.00	134.28	(Yan and Lu, 1995)
Gansu	13	31.00	1820.00	671.32	(Ren et al., 2006)
Guangxi	15	4.00	128.70	52.49	(Yan and Lu, 1995; Tang et al., 2002)
Guizhou	101	7.00	937.00	152.62	(Zhuang et al., 2000; Tang et al., 2002; Wu et al., 2008)
Hebei	5	20.00	111.00	45.80	(Zhao et al., 2002)
Heilongjiang	1	219.80	219.80	219.80	(Tang et al., 2002; Ren et al., 2006)
Henan	10	22.53	367.46	101.39	(Yan and Lu, 1995; Guo et al., 2005)
Hubei	9	4.00	100.00	49.53	(Yan and Lu, 1995)
Hunan	7	4.00	690.00	266.01	(Wang and Mo, 1999; Tang et al., 2002)
Inner Mongolia	10	12.70	510.00	149.43	(Tang et al., 2002; Guo et al., 2005; Li et al., 2008)
Jiangsu	3	3.90	188.00	95.95	(Tang et al., 2002; Xiu and Wen, 2004; Ren et al., 2006)
Jiangxi	21	8.00	224.00	79.59	(Tang et al., 2002)
Jilin	10	3.30	270.90	84.39	(Ma et al., 2000; Tang et al., 2002; Ren et al., 2006)
Liaoning	6	7.00	200.34	120.56	(Tang et al., 2002; Guo et al., 2005; Ren et al., 2006)
Ningxia	16	7.75	209.50	48.49	(Zhao et al., 2002)
Qinghai	4	22.08	212.00	82.54	(Ren et al., 2006)
Shaanxi	31	6.39	3950.00	398.87	(Dou et al., 1998; Yang et al., 2008a; Yang et al., 2008b)
Shandong	19	9.00	239.50	87.06	(Yan and Lu, 1995; Tang et al., 2002; Guo et al., 2005)
Shanxi	64	0.20	1624.00	80.90	(Tang et al., 2002; Guo et al., 2005)
Sichuan	14	7.20	412.00	121.37	(Tang et al., 2002; Zhuang et al., 2003)
Xinjiang	99	2.00	501.00	52.18	(Cui et al., 2004; Zhou et al., 2010)
Yunnan	9	31.00	125.30	51.41	(Tang et al., 2002; Guo et al., 2005; Dai et al., 2009)
Zhejiang	3	28.00	41.24	32.71	(Li et al., 1993; Tang et al., 2002)

[a] Hong Kong, Macao, Taiwan are not included in this table, Guangdong, Hainan, Shanghai, Tianjin and Tibet do not produce raw coal.

[b] Beijing lack of corresponding date, in this study, we choose the Mn content of Hebei instead.

Table S5 Co Content of Raw Coal as Mined in China, by Province

Provinces[c]	Number of samples	Minimum (μg/g)	Maximum (μg/g)	Arithmetic mean (μg/g)	Literature cited
Anhui	97	1.32	65.70	12.12	(Tang et al., 2002; Wu, 2006; Chen et al., 2009)
Beijing[d]	/	/	/	8.91	/
Chongqing	38	1.38	90.30	13.38	(Zhao et al., 2002; Bai et al., 2007)

Fujian	4	1.24	15.50	7.55	(Yan and Lu, 1995; Wang et al., 1997; Xu et al., 2001)
Gansu	3	1.54	15.90	7.05	(Ren et al., 2006)
Guangxi	35	2.24	19.90	7.05	(Yan and Lu, 1995; Wang et al., 1997; Xu et al., 2001; Zeng et al., 2005)
Guizhou	148	0.40	119.00	11.91	(Zhuang et al., 1999; Zhuang et al., 2000; Tang et al., 2002; Yang, 2006; Wu et al., 2008)
Henan	9	3.25	12.77	5.93	(Yan and Lu, 1995; Xu et al., 2001; Tang et al., 2002)
Hubei	13	3.00	45.00	8.91	(Yan and Lu, 1995; Xu et al., 2001)
Hebei	38	1.00	24.40	6.80	(Wang et al., 1997; Zhuang et al., 1999; Tang et al., 2002; Dai et al., 2003; Tang et al., 2005; Tang et al., 2009)
Heilongjiang	7	5.60	25.50	12.42	(Tang et al., 2002; Ren et al., 2006)
Hunan	12	0.80	18.50	6.15	(Wang and Mo, 1999; Tang et al., 2002)
Inner Mongolia	99	0.20	28.20	4.08	(Wang et al., 1997; Tang et al., 2002; Dai et al., 2003; Li et al., 2008)
Jilin	13	4.98	38.50	10.91	(Ma et al., 2000; Ren et al., 2006)
Jiangsu	3	1.30	20.10	11.20	(Tang et al., 2002; Xiu and Wen, 2004)
Jiangxi	20	1.00	13.00	5.48	(Xu et al., 2001; Tang et al., 2002)
Liaoning	24	3.60	53.66	13.59	(Kong et al., 2001; Tang et al., 2002; Ren et al., 2004)
Ningxia	18	0.88	22.60	7.29	(Zhao et al., 2002; Bai, 2003)
Qinghai	4	2.19	4.03	2.85	(Zhao et al., 2002; Bai, 2003)
Sichuan	21	0.80	47.60	9.39	(Tang et al., 2002; Zhuang et al., 2003)
Shandong	73	0.34	46.30	5.89	(Yan and Lu, 1995; Huang et al., 2000; Xu et al., 2001; Tang et al., 2002)
Shaanxi	34	0.94	32.90	8.65	(Dou et al., 1998; Tang et al., 2002; Yang et al., 2008a; Yang et al., 2008b)
Shanxi	69	0.40	28.30	4.82	(Wang et al., 1997; Zhuang et al., 1999; Tang et al., 2002; Dai et al., 2003)
Xinjiang	62	0.45	25.80	6.63	(Tang et al., 2002; Zhou et al., 2010)
Yunnan	40	1.79	37.86	11.84	(Tang et al., 2002; Dai et al., 2009; Hu et al., 2009)
Zhejiang	3	2.65	7.39	4.64	(Li et al., 1993; Tang et al., 2002)

[c] Hong Kong, Macao, Taiwan are not included in this table, Guangdong, Hainan, Shanghai, Tianjin and Tibet do not produce raw coal.

[d] Beijing lack of corresponding date, in this study, we choose the Co content of Hebei instead.

Table S6 Cu Content of Raw Coal as Mined in China, by Province

Province[e]	Number of	Minimum (μg/g)	Maximum (μg/g)	Arithmetic mean	Literature cited

	samples			(μg/g)	
Anhui	85	5.03	140.00	36.21	(Tang et al., 2002; Wu, 2006; Chen et al., 2009)
Beijing[f]	/	/	/	27.37	/
Chongqing	28	14.50	156.00	42.57	(Zhuang et al., 2003; Bai et al., 2007; Zhu and Li, 2009)
Fujian	4	21.60	59.00	38.48	(Yan and Lu, 1995; Xu et al., 2001)
Gansu	1	7.25	7.25	7.25	(Bai, 2003; Ren et al., 2006)
Guangxi	45	3.00	69.00	25.79	(Yan and Lu, 1995; Xu et al., 2001; Zeng et al., 2005)
Guizhou	131	0.90	370.00	55.04	(Zhuang et al., 2000; Yang, 2006; Wu et al., 2008; Cheng et al., 2013)
Henan	8	23.30	60.37	40.86	(Yan and Lu, 1995; Xu et al., 2001; Tang et al., 2002)
Hubei	9	19.00	81.00	33.89	(Yan and Lu, 1995; Xu et al., 2001)
Hebei	31	6.90	78.40	27.37	(Zhuang et al., 1999; Tang et al., 2002; Dai et al., 2003; Tang et al., 2005; Tang et al., 2009)
Heilongjiang	12	4.10	69.00	15.62	(Tang et al., 2002; Ren et al., 2006)
Hunan	5	4.28	51.50	25.79	(Wang and Mo, 1999; Tang et al., 2002)
Inner Mongolia	93	1.60	92.20	18.63	(Tang et al., 2002; Dai et al., 2003; Li et al., 2008)
Jilin	10	5.00	98.70	28.17	(Ma et al., 2000; Tang et al., 2002)
Jiangsu	2	21.60	76.30	48.95	(Tang et al., 2002; Xiu and Wen, 2004)
Jiangxi	20	7.00	60.70	21.13	(Xu et al., 2001; Bai, 2003)
Liaoning	19	7.90	85.00	30.38	(Kong et al., 2001; Ren et al., 2004)
Ningxia	4	1.49	8.07	4.52	(Zhao et al., 2002; Bai, 2003)
Qinghai[g]	/	/	/	15.71	/
Sichuan	12	11.20	65.90	33.52	(Tang et al., 2002; Bai, 2003; Zhuang et al., 2003)
Shandong	37	2.64	238.00	34.78	(Yan and Lu, 1995; Liu et al., 2001; Xu et al., 2001; Tang et al., 2002)
Shaanxi	31	5.60	164.00	31.93	(Dou et al., 1998; Tang et al., 2002; Yang et al., 2008a; Yang et al., 2008b)
Shanxi	57	0.00	264.00	27.89	(Zhuang et al., 1999; Tang et al., 2002; Dai et al., 2003)
Xinjiang	96	0.80	36.00	6.58	(Zhao et al., 2002; Bai, 2003; Zhou et al., 2010)
Yunnan	24	0.00	169.00	59.38	(Tang et al., 2002; Dai et al., 2009; Hu et al., 2009)
Zhejiang	1	93.28	93.28	93.28	(Zhao et al., 2002; Bai, 2003)

[e] Hong Kong, Macao, Taiwan are not included in this table, Guangdong, Hainan, Shanghai, Tianjin and Tibet do not produce raw coal.

[f,g] Beijing and Qinghai lack of corresponding date, in this study, we choose the Cu content of

Hebei and the average Cu content of surrounding province (Gansu, Sichuan and Xinjiang) instead, respectively.

Table S7 Zn Content of Raw Coal as Mined in China, by Province

Provinces[h]	Number of samples	Minimum (μg/g)	Maximum (μg/g)	Arithmetic mean (μg/g)	Literature cited
Anhui	100	1.00	112.00	26.17	(Tang et al., 2002; Wu, 2006; Chen et al., 2009; Li et al., 2011)
Beijing[i]	/	/	/	49.54	/
Chongqing	26	1.00	39.00	23.41	(Zhao et al., 2002; Zhuang et al., 2003)
Fujian	4	90.00	299.00	174.75	(Yan and Lu, 1995; Wang et al., 1997)
Gansu	2	6.40	54.30	30.35	(Ren et al., 2006)
Guangxi	38	1.41	212.00	56.88	(Yan and Lu, 1995; Wang et al., 1997; Zeng et al., 2005)
Guizhou	157	0.79	561.00	56.97	(Zhuang et al., 1999; Zhuang et al., 2000; Yang, 2006; Li et al., 2011; Wei et al., 2012; Cheng et al., 2013)
Henan	8	10.41	60.00	31.93	(Yan and Lu, 1995)
Hubei	11	5.00	384.00	63.46	(Yan and Lu, 1995)
Hebei	40	5.13	131.00	49.54	(Zhuang et al., 1999; Tang et al., 2002; Dai et al., 2003; Tang et al., 2005; Tang et al., 2009)
Heilongjiang	2	21.40	33.00	27.20	(Ren et al., 2006)
Hunan	4	19.80	158.00	60.35	(Zhao et al., 2002)
Inner Mongolia	97	23.90	257.00	43.18	(Wang et al., 1997; Dai et al., 2003; Li et al., 2008)
Jilin	14	5.40	360.00	79.71	(Ma et al., 2000; Tang et al., 2002)
Jiangsu	2	17.20	18.94	18.07	(Tang et al., 2002; Xiu and Wen, 2004)
Jiangxi	17	3.40	173.00	92.12	(Zhao et al., 2002)
Liaoning	20	22.00	310.00	70.71	(Kong et al., 2001; Tang et al., 2002; Ren et al., 2004)
Ningxia	9	7.30	73.96	21.60	(Song et al., 2011)
Qinghai[j]	/	/	/	30.89	/
Sichuan	13	22.30	99.50	45.65	(Zhao et al., 2002; Zhuang et al., 2003)
Shandong	62	2.67	68.70	16.38	(Yan and Lu, 1995; Huang et al., 2000; Liu et al., 2001; Tang et al., 2002)
Shaanxi	33	8.75	1511.00	114.64	(Dou et al., 1998; Yang et al., 2008a; Yang et al., 2008b)
Shanxi	62	0.56	864.85	65.05	(Wang et al., 1997; Zhuang et al., 1999; Tang et al., 2002; Dai et al., 2003)
Xinjiang	65	4.00	112.00	16.55	(Tang et al., 2002; Zhou et al., 2010)
Yunnan	40	0.00	204.00	59.11	(Dai et al., 2009; Hu et al., 2009)

Zhejiang	1	14.81	14.81	14.81	(Zhao et al., 2002)

[h] Hong Kong, Macao, Taiwan are not included in this table, Guangdong, Hainan, Shanghai, Tianjin and Tibet do not produce raw coal.

[i,j] Beijing and Qinghai lack of corresponding date, in this study, we choose the Zn content of Hebei and the average Zn content of surrounding province (Gansu, Sichuan and Xinjiang) instead, respectively.

Table S8 Averaged concentrations of Hg, As, Se, Pb, Cd, Cr, Ni, Sb, Mn, Co, Cu and Zn in coals as consumed by province (unit: μg/g).

Provinces	Hg	As	Se	Pb	Cd	Cr	Ni	Sb	Mn	Co	Cu	Zn
Anhui	0.40	2.94	7.19	13.82	0.16	30.37	19.16	0.28	34.85	11.37	36.36	27.00
Beijing	0.17	4.02	3.59	26.74	0.66	23.39	15.27	1.01	75.49	5.16	27.82	63.24
Chongqing	0.32	5.74	3.70	29.91	1.19	28.44	21.06	2.05	73.65	13.29	43.67	26.11
Fujian	0.12	4.48	2.32	15.23	0.29	18.53	12.93	0.37	86.32	6.55	29.83	68.99
Gansu	0.23	3.74	0.43	7.48	0.08	21.31	16.30	0.72	166.54	6.50	8.05	27.64
Guangdong	0.14	4.04	2.78	19.20	0.53	18.67	12.29	1.41	99.33	6.09	27.99	59.28
Guangxi	0.14	6.31	1.75	14.04	0.22	44.78	9.73	1.86	52.55	6.78	23.12	38.71
Guizhou	0.39	6.69	3.81	23.86	0.79	28.60	22.87	6.00	153.52	11.88	55.20	57.30
Hainan	0.09	2.07	2.08	14.16	0.40	11.64	8.32	0.61	68.33	5.23	21.91	46.98
Hebei	0.19	5.17	1.94	27.45	0.25	20.71	10.50	0.68	104.34	5.08	22.95	49.06
Heilongjiang	0.16	4.39	0.97	23.76	0.12	14.97	9.46	0.76	187.49	9.30	17.24	35.33
Henan	0.19	2.64	4.58	19.47	0.59	24.61	12.87	0.61	96.01	5.77	37.72	41.57
Hubei	0.21	3.85	4.22	33.40	0.70	31.65	17.70	2.26	98.98	8.00	32.77	92.46
Hunan	0.14	8.10	3.80	25.46	0.66	31.66	13.72	1.34	200.45	5.75	27.19	59.80
Inner Mongolia	0.21	5.57	1.13	25.54	0.11	12.97	6.63	0.69	142.62	4.18	18.60	42.68
Jiangsu	0.25	4.13	3.47	26.28	0.44	24.72	15.73	1.50	103.15	7.38	30.50	64.74
Jiangxi	0.18	5.25	6.19	22.91	0.61	33.52	19.09	1.71	88.56	6.28	27.96	81.93
Jilin	0.28	7.77	2.38	26.86	0.15	18.10	11.33	0.86	128.46	8.23	22.62	57.68
Liaoning	0.18	5.42	1.18	21.94	0.16	20.25	16.05	0.77	130.45	9.77	24.51	56.64
Ningxia	0.21	3.67	4.22	15.44	1.06	11.88	11.46	0.37	51.87	7.00	7.19	26.70
Qinghai	0.24	2.89	1.16	11.44	0.26	26.46	11.93	0.77	75.08	3.81	13.29	28.88
Shaanxi	0.21	3.85	3.51	33.16	0.78	30.63	18.11	2.70	102.92	8.48	29.21	104.21
Shandong	0.19	5.05	2.76	22.77	0.37	18.26	15.05	0.71	108.20	4.97	27.19	38.62

Shanghai	0.28	4.92	2.73	24.01	0.20	17.07	10.94	0.68	118.85	5.76	25.58	39.93
Shanxi	0.16	3.76	3.64	24.81	0.70	20.74	14.96	1.08	79.91	4.88	27.06	63.26
Sichuan	0.29	5.45	3.27	29.64	1.74	34.39	19.63	1.76	110.71	9.90	35.28	49.16
Tianjin	0.17	3.98	3.65	26.52	0.69	22.39	15.14	1.05	76.65	5.09	27.84	63.76
Xinjiang	0.06	3.00	0.25	2.93	0.12	7.88	8.24	0.67	53.35	6.62	6.71	16.83
Yunnan	0.36	8.73	1.58	41.73	0.80	71.66	24.26	1.19	55.82	11.90	59.63	59.34
Zhejiang	0.20	4.05	3.41	27.76	0.60	24.73	15.81	1.63	95.01	6.55	28.43	74.94

Table S9 Removal efficiencies of 12 HMs through coal cleaning and coke process

Categories	Hg	As	Se	Pb	Cd	Cr
Coal cleaning	50.0%	54.0%	30.0%	36.3%	32.2%	58.0%
Coking process	90%	30%	40%	31.5%	20%	24%
Category	Ni	Sb	Mn	Co	Cu	Zn
Coal cleaning	58.5%	35.7%	68.2%	39.3%	31.8%	48.6%
Coking process	9.7%	70%	7.6%	7.1%	20%	26.1%

Table S10 Release rates of Mn, Co, Cu and Zn from coal-fired facilities.

Categories	Release Rates (%)[k]				Literature cited
	Mn	Co	Cu	Zn	
Pulverized-coal boiler	91.70			94.70	(Jin et al., 2003)
	67.00	88.00			(Nodelman et al., 2000)
	57.00	90.00			
	58.00	62.00			(Llorens et al., 2001)
	94.40	92.86	92.31		(Benson et al., 1995)
	86.00	94.00	93.00	96.00	(Xu et al., 2004)
				84.00	(Álvarez-Ayuso et al., 2006)
Stoker fired boiler	16.24	42.44	25.69	33.34	(Wang et al., 1996)
	6.30	11.40			(Zhang et al., 2003)
	26.00	21.88			(Song et al., 2006a)
				15.00	(He et al., 2005)
				5.00	
				12.00	
Fluidized-bed furnace	47.70	57.50	50.20	51.40	(Reddy et al., 2005)
	50.20	55.30	43.40	49.70	
	42.70	56.20	45.75	44.80	
	64.29	66.70	83.30		(Benson et al., 1995)
		76.45	78.90	82.01	(Klika et al., 2001)
		82.29	87.26	77.94	
		47.00	30.00		(Bartoňová and Klika, 2009)
		61.00	68.00		
	28.00				(Zajusz-Zubek and Konieczyński, 2003)
Coke furnace	23.00	22.00	11.00		(Chen et al., 2008)
	38.00	36.00	22.00		
	40.00	37.00	33.00		
	12.00				(Guo et al., 2004)
				58.00	(Helble et al., 1996)
				30.00	(Wei et al., 2012)

[k] the release rate of Hg, As, Se, Pb, Cd, Cr, Ni and Sb from different combustion boilers can be referred in our previously studies (Tian et al., 2010; Tian et al., 2011; Tian et al., 2012a; Tian et al., 2012b).

Table S11. Removal efficiencies of Mn, Co, Cu and Zn by different control devices

Categories	Release Rates (%)[1]				Literature cited
	Mn	Co	Cu	Zn	
	93.00	97.00	97.00		(Benson et al., 1995)
	95.00				
	97.90	97.20		92.90	(Ondov et al., 1979)
	99.90	99.80			
	99.10	99.80			(Nyberg et al., 2009)
ESPs	97.20	99.40			
		97.70			
	98.50	98.20			(Helble, 2000)
	86.00	94.00	93.00	96.00	(Xu et al., 2004)
		90.02			(Han et al., 2002)
	87.00	93.00	97.75	97.50	(Nodelman et al., 2000)
FFs	98.00	99.00			
	99.70	99.90			(Nyberg et al., 2009)
	99.80	99.90			
Cyclone	67.00	72.00	60.00	64.00	(Gogebakan and Selçuk, 2009)
Wet scrubber	98.97	99.82	98.97	99.03	(Ondov et al., 1979)
	65.79	76.19	55.56	29.09	(Córdoba et al., 2012a)
	37.50	66.67	86.49	52.50	
	72.08	78.95	24.56	71.38	(Córdoba et al., 2012b)
WFGD		68.93	35.26	80.00	
		32.30	27.27		(Tang et al., 2013)
		41.95	22.17		
		32.88	31.29		

[1] the removal efficiencies of Hg, As, Se, Pb, Cd, Cr, Ni and Sb by different control device can be referred in our previously studies (Tian et al., 2010;Tian et al., 2012a;Tian et al., 2012b;Tian et al., 2011).

Table S12 TSP emission factors for vehicle brake and tyre wear

Vehicle categories	TSP emission factor (g/km)	Uncertainty range(g/km)
TSP emission factors for vehicle tyre wear		
Two-wheel vehicles	0.0046	0.0042–0.0053
Passenger cars	0.0107	0.0067–0.0162
Light-duty trucks	0.0169	0.0088–0.0217
Heavy-duty vehicles	0.0412	0.0227–0.0898
TSP emission factors for vehicle brake wear		
Two-wheel vehicles	0.0037	0.0022 –0.0050
Passenger cars	0.0075	0.0044 –0.0100
Light-duty trucks	0.0117	0.0088 –0.0145
Heavy-duty vehicles	0.0365	0.0235 –0.0420

Table S13 Composition of tyre and brake wear in term of heavy metals, ppm

Element	tyre			brake		
	mean	min	max	mean	min	max
As	3.8	1.6	6	67.5	10	130
Se	20	/	/	20	/	/
Pb	176	6.3	670	6072	120	20000
Cd	4.7	1.4	9	22.4	1.5	57
Cr	23.8	2	61	2311	115	8050
Ni	29.9	2.4	63	327	80	600
Sb	2	/	/	10000	/	/
Mn	51	2	100	2460	1700	3220
Co	12.8	0.9	24.8	6.4	/	/
Cu	174	1.8	490	51112	370	142000
Zn	7434	430	13494	8676	270	21800

Table S14 Parameter values used in the transformed normal distribution function computation of the variation of heavy metals emission factors over time

Elements	Parameters	Hg	As	Se	Pb	Cd	Cr	Ni	Sb	Mn	Cu	Zn
Copper	ef_a	27.50	3333.33	300.00	4000.00	1250.00	25.00	5000.00	336.67	100.00	8333.33	6000.00
	ef_b	8.50	100.00	15.00	200.00	50.00	1.00	50.00	10.10	4.50	250.00	300.00
	S	40	30	30	30	30	30	30	30	30	30	30
Lead	ef_a	43.60	400.00	330.00	8000.00	500.00	57.50	166.67	506.67	/	83.33	680.00
	ef_b	6.00	1.00	16.50	200.00	5.00	2.30	5.00	15.20	/	5.00	20.00
	S	40	30	30	30	30	30	30	30	/	30	30
Zinc	ef_a	75.00	600.00	66.67	2900.00	500.00	39.00	68.00	200.00	/	420.00	16000.00
	ef_b	17.00	5.00	10.00	50.00	5.00	1.17	1.36	6.00	/	25.00	500.00
	S	40	30	30	30	30	30	30	30	30	30	30
Gold (large scale)	ef_a	520	/	/	/	/	/	/	/	/	/	/
	ef_b	25	/	/	/	/	/	/	/	/	/	/
	S	36	/	/	/	/	/	/	/	/	/	/
Mercury mining	ef_a	182	/	/	/	/	/	/	/	/	/	/
	ef_b	45	/	/	/	/	/	/	/	/	/	/
	S	36	/	/	/	/	/	/	/	/	/	/
Iron	ef_a	0.06	3.50	0.26	3.50	1.60	2.67	12.00	0.40	0.83	20.00	57.14
	ef_b	0.04	0.08	0.01	0.07	0.02	0.08	0.12	0.00	0.08	0.40	4.00
	S	40	30	30	30	30	30	30	30	30	30	30
Steel	ef_a	0.05	0.56	0.12	74.32	2.47	4.11	3.15	0.20	129.27	5.79	190.00
	ef_b	0.01	0.01	0.00	1.49	0.02	0.12	0.03	0.00	2.02	0.12	6.05
	S	40	30	30	30	30	30	30	30	30	30	30

Cement	ef_a	0.10	13.94	2.54	54.43	2.28	11.79	8.12	/	55.34	24.20	57.20
	ef_b	0.02	0.07	0.01	0.38	0.01	0.05	0.04	/	0.28	0.12	0.29
	S	35	25	25	25	25	25	25	/	25	25	25
MSW	ef_a	2.80	2.14	0.50	107.00	5.45	4.49	3.93	6.00	9.00	14.00	60.00
	ef_b	0.06	0.05	0.01	0.12	0.01	0.04	0.09	3.00	0.21	0.13	0.11
	S	32	28	28	28	28	28	28	28	28	28	28

Table S15 Abatement efficiencies for nonferrous metals smelting (Pacyna et al., 2002)

Elements	Efficiency, %	95% confidence interval	
		lower, %	upper, %
Hg	0	0	67
As	97	91	99
Se	85	55	95
Pb	95	85	98
Cd	99	96	100
Cr	90	70	97
Ni	97	90	99
Cu	94	81	98
Zn	80	40	93

Table S16 Heavy metals emission factors for non-coal combustion sources: temporal, and process variations

Categories		Durable year	Hg	As	Se	Pb	Cd	Cr	Literature cited
Liquid fuel	Crude oil	1949-2012	0.01	0.17	0.09	0.19	0.05	0.11	(de Souza et al., 2006; UK, 2013)
combustion (g t^{-1} fuel	Fuel oil for stationary	1949-2012	0.01	0.17	0.09	0.19	0.05	0.11	(de Souza et al., 2006; UK, 2013)

combustion)	sources								
	Kerosene for stationary sources	1949-2012	0.0019	0.02	0.0115	0.000009	0.0000504	0.0000504	(US EPA, 1996a)
	Diesel oil for stationary sources	1949-2012	0.0019	0.02	0.0115	0.000009	0.0000504	0.0000504	(US EPA, 1996c)
	Gasoline	1949-2012	0.06	0.02	0.3	/	0.01	0.01	(UK, 2013)
	Diesel oil for transportation	1949-2012	0.06	0.02	0.3	0.0325	0.04	0.04	(Wang et al., 2003b; UK, 2013)
	Kerosene for transportation	1949-2012	/	/	0.14	0.06	0.01	0.01	(UK, 2013)
Nonferrous metal smelting (g t^{-1} nonferrous metal production)	Primary Al	1949-1996	/	/	/	/	0.197	/	(UK, 2000, 2013)
		2012	/	/	/	/	0.1	/	
	Secondary Al	1949-1996	0.032	0.319	/	3.734	0.175	0.841	
		2012	0.0161	0.162	/	1.896	0.0887	0.427	
	Primary Cu	Pre-1900	27.5	3333	300	4000	1250	25	(Habashi, 1978; Nriagu, 1979; Pacyna, 1984; Nriagu and Pacyna, 1988; Skeaff and Dubreuil, 1997; EC, 2001b; Pacyna and Pacyna, 2001; Pacyna et al., 2002; Theloke et al., 2008; Streets et al., 2011; EEA, 2009, 2013)
		2012	8.5	100	15	200	50	1	
	Secondary Cu	1949-1996	1	2	5	90	3	1	
		2012	0.4	0.8	2	36	1.2	0.4	
	Primary Pb	Pre-1900	43.6	400	330	8000	500	57.5	
		2012	6	1	16.5	200	5	2.3	
	Secondary Pb	1949-1996	1	0.5	/	100	2.5	1.773	
		2012	0.4	0.2	/	40	1	0.709	
	Primary Zn	Pre-1900	75	600	67	2900	500	39.0	
		2012	17	5	10	50	5	1.170	

	Secondary Zn	1949-1996	0.013	0.945	/	10.439	5.515	1.799	
		2012	0.0065	0.48	/	5.3	2.8	0.913	
	Ni smelting	1949-1996	/	/	/	/	/	/	/
		2012	/	/	/	/	/	/	
	Sb smelting	1949-1996	/	/	/	/	/	/	/
		2012	/	/	/	/	/	/	
	Gold (large scale)	Pre-1900	520	/	/	/	/	/	(Hylander and Meili, 2005; Pacyna, 2006; Pacyna, 2010; Pirrone, 2010; Streets, 2011)
		2012	25	/	/	/	/	/	
	Mercury mining	Pre-1900	182	/	/	/	/	/	
		2012	45	/	/	/	/	/	
Ferrous metals smelting (g t^{-1} ferrous metal production)	Pig iron	Pre-1900	0.06	3.5	0.26	3.5	1.6	2.7	(Nriagu, 1979; Pacyna, 1984; Nriagu and Pacyna, 1988; Kakareka et al., 1998; UK, 2000; EC, 2001a; Theloke et al., 2008; EEA, 2009; Pirrone et al., 2010; Streets et al., 2011; UK, 2013)
		2015	0.04	0.08	0.013	0.0699	0.016	0.08	
	Steel produced	Pre-1900	0.05	0.5584	0.12	74.3	2.5	4.1	
		2012	0.008	0.011168	0.003	1.5	0.025	0.123	
The output of Non-metallic minerals manufacturing in China, 2000-2012 (g t^{-1} material production)	Glass	1949-1996	0.124	0.248	49.556	24.8	0.372	6.194	(EEA, 2000; EC, 2001c)
		2012	0.050	0.101	20.153	10.1	0.151	2.519	
	Cement	Pre-1900	0.1	13.94	2.54	54.4	2.28	11.8	(Nriagu and Pacyna, 1988; Passant et al., 2002; NPI, 2008; Streets et al., 2011; US EPA, 2012)
		2012	0.0202	0.0697	0.0127	0.38	0.0114	0.0511	
	Brick	1949-1996	0.044	0.059	0.104	0.068	0.007	0.023	(US EPA, 1996b; NPI, 1998)
		2012	0.015	0.020	0.036	0.023	0.002	0.008	
Municipal solid waste	MSWI	Pre-1900	2.8	2.14	0.5	107	5.45	4.49	(Nriagu, 1979; Pacyna, 1984; Nriagu and Pacyna, 1988; US

incineration (g t^{-1}waste)		2016	0.060	0.053	0.0117	0.118	0.012	0.037	EPA, 1996a; UK, 2000)
Biomass burning (g t^{-1} residue)	Crop straw	1949-2012	0.008	0.058	0.036	0.865	0.049	0.22	(US EPA, 1996; Li et al., 2007; EEA, 2013; UK, 2013)
	Firewood	1949-2012	0.03	0.03	0.09	0.91	0.08	0.9	
Category		Durable year	Ni	Sb	Mn	Co	Cu	Zn	Literature cited
Liquid fuel combustion (g t^{-1} fuel combustion)	Crude oil	1949-2012	10.6	/	0.223	0.151	0.460	1.035	(de Souza et al., 2006; UK, 2013)
	Fuel oil for stationary sources	1949-2012	10.6	/	0.223	0.151	0.460	1.035	(de Souza et al., 2006; UK, 2013)
	Kerosene for stationary sources	1949-2012	0.06	/	0.0504	0.101	0.030	0.489	(US EPA, 1996a)
	Diesel oil for stationary sources	1949-2012	0.06	/	0.0504	0.101	0.030	0.489	(US EPA, 1996c)
	Gasoline	1949-2012	0.04	/	0.004	0.002	0.02	0.0275	(UK, 2013)
	Diesel oil for transportation	1949-2012	0.04	/	0.040	0.0151	0.221	0.234	(Wang et al., 2003b; UK, 2013)
	Kerosene for transportation	1949-2012	0.03	/	0.004	0.002	0.034	0.01	(UK, 2013)
Nonferrous metal smelting (g t^{-1} nonferrous metal	Primary Al	1949-1996	19.7	/	/	/	/	19.7	(UK, 2000; 2013)
		2012	10	/	/	/	/	10	
	Secondary Al	1949-1996	0.802	/	1.16	/	3.2	15.2	
		2010	0.407	/	0.588	/	1.621	7.734	
	Primary Cu	Pre-1900	5000	337	100	/	8333	6000	(Nriagu, 1979; Pacyna, 1984;

production)		2012	50	10.1	4.5	/	250	300	Nriagu and Pacyna, 1988; Skeaff and Dubreuil, 1997; EC, 2001a; Pacyna and Pacyna, 2001; Pacyna et al., 2002; Theloke et al., 2008; EEA, 2009, 2013)
	Secondary Cu	1949-1996	1	3	/	/	100	200	
		2012	0.4	1.2	/	/	40	80	
	Primary Pb	Pre-1900	167	507	/	/	83	680	
		2012	5	15.2	/	/	5	20	
	Secondary Pb	1949-1996	/	/	/	/	1	20	
		2012	/	/	/	/	0.4	8	
	Primary Zn	Pre-1900	68	200	/	/	420	16000	
		2012	1.36	6	/	/	25	500	
	Secondary Zn	1949-1996	0	0	/	/	/	270	
		2012	0	0	/	/	/	137.1	
	Ni smelting	1949-1996	900	/	/	/	/	/	(Nriagu, 1979; Tian et al., 2012b)
		2012	360	/	/	/	/	/	
	Sb smelting	1949-1996	/	173	/	/	/	/	(Tian et al., 2012c)
		2012	/	70	/	/	/	/	
Ferrous metals smelting (g t^{-1} ferrous metal production)	Pig iron	Pre-1900	12	0.4	0.830	/	20	57.143	(Nriagu, 1979; Pacyna, 1984; Nriagu and Pacyna, 1988; Kakareka et al., 1998; UK, 2000; EC, 2001a; Theloke et al., 2008; EEA, 2009; Pirrone et al., 2010; UK, 2013)
		2015	0.12	0.004	0.082	/	0.4	4	
	Steel produced	Pre-1900	3.2	0.2	129	/	5.790	190	
		2012	0.032	0.004	2.016	/	0.116	6.0	
Non-metallic minerals manufacturing (g t^{-1} material production)	Glass	1949-1996	5.0	/	/	/	1.239	24.8	(EEA, 2000; EC, 2001c)
		2012	2.015	/	/	/	0.504	10.1	
	Cement	Pre-1900	8.1	/	55.3	/	24.2	57.2	(Nriagu and Pacyna, 1988; Passant et al., 2002; NPI, 2008; US EPA, 2012)
		2012	0.0406	/	0.277	/	0.121	0.286	

	Brick	1949-1996	0.033	0.012	0.132	0.001	/	/	(US EPA, 1996c; NPI, 1998)
		2012	0.011	0.004	0.045	0.0003	/	/	
Municipal solid waste incineration (g t^{-1}waste)	MSWI	Pre-1900	3.93	6	9	/	14	60	(Nriagu, 1979; Pacyna, 1984; Nriagu and Pacyna, 1988; US EPA, 1996b; UK, 2000)
		2016	0.086	3	0.208	/	0.127	0.109	
Biomass burning (g t^{-1} residue)	Crop straw	1949-2012	0.177	0.019	0.0955	0.0045	0.1	0.028	(US EPA, 1996a; Li et al., 2007; EEA, 2013; UK, 2013)
	Firewood	1949-2012	0.98	0.0728	0.652	0.0045	0.1	1.25	

Table S17 Abatement efficiencies for iron and steel production (Kakareka et al., 1998)

Elements	Efficiency, %	95% confidence interval	
		lower, %	upper, %
Pb	96	93	98
Cd	96	91	98
Ni	94	88	97
Zn	95	90	98

Table S18 The emission limits of air pollutants of relative industrial process in China and developed regions, mg/m^3

Pollutants	GB 25467-2010	GB 25466-2010	GB 30770-2014	GB 25465-2010	GB 28663-2012	GB 28664-2012	GB 4915-2013	GB 29620-2013	GB 26453-2011	GB 18485-2014	EU 2000/76/EC	EU 2001/80/EC
PM	80	80	30	20-100	50	50-100	30	100	50	20	10~30	50~100
SO_2	400	400	400	400	100	/	200	400	400	80	50	200~850
NO_X	/	/	200	/	300	/	400	/	700	250	200	200~400
As & compounds	0.4	/	0.5	/	/	/		/	/	/	/	/

Pb & compounds	0.7	/	0.5	/	/	/		/	/	/	/	/
Hg & compounds	0.012	0.05	0.01	/	/	/	0.05	/	/	0.05	0.05	/
Cd & compounds	/	/	0.05	/	/	/		/	/		0.05	/
(Cd+Tl) & their compounds	/	/	/	/	/	/	/	/	/	0.1	0.05~0.1	/
(Sb+As+Pb+Cr+Co+Cu+Mn +Ni) & their compounds	/	/	/	/	/	/	/	/	/	1.0	0.5~1.0	/

Table S19 Data source of activity data for the main heavy metals emitting sectors in China

Emission sectors	Data Sources
Coal consumption by power plants	China Electric Power Yearbook
	China Editorial Power Industry Statistics
Coal consumption by industrial boilers	China Mechanical Industry Yearbook
	China Coal Industry Yearbook
	China Energy Statistical Yearbook
Coal consumption by residential sectors	China Energy Statistical Yearbook
Coal consumption by other sectors	China Energy Statistical Yearbook
Biomass burning	China Statistical Yearbook
Liquid fuels combustion	China Energy Statistical Yearbook
Nonferrous metals smelting	The Yearbook of Nonferrous Metals Industry of China
Non-metallic minerals manufacturing	China Cement Almanac
Ferrous metal smelting	China Steel Yearbook
Municipal solid waste incineration	China Energy Statistical Yearbook
Brake and tyre wear	China Automotive Industry Yearbook
	China's Auto Market Almanac

Table S20 Selected parameters showing method and assumption for uncertainty analysis

Categories	Parameter description	Distribution	Sources or methods
Coal combustion sources			
Coal consumption	power plant	Normal (CV: 5%)	(Zhao et al., 2008)
	Industrial sectors	Normal (CV: 5%)	(Tian et al., 2012a)
	Residential sectors	Normal (CV: 14%)	(Tian et al., 2012a)
	Other sectors	Normal (CV: 16%)	(Tian et al., 2012a)
Release rate	Pulverized-coal boiler	Triangular	Data fitting
	Stoker fired boiler	Triangular	Data fitting
	Fluidized-bed furnace	Triangular	Data fitting
	Coke furnace	Triangular	Data fitting
Removal efficiency	ESPs	Normal (CV: 5%)	Subject judgment
	EFs	uniform	Data fitting
	Wet scrubber	Triangular	Subject judgment
	Cyclone	Normal (CV: 20%)	Subject judgment
	Wet-FGD	Triangular	Subject judgment
	Coal washing	Uniform	Data fitting
Non-coal combustion sources			
Biomass burning	Biofuel consumption	Normal (CV: 20%)	(Zhao et al., 2011)
	Emission factors	Triangular	(Zhao et al., 2011)
	Ratio of biomass burning	Normal (province dependent)	(Zhao et al., 2011)
	straw-to-crop ratio	Uniform (product dependent)	(Zhao et al., 2011)
Liquid fuel	Liquid fuel consumption	Normal (CV: 5%)	(Zhao et al., 2011)

combustion	Emission factors	Normal (CV: 25%)	Subject judgment
Nonferrous metal	Nonferrous metal production	Normal (CV: 5%)	(Zhao et al., 2011)
smelting	Emission factors	Triangular	Data fitting
Non-metallic minerals	Output of Cement/ glass / brick	Normal (CV: 20%)	Subject judgment
manufacturing	emission factors (cement, glass)	Normal (CV: 25%)	Subject judgment
	emission factors (brick)	Normal (CV: 30%)	Subject judgment
Ferrous metal	Pig iron and steel yield	Normal (CV: 15%)	(Zhao et al., 2011)
smelting	Emission factors	Triangular	Subject judgment
Municipal solid	MSW consumption	Normal (CV: 20%)	Subject judgment
waste incineration	Emission factors	Normal (CV: 20%)	Subject judgment
Brake and tyre	Vehicle number	Normal (CV: 5%)	(Zhao et al., 2011)
wear	Average vehicle mileage	Normal (CV: 5%)	(Zhao et al., 2011)
	TSP emission factors	Uniform	Data fitting
	Heavy metal content	Triangular	Data fitting

Table S21 The atmospheric concentrations of As, Pb, Cr and Cu in $PM_{2.5}$ in Beijing during 2000 to 2012

Year	Atmospheric concentration (ng m^{-3})				Literature cited
	As	Pb	Cr	Cu	
2000	30.0	300.0	20.0	30.0	(Song et al., 2006b)
2001		170.0		50.0	(Duan et al., 2006)
2002	38.3	218.3	26.7	58.3	(Sun et al., 2004)
2003	38.3	218.3	26.7	58.3	(Sun et al., 2004)
2005	16.0	189.5	50.0	53.0	(Yu et al., 2012; Zhang et al., 2012)
2006	20.2	173.3	73.6	43.1	(Cui et al., 2008; Yang et al., 2008c, d; Yu et al., 2012)
2007	19.0	189.7	31.7	51.3	(Gao, 2012; Wang et al., 2010; Yu et al., 2012; Zhang et al., 2010)
2008	7.6	67.5	5.8	25.8	(Mu et al., 2010; Yu et al., 2012; Zhang, 2012; Zhang et al., 2010)
2009	17.2	135.5	13.6	40.0	(Tao et al., 2014; Zhao et al., 2013)
2010	22.8	142.7	16.4	36.8	(Tao et al., 2014; Yu et a., 2013)
2011	15.6		13.4	47.7	(Wang et al., 2014)
2012	23.4	158.0	24.6	54.7	(Guo, 2014; Yang et al., 2015; Zhang et al., 2014)

Table S22 Uncertainties in the sectoral emissions of heavy metals in China in 2010

Categories	Hg	As	Se	Pb
Coal-fired power plant	139.4 (-21.5%, 25.7%)	406.4 (-22.6%, 25.2%)	538.6 (-25.5%, 22.5%)	833.0 (-25.6%, 25.9%)
Coal-fired industrial boiler	171.0 (-44.6%, 60.7%)	821.3 (-49.4%, 53.4%)	965.1 (-49.3%, 56.6%)	5449.5 (-57.5%, 58.7%)
Coal-fired residential sector	7.4 (-57.4%, 55.7%)	10.9 (-54.2%, 64.7%)	74.3 (-57.3%, 65.7%)	417.2 (-53.2%, 65.3%)
Coal-fired other sector	22.4 (-66.1%, 71.4%)	485.2 (-70.2%, 78.6%)	282.9 (-69.4%, 71.9%)	1224.1 (-57.0%, 80.9%)
Biomass burning	9.1 (-47.5%, 48.4%)	34.0 (-48.3%, 66.6%)	33.3 (-50.2%, 42.3%)	585.6 (-57.8%, 56.4%)
Liquid fuel combustion	15.9 (-62.9%, 63.6%)	11.8 (-48.2%, 41.3%)	83.5 (-56.0%, 51.3%)	678.4 (-89.3%, 68.2%)
Nonferrous metal smelting	222.5 (-45.2%, 54.8%)	338.9 (-57.1%, 67.6%)	146.7 (-39.5%, 37.4%)	1602.9 (39.6%, 37.8%)
Ferrous metal smelting	29.9 (-52.0%, 52.1%)	57.8 (-55.1%, 61.7%)	9.9 (-47.2%, 58.4%)	1047.3 (-53.5%, 72.3%)
Non-metallic minerals manufacturing	52.9 (-81.6%, 98.5%)	152.4 (-90.2%, 107.5%)	744.0 (-95.1%, 103.2%)	1085.0 (-82.3%, 98.7%)
Municipal solid waste incineration	1.6 (-69.4%, 67.9%)	1.2 (-65.3%, 75.1%)	0.3 (-55.6%, 64.9%)	3.8 (-65.3%, 69.2%)
Brake and tyre wear	/	3.1 (-80.9%, 97.3%)	1.9 (-81.0%, 159.6%)	267.7 (-89.0%, 157.6%)
Total emission	672.1 (-34.2%, 46.7%)	2322.9 (-36.4%, 48.8%)	2880.5 (-39.1%, 50.6%)	13194.5 (-32.7%, 46.9%)

Categories	Cd	Cr	Ni	Sb
Coal-fired power plant	15.5 (-19.0%, 27.0%)	598.3 (-27.2%, 26.4%)	537.8 (-19.1%, 23.7%)	97.9 (-29.4%, 23.9%)
Coal-fired industrial boiler	104.0 (-50.1%, 55.8%)	5317.6 (-61.9%, 53.8%)	1005.8 (-46.8%, 52.3%)	214.8 (-51.6%, 63.6%)
Coal-fired residential sector	3.7 (-58.6%, 60.4%)	58.9 (-56.6%, 67.1%)	33.7 (-58.2%, 68.8%)	1.0 (-58.9%, 77.9%)
Coal-fired other sector	25.7 (-62.5%, 66.5%)	773.7 (-57.5%, 73.0%)	185.7 (-69.8%, 80.0%)	105.9 (-61.1%, 78.7%)
Biomass burning	38.0 (-41.5%, 52.5%)	263.1 (-45.3%, 59.6%)	255.4 (-46.5%, 54.6%)	21.9 (-60.7%, 65.1%)
Liquid fuel combustion	9.6 (-61.0%, 57.9%)	11.9 (-47.7%, 50.3%)	422.1 (-58.2%, 67.9%)	/
Nonferrous metal smelting	200.6 (-54.5%, 56.7%)	19.7 (-35.9%, 37.3%)	422.5 (-39.9%, 38.9%)	122.6 (-38.1%, 33.6%)
Ferrous metal smelting	28.3 (-48.7%, 56.7%)	131.3 (-40.7%, 47.5%)	102.7 (-55.0%, 45.8%)	5.4 (-49.0%, 53.0%)
Non-metallic minerals	28.8 (-80.6%, 90.0%)	190.2 (-85.3%, 93.6%)	155.4 (-90.4%, 86.9%)	3.4 (-96.9%, 106.2%)

manufacturing				
Municipal solid waste incineration	0.3 (-66.5%, 76.2%)	0.9 (-58.8%, 68.3%)	2.0 (-59.3%, 76.0%)	69.5 (-75.7%, 71.0%)
Brake and tyre wear	1.2 (-77.1%, 120.7%)	99.6 (-97.0%, 171.5%)	15.5 (-70.9%, 85.4%)	425.7 (-91.1%, 170.0%)
Total emission	455.8 (-35.7%, 49.3%)	7465.2 (-33.8%, 47.8%)	3138.6 (-38.2%, 49.4%)	1068.1 (-35.6%, 48.3%)

Categories	Mn	Co	Cu	Zn
Coal-fired power plant	3072.1 (-23.5, 29.1%)	164.9 (-33.5%, 25.2%)	1477.5 (-21.6%, 28.7%)	2367.1 (-23.0%, 31.5%)
Coal-fired industrial boiler	4472.2 (-43.6%, 45.1%)	517.4 (-39.8%, 43.5%)	2004.4 (-55.0%, 56.9%)	4449.7 (-51.6%, 49.0%)
Coal-fired residential sector	43.4 (-57.8%, 56.7%)	5.3 (-62.6%, 58.4%)	24.0 (-61.3%, 57.0%)	284.6 (-59.7%, 57.0%)
Coal-fired other sector	2130.7 (-60.6%, 74.7%)	218.8 (-59.7%, 63.2%)	933.0 (-65.2%, 73.4%)	1058.8 (-65.6%, 65.2%)
Biomass burning	158.8 (-52.3%, 53.4%)	3.0 (-45.7%, 56.3%)	66.8 (-45.7%, 46.3%)	227.4 (-54.5%, 63.6%)
Liquid fuel combustion	15.6 (-51.4%, 53.8%)	8.6 (-46.7%, 55.1%)	56.4 (-60.9%, 52.2%)	81.5 (-62.6%, 44.4%)
Nonferrous metal smelting	15.8 (-59.8%, 56.5%)	/	990.1 (-66.8%, 53.5%)	3960.2 (-56.3%, 41.6%)
Ferrous metal smelting	1431.4 (-56.9, 68.2%)	/	331.2 (-43.7%, 48.5%)	6421.2 (-56.4%, 43.9%)
Non-metallic minerals manufacturing	563.5 (-89.9%, 85.7%)	0.3 (-93.3%, 94.1%)	247.6 (-88.7%, 103.3%)	889.8 (-86.4%, 94.2%)
Municipal solid waste incineration	4.9 (-87.9%, 60.9%)	/	3.1 (-61.4%, 66.5%)	3.1 (-79.2%, 61.9%)
Brake and tyre wear	107.4 (-45.1%, 61.8%)	0.9 (-79.7%, 159.8%)	2184.8 (-93.4%, 139.4%)	760.2 (-84.3%, 113.9%)
Total emission	12015.9 (-32.2%, 42.0%)	919.2 (-38.1%, 41.4%)	8318.8 (-37.5%, 50.8%)	20503.7 (-32.2%, 45.5%)

Figures

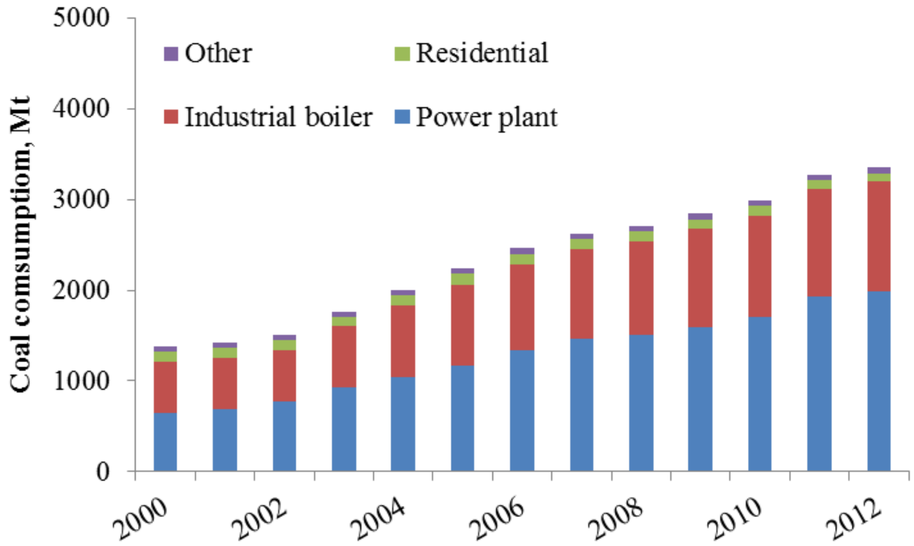

Fig. S1. Coal consumption by different sectors in China, 2000-2012

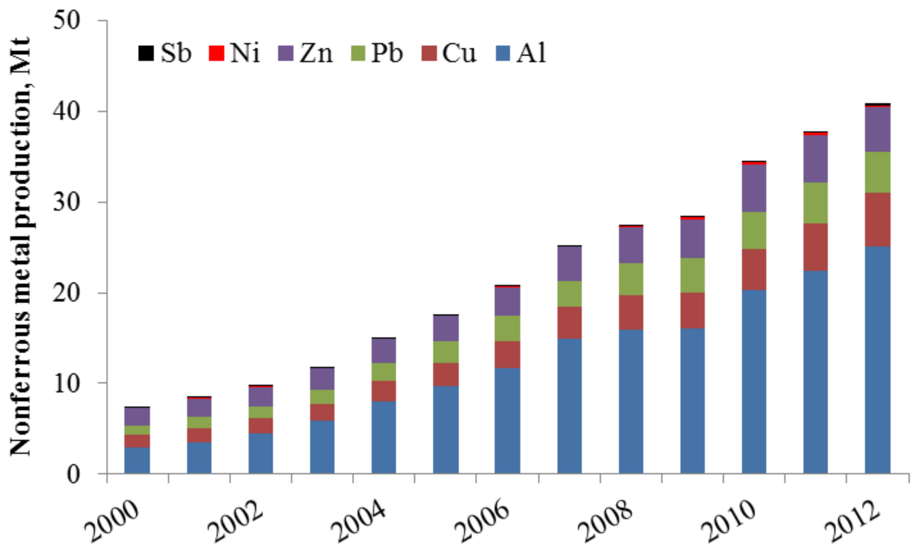

Fig. S2. The output of nonferrous metals in China, 2000-2012

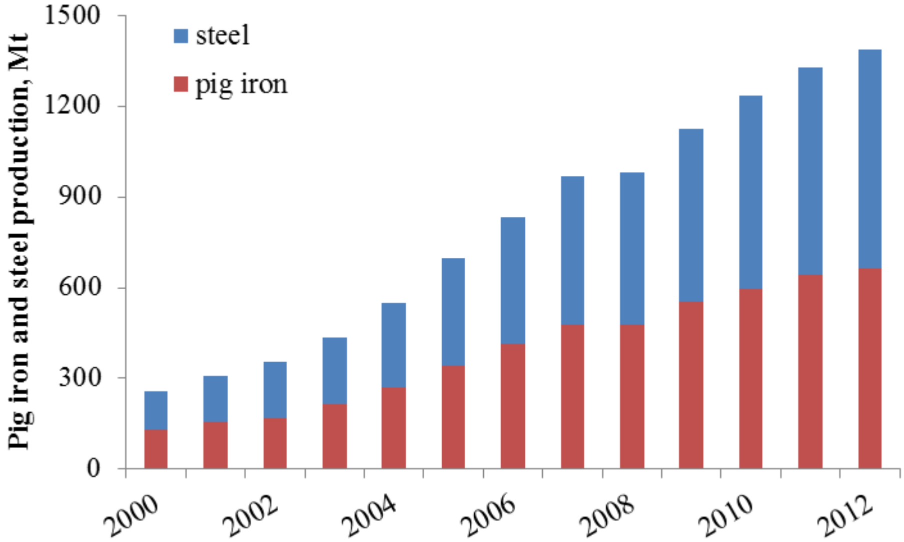

Fig. S3. The output of pig iron and steel products in China, 2000-2012

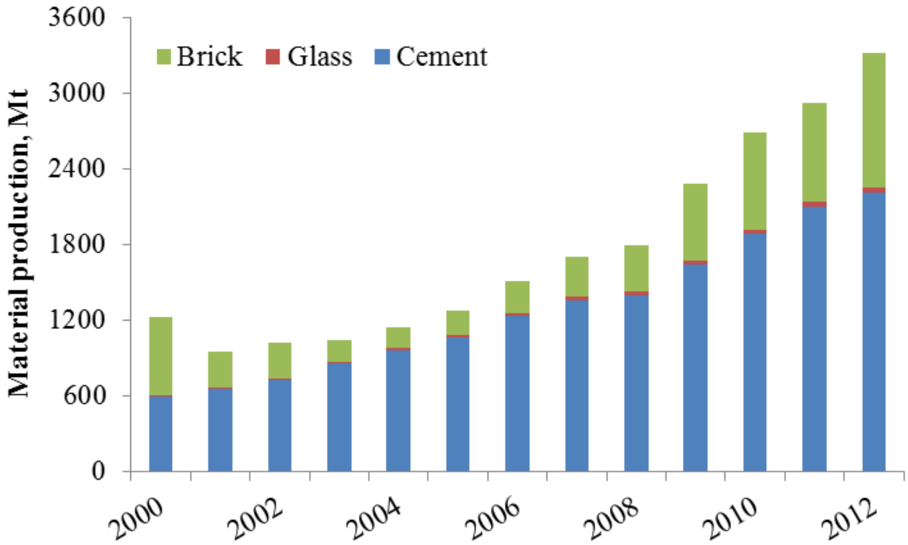

Fig. S4. The output of non-metallic minerals manufacturing in China, 2000-2012

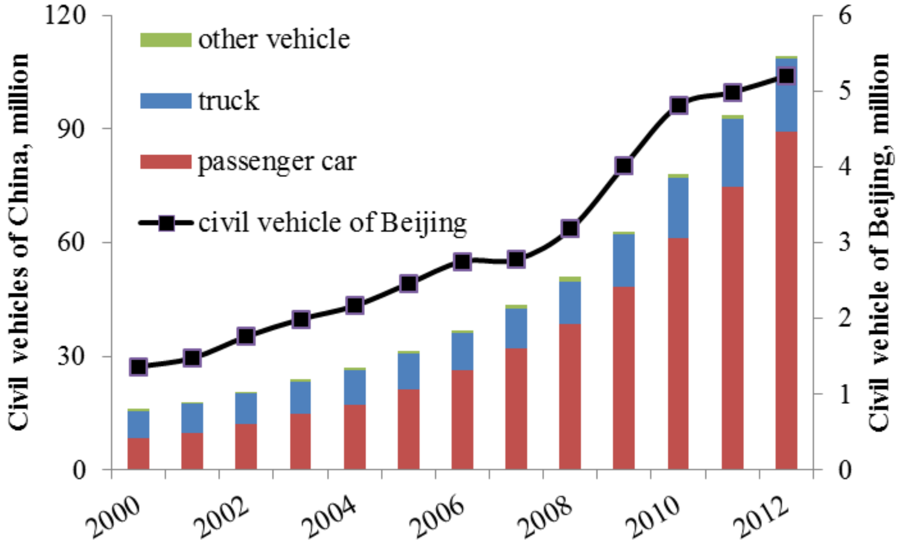

Fig. S5. The number of civil vehicles in China and Beijing, 2000-2012

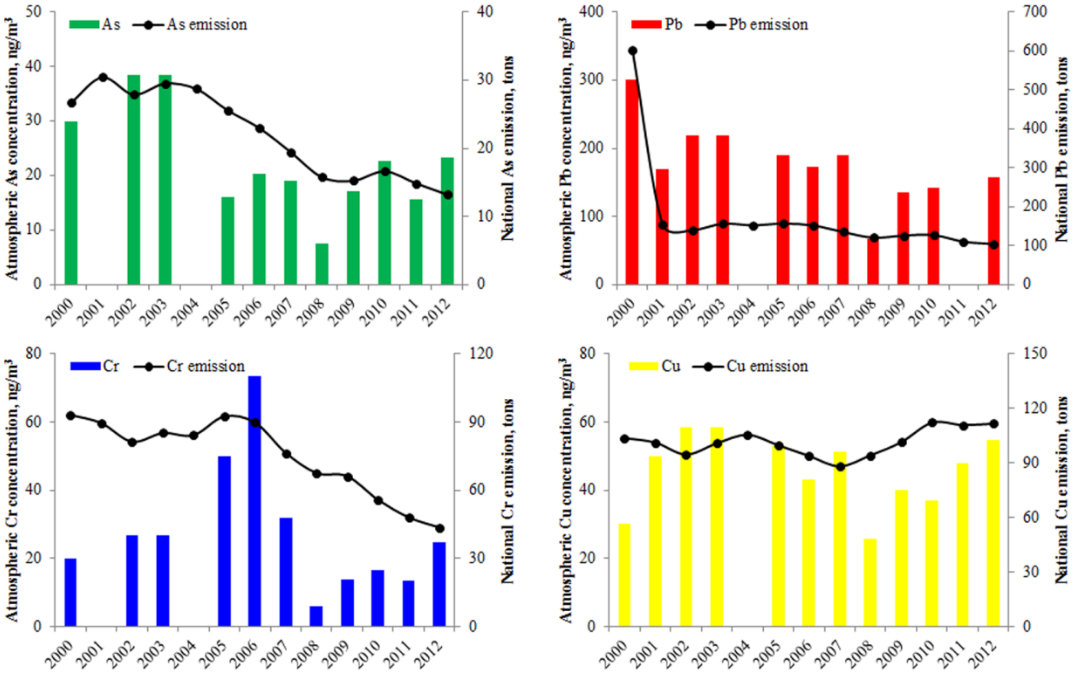

Fig. S6. Comparison between historical HM (As, Cr, Pb, and Cu) emissions and temporal variation of atmospheric concentrations in $PM_{2.5}$ in Beijing during 2000-2012

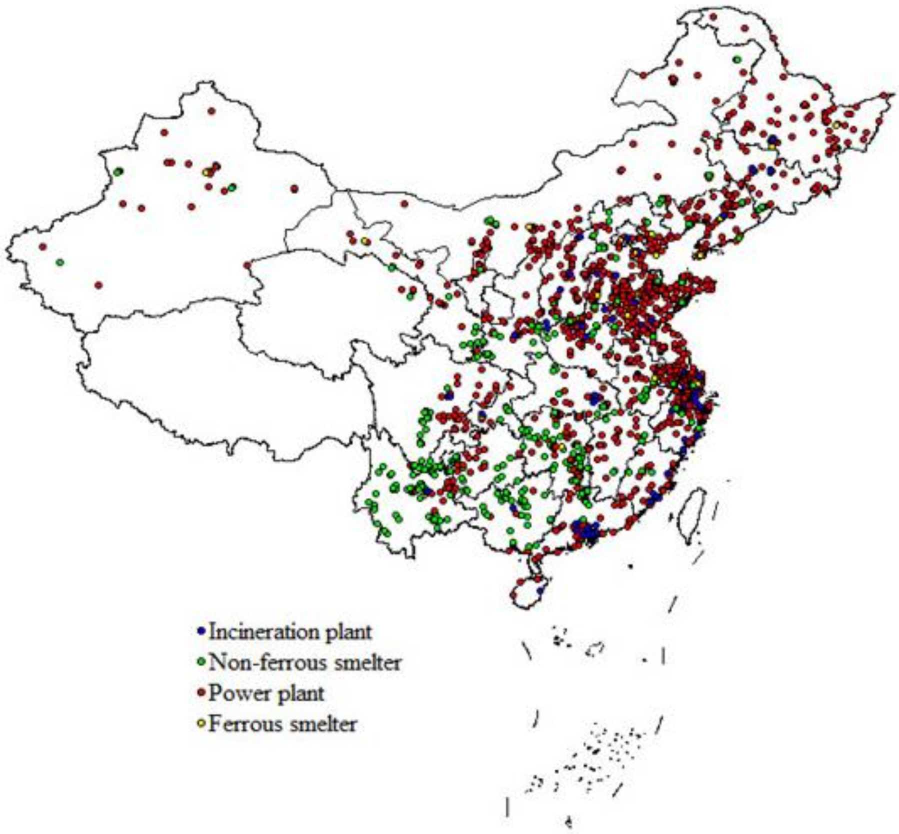

Fig. S7 The distribution of point sources in China

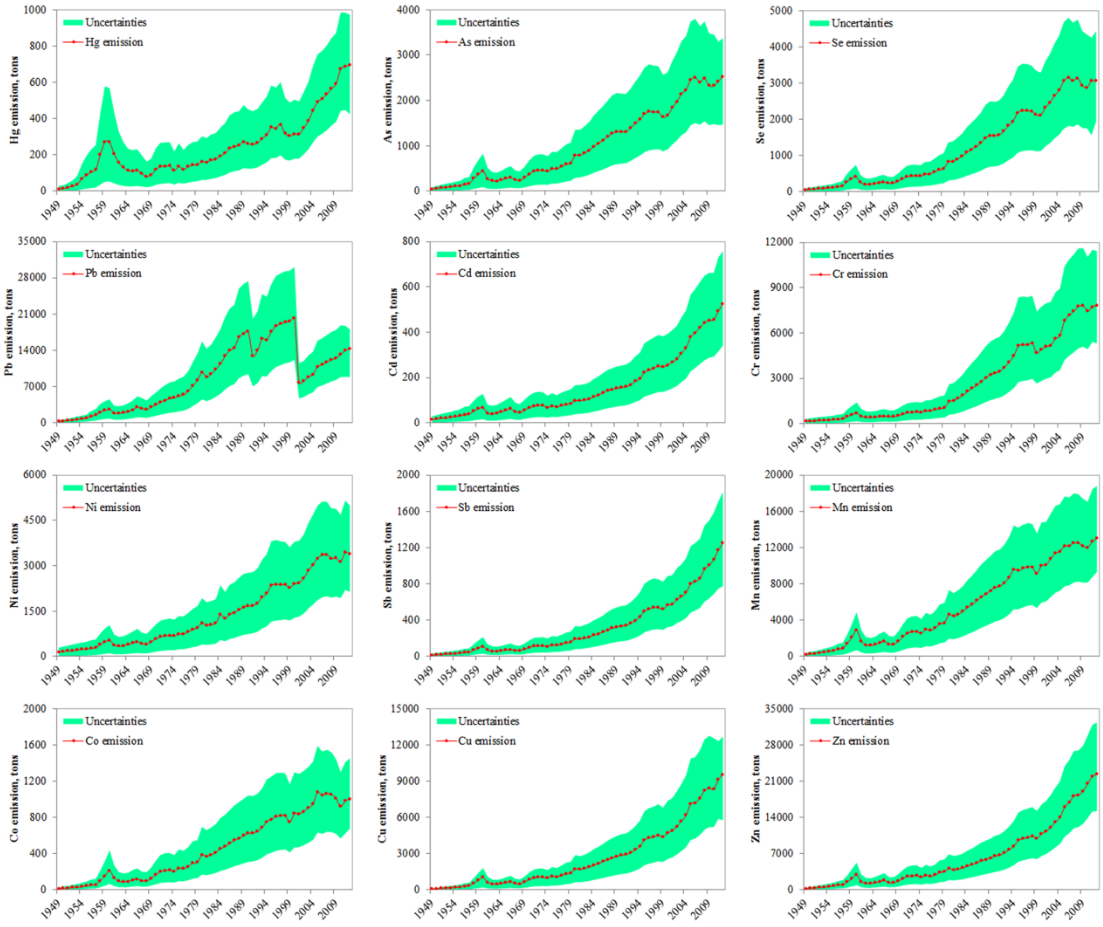

Fig. S8. The uncertainty bounds for China's anthropogenic atmospheric emissions of twelve HMs during 1949 to 2012

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