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Review

Atmospheric heavy metals and Arsenic in China: Situation, sources and control policies

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HIGHLIGHTS

- Atmospheric heavy metals of 44 cities in China were reviewed, the pollution of Cr, As and Cd were rather serious.
- Research on emission characteristics and sources of atmospheric heavy metals in China were reviewed.
- Coal burning, iron and steel industry and vehicle emission are important sources in China.
- Control policies and effects in China were reviewed, and further works for atmospheric heavy metals control were suggested.

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ABSTRACT

In recent years, heavy metal pollution accidents were reported frequently in China. The atmospheric heavy metal pollution is drawing all aspects of attention. This paper summarizes the recent research results from our studies and previous studies in recent years in China. The level, temporal variation, seasonal variation and size distribution of the heavy metals of atmospheric Lead(Pb), Vanadium(V), Manganese(Mn), Nickel(Ni), Chromium(Cr), Cadmium(Cd), Copper(Cu), Zinc(Zn) and Arsenic(As) were characterized in China. The emission characteristics and sources of atmospheric heavy metals and As in China were reviewed. Coal burning, iron and steel industry and vehicle emission are important sources in China. Control policies and effects in China were reviewed including emission standards, ambient air quality standards, phase out of leaded gasoline and so on, and further works for atmospheric heavy metals control were suggested. The comprehensive heavy metals pollution control measures and suggestions were put forward based on the summarization of the development and experience of the atmospheric heavy metal pollution control abroad.

1. Introduction

Atmospheric heavy metals and As can not only result in human dysfunctions and various diseases, but also impose a long term burden on biogeochemical cycling in the ecosystem (Kelly et al., 1996; Nriagu, 1988; Nriagu and Pacyna, 1988). Among them, Cr, As, Ni, Pb, Zn, Cu, V and Cd are carcinogenic, Cd and As are potentially mutagenic and Pb and Hg are fetal toxic (Cheng, 2003; He et al., 2001). Natural emissions (crustal minerals, forest fires and oceans), traffic and industrial emissions (combustion of fossil fuel and industrial metallurgical processes) are the principal sources of atmospheric heavy metals (Park et al., 2008; Fang et al., 2010; Cheung et al., 2011).

Due to the activity of human beings, high anthropogenic emissions of heavy metals enter into the biosphere in China and these emissions far exceed those of other countries and also show an increasing trend (Pacyna and Pacyna, 2001). In recent years, heavy metal pollution accidents have been reported frequently in China (Zhou, 2011). According to the statistics of the Ministry of Environmental Protection (MEP) of China, there were 12 cases of heavy metals accidents, resulting in 4035 blood lead level (BLLs) exceedances and 182 Cd exceedances in 2009. In February 2011, to cope with the situation, the State Council officially approved the 12th Five-Year Plan for comprehensive prevention and control of heavy metals pollution. The atmosphere is one of the major pathways of heavy metal dispersion in the environment; however, little attention has been paid to it in China. Firstly, the atmospheric heavy metals pollution situation is unclear, and routine monitoring of atmospheric heavy metals has not been carried out; Secondly, the emission sources of atmospheric heavy metals is unclear, and the

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first national census of pollution sources carried out in 2007 did not include atmospheric heavy metal emissions; Thirdly, emission standards and ambient air quality standards are not suitable to pollution situation in China, and most emission standards exclude atmospheric heavy metals and the ambient air quality standard only includes Pb; Fourthly, control technologies of atmospheric heavy metals are few, and there are no Best Available Technology (BAT) documents for atmospheric heavy metals in China.

Many studies on atmospheric heavy metals and their sources have been conducted in China and abroad in recent years. To fully understand atmospheric heavy metals in China, the status, sources, distribution characteristics, control technologies and control policies are reviewed in this study.

2. Literature sources

Information from scientific journals, university journals and governmental releases is compiled focusing mainly on Pb,V, As, Cr, Ni, Mn, Cd, Cu and Zn. As is not a heavy metal. Considering its health effect we discussed it with other heavy metals. Hg had been well reviewed recently and therefore didn't include in this study (Fu et al., 2012; Streets et al., 2005) .Three major online databases were used: (1) Science Citation Index Expanded (1900-present, ISI Web of Knowledge); (2) ScienceDirect; (3) China National Knowledge Infrastructure (CNKI). An Endnote database was assembled. We couldn't get heavy metals directly from the paper in some cities and calculated those value from the original data. Considering there are dozens of literature in some cities such as Beijing, Shanghai and Guangzhou et al., we only choose the most recent data with high quality and representativeness data here.

The concentration data collected here are from studies of $PM_{2.5}$, PM_{10}, TSP and dust. This can lead to some confusion on the comparison of concentrations and the comparison between cities according to these data should be done very carefully; however, since studies indicated most heavy metals were enriched in fine particles (Duan et al., 2012; Lu et al., 2012) and most data in this paper are from $PM_{2.5}$ and PM_{10}, it is believed that the data can represent the atmospheric heavy metals situation in China.

3. Results and discussion

3.1. *Characteristics of atmospheric heavy metals and As*

3.1.1. *Levels in China*

A collection of atmospheric heavy metals and As in 44 major cities in China during the last 10 years was obtained, including both urban and background data (Table 1 and Table 2). Concentration of the surveyed heavy metals and As is shown as box plots in Fig.1 and the spatial distribution of heavy metal in China is illustrated in Fig.2.

According to Table 1, the average concentration of atmospheric Pb is 261.0 ± 275.7 ng m^{-3} in China, which fall below the limits of the current national ambient air quality standard (NAAQS) of China (GB3095-1996) of 1000 ng m^{-3}, new NAAQS (GB3095 -2012) and the World Health Organization (WHO) of 500 ng m^{-3} (Table 3). This can be attributed mainly to the nationwide prohibition of leaded gasoline in China since July 1st, 2000. A Previous study by Li et al. (2000) has shown that, in the year of 1998, when leaded gasoline was prohibited in Tianjin, the average annual concentration of Pb was 440 ng m^{-3}, a decrease by 74% compared with 1720 ng m^{-3} .in 1994. However, as Table 1 shows, there are still some cities, such as Zhengzhou, Hangzhou and Shaoguan city which exceed the limits of NAAQS (GB3095 -2012) and WHO of 500 ng m^{-3}. It suggests that other than vehicle emissions, other Pb sources can not be ignored. Coal combustion emissions are also a major source of emissions for anthropogenic Pb sources (Duan et al., 2012).

The concentration of atmospheric V is 17.9 ± 16.5 ng m^{-3} in China (Table 1), much lower than the limit of WHO of 1000 ng m^{-3}. It indicates that the pollution of atmospheric V is not serious in China. There is no limit of V in both current and new NAAQS (GB3095—1996, GB3095 -2012).

The concentration of atmospheric As is 51.0 ± 67.0 ng m^{-3} in China, which is much higher than the limit of the new NAAQS (GB3095 -2012) in China (6 ng m^{-3}) and the limit of WHO (6.6 ng m^{-3}) (Table 3). All cities, except for Lhasa city exceed the limit of 6 ng m^{-3} of the new NAAQS (GB3095 -2012) in China. The highest levels of As were found in Zhengzhou, Yinchuan, Harbin and Foshan. It indicates As pollution is very serious in China.

The concentration of atmospheric Mn is 198.8 ± 363.4 ng m^{-3} in China, which is close to the limit of WHO of 150 ng m^{-3}. The highest levels of Mn were found in Hefei, Shijiazhuang and Zhengzhou, which are much higher than the WHO limit (Table 3). The concentration of atmospheric Ni is 29.0 ± 39.4 ng m^{-3} in China, more than the limit of WHO of 25 ng m^{-3}. The concentrations of atmospheric Ni in some cities such as Urumqi, Harbin and Shijiazhuang are much higher than the limit of WHO. There are no limits of Mn and Ni in both current and new NAAQS (GB3095-1996, GB3095 -2012) (Table 2).

Most atmospheric Cr exists in two forms of inorganic Cr (III) and Cr (VI). Of them, Cr (VI) can do harm to the kidneys and heart, and have a carcinogenic effect. Both new NAAQS (GB3095 -2012) of China and the limit set by the WHO only list out the Cr (VI) concentration limit; however, generally the data reported in the literature is of total Cr and few data on the Cr (VI). The total concentration of atmospheric Cr is 85.7 ± 110.9 ng m^{-3} in China. The limit of China's new NAAQS (GB3095 -2012) and WHO are 0.025 ng m^{-3} and 0.25 ng m^{-3} respectively (Table 3). Cities with the highest concentrations of atmospheric Cr are Shaoguan, Tianjin and Shijiazhuang. The studies on the forms of atmospheric Cr are limited, and atmospheric Cr (VI) require further study.

The concentration of atmospheric Cd is 12.9 ± 19. 6 ng m^{-3} in China, two times higher than the new NAAQS in China and the WHO limit of 5 ng m^{-3} (Table 2), and the most serious levels of pollution occur in cities such as Chongqing, Foshan and Zhengzhou.

The concentration of atmospheric Cu is 117.0 ± 163.3 ng m^{-3} in China and the most serious levels of pollution occurs in some cities such as Huizhou, Shaoguan and Zhengzhou. The concentration of atmospheric Zn is 424.5 ± 336.6 ng m^{-3} in China.

3.1.2. *Size distribution*

Knowledge of the size distribution of atmospheric heavy metals is not only vital in understanding its effects on human health, as well as its sources and transformation processes during atmospheric transport, but also for estimating the dry deposition of the aerosol (Duan et al., 2007; Tan et al., 2009). However, few studies have focused on fine size distributions of atmospheric heavy metals in China (Duan et al., 2012; Lu et al., 2012). Studies showed that Pb, Cd, As and Zn show mostly in accumulation mode; V, Mn and Cu exist mostly in both coarse and accumulation modes; and Ni and Cr exist in all of the three modes. By estimation, the ratios of atmospheric heavy metals in $PM_{2.5}$ to those in PM_{10} in Beijing, China ranked as Pb (88.5%) > Cd (81.8%) > Zn (81.5%) > As (77.1%) > Cu (75.9%) > Cr (71.7%) > Ni (67.9%) > Mn (63.3%) > V (46.8%) (Fig. 3). If left to suspend in the atmosphere for a long time, heavy metal bonded $PM_{2.5}$ can lead to regional pollution by long-range transportation, and enhance the dangers to human health. On the other hand, the activity of heavy metals is affected by particle size. Some studies suggest that the activity of Cd and As increases as the particle size reduces, and Pb and Cr activity is stable in particle of different sizes (Lu et al., 2012).

Table 1
Heavy metals and As in aerosol in China (ng m^{-3}).

	Pb	V	As	Mn	Ni	Cr	Cd	Zn	Cu
Beijing	195.0	25.0	15.0	75.0	20.0	45.0	50.0	295.0	60.0
Hefei	199.0			555.0	38.7	74.3		506.0	121.0
Chongqing	320.0	43.0	37.0	147.0	30.0	147.0	63.0	593.0	53.0
Guangzhou	417.3	19.7	39.2	134.6		53.6	10.4	1220.0	173.6
Nanning	184.3		22.7			60.3	4.7	366.6	22.1
Guiyang	68							26	65
Shijiazhuang	462.0	78.8		577.0	73.0	321.0		1656.0	162.0
Harbin	200.0	10.0	120.0	100.0	80.0	90.0		460.0	90.0
Zhengzhou	1572.0		185.2	781.4	40.6	128.4	47.4		337.0
Wuhan	415.6	11.5	46.9	155.6	6.5	14.0	9.0	604.4	36.9
Changsha	92.5			33.3	38.9			171.2	138.9
Nanjing	190.0		85.0	225.0				415.0	
Nanchang	237.1		19.4	7.7	15.4	11.6		1.1	2.1
Shenyang	346.0		30.2	40.1	26.9	35.5	1.9	388.0	56.4
Hohhot	248.0	13.0		186.0	20.0			452.0	56.0
Yinchuan	143.3		200.0	107.5				190.0	
Jinan	76.5	18.8	19.9	85.6	47.3	57.9		350.8	105.2
Shanghai	108.5	10.3	30.8	60.3	10.0	27.3	2.9	418.5	35.5
Xi’an	230.5		10.6	687.0		167.3		421.5	95.0
Taiyuan	106.8	7.4	38.4	105.3	39.9	69.9		363.1	31.5
Chengdu	182.2		5.9	87.1	3.7	11.3	6.6	387.8	29.2
Taipei	66.5			15.5		328.0		125.5	24.5
Tianjin	291.0	12.0		220.0	25.0	352.0			487.0
Urumqi	67.1			76.9	213.6		3.2		215.3
Lasa	37.0	4.8	1.8	27.0	7.2	19.0	0.5	81.0	9.1
Hangzhou	370.0	20.0	120.0	130.0	20.0	20.0	10.0	550.0	130.0
Hongkong	76.2	4.2	3.9	13.0	5.9	3.7		0.3	21.3
Dalian	193.0		12.3	41.0	7.0	26.4	1.6	189.0	19.6
Qingdao	166.0	30.7		245.2	15.3		2.5	280.3	32.4
Himalayas	2.9	1.0	1.0	3.6	1.3	0.8		5.3	4.8
Foshan	765.3	43.7	96.9	170.9			60.5	1076.0	192.4
Shenzhen	291.2	10.2	28.6	98.3		24.1	13.4	419.0	275.0
Jinzhou	264.0		29.3	49.9	18.3	35.3	3.4	153.0	25.2
Fushun	218.0		12.1	40.6	5.7	22.3	2.3	127.0	21.2
Anshan	376.0		36.9	47.4	11.4	40.8	5.9	509.0	73.3
Zhaoqing	216.2	15.4	31.8	–	–	–	–	432.1	60.6
Shaoguan	960.0	10.0	10.0	200.0	40.0	430.0		790.0	360.0
Hengyang	381.7	5.3	–	43.3	–	–	–	268.3	30.6
Xiamen	119.0	15.0	7.0	57.0	9.0	90.0	9.0	508.0	19.0
Huizhou	203.6	19.1	16.3	76.4	–	51.8	4.0	830.0	884.0
Fuzhou	39.6	3.7	22.5	47.5	4.2	15.7	–	281.2	179.7
Lanzhou	102.7	–	–	–	9.6	29.6	0.305	290.9	84.0
Yulin	23.4	23.9	–	62.0	13.7	25.0	0.4	47.8	9.9
Panzhihua	50	0	0.03	370	20	80	0	260	30
Mean	261.0	17.9	51.0	198.8	29.0	85.7	13.2	424.5	117.0
STD	275.7	16.5	67.0	363.4	39.4	110.9	19.7	336.6	163.3

3.1.3. *The seasonal variation*

Due to meteorological conditions, sources and transportation, the seasonal variation of the atmospheric heavy metals changes significantly. The concentrations of atmospheric heavy metals were generally high in winter and low in the summer (Duan et al., 2012). The low concentrations in summer were mainly due to the high temperature, rainfall and the relatively strong diffusion capacity. The high concentrations were found in winter, which suggested the increased emissions and unfavorable meteorological conditions in winter time. The consumption of coal increases in winter. The concentration of atmospheric particulate of As, Se, Mo, Cd during the winter heating period were higher than those in no-heating period by more than 2 times,due to the enhancement of combustion sources (Yang et al., 2003). Studies on background atmosphere in remote inner Asia demonstrated that high heavy metal concentrations occur in the summer but with greater EF (enrichment factors) values in the winter (Wu et al., 2009).

On the contrary, atmospheric deposition fluxes of heavy metals tended to be higher in the summer than in the winter. Studies shows atmospheric deposition of Cr, Cu and Zn exhibited statistically significant seasonal differences in the Pearl River Delta, China, and the mean atmospheric deposition of these metals was significantly elevated compared with those reported in North American and Europe (Wong et al., 2003).

3.2. *Emission characteristics and sources of atmospheric heavy metals and As in China*

Studies have shown that atmospheric heavy metals can from varieties of sources, and the sources may be very different for different heavy metals (Table 4). Though the pollution is serious in China, atmospheric heavy metals were not included in the first national census of pollution sources in China carried out in 2007. However, in recent years, researchers have carried out a lot of investigations on heavy metal emissions from anthropogenic sources in China, and the emission volume for some heavy metals were estimated.

Anthropogenic Hg emission has increased from 552.2 tons in 1995 to 695.6 tons in 2003, with an average annual growth rate of 2.9% in China (Streets et al., 2005). Studies (Tian et al., 2012a, 2012b, 2010) have estimated that Cd, Cr and Pb from coal-burning increased rapidly from 31.14 tons, 1019.07 tons and 2671.73 tons

Table 2
Summary of atmospheric particulate heavy metals experiment in China.

City	Sampling site	References	Particle size	Sampling period	Sampling numbers	Analysis
Beijing	Tsinghua university (Urban)	(Zhao, 2010)	$PM_{2.5}$	2005.3–2006.2	94	ICP/MS
Hefei	Urban	(Chen et al., 2001)	Dust	One year	–	X-ray fluorescence
Chongqing	Jiangbei(Urban)	(Zhao, 2010)	$PM_{2.5}$	2005.3–2006.2	113	ICP/MS
Guangzhou	Tianhe district	Not published	$PM_{2.5}$	2010.6–2010.12	87	ICP/MS
Nanning	4 Urban,industry, background	(Tang, 2009)	TSP	–	60	HG-AFS, ICP-AES
Guiyang	Urban	(Ji et al., 2006a)	TSP	–	6	ICP/MS
Shijiazhuang	8 sampling sites	(Wang, 2004)	PM_{10}	2000.1–2001.1	–	X-ray fluorescence
Harbin	Urban	(Yuan et al., 2009)	PM_{10}	2005.8–2006.4	105	ICP/AES
Zhengzhou	Urban	(Li et al., 2011)	TSP	2005.8–2005.10	90	ICP/AES
Wuhan	Urban, industrial	(Lv et al., 2006)	PM_{10}	2003.9–2004.9	119	ICP-AES and ICP-MS
Changsha	rural	(Li et al., 2010)	PM_{10}	2008.7–2008.10	60	X-ray fluorescence
Nanjing	Urban, Rural	(Fan et al., 2005)	$PM_{2.5}$	2002.12–2003.10	40	X-ray fluorescence
Nanchang	Urban	(Peng et al., 2009)	$PM_{2.5}$	2007.7–2007.8	60	ICP/AES
Shenyang	4,Urban	(Yu et al., 2008)	$PM_{2.5}$	2007.10	120	ICP/MS
Hohhot	6 sampling sites	(Li and Lu, 2010)	PM_{10}	–	60	ICP/AES
Jinan	4 Urban	(Hao, 2005)	$PM_{2.5}$	2004.3–2004.4	76	ICP/MS
Shanghai	2 Urban,2 Rural	(Chen et al., 2008)	$PM_{2.5}$	2004.3–2005.3	52	ICP/MS
Xi'an	Urban	(Han et al., 2006)	dust	1998,1999,2000	65	Atomic Absorption Spectroscopy
Taiyuan	Urban	(Liu, 2006)	TSP	2004.9–2004.12	–	X-ray fluorescence
Chengdu	Urban	(Wang et al., 2010)	$PM_{2.5}$	2009.4–2009.5	30	X-ray fluorescence
Taipei	Suburban,rural	(Fang et al., 1999)	$PM_{2.5}$,PM_{10}	1998.6–1998.8	40	Flame atomic absorption spectrophotometer
Tianjin	7 sampling site	(Ji et al., 2006b)	PM_{10}	2000.12–2001.5	210	X-ray fluorescence
urumqi	Urban,industry, Rural	(Sun, 2009)	$PM_{2.5}$,PM_{10}	2007.7–2008.5	40	Atomic Absorption Spectroscopy
Lhasa	Jinzhu West Road (Urban)	(Cong et al., 2011)	PM_{10}	2007.9–2008.8	59	ICP-MS
Hangzhou		(Cao et al., 2009)	PM_{10}	2001.9–2002.8	176	PIXE,AES
Hongkong	Urban,industry, background	(Ho et al., 2003)	$PM_{2.5}$,PM_{10}	2000.11–2001.2	70	ICP-MS
Dalian	Urban	(Yu et al., 2008)	TSP, $PM_{2.5}$,PM_{10}			
Qingdao	Urban	(Chen et al., 2004)	TSP	2001.6–2005.5	23	ICP-OES
Himalayas	Background			2006.6–2006.12	7	ICP/MS
Foshan	Urban	Not published	$PM_{2.5}$	2010.6–2010.12	97	ICP/MS
Shenzhen	Rural	Not published	$PM_{2.5}$	2010.6–2010.12	63	ICP/MS
Jinzhou	4,Urban	(Yu et al., 2008)	$PM_{2.5}$	2007.10	120	ICP/MS
Zhaoqing	Rural	(Yang et al., 2009)	$PM_{2.5}$	2006.6–2006.12	30	ICP/MS
Shaoguan	Urban	(Luo, 2006)	TSP	2004.7–2005.5	115	ICP/MS
Hengyang	Urban	(Xie et al., 2002)	$PM_{2.0}$	2001.7–2002.1	7	ICP/AES
Xiamen	Urban	(Zhuang, 2007)	PM_{10}	2004.12–2005.7	25	WD·XRF
huizhou	Rural	Not published	$PM_{2.5}$	2010.6–2010.12	89	ICP/MS
Fuzhou	Urban	(Xu et al., 2012)	$PM_{2.5}$	2007.1–2008.3	40	PIXE analysis
Lanzhou	Urban	(Lin et al., 2012)	$PM_{2.5}$	2010.7–2011.1	8	Atomic Absorption Spectroscopy
Yulin	Zhenbeitai	(Arimoto et al., 2004)	$PM_{2.5}$	2001.3	23	ICP/MS
Panzhihua	Traffic and residential area	(Xue et al., 2010)	PM_{10}	2007.2–2007.8	258	ICP/MS

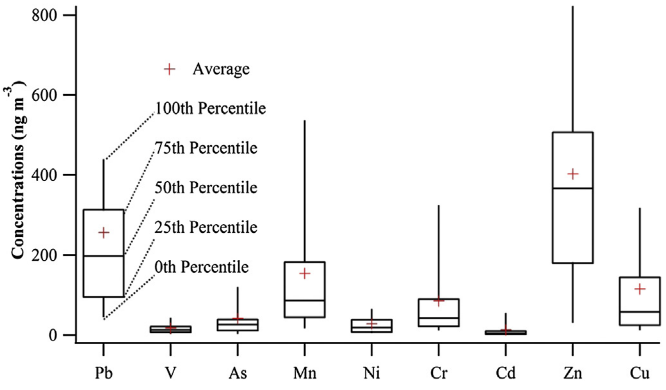

Fig. 1. Box plots of heavy metal and As concentrations in China. The 0th, 25th, 50th, 75th, 100th and average value are indicated.

in 1980 to 261.52 tons, 8593.35 tons and 12,561.77 tons in 2008, respectively; As from coal-burning increased rapidly from 635.57 tons in 1980–2205.50 tons in 2007 (Tian et al., 2010); anthropogenic atmospheric Ni emissions increased rapidly from 1096.07 tons in 1980–3933.7 in 2009 tons with an average annual growth rate of 4.5%(Tian et al., 2012b). Combustion of fossil fuels are the main anthropogenic V emission accounting for 95.5% (Oil:85.0%, Coal:10.5%). Since 1978 anthropogenic V emission have increased 4 times to 31,800 tons in 2010 in China (Zheng and Teng, 2012). In addition to the coal-fired sources, heavy metal emissions from the iron and steel industry are still important. According to the Best Available Techniques reference document on the Production of Iron and Steel of the European Commission (http://eippcb.jrc.ec.europa.eu/reference/), in iron and steel industries, in particular, the sintering process will emit lots of Pb, Hg, Zn and other heavy metal pollutants, and other processes such as ironmaking and steelmaking also emit fugitive dust containing high concentration of heavy metals. Lv et al. (2006) carried out source apportionment of

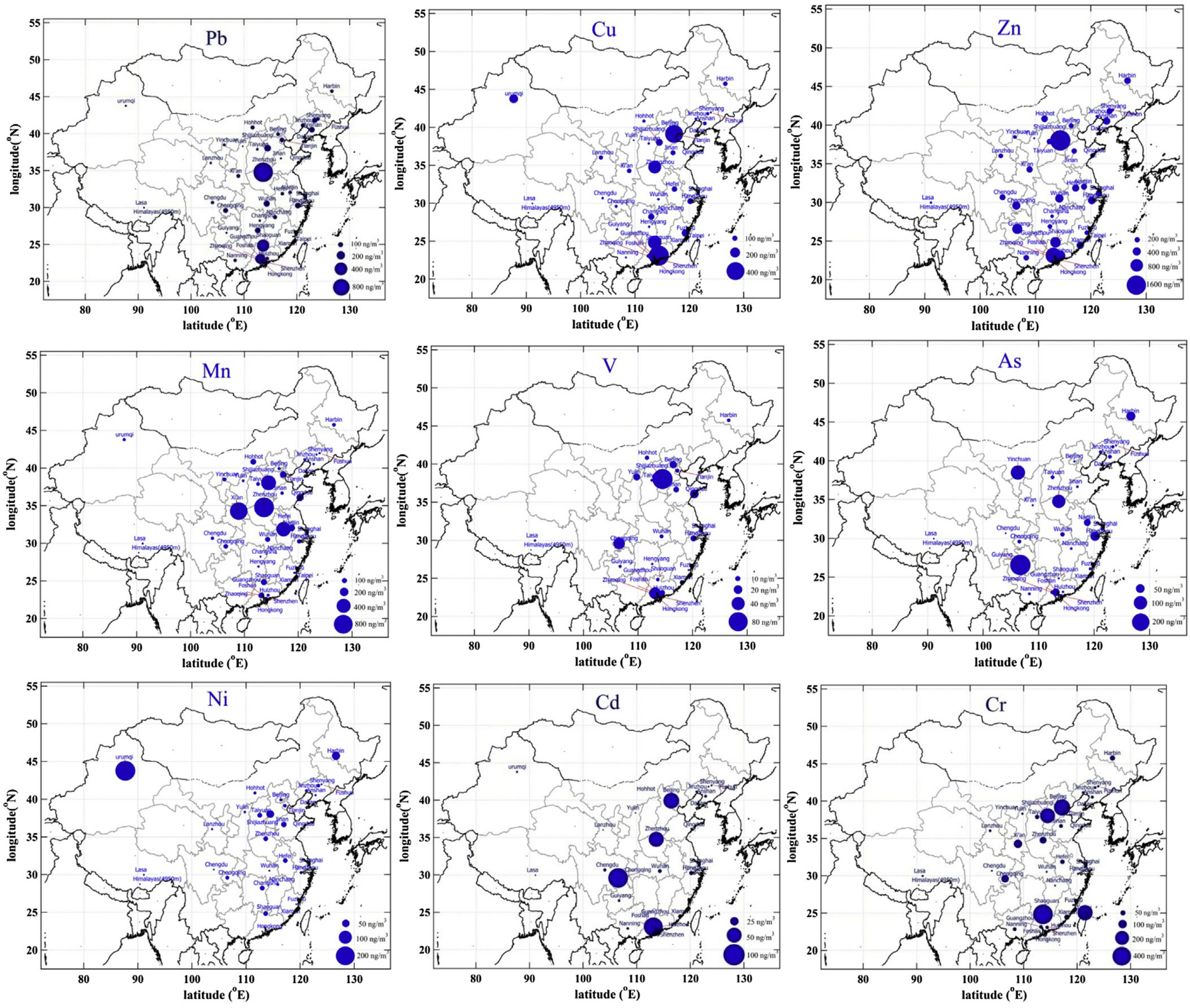

Fig. 2. The distribution of heavy metals and As in China.

atmospheric heavy metals in Wuhan. It indicated that smelting and iron and steel industries are the main atmospheric sources of Pb and Cd, As and Cr in the industrial zone, and the pollutants will affect the air quality in urban Wuhan city by transportation. As the largest producer of steel in the world, the production of crude steel in China has reached 626.7 million tons in 2010, accounting for 44 percent of world production. Iron and steel enterprises in China in 2008 has amounted to more than 8000, and most enterprises are located in densely populated peri-urban areas, so atmospheric heavy metals pollution in urban cities caused by iron and steel industries should be seriously considered in China.

Vehicle emission is another important source of urban atmospheric heavy metals pollution, and the heavy metals can be from six sources (Guo et al., 2008; Harrison et al., 2003): 1) direct emission of Pb, Zn, Cu and Cd bounded particles from exhaust; 2) reentrainment dust enriched with Pb, Zn and Cu by traffic; 3) fuel additives containing Pb and Mn; 4) lubricating oil additives containing Zn and Cd; 5) emissions of Zn, Cd, Pb and Cu from tyre and brake wearing; 6) wearing and corrosion of auto parts such as Zn from anti-corrosion galvanized automobile sheet.

Other possible sources of atmospheric heavy metals can also include the cement industry, the ceramic industry, waste

Table 3
Limits for heavy metals and As in China, WHO and other regions and countries (ng m^{-3}).

	Hg	Pb	V	As	Mn	Ni	Cr(VI)	Cd	Year
NAAQS GB3095-1996		1000							1996
NAAQS GB3095-2012	50	500		6			0.025	5	2012
WHO	1000	500	1000	6.6[a]	150	25[a]	0.25[a]	5	2000
EU(DIRECTIVE 2004/107/EC)				6		20		5	2004
India		500		6		20			2009

[a] concentrations corresponding to an excess lifetime risk of 1:100,000.

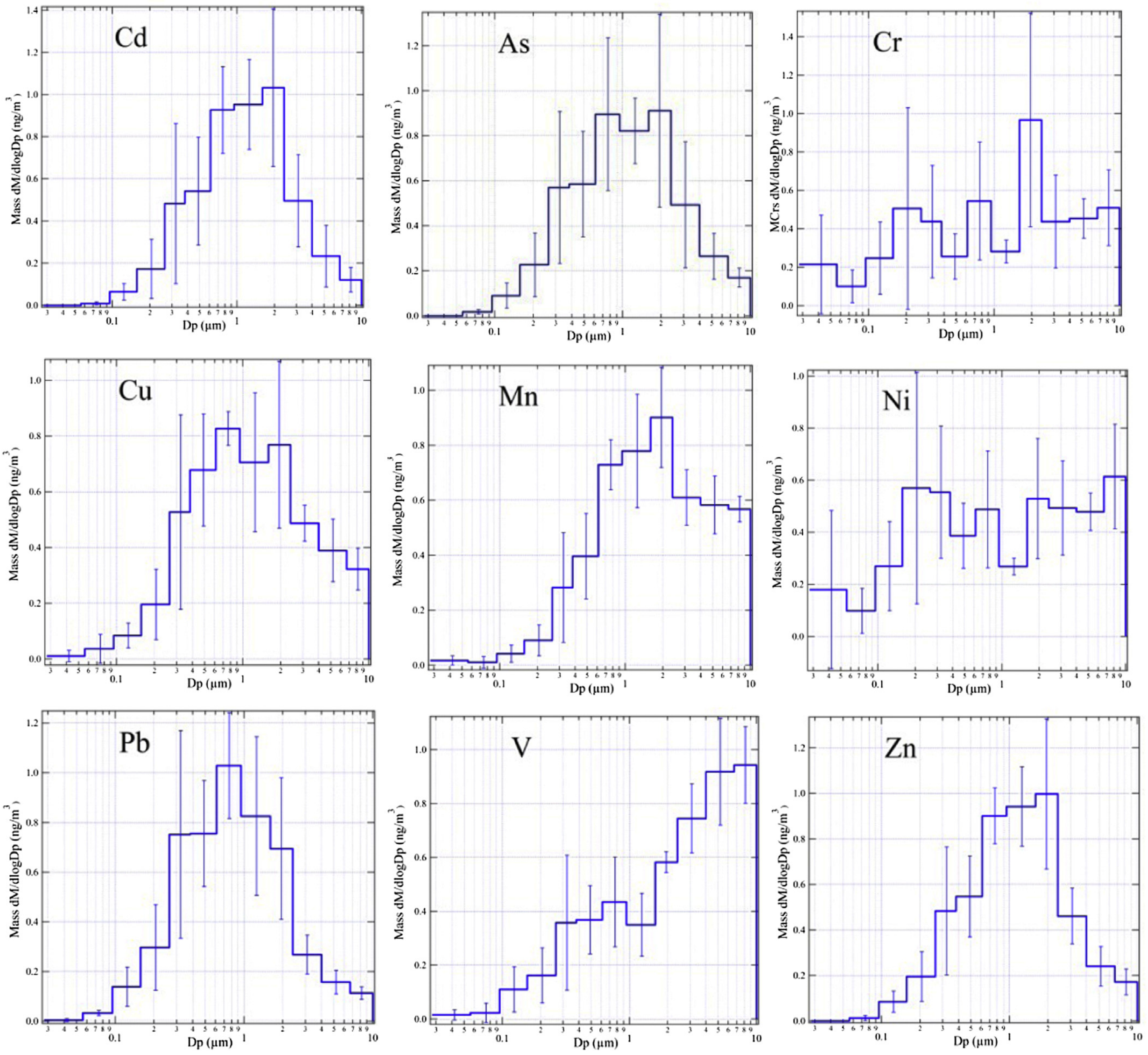

Fig. 3. Size distribution of atmospheric heavy metals and As in Beijing (The error bars represent one standard deviation for the four sets of samples.).

incineration, and nonferrous metal smelting etc; however, the studies on atmospheric heavy metal inventories of these sectors are still very limited in China.

3.3. *Control situations and prospects of atmospheric heavy metals in China*

In 1998, under the framework of the 1979 Geneva Convention on Long-Range Transboundary Air Pollution (CLRTAP), UNECE signed the Aarhus Protocol on Heavy Metals. It targets three particularly harmful metals: Cd, Pb and Hg. The WHO published Guidelines for Air Quality in 2000, which determined the guideline limits of the major heavy metals such as Hg, Pb, V, As, Mn, Ni, Cr(VI) and Cd. Thereafter, many countries began to strengthen the control of atmospheric heavy metals pollution, and amended ambient air quality standards to incorporate heavy metals. In 2004, the EU issued DIRECTIVE 2004/107/EC, which relating to As, Cd, Hg, Ni and polycyclic aromatic hydrocarbons in ambient air, and determined their target values for As, Cd, Ni and benzo(a)pyrene in 2012; In 2009, India amended its ambient air quality standards, and the limits of As and Ni were added, and Pb tightened.

Control of atmospheric heavy metals has not been paid sufficient attention by the government of China, and the main control measurements are emission standards and ambient air quality standards. The current ambient air quality standard was promulgated in 2000, and only Pb included in the standard limits. Before 2008, atmospheric heavy metals including Cr, Pb, Hg, Cd, B, Ni and Tin have only been included in emission standards of integrated emission standards of air pollutants (GB16297-1996). However, since the limits of GB16297-1996 were not based on industry sectors and were rather relaxed, and the monitoring capacity of local environmental protection departments was weak, atmospheric heavy metals pollution was not well controlled except for Pb from vehicle emission.

Table 4
Major sources of some heavy metals and As.

Heavy metal	Sources	References
Cu	Vehicle emission	(Xia and Gao, 2011)
	Smelting furnace burning	(Yang et al., 2003)
Pb	Steel, plastics and pigments production	(Li et al., 2012)
	Contaminated soil and other particles re-enter the atmosphere	(Sun et al., 2006)
	Coal-fired boiler and furnace burning	(Tian et al., 2010)
	Lead gasoline	(Yang et al., 2003)
Cd	Steel, plastics and pigments production	(Tian et al., 2010)
	Lubricating oil	(Aucelio et al., 2007)
	Some tire wear	(Hjortenkrans et al., 2007)
Zn	Waste incineration	(Hopke, 1991)
	Vehicle emission and fly ash from coal burning	(Chow et al., 2004)
	Lubricating oil	(Aucelio et al., 2007)
	Steel smelting	(Querol et al., 2006)
	Rubber tire wear	(Hjortenkrans et al., 2007)
	Burning of incinerators, coal-fired boiler, furnace	(Yang et al., 2003)
Ni	Petroleum and coal combustion	(Tian et al., 2012a)
Cr	Coal and oil combustion	(Tian et al., 2010)
V	Natural rock weathering	(Hope, 1994)
	Combustion of fossil fuels such as coal and oil;	(Lin et al., 2005)
	Mining and smelting of vanadium	(Hope, 1997)
Mn	Steel smelting	(Querol et al., 2006)
	Coal combustion	(Kauppinen and Pakkanen, 1990)
	Gasoline antiknock additive	(Loranger and Zayed, 1995)
As	Smelting furnace	(Yang et al., 2003)
	Coal combustion	(Tian et al., 2010)

June 1, 1997, leaded gasoline was first prohibited in eight urban districts of Beijing, and then, the production of leaded gasoline and the use of leaded gasoline were prohibited nationwide on January 1, 2000 and July 1, 2000, respectively. Studies such as (Wang et al., 2003) have shown that after the prohibition of leaded gasoline, Pb pollution in Chinese cities has improved remarkably, and it brought great public health benefits. A research (Yan and Shen, 2006) lasted decades shows that, after implementation of unleaded gasoline in 1997 in Shanghai, the ratios of blood lead levels (BLLs) exceeding 100ug L^{-1} for children of 1–6 years have dropped from 37.8% to 3.9%. Previous studies (He et al., 2009; Wang and Zhang, 2006) reviewed BLLs in children during periods of 1995–2003 and 2004–2007 in China. Studies suggest that the mean BLLs of children in China have decreased from 92.9ug L^{-1} to 80.7ug L^{-1}, and the prevalence of elevated BLLs (>=100ug L^{-1}) has decreased from 33.8% to 23.9% from the period of 1995–2003 to 2004–2007.

In recent years, with the continuous occurrence of heavy metal pollution accidents, increasing emphasis has been put on heavy metal pollution control in China. Since 2010, the MEP of China sped up the revision of the system of air pollutant emission standards, and strengthened the control of heavy metal pollutant emissions. As Table 5 shows, the emission standards on the ceramic industry, nonferrous metallurgy of Pb, Zn, Cu, Ni, V, glass industry and power plant were promulgated, and atmospheric heavy metals were included in. In addition to the emission standards, a new NAAQS (GB3095 -2012) was promulgated in China in 2012, and it will be formally implemented in January 1, 2016. In addition to tightening the Pb limit, for the first time, NAAQS (GB3095 -2012) will be included in the limits of Cd, Hg, As, Cr(VI) in the Appendix as a reference for local governments to set up local ambient air quality standards in due time.

As for emission control technology, atmospheric heavy metals were generally controlled by a dust control measurement in China. Studies have shown that emission control facilities, such as electrostatic precipitators, bag filter, cyclone, wet precipitators, wet desulfurization and denitrification processes are capable of heavy metals removal to some extent (Tian et al., 2012a). In recent years, the consumption of coal increased rapidly in China; however, the emissions of atmospheric heavy metals increased much slower than coal consumption, especially in the power sector, which has benefited from significant progress of flue gas desulfurization and dust removal in power sector since the 11th Five-plan. By 2010, the proportion of the desulfurization for power units had increased from 14% in 2005 to 86%, and high efficiency electrostatic precipitator facilities had been installed universally. A strengthened standard for power plants (GB 13223-2011) was issued in 2011 in China. It is one of the most stringent standards for PM emission, and it will further reduce the emissions of atmospheric heavy metals in the power sector. In the near future, the revision of standards of cement industry, iron and steel industry can be expected, which will further strengthen the control of atmospheric heavy metals emissions in these sectors in China.

It is estimated that there are about 585 thousand industry boils, which consumed about 0.65 billion tons raw coal in China in 2009

Table 5
Limits for heavy metals and As of emission standards in China promulgated after the year of 2010(mg Nm^{-3}).

Emission standard\Limits	Hg	Pb	As	Ni	Cd	Others
Emission standard of pollutants for ceramics industry (GB 25464-2010)		0.5[a]; 0.1[b]		0.5[a]; 0.2[b]	0.5[a]; 0.1[b]	
Emission standards of pollutants for lead and zinc industry (GB 25466-2010)	1[a]; 0.05[b]	10[a] 8[b]				
Emission standard of pollutants for copper, nickel, cobalt industry (GB 25467-2010)	0.012[a]; 0.012[b]; 0.0012[c]	0.7[a] 0.7[b] 0.006[c]	0.5[a] 0.4[b] 0.01[c]	4.3[a] 4.3[b] 0.04[c]		
Emission standard of air pollutants for thermal power plants (GB 13223-2011)	0.03					
Discharge standard of pollutants for Vanadium Industry (GB 26452-2011)	0.7–1.5[a] 0.5–1.0[b] 0.006[c]					
Emission standard of air pollutants for flat glass industry (GB 26453-2011)						Tin 8.5[a] 5[b]

[a] For existing facility.
[b] For new facility.
[c] For enterprise boundary.

(Yu and He, 2012). With backward control technologies, industry boils consume 23.4% total coal consumption; however emit 41.6% total dust in China now. The emission standard for new coal-burning boiler (GB13271-2001) is 80–250 mg m^{-3} for dust emission. However, facing serious PM pollution, on the one hand, governments issue more strict local emission standards for new coal-burning boilers in some large cities, such as Beijing (DB11/139-2007, 10 mg m^{-3}), Shanghai (DB31/387-2007, 80–120 mg m^{-3}), Lanzhou (DB62/1922-2010; 50–120 mg m^{-3}) and Shijiazhuang (DB13/841-2007, 50–100 mg m^{-3}); on the other hand, governments begin to demolish small and medium boilers or switching from coal to gas for boilers. For example, the consumption of nature gas has increased from 0.75 billion (1999) to 3.59 billion (2006), and to 6.90 billion (2011) cubic meters in Beijing, meanwhile the consumption of raw coal has decreased slightly from 26.51 million (1999) to 26.35 million (2010) tons in Beijing. According to the government's plan, the consumption of raw coal will further decrease to 15.0 million and 10.0 million tons in 2015 and 2020 respectively in Beijing. These steps will be much helpful to control heavy metals from industry boils.

3.4. Further works for atmospheric heavy metals control

Though in recent years, lots of research have focused on the atmospheric levels, emission characteristics and inventories of atmospheric heavy metals, and the government has taken measurement to control atmospheric heavy metals by strengthening both NAAQS and emission standards in China, the studies are still insufficient on issues of atmospheric levels of atmospheric heavy metals, particularly for Hg and Cr(VI), the detail inventory of atmospheric heavy metals, and BAT and emission standards for industries. Some further works are suggested as follow: 1) Carry out investigative monitoring for levels of atmospheric heavy metals including Hg and Cr(VI), and make clear the regional characteristics of atmospheric heavy metal pollution in China, providing the basis for risk control; 2) Include atmospheric heavy metals in the national census of pollution sources; Carry out monitoring in industrial sectors to ensure the production and emission amounts of heavy metals; 3) Establish BAT reference documents based on up-to-date technologies, and provide technical support for atmospheric heavy metal control; 4) Strengthen the preparation and amendment of the atmospheric emission standards for heavy metals, and set up an emission standard system for heavy metals; 5) Strengthen the amendment of NAAQS, and gradually included heavy metals affecting human health into basic limits of NAAQS, according to the WHO guideline values and the pollution situation in China.

4. Conclusion

According to literature statistics of atmospheric heavy metals and As of 44 cities over the past decade in China, compared with the WHO guideline values, the pollution of Cr, As and Cd were rather serious, V was much lower than the WHO guideline. Compared with overseas cities, the levels of atmospheric heavy metals in China are much higher than those of developed countries such as the United States and the European Union and slightly lower than those of cities in India and Pakistan. In addition to tightening the Pb limit, for the first time, NAAQS (GB3095 -2012) includes the limits of Cd, Hg, As, Cr(VI) in the Appendix as a reference for local governments to set up local ambient air quality standards. Benefitting from significant progress of flue gas desulfurization and dust removal in the power sector since the 11th Five-Year plan, the emissions of atmospheric heavy metals increased at a much slower rate than coal consumption. Further works such as investigative monitoring in both the industrial sectors and ambient air, establishment of BAT reference documents based on up-to-date technologies, the preparation and amendment of the atmospheric emission standards and the amendment of NAAQS for heavy metals are suggested.

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