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Pollution characteristics of atmospheric dustfall and heavy metals in a typical inland heavy industry city in China

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ABSTRACT

Through field sampling of atmospheric dustfall in regions of Zhuzhou City, China for a period of one year, the deposition fluxes of atmospheric dustfall and five heavy metals contained inside, including Cr, As, Cd, Hg and Pb, were analyzed. Meanwhile the enrichment factor and index methods were used to analyze the pollution characteristics of heavy metals of atmospheric dustfall in Zhuzhou. The annual deposition flux of atmospheric dustfall in Zhuzhou was 50.79 g/(m^2·year), while the annual deposition fluxes of Cr, As, Cd, Hg and Pb were 9.80, 59.69, 140.09, 0.87 and 1074.91 mg/(m^2·year), respectively. The pollution level of atmospheric dustfall in Zhuzhou was relatively lower compared with most other cities in China, but the deposition fluxes of As, Cd, Hg and Pb in atmospheric dustfall in Zhuzhou were much higher than that in most cities and regions around the world. Cd is the typical heavy metal element in atmospheric dustfall in Zhuzhou, and both the enrichment factor and pollution index of Cd were the highest. Cd, Hg, Pb and As in atmospheric dustfall were mainly from human activities. According to the single-factor index, Nemerow index and pollution load index analyses, the atmospheric dustfall in Zhuzhou could easily cause severe heavy metal pollution to urban soil, and the most polluting element was Cd, followed by Pb, As and Hg. Only the pollution level of Cr lay in the safety region and mainly originated from natural sources.

Introduction

Atmospheric dustfall refers to the particles that settle on the ground by gravity under natural conditions. It is one of the most complicated and harmful pollutants in the atmosphere, not only a harmful substance, but a carrier and a reaction bed for other pollutants, greatly increasing the potential harm of particulate matter to human beings and organisms (Bermudez et al., 2012; Bi et al., 2006; Lee et al., 2005). Heavy metals characterized by enrichment, toxicity and persistence are released from various industrial activities such as mining, smelting and processing (Dragović et al., 2008; Zhang et al., 2012). Their long-term existence in the dustfall has posed a great threat to the environment (Nriagu, 1988; Wong et al.,

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2003). Atmospheric dustfall is one of the most important exogenous factors for the input and enrichment of heavy metals into the ecosystem, especially for the soil subsystem (Fernández-Olmo et al., 2015; Shi et al., 2012; Theodosi et al., 2013). The precipitation of dust is closely related to dust source, collection method, and sampling position and so on (Bao et al., 2012). The source of atmospheric dustfall can be divided into natural source, anthropogenic source and mixed source. The identification of different sources of pollution and their contribution rates can be classified into two categories: source model and receptor model (Manoli et al., 2004). Over the past decades, the studies of atmospheric dustfall have received widespread attention and have been widely conducted in urban or rural areas at home and abroad (Cao et al., 2003; Fernández-Olmo et al., 2015; Pan and Wang, 2015; Soriano et al., 2012; Zhang and Iwasaka, 2004). Fernández-Olmo et al. (2015) conducted a comparative study of trace element deposition fluxes in urban, industrial, rural, and traffic sites of the Cantabria region (Northern Spain) and demonstrated that the most enriched elements were Cd, Zn, and Cu at all sites, while V, Ni, and Cr were less enriched. Bermudez et al. (2010) analyzed the levels and sources of heavy metals in topsoils from different functional areas in the region of Cordoba (Argentina). Li et al. (2010) identified the pollution sources of dustfall in Tongling City, the results showed that the main sources are the smelting dustfall, mining dustfall and coal combustion dustfall. Yin (2006) studied the distribution of heavy metals in atmospheric dustfall of nine cities (Beijing, Taiyuan and so on) by applying the element enrichment factor method. The results showed that the concentrations of Cd, As, Pb, Zn and Cu in the atmospheric dustfall are much higher than other elements; moreover, large cities and heavy industrial cities are more seriously polluted with heavy metals than small and medium-sized industrial cities.

With the continuous development of urbanization and industrialization in China, human activities (especially industrial activities) have resulted in a significant increase in urban atmospheric dustfall and heavy metal contents. Heavy metals such as Pb, Cd, As and Cr in atmospheric dust are very harmful to the human body, of which Cd and As are carcinogens. Zhuzhou City, belonging to the Chang-Zhu-Tan Metropolitan Region (a group of cities, in which Changsha City, Zhuzhou City and Xiangtan City are the core cities), is the second largest city in Hunan province, situated in central mainland China with local weather greatly influenced by the prevalence of subtropical humid monsoon. Zhuzhou is a typical heavy industry base and one of the most important transportation hubs in inland China, and has been undergoing accelerated industrialization and urbanization over the past decades which also led to severe air pollution. Therefore, in recent years, many scientists have chosen Zhuzhou as a typical inland research area to study its environmental pollution status, but so far, the researches on Zhuzhou are limited to the analysis of the heavy metal pollution in the soil and water; the research into the atmosphere is very few and the current studies on the distribution characteristics of heavy metals in atmospheric dustfall of China mainly focus on mega-cities like Beijing and Shanghai. Therefore, it is of importance to conduct field measurements in inland city like Zhuzhou on atmospheric heavy metals pollution, and studying the composition, distribution and transmission of atmospheric dustfall, especially the quantity and potentially hazard of heavy metal elements, is of great significance to the study of urban air pollution and human health in Zhuzhou. Based on the regional environmental situation analysis, this paper systematically studied atmospheric dustfall fluxes and typical heavy metal pollutions in Zhuzhou City for a long period of one year, and provided scientific basis for the establishment and improvement of municipal dustfall-related environmental pollution control policy in Zhuzhou, especially for the heavy-metal pollution prevention and remediation.

1. Experimental methods

1.1. Sampling site and time

According to different urban functional areas in Zhuzhou, a total of 12 sampling points for collecting atmospheric dustfall were determined (Table 1), representing cultural and educational area, residential area, industrial area, mixed area, typical farmland and other type of functional areas. During the sample collection period, the eastern part of Zhuzhou administrative district had not been developed, so only samples in the western urban area were collected. The results of this experiment could represent the whole situation of

Table 1 – Location of sampling sites in Zhuzhou City.

Site no.	Site name	Coordinates (longitude & latitude)	Notes
S1	Zhuye Hospital	E113°05′43″ N27°53′12″	Industrial area
S2	The No. 4 High School of Zhuzhou	E113°10′00″ N27°52′00″	Cultural and educational area
S3	Tiantai Shanzhuang Hotel	E113°08′05″ N27°49′28″	Mixing area
S4	The No. 12 High School of Zhuzhou	E113°04′16.2″ N27°53′08.2″	Cultural and educational area
S5	Haili Chemical Company	E113°05′03.4″ N27°51′52.5″	Industrial area
S6	The Government of Shifeng District	E113°06′42.8″ N27°52′40.2″	Residential area
S7	The No. 8 High School of Zhuzhou	E113°06′48″ N27°52′10″	Cultural and educational area
S8	Hexi Sewage Treatment Plant	E113°03′2.6″ N27°50′55.3″	Typical farmland
S9	Jianning High School	E113°10′5.9″ N27°48′10.6″	Typical farmland
S10	Hunan Haohua Chemical Company	E113°05′50.6″ N27°52′30.0″	Industrial area
S11	Hunan Jingshi Company	E113°06′5.3″ N27°52′12.9″	Industrial area
S12	Southern Industrial and Trade Park (with Sports Center)	E113°06′5.8″ N27°50′29.1″	Residential area

dustfall in urban Zhuzhou. The measurement campaign was conducted once a month in Zhuzhou from November 2011 to November 2012.

1.2. Sample collection and analysis method

The dust fall collector used for sampling is a cylindrical dust reduction container with a diameter of 15 ± 0.5 cm and a height of 30 cm as specified in the gravimetric method for determination of ambient air dustfall (China National Standards: GB/T 15265-94). Before sampling, the 80-mL ethylene glycol was added to fill the cylinder bottom. The samples of atmospheric dustfall were collected once a month at the end of each month. After collecting, they were transferred to plastic bottle, sealed and then brought back to the laboratory for analysis, at the meantime the information such as sampling point, sampling time and cylinder number are recorded. The collected samples were undergone the process of solid–liquid separation by using mixed fiber microporous membrane. The solid part of filtration were then placed in Teflon digestion tank inside which 6-mL HNO_3, 2-mL H_2O_2 and 0.1-mL HF were added and mixed. The mixed sample was then heated in the oven (185°C) and then reduced to 50 mL. The contents of Cr, As, Cd, Hg, Pb and other elements in the sample were determined by Inductively Coupled Plasma Mass Spectrometry (ICP-MS, Agilent 7700x). With standard addition method, the recovery rate of each element was between 95%–105%, and the relative standard error of repeated tests was less than 2%.

1.3. The calculation method of deposition flux and factor analyses

1.3.1. Deposition fluxes of atmospheric dustfall and heavy metals

The equation for calculating deposition flux of atmospheric dustfall and heavy metal are expressed as follows:

$$Q = M/(S \times d) \times D \tag{1}$$

$$Qt = (W \times Cs + V \times Ci)/(S \times d) \times D \tag{2}$$

where, Q (g/(m^2·year) or g/(m^2·month)) is the deposition flux; S (m^2) is the total area of dust reduction cylinder placed at the sampling site; M (g) is the total amount of deposition; d is the sampling days; D is the day numbers in one month or year; Qt (μg/(m^2·year) or μg/(m^2·month)) is the deposition flux of an element; W (g) is the weight of the dustfall precipitation; Cs (mg/kg) is mass concentration of an element in precipitation part; V (L) is the volume of atmospheric dustfall solution; Ci (μg/L) is the mass concentration of an element in the solution part.

1.3.2. Enrichment factor (EF)

The enrichment factor is used to determine if the elemental concentration is enriched from natural or anthropogenic sources. The EF calculation is expressed as follows:

$$EF = (C_i/C_n)_s/(C_i/C_n)_b \tag{3}$$

where, C_i is the concentration of the examined element i; C_n is the concentration of the reference element (Ti, Al and Fe are commonly used as reference); s and b represent the sample and background value, respectively. If $EF \leq 10$, the element is not enriched and mainly from the crust and other natural sources, while if $EF > 10$, the element is enriched and mainly from anthropogenic pollution.

1.3.3. The index methods

The single-factor pollution index method, the Nemerow's index method and the pollution load index are used to assess the pollution level of heavy metals, as illustrated in The Technical Specification for Soil Environmental Monitoring [HJ/T166-2004] of China, the calculations are expressed as follows:

$$P_i = C_i/S_i \tag{4}$$

where, P_i is the single-factor pollution index value of heavy metal i in atmospheric dustfall; C_i is the measured value of heavy metal i; S_i is the standard value of heavy metal i based on the three-grade classification value in China National Environmental Quality Standard for Soils.

$$NI_i = \sqrt{\left[(Pi_{avg})^2 + (Pi_{max})^2\right]/2} \tag{5}$$

where, NI_i is the Nemerow comprehensive pollution index value of heavy metal i in atmospheric dustfall; Pi_{avg} and Pi_{max} are the average and maximum value of the single-factor pollution index of heavy metal i at the sampling site.

$$\begin{aligned} CF_i &= C_i/C_{oi} \\ PLI &= \sqrt[n]{CF_1 \times CF_2 \times \ldots \times CF_n} \\ PLI_{zone} &= \sqrt[m]{PLI_1 \times PLI_2 \times \ldots \times PLI_m} \end{aligned} \tag{6}$$

where, PLI and PLI_{zone} are the pollution load index of heavy metals in atmospheric dustfall at sampling site and in the region; C_i and C_{oi} (mg/kg) are the measured value and evaluation standard value of the heavy metal i in the atmospheric dustfall; CF_i is the highest pollution coefficient of a single heavy metal element i in the atmospheric dustfall; n is the number of heavy metals in the atmospheric dustfall; m is the number of sampling sites.

The three index values (hereafter abbreviated as PNP) including P_i, NI_i, PLI can be used to divide the pollution levels under same standard classification values, the boundary values are 0.7, 1.0, 2.0, 3.0 respectively for five-level classification as follows in Table 2.

2. Results and discussion

2.1. Deposition fluxes

The annual deposition flux of atmospheric dustfall in Zhuzhou was 50.79 g/(m^2·year) in average and the order of annual

Table 2 – Standard values for pollution grading.

Grade	PNP range	Pollution level
I	PNP ≤ 0.7	Security (clean)
II	0.7 < PNP ≤ 1.0	Warning grade (clean)
III	1.0 < PNP ≤ 2.0	Slight pollution
IV	2.0 < PNP ≤ 3.0	Moderate pollution
V	PNP > 3.0	Severe pollution

PNP were the abbreviations of three index values (P_i, NI_i, PLI).

deposition flux of atmospheric dustfall among 12 sampling sites was S5 > S10 > S11 > S4 > S12 > S1 > S6 > S8 > S9 > S7 > S2 > S3 (Fig. 1). The deposition flux in spring and autumn were higher than that in winter and summer. The peak values of the monthly deposition flux were 5.61 and 5.91 g/(m^2·month) in April and September, respectively (Fig. 2). The April peak may be caused by a large number of fine particles falling to the ground due to the scouring effect of rainwater during the Meiyu season. September was the beginning of the new school year, the high traffic flow resulted in the increase of ground dust and probably formed peak value. In addition, the annual deposition fluxes of Cr, As, Cd, Hg and Pb in atmospheric dustfall were 9.80, 59.69, 140.09, 0.87 and 1074.91 mg/(m^2·year), respectively. Compared with other sites, S5 site had the highest annual deposition flux of five heavy metals. The variation trend of Cd and Pb was basically consistent, which formed two peaks in February and November, the maximum peak values were 33.71 (Cd) and 163.04 (Pb) mg/(m^2·month), and both the minimum values appeared in June, which were 1.44 (Cd) and 23.21 (Pb) mg/(m^2·month). It can be speculated that Cd and Pb in atmospheric dustfall had the same pollution sources. Cd and Pb are the representative elements of nonferrous metallurgy emission, and coal combustion and traffic pollution are also important factors (Liu et al., 2010; Xiao et al., 2008). According to local survey, in November, a number of nonferrous smelters in Zhuzhou increased productions to achieve the annual gross output value, and in February, the traffic volume of motor vehicles during the China's Spring Festival was very high, thus leading to the peak values of Cd and Pb deposition fluxes in November and February, respectively. The monthly deposition fluxes of As were 11.30 mg/(m^2·month) and 12.11 mg/(m^2·month) in November and December, which were significantly higher than the other months. It was mainly related to the emissions of coal-fired processes (especially for heating in winter and higher coal-fired power generation demand in winter). The deposition fluxes of Cr were smaller than other three heavy metals (As, Cd, Pb) and only ranged from 0.65–13.1 mg/(m^2·month). The monthly deposition fluxes trend of Hg was similar to that of the atmospheric dustfall, its peak values in April and September were 143.43 μg/(m^2·month) and 119.74 μg/(m^2·month), respectively.

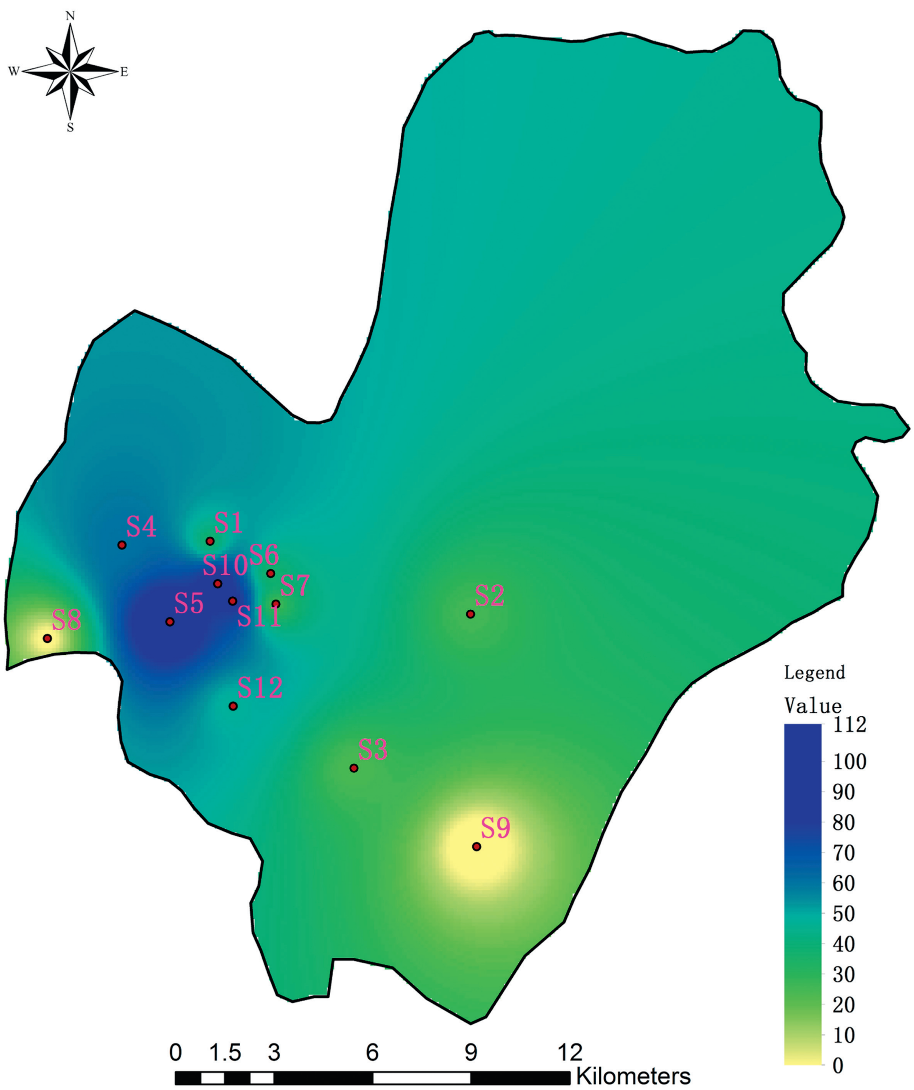

Fig. 1 – Distribution of annual deposition flux of atmospheric dustfall in Zhuzhou City (unit: g/(m^2·year)).

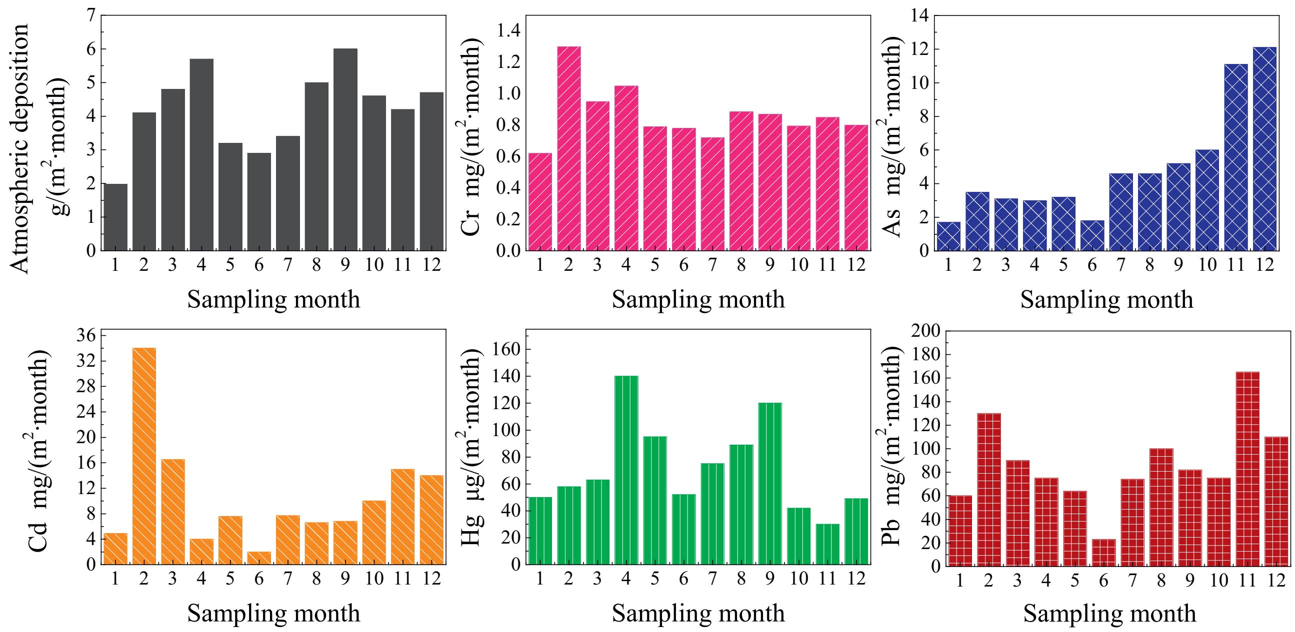

Fig. 2 – Monthly deposition flux of atmospheric dustfall and heavy metals in Zhuzhou.

2.2. Comparison with other cities

In order to assess the pollution levels of atmospheric dustfall in Zhuzhou City, the deposition fluxes of atmospheric dustfall, and its heavy metals in some cities in China were compared (Table 3). The deposition flux of atmospheric dustfall in Zhuzhou city in 2012 was significantly lower than that in other cities such as Anshan (Xing et al., 2010), Shenyang (Lin et al., 2008), and Chaihe Basin (Ge et al., 2012), and much lower than that of road dustfall in Beijing (Y. Tang et al., 2011) and Huhhot (Fan et al., 2011) as well. While there is minor difference in urban atmospheric dustfall deposition of Beijing (Li, 2010) and Urumqi (Liu, 2008) compared with Zhuzhou's. Therefore, the pollution level of atmospheric dustfall in Zhuzhou was relatively lower compared with most other cities reported in China.

The deposition fluxes of heavy metals in atmospheric dustfall in some cities and regions in China and other countries are summarized in Table 4, and indicated that the annual deposition fluxes of As, Cd, Hg and Pb in atmospheric dustfall in Zhuzhou City in 2012 were much higher than that in most cities and regions. Except for Beijing (Cong et al., 2008), the deposition flux of Cd in Zhuzhou was significantly higher than remaining cities and regions in Table 4. It can be seen that Cd is a typical pollution element in atmospheric dustfall in Zhuzhou and should be controlled firstly. Secondly, Pb, Hg and As also need to be strictly controlled due to their much higher levels in urban Zhuzhou. In addition, the pollution level of Cr in Zhuzhou was not serious, significantly lower than that of Harbin (J. Tang et al., 2011) and not much different from that of other cities and regions in China and other countries.

2.3. Factor analyses of heavy metals in atmospheric dustfall

In order to analyze the enrichment characteristics and pollution sources of heavy metals of atmospheric dustfall in Zhuzhou, the enrichment factor (*EF*) method (Fang et al., 2006; Odabasi et al., 2002; Zhang et al., 2014) was used, and Ti was selected as a reference element. The enrichment factors of Cr, As, Cd, Hg and Pb in atmospheric dustfall in Zhuzhou (Fig. 3) demonstrated that the enrichment factor of Cd was the largest in 12 sampling sites in Zhuzhou, with an average value of 2690.61, followed by Hg, Pb and As, with an average value of 268.39, 262.53 and 39.22, respectively, which were significantly higher than 10. It can be seen that Cd, Hg, Pb and

Table 3 – Domestic deposition fluxes of atmospheric dustfall in China (unit: g/(m^2·year)).

Sampling sites	Period	Item of monitoring	Deposition flux	Reference
Zhuzhou	2012	Atmospheric dustfall	50.79	(This study)
Anshan	2008	Atmospheric dustfall	379.2	(Xing et al., 2010)
Shenyang	2006	Atmospheric dustfall	276.6	(Lin et al., 2008)
Beijing	2009	Road dustfall	225.5	(Y. Tang et al., 2011)
	2008–2009	Atmospheric dustfall	58.4	(Li, 2010)
Huhhot	2008–2009	Road dustfall	313.8	(Fan et al., 2011)
Urumqi	2007	Atmospheric dustfall	69.99	(Liu, 2008)
Chaihe Basin	2010–2011	Atmospheric dustfall	339.66	(Ge et al., 2012)

Table 4 – Deposition fluxes of heavy metals in different cities (mg/(m^2·year)).

Sampling sites	Period	Deposition fluxes of heavy metals					References
		Cr	As	Cd	Hg	Pb	
Zhuzhou	2012	9.80	59.69	140.09	0.87	1074.91	This study
Beijing	2005–2006	11.86	2.92	236	0.02	22.00	(Cong et al., 2008)
Changchun	2006–2007	10.67	4.79	0.25	0.03	12.31	(Yang et al., 2009)
Anshan	2008	4.545	0.378	0.026		4.277	(Xing et al., 2010)
Harbin	2008–2009	39.09	7.78	0.67	0.13	52.74	(J. Tang et al., 2011)
Tianjin	2007–2010	9.77	5.51	0.56		31.1	(Pan and Wang, 2015)
Baoding	2007–2010	8.09	8.69	0.98		45.8	(Pan and Wang, 2015)
Tanggu	2007–2010	12.21	3.13	0.50		37.3	(Pan and Wang, 2015)
Belgrade	2002–2006	4.5		0.60		59.5	(Mijić et al., 2010)
Brisbane	2007–2008	1.8	0.97	0.32		5.9	(Huston et al., 2012)
Huelva (urban)	2008–2011	4.4	1.6	0.27		9.3	(Castillo et al., 2013)
Huelva (industrial)	2008–2011	4.9	4.7	0.55		27.4	(Castillo et al., 2013)
Venice	2001–2003	15.7	63.0	33.0		52.0	(Rossini et al., 2010)
Santander	2009–2013	5.2	0.30	0.10		4.5	(Fernández-Olmo et al., 2015)
Maliaño	2012–2013	11.6	0.60	0.20		18.1	(Fernández-Olmo et al., 2015)
Cabezón de la Sal	2012–2013	1.8	0.40	0.24		6.8	(Fernández-Olmo et al., 2015)
Bárcena Mayor	2012–2013	1.3	0.20	0.09		2.5	(Fernández-Olmo et al., 2015)
Tokyo	2001–2002			0.058	0.01	3.395	(Sakata and Marumoto, 2004)

As mainly came from human activities in Zhuzhou. The enrichment factor of Cr did not reach 10 (the average value of 12 sampling points was 3.88, the range was 2.54–4.93), indicating that Cr in Zhuzhou's atmospheric dustfall mainly came from natural sources. In addition, the enrichment factors of As, Cd, Hg and Pb in S5 sampling site were consistently the largest among all the sampling sites and showed that the S5 site was the most affected area by human activities. According to the correlation analysis of Cr, As, Cd, Hg and Pb (Table 5), it can be seen that the correlation coefficient between As, Cd, Hg and Pb were all higher than 92.6%, especially the correlation coefficient between As and Pb was the highest, reaching 99.8%. It indicated that the heavy metals in Zhuzhou's atmospheric dustfall have the same source of pollution, expect for Cr. Therefore, the management and treatment of specific pollution sources could effectively improve the air quality of Zhuzhou in term of heavy metal pollution.

Furthermore, the index methods (the single-factor pollution index method, the Nemerow's index method and the pollution load index) (Wang et al., 2015) were used to analyze pollution degree of heavy metals of atmospheric dustfall in Zhuzhou. The single-factor pollution index value and pollution grading distribution of Cr, As, Cd, Hg and Pb in

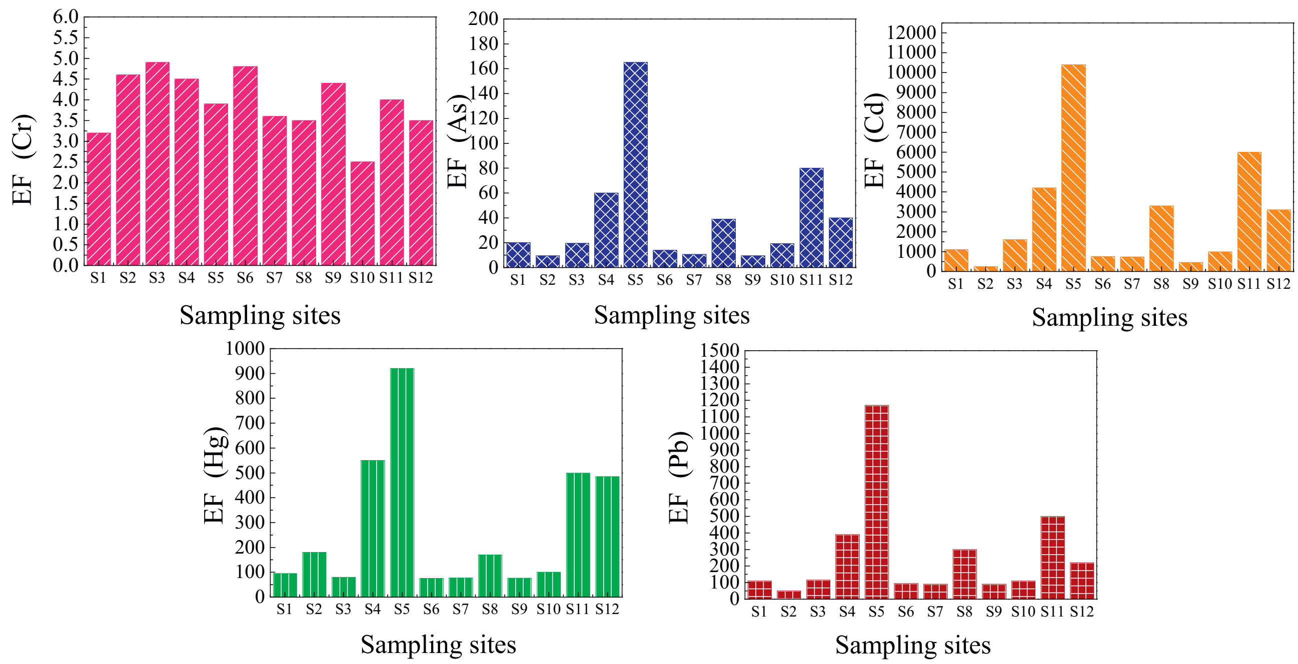

Fig. 3 – Enrichment factors of heavy metals in atmospheric dustfall at 12 sampling sites.

Table 5 – Correlation coefficients between heavy metals.

/	Cr	As	Cd	Hg	Pb
Cr	1				
As	−0.064	1			
Cd	−0.071	0.994	1		
Hg	−0.056	0.938	0.937	1	
Pb	−0.063	0.998	0.991	0.926	1

Table 6 – Single factor pollution index of heavy metals in atmospheric dustfall in each sampling site of Zhuzhou.

Sampling sites	P_{Cr}	P_{As}	P_{Cd}	P_{Hg}	P_{Pb}
S1	0.6	4.9	93.1	3.3	4.7
S2	0.8	2	18	6.3	2.2
S3	1	5.1	143.7	3.4	5.8
S4	0.9	16.1	401.3	21.7	18.8
S5	0.5	31.3	702.4	25.9	40.5
S6	0.9	3.1	60.8	2.4	3.9
S7	0.8	3.3	74.5	3.6	4.5
S8	0.6	8.1	246.3	5.7	11.4
S9	0.7	1.8	32.1	2.5	2.9
S10	0.5	4.4	90.1	3.7	5.5
S11	0.7	20	504.5	17.3	24.2
S12	0.6	10.6	276.7	19	12.2
Average	0.7	9.2	220.3	9.6	11.4

atmospheric dustfall at 12 sampling sites (144 samples in total) in Zhuzhou are shown in Table 6 and Fig. 4, which demonstrated that the most serious pollution element was Cd, the average index value of Cd in the entire city reached 220.3, far greater than the worst Grade-V pollution level; followed by Pb, Hg, As, with the single-factor pollution index of 11.4, 9.6, 9.2, also reaching the worst pollution level (Grade-V). Only the single-factor pollution index of Cr reached the level of security (Grade-I). According to the analyses of all the 144 dustfall samples from 12 sampling sites in Zhuzhou, the pollution indices of Cd were all in Grade-V, and the sample numbers of Pb, As and Hg reaching Grade-V were all more than 50%, which were 77.08%, 65.97% and 55.56%, respectively. Obviously, the heavy metal pollution of atmospheric dustfall

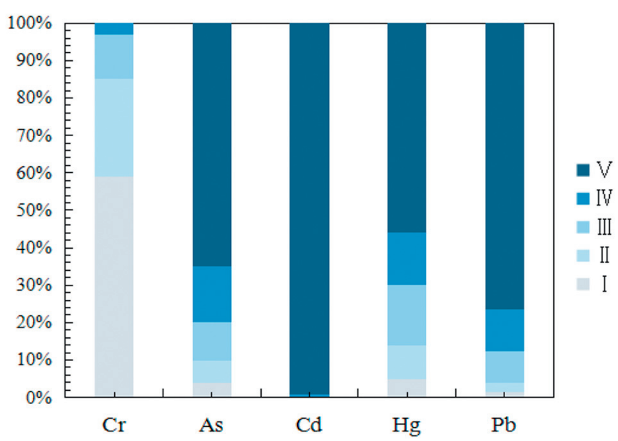

Fig. 4 – Pollution level distribution of heavy metals in atmospheric dustfall in Zhuzhou. (Grade I to IV represent five pollution levels as shown in Table 2).

Table 7 – Nemerow comprehensive pollution index (NI) of heavy metals in atmospheric dustfall in Zhuzhou.

Sampling sites	NI_{Cr}	NI_{As}	NI_{Cd}	NI_{Hg}	NI_{Pb}	Comprehensive pollution index
S1	0.67	9.48	225.50	5.66	7.88	49.84
S2	1.07	4.40	29.23	29.12	3.35	13.43
S3	1.83	6.68	308.74	6.67	8.77	66.54
S4	1.58	41.47	1472.35	53.25	61.94	326.12
S5	0.76	46.89	1121.22	46.68	59.42	254.99
S6	1.58	6.10	103.66	4.70	5.19	24.25
S7	1.04	4.90	143.31	13.59	8.56	34.28
S8	0.66	24.81	377.25	16.17	18.97	87.57
S9	0.92	2.58	69.87	7.22	5.81	17.28
S10	0.84	7.76	167.13	7.40	8.74	38.37
S11	0.96	32.85	970.88	36.81	57.53	219.80
S12	0.84	21.65	505.07	52.77	18.03	119.67
Average	1.06	17.47	457.85	6.77	8.05	98.24

in Zhuzhou was very serious and brought severe heavy metal pollution into soil. The similar evaluation results were obtained by the Nemerow comprehensive index method and the pollution load index method (Tables 7, 8), also showing the atmospheric dustfall in Zhuzhou caused serious heavy metal pollution to the soil. The main heavy metal pollution elements were still Cd; Pb, As and Hg also had very high pollution indices. Only for Cr, the whole city of Zhuzhou (all the sampling sites) was much less polluted by that element and was at a safety grade (Grade-I).

3. Conclusions

(1) The annual deposition flux of atmospheric dustfall in Zhuzhou was 50.79 g/(m^2·year), and the annual deposition fluxes of Cr, As, Cd, Hg and Pb were 9.80, 59.69, 140.09, 0.87 and 1074.91 mg/(m^2·year), respectively. The S5 sampling site (Haili Chemical Industry, in industrial area) had both the highest annual deposition fluxes of atmospheric dustfall and five heavy metals among the 12 sampling sites. The pollution level of atmospheric dustfall in Zhuzhou was lower compared with most other cities in China. But the deposition fluxes of Cd, Pb,

Table 8 – Pollution load index (PLI) of heavy metals in atmospheric dustfall in Zhuzhou.

Sampling sites	PLI_{Cr}	PLI_{As}	PLI_{Cd}	PLI_{Hg}	PLI_{Pb}
S1	0.54	3.19	74.94	2.49	3.86
S2	0.76	1.43	14.46	3.08	1.87
S3	0.91	4.66	100.79	2.75	5.47
S4	0.76	10.21	240.14	13.69	11.51
S5	0.50	29.48	612.00	21.61	34.02
S6	0.73	2.44	51.52	1.81	3.55
S7	0.83	2.79	57.07	2.09	3.88
S8	0.57	5.08	198.29	3.99	9.14
S9	0.63	1.21	21.61	1.54	2.19
S10	0.44	3.71	74.28	2.48	4.99
S11	0.65	14.50	327.92	12.76	17.34
S12	0.62	8.62	225.54	10.57	11.53
PLI_{zone}	0.65	4.73	100.88	4.37	6.35

As and Hg in atmospheric dustfall in Zhuzhou were much higher than that in most cities and regions around the world.

(2) According to the enrichment factor and three types of index methods analyses, the similar results indicated that the atmospheric dustfall in Zhuzhou was easy to cause serious heavy metal pollution to urban soil. Cd was a typical pollution element in atmospheric dustfall in Zhuzhou City and had both the highest enrichment factor and pollution index, therefore should be controlled first. Secondly, Pb, As and Hg in Zhuzhou also need to be strictly controlled due to their high pollution indices. The elements of Cd, Pb, As and Hg in the atmospheric dustfall were mainly from human activities. Only the pollution level of Cr lay in the safety region and mainly originated from natural sources. The heavy metals in Zhuzhou's atmospheric dustfall had the same source of pollution except for Cr. Therefore, the management and treatment of specific pollution sources can effectively improve the air quality of Zhuzhou in term of heavy metal pollution.

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