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Original Research

Differences in the Risk Assessment of Dustfall Heavy Metals between Industrial and Non-Industrial Areas in China

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Abstract

Heavy metal contamination is widespread across China, but the differences of dustfall heavy metals between industrial and non-industrial areas of the country remain incompletely understood. The research areas were firstly divided into industrial and non-industrial ones. The pollution status of dustfall heavy metals was comprehensively evaluated using chronological difference and ecological and health risks. The results showed that (1) As, Hg, Cd, Pb and Zn concentrations were significantly increased in industrial areas, whereas no significant change was observed for most heavy metals (except Pb and Zn) in non-industrial areas. The heavy metal concentrations were significantly higher in industrial areas than in non-industrial areas, which was largely associated with China's industrial production mode and mining activities. (2) Cd had the highest ecological risk (E_r^i), and Cr and Ni had the lowest E_r^i throughout China. The E_r^i of As, Hg, Cu, Pb and Zn were more serious in industrial areas than in non-industrial areas. (3) The non-carcinogenic risk index of children was higher than that of adults in all exposure pathways in non-industrial and industrial areas, the carcinogenic risk index of four carcinogenic heavy metals (As, Cd, Cr and Ni) was within 10^{-6}-10^{-4}, and the carcinogenic risk was negligible.

Keywords: dustfall, ecological risk, risk assessment, heavy metal

Introduction

Heavy metals refer to metallic chemical elements that have a relatively high density, including As, Hg, Cd, Cr, Cu, Ni, Pb and Zn; these metals are usually toxic for living organisms even at low concentrations [1-3]. Heavy metals in emissions can return to the soil and water through atmospheric depositional processes via dry and wet precipitation [4]. They have strong capacities to migrate, enrich and contaminate and enter the human body through ingestion, inhalation and dermal contact, exerting a negative influence on human health [5, 6]. Therefore, detailed investigations on the environmental impact of heavy metals in urban environments are of great importance.

Generally, street dust, atmospheric dust and foliar dust are classified as dustfall. At present, many studies

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have made significant progress in analysing pollution sources and distribution characteristics and have conducted pollution and risk assessment of dustfall heavy metals in some countries [7-12]. Investigating dustfall heavy metals is undoubtedly important, especially in developing countries undergoing industrialisation and urbanisation with high energy consumption and high emissions. Conducting a nationwide sampling collection is difficult due to the limitations of sampling period and confined regions. Therefore, the statistical analysis of dustfall heavy metals should be conducted in various regions as a holistic study of the whole nation. Wang et al. [13] and Zhang et al. [14] conducted a provincial spatial distribution analysis in China by using statistics and revealed that heavy metals are distributed in cities mainly located in southern, central and southeast coastal areas of China and mainly influenced by mining activities. However, the differences of dustfall heavy metals between industrial and non-industrial areas remain incompletely understood because the results might overestimate the impact of heavy metals in non-industrial areas or underestimate the effect of heavy metals in industrial areas.

For example, Wang et al. [15] found that the Cd, Pb and Zn concentrations of dustfall in Baoji were 5.5, 408.4 and 715.1 mg/kg, respectively, but Liu et al. [16] found that the concentrations of these metals were 62.1, 4201.7 and 5264.1 mg/kg, respectively. The discrepancy was due to the difference in sample locations. The sampling areas involved in the study by Liu et al. [16] were surrounded by industrial areas, whereas those related to the investigation performed by Wang et al. [15] were scattered in commercial, traffic and industrial areas. The application of average of mathematical statistics to evaluate the pollution degree of dustfall heavy metals is inaccurate under the absence of an evident distinction between industrial and non-industrial areas. In the present work, the study areas were divided into industrial and non-industrial ones for the first time to provide more precise data of dustfall heavy metals.

In addition, chronological difference and ecological and health risk assessments were adopted to understand the distribution characteristics of heavy metals and to estimate the influence of heavy metals on humans and the environment in non-industrial and industrial areas. The results will help to accurately assess the pollution status of dustfall heavy metals and to take measures for pollution control.

Material and Methods

Data Source and Processing

Data were obtained from the Web of Science, Springer Link, Science Direct, China National Knowledge Infrastructure, Wanfang Data Knowledge Service Platform and China Science and Technology Journal Database. The following terms were used for retrieval: 'atmospheric dust', 'dust', 'dustfall', 'heavy metal' and 'health risk assessment'. Sampling time was from 2006 to 2016, and if the sampling time was not clearly reported, the publication time minus two years as the sampling time. Data in Jiangxi, Tibet, Yunnan, Hainan, Hong Kong, Macao and Taiwan were unavailable.

Statistical Analysis of Data

Given that studies on atmospheric dust are incomplete, dustfall referred to all kinds of dust that settled on the surface, including atmospheric dust, road dust, foliar dust and street dust. Excel 2016, SPASS 22 and Origin 8.0 were used for the statistical analysis of data. When the data significantly obeyed a normal distribution at the level of 0.05, the arithmetic mean value was adopted. If the data significantly obeyed a lognormal distribution, the value was expressed with geometric average. When the data obeyed the skewed distribution, the median was taken [17].

The research areas were divided into industrial and non-industrial ones, with the latter including commercial, cultural and educational, residential, transportation, and tourist areas. The background values of heavy metals were referred from the China National Environmental Monitoring Center. The heavy metal concentrations in dustfall are shown in Tables 1, S1 and S2.

Research Methods

The ecological risk index and health risk assessment were used to evaluate the dustfall heavy metals. The health risk assessment was evaluated using the human exposure risk assessment model recommended by the USEPA, which could quantify the daily metal intake from contaminated dust, carcinogenic risk and non-carcinogenic risk for both children and adults.

Ecological Risk Index

The ecological risk index was proposed by Hakanson [18] in 1980. This method has been widely applied to evaluate the harm of heavy metals in sediments because the ecological risk is related not only to the concentration of heavy metals but also to their toxicological characteristics [19, 20]. The calculation formulas of the method are as follows:

$$C_r^i = \frac{c_i}{c_s} \tag{1}$$

$$E_r^i = T_i \times C_r^i \tag{2}$$

where C_r^i is the pollution coefficient for a certain heavy metal, which can reflect the pollution characteristics of

Table 1. Dustfall heavy metals in the non-industrial (NA) and industrial areas (IA) (mg/kg)

Element		As	Hg	Cd	Cr	Cu	Ni	Pb	Zn
Background value		11.2	0.1	0.1	61.0	22.6	26.9	26.0	74.2
Maximum	NA	333.6	1.1	14.8	436.8	696.1	91.0	559.9	3479.1
	IA	3912.1	27.6	195.0	1591.8	1674.2	248.2	5287.0	15343.3
Minimum	NA	1.1	0.0	0.1	9.4	22.7	16.4	17.8	80.8
	IA	4.4	0.1	0.0	39.7	24.5	6.5	20.1	89.5
Arithmetic mean	NA	37.1	0.3	3.2	131.4	118.7	42.5	171.9	610.8
	IA	362.4	3.1	22.1	210.9	267.7	51.6	860.6	2338.7
Geometric mean	NA	18.6	0.2	2.0	105.5	93.0	38.9	127.1	449.4
	IA	42.2	0.7	5.3	139.5	143.0	41.4	323.7	1180.1
Median	NA	15.8	0.2	2.4	105.6	86.3	41.1	119.4	522.2
	IA	26.2	0.4	4.7	139.9	113.3	42.0	257.0	1252.5
SD	NA	62.6	0.3	3.1	97.9	106.2	18.2	131.4	574.0
	IA	932.1	7.8	37.4	276.5	382.9	45.2	1362.0	2802.6
CV(%)	NA	169%	93%	96%	74%	89%	43%	76%	94%
	IA	257%	248%	169%	131%	143%	88%	158%	120%
Distribution type	NA	Lognormal	Lognormal	Lognormal	Skewness	Skewness	Lognormal	Lognormal	Lognormal
	IA	Skewness	Lognormal	Lognormal	Lognormal	Skewness	Skewness	Lognormal	Lognormal

the investigated region but cannot reveal the ecological effects and hazards. C_i is the measured values of heavy metals in the sediments. C_s is the background values of heavy metals. E_r^i is the ecological risk index of each heavy metal, and T_i is the response coefficient for the toxicity of single heavy metals. The toxic response factors of metals are as follows: Hg = 40, Cd = 30, As = 10, Cu = Pb = Ni = 5, Cr= 2 and Zn = 1 [18, 21]. According to Hakanson [18], the ecological risk index is categorized into five levels as follows: low ($E_r^i < 40$), moderate ($40 \leq E_r^i < 80$), considerable ($80 \leq E_r^i < 160$), high ($160 \leq E_r^i < 320$) and serious ($320 \leq E_r^i$).

Daily Intake Estimation of Heavy Metals

According to the migration and transformation of pollutants in the environment, heavy metals enter the human body mainly through the following pathways: ingestion, inhalation and dermal contact. The risk of Hg being absorbed by outdoor steam also needs to be considered. The average daily exposure dose (ADD_{ing}, ADD_{inh}, ADD_{dermal}, ADD_{vapour}, mg/kg·day) of heavy metals via various pathways was determined using the following equations:

$$ADD_{ing} = c \times \frac{IngR \times EF \times ED}{BW \times AT} \times 10^{-6} \quad (3)$$

$$ADD_{inh} = c \times \frac{InhR \times EF \times ED}{PEF \times BW \times AT} \quad (4)$$

$$ADD_{dermal} = c \times \frac{SA \times SL \times ABS \times EF \times ED}{BW \times AT} \times 10^{-6} \quad (5)$$

$$ADD_{vapour} = c \times \frac{InhR \times EF \times ED}{VF \times BW \times AT} \quad (6)$$

where *IngR* is the ingestion rate (200 and 100 mg/day for children and adults, respectively), *InhR* is the inhalation rate (7.6 and 12.8 m^3/day for children and adults, respectively), *EF* is the frequency of exposure (350 days/year), *ED* is the exposure duration (6 and 24 years for children and adults, respectively), *SA* is the exposed skin area (2800 and 5700 cm^2 for children and adults, respectively), *SL* is the skin adherence factor (0.2 and 0.07 mg/(cm^2 day) for children and adults, respectively), *ABS* is the dermal absorption factor (0.001 for all elements), *PEF* is the particle emission factor (1.36×10^9 m^3/kg for all elements), *BW* is the average body weight (15 and 70 kg for children and adults, respectively), *AT* is the averaging time (non-carcinogens, *ED* × 365 days; carcinogens, 70 × 365 days), and *VF* is the volatilization factor (32675.6 m^3/kg for Hg) [22].

Health Risk Assessment of Heavy Metals

Based on the calculation of the exposure dose via the possible exposure pathways, the hazard quotient (*HQ*) was calculated for each exposure pathway (Eqs. (7) and (8)). *RfD* is the corresponding reference dose for individual heavy metals and exposure pathways. The values are given in Table S3.

$$HQ_{i,j} = ADD_{i,j}/RfD_{i,j} \tag{7}$$

$$HI = \sum_{i=1}^{n}\sum_{j=1}^{m} HQ_{i,j} \tag{8}$$

where $HQ_{i,j}$ is the non-carcinogenic risk value of heavy metal i to human body through the j pathway. The non-carcinogenic risk index (HI) was the potential health risk of heavy metals via multiple pathways. When $HQ_{i,j}$<1 or HI<1, the risk is generally considered small or neglected; when $HQ_{i,j}$>1 or HI>1, humans are considered to tolerate the non-carcinogenic risk [23]. $ADD_{i,j}$ means the average daily dose exposure of heavy metal i through the j pathway, and $RfD_{i,j}$ indicates the reference dose of non-carcinogenic risk of heavy metal i through the j pathway.

Among the several heavy metals, As, Cd, Cr and Ni pose carcinogenic risks to human health, and the risks are closely related to the corresponding carcinogenic slope factor (SF). Given that SF is available only for the inhalation exposure pathway, the carcinogenic risk was calculated from exposure to inhalation route in the present study:

$$Risk_i = ADD_{inh} \times SF \tag{9}$$

$$Risk_T = \sum Risk_i = \sum(ADD_{inh} \times SF) \tag{10}$$

where $Risk_i$ is the individual carcinogenic risk of heavy metals and $Risk_T$ is the total carcinogenic risk of multiple carcinogenic heavy metals. The SF_{inh} values of As, Cd, Cr and Ni are 1.5, 6.3, 42.0 and 0.84, respectively. Carcinogenic risks exceeding 1×10^{-4} are regarded as unacceptable, risks below 1×10^{-6} are considered to pose no significant health effects, and risks within the range of 10^{-6}–10^{-4} may pose a carcinogenic health risk [24].

Results and Discussion

Heavy Metal Concentrations in the Dustfall of Various Countries

The heavy metal concentrations of dustfall collected in China and other countries worldwide are shown in Table 2. According to the data analysis in Table 2, both in China and other countries, the dustfall heavy metals (As, Hg, Cd, Cr, Cu, Ni, Pb and Zn) are generally polluted to a certain extent. The heavy metal concentrations were higher than the Chinese soil background values in industrial and non-industrial areas. However, the concentrations of As, Hg, Cd, Cu, Pb and Zn in industrial and non-industrial areas were greatly different. For example, the concentrations of As, Hg, Cd, Cu, Pb and Zn were 26.2, 0.7, 5.3, 113.3, 323.7 and 1180.1 mg/kg in industrial areas and 18.6, 0.2, 2.0, 86.3, 127.1 and 449.4 mg/kg in non-industrial areas, respectively. The concentrations of all heavy metals were significantly higher in industrial areas than in non-industrial areas. The difference was largely associated with China's industrial production mode and mining activities.

Moreover, the concentrations of Cd, Cr, Cu and Zn in non-industrial areas of China were much higher than those in India, Iran, Spain and Bangladesh. The concentrations of Cr, Cu, Ni, Pb and Zn in non-industrial areas of China were lower than those in Singapore. The concentrations of As and Pb in non-industrial areas of India were much higher than those in China. However, the concentrations of As and Pb in industrial areas of China were much higher than those in all other countries. The concentration of Ni in China was not different from that in other countries except Singapore. The concentrations of Cr, Cu, Ni and Zn in China were much lower than those in Singapore, which may be attributed to the local metal-related industries such as hardware and stainless steel in Singapore.

Chronological Difference of Heavy Metals in China

The data were divided into two periods (2006-2010 and 2011-2016) in China over the past years to better understand the changes of dustfall heavy metals (Fig. 1). The heavy metal concentrations in non-industrial areas showed no obvious change or decrease in recent years. Conversely, the concentrations of heavy metals (except Cr, Cu and Ni) in industrial areas increased significantly. The decrease in heavy metal concentrations in non-industrial areas suggests that the projects of heavy-polluting enterprises, which relocated and centralized in industrial parks, took a positive effect during the period of the '11th Five-Year Program' and '12th Five-Year Program' [45-47]. According to previous studies, As is mainly derived from coal burning [48]; Cu originates from brake abrasion in urban environments; Cd pollution mainly comes from the aging of automobile tires, gasoline use, car body wear and brake lining wear [49]; Cr mainly comes from vehicle emissions [50]. By the end of 2018, the number of civilian cars in China reached 240.28 million, which explained the subtle change of As, Cd, Cr and Cu in non-industrial areas.

In industrial areas, although the emission standards for pollutant concentrations have been strictly limited recently, the total amount of pollutants discharged continues to increase, which is closely associated with the clustering of industries in China. The concentrations of As, Hg, Cd, Pb and Zn in industrial areas increased, which were largely affected by the huge amount of coal burning and smelting emissions [51, 52]. As signature elements of metal smelting, Cr and Ni were largely influenced by alloy industrial emissions [48, 52, 53]. The concentrations of Cr and Ni decreased in industrial areas, suggesting that the management of dust emission in China's high-energy industries (e.g. iron and steel

Table 2. Dustfall heavy metals in non-industrial (NA) and industrial areas (IA) of various countries (mg/kg)

Study area	Areas	As	Hg	Cd	Cr	Cu	Ni	Pb	Zn	Reference
China	NA	18.6	0.2	2.0	105.6	86.3	38.9	127.1	449.4	This study
	IA	26.2	0.7	5.3	139.5	113.3	42.0	323.7	1180.1	
Soil background value (China)	NA/IA	11.2	0.1	0.1	61.0	22.6	26.9	26.0	74.2	-
U.S.	NA	-	-	-	95.0	105.0	-	73.0	240.0	[25]
	IA	-	151.5	0.1	597.5	23.7	47.5	3.0	474.6	[26]
Mexico	NA	16.4	-	4.2	86.4	26.3	4.7	46.0	388.0	[27]
	IA	10.1	-	-	101.5	167.0	52.8	226.9	649.4	[28]
Korea	NA	-	-	1.9	-	134.7	20.4	111.3	180.0	[29]
	IA	-	-	8.0	-	142.4	52.1	217.8	132.0	[30]
India	NA	95.6		0.7	29.3	42.3	-	202.7	303.2	[31,32]
	IA	3.6	-	0.3	64.4	57.0	40.3	81.0	280.5	[32,33]
Iran	NA	-	0.4	0.7	53.1	84.5	46.5	110.8	237.2	[34-36]
	IA	-	-	0.1	49.1	30.2	61.9	11.9	123.0	[36]
Spain	NA	-	-	1.2	25.9	57.5	37.8	78.2	128.6	[37-39]
	IA	-	-	3.6	129.0	120.3	74.3	210.8	398.3	[38,39]
Singapore	NA	-	-	0.5	195.7	335.2	53.6	145.5	995.1	[40]
	IA	-	-	2.4	823.5	4899.0	194.8	309.0	1672.5	[40,41]
Turkey	NA	-	-	-	-	111.0		177.0	245.0	[42]
	IA	-	-	2.5	29.0	36.9	44.9	74.8	112.0	[43]
Bangladesh	NA	5.7	-	-	93.7	27.3	24.3	44.7	105.3	[44]
	IA	7.0	-	-	136.0	105.0	35.0	54.0	169.0	

industry, smelting industry, etc.) has been successfully achieved in recent years.

In conclusion, limiting the concentration of pollutants was far from enough in the process of pollution control. Based on the dual control of total energy consumption and pollutant discharge, the industrial structure needs to be optimised to prevent the serious regional pollution caused by industrial concentration.

Ecological Risk Assessment of Heavy Metals

The ecological risk of dustfall heavy metals in industrial and non-industrial areas is shown in Fig. 2. In most areas of China, the pollution of Cd was more serious than that of other heavy metals, whereas the pollution of Cr and Ni was less serious than that of other heavy metals, which present the same distribution patterns in non-industrial and industrial areas. Dustfall heavy metals were mainly concentrated in China's central and eastern regions, and the ecological risk of dustfall heavy metals in industrial areas was generally higher than that in non-industrial areas.

In non-industrial areas, Hg and Cd caused higher ecological hazards than other heavy metals. The E_r^i values of Hg in Inner Mongolia, Guangdong, Guizhou and Sichuan were 337.14, 525.71, 400 and 628.57, respectively. The E_r^i values of Hg in Shanghai, Jiangsu, Shanxi and Heilongjiang were under considerable ecological hazards. The E_r^i value of Cd was serious in non-industrial areas (except in Inner Mongolia, Ningxia and Xinjiang). The ecological hazards caused by As were high in Jilin, considerable in Shaanxi and moderate in Guangdong and Henan. In industrial areas, the E_r^i values of Cr and Ni were lower than 40, indicating slight ecological risk. As caused ecological hazards in Henan and Guangxi, which posed serious ecological risk level ($E_r^i \geq 320$). The E_r^i value of As in Guangdong was 116.07, posing a high ecological risk. The E_r^i values of As in Shaanxi and Jilin were within $40 \leq E_r^i < 80$, indicating a moderate ecological risk. The E_r^i values of Hg in Fujian, Zhejiang, Liaoning, Guizhou and Guangxi were ≥320, indicating serious ecological hazards, whereas those in Jiangsu and Hebei were $160 \leq E_r^i < 320$, posing high ecological hazards. In Xinjiang and Shaanxi, the E_r^i values of Hg were $40 \leq E_r^i < 80$, indicating moderate ecological hazards. The ecological hazards caused by Pb were mainly distributed in Shaanxi, Henan, Chongqing and Tianjin with $E_r^i \geq 320$. The maximum E_r^i value of Zn in Hubei was 117.69, posing a considerable ecological risk. Cd caused the main ecological hazards in industrial areas (except in Ningxia, Shaanxi and Shandong); however, the E_r^i value of Cd in non-industrial areas was more than 320, indicating that Cd posed more ecological risk in non-industrial areas than in industrial areas.

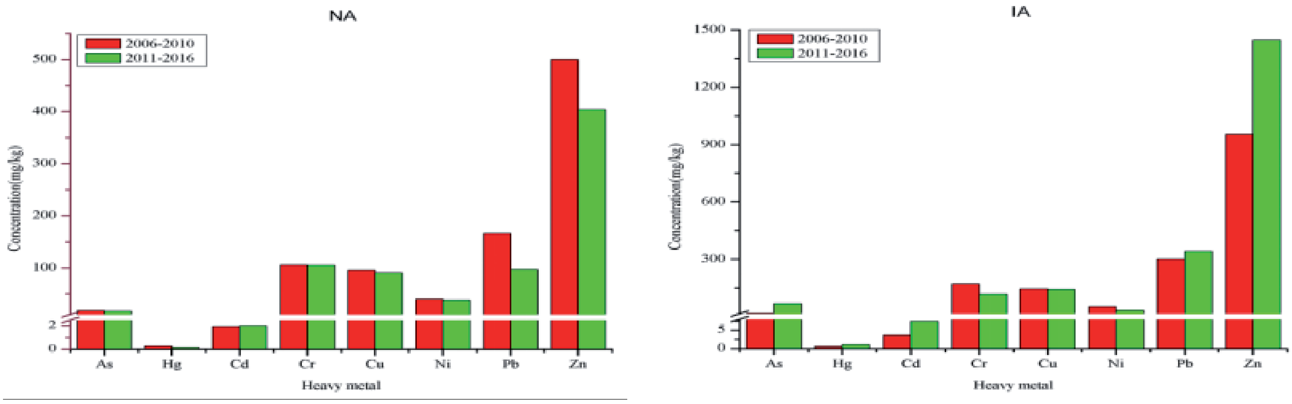

Fig. 1. Changes of dustfall heavy metals in non-industrial (NA) and industrial areas (IA) of China.

Additionally, the ecological pollution levels of dustfall heavy metals (except Hg and Cd) in non-industrial areas ranged from low to moderately polluted, whereas those in industrial areas mostly ranged from considerable to serious, except Cr and Ni with low ecological pollution level.

Daily Exposure Doses of Heavy Metals Via Various Pathways

The exposure doses of dustfall heavy metals are shown in Table 3.

The exposure doses of dustfall heavy metals are shown in Table 3. Amongst the three contact exposure pathways, ingestion was the main exposure pathway, followed by dermal contact and inhalation (Table 3). The non-carcinogenic exposure dose for children was higher than that for adults in all three exposure pathways, which was consistent with the results of Fang et al. [54]. The outdoor vapour inhalation of Hg was the most important exposure pathway. The exposure doses of children and adults in non-industrial areas were 3.42×10^{-6} mg/(kg·day) and 1.23×10^{-6} mg/(kg·day), respectively, and those in industrial areas were 1.02×10^{-6} mg/(kg·day) and 3.70×10^{-5} mg/(kg·day), respectively, which were consistent with the results of Zheng et al. [55]. The order of the non-carcinogenic exposure dose by the three pathways for children and adults in non-industrial and industrial areas was Zn>Pb> Cr>Cu>Ni>As>Cd>Hg and Zn>Pb>Cu >Cr>As>Ni>Cd>Hg, respectively. Zn and Hg had the maximum and minimum exposure doses, respectively, which were consistent with the results of heavy metal concentrations in non-industrial and industrial areas.

The carcinogenic exposure doses of As, Cd, Cr and Ni for children were lower than those for adults both in

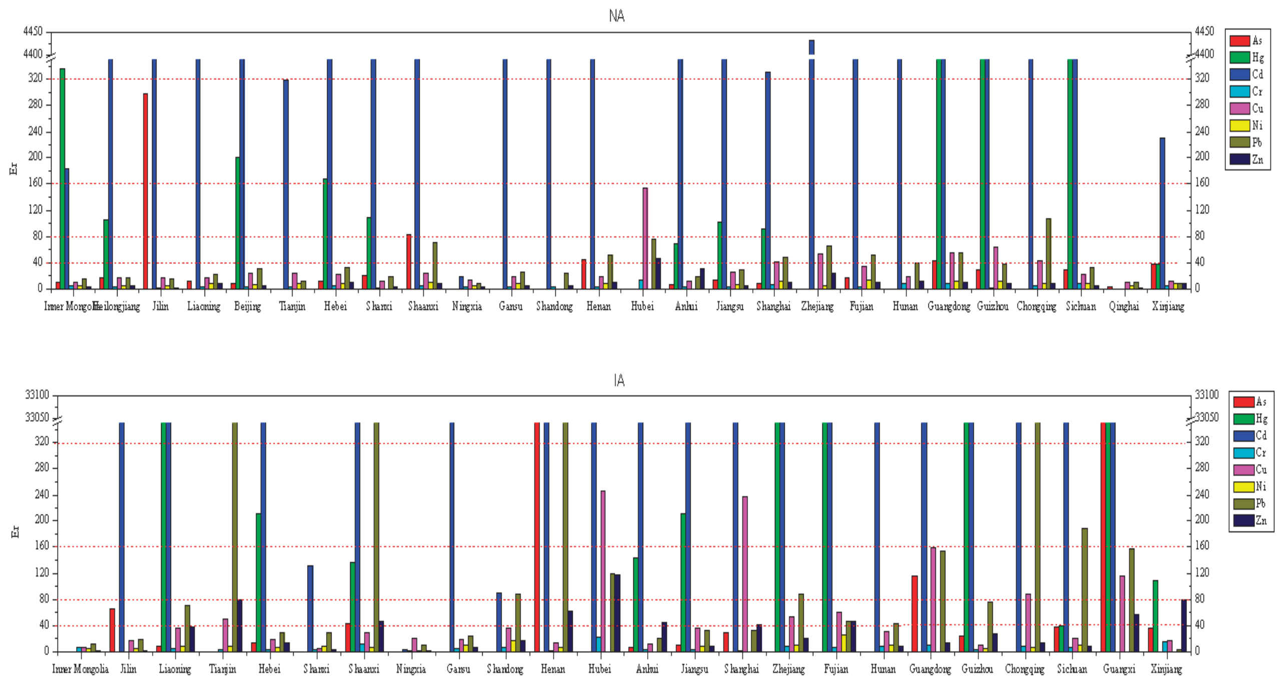

Fig. 2. Ecological risk index of dustfall heavy metals in non-industrial (NA) and industrial areas (IA) of China.

Table 3. Exposure doses of heavy metals in dustfall via different exposure pathways in China [mg/(kg·day)].

Element	Areas	Concentration	Ingestion		Inhalation		Dermal contact		Vapor		ADD_C	
			Adults	Children	Adults	Children	Adults	Children	Adults	Children	Adults	Children
As	NA	18.6	2.6×10^{-5}	2.4×10^{-4}	2.4×10^{-9}	6.6×10^{-9}	1.4×10^{-7}	4.9×10^{-7}			8.3×10^{-10}	5.7×10^{-10}
	IA	42.2	5.8×10^{-5}	5.4×10^{-4}	5.4×10^{-9}	1.5×10^{-8}	3.1×10^{-7}	1.1×10^{-6}			1.9×10^{-9}	1.3×10^{-9}
Hg	NA	0.2	3.2×10^{-7}	2.9×10^{-6}	3.0×10^{-11}	8.2×10^{-11}	1.7×10^{-9}	6.1×10^{-9}	1.2×10^{-6}	3.4×10^{-6}		
	IA	0.7	9.5×10^{-7}	8.8×10^{-6}	9.0×10^{-11}	2.5×10^{-10}	5.1×10^{-9}	1.8×10^{-8}	3.7×10^{-6}	1.0×10^{-5}		
Cd	NA	2.0	2.7×10^{-6}	2.5×10^{-5}	2.5×10^{-10}	7.0×10^{-10}	1.5×10^{-8}	5.2×10^{-8}			8.7×10^{-11}	6.0×10^{-11}
	IA	5.3	7.2×10^{-6}	6.8×10^{-5}	6.8×10^{-10}	1.9×10^{-9}	3.9×10^{-8}	1.4×10^{-7}			2.3×10^{-10}	1.6×10^{-10}
Cr	NA	105.5	1.4×10^{-4}	1.4×10^{-3}	1.4×10^{-8}	3.8×10^{-8}	7.8×10^{-7}	2.8×10^{-6}			4.7×10^{-9}	3.2×10^{-9}
	IA	139.5	1.9×10^{-4}	1.8×10^{-3}	1.8×10^{-8}	5.0×10^{-8}	1.0×10^{-6}	3.7×10^{-6}			6.2×10^{-9}	4.3×10^{-9}
Cu	NA	93.0	1.3×10^{-4}	1.2×10^{-3}	1.2×10^{-8}	3.3×10^{-8}	6.9×10^{-7}	2.5×10^{-6}				
	IA	143.0	2.0×10^{-4}	1.8×10^{-3}	1.9×10^{-8}	5.1×10^{-8}	1.1×10^{-6}	3.8×10^{-6}				
Ni	NA	38.9	5.3×10^{-5}	5.0×10^{-4}	5.0×10^{-9}	1.4×10^{-8}	2.9×10^{-7}	1.0×10^{-6}			1.7×10^{-9}	1.2×10^{-9}
	IA	41.4	5.7×10^{-5}	5.3×10^{-4}	5.3×10^{-9}	1.5×10^{-8}	3.1×10^{-7}	1.1×10^{-6}			1.8×10^{-9}	1.3×10^{-9}
Pb	NA	127.1	1.7×10^{-4}	1.6×10^{-3}	1.6×10^{-8}	4.5×10^{-8}	9.4×10^{-7}	3.4×10^{-6}				
	IA	323.7	4.4×10^{-4}	4.1×10^{-3}	4.2×10^{-8}	1.2×10^{-7}	2.4×10^{-6}	8.6×10^{-6}				
Zn	NA	449.4	6.2×10^{-4}	5.7×10^{-3}	5.8×10^{-8}	1.6×10^{-7}	3.3×10^{-6}	1.2×10^{-5}				
	IA	1180.1	1.6×10^{-3}	1.5×10^{-2}	1.5×10^{-7}	4.2×10^{-7}	8.7×10^{-6}	3.1×10^{-5}				

Note. ADD_C = Carcinogenic exposure dose.

Table 4. Non-carcinogenic risk index of dustfall heavy metals in non-industrial (NA) and industrial areas (IA) of China.

Element	Areas	$HQ_{i,j}$								HI		Risk	
		Ingestion		Inhalation		Dermal		Vapor					
		Adults	Children	Adults	Children	Adults	Children	Adults	Children	Adults	Children	Adults	Children
As	NA	8.5×10^{-2}	7.9×10^{-1}	8.0×10^{-6}	2.2×10^{-5}	1.1×10^{-3}	4.0×10^{-3}			8.6×10^{-2}	8.0×10^{-1}	1.2×10^{-9}	8.6×10^{-10}
	IA	1.9×10^{-1}	1.8	1.8×10^{-5}	5.0×10^{-5}	2.5×10^{-3}	9.1×10^{-3}			2.0×10^{-1}	1.8	2.8×10^{-9}	1.9×10^{-9}
Hg	NA	1.1×10^{-3}	9.8×10^{-3}	3.5×10^{-7}	9.6×10^{-7}	8.1×10^{-5}	2.9×10^{-4}	1.4×10^{-2}	4.0×10^{-2}	1.1×10^{-3}	1.0×10^{-2}		
	IA	3.2×10^{-3}	2.9×10^{-2}	1.0×10^{-6}	2.9×10^{-6}	2.4×10^{-4}	8.7×10^{-4}	4.3×10^{-2}	1.2×10^{-1}	3.4×10^{-3}	3.0×10^{-2}		
Cd	NA	2.7×10^{-3}	2.5×10^{-2}	2.5×10^{-7}	7.0×10^{-7}	1.5×10^{-3}	5.2×10^{-3}			4.2×10^{-3}	3.0×10^{-2}	5.5×10^{-10}	3.5×10^{-10}
	IA	7.2×10^{-3}	6.8×10^{-2}	6.8×10^{-7}	1.9×10^{-6}	3.9×10^{-3}	1.4×10^{-2}			1.1×10^{-2}	8.2×10^{-2}	1.5×10^{-9}	1.0×10^{-9}
Cr	NA	4.8×10^{-2}	4.5×10^{-1}	4.7×10^{-4}	1.3×10^{-3}	1.3×10^{-2}	4.7×10^{-2}			6.2×10^{-2}	5.0×10^{-1}	2.0×10^{-7}	1.4×10^{-7}
	IA	6.4×10^{-2}	5.9×10^{-1}	6.3×10^{-4}	1.7×10^{-3}	1.7×10^{-2}	6.2×10^{-2}			8.2×10^{-2}	6.6×10^{-1}	2.6×10^{-7}	1.8×10^{-7}
Cu	NA	3.2×10^{-3}	3.0×10^{-2}	3.0×10^{-7}	8.2×10^{-7}	5.7×10^{-5}	2.1×10^{-4}			3.3×10^{-3}	3.0×10^{-2}		
	IA	4.9×10^{-3}	4.6×10^{-2}	4.6×10^{-7}	1.3×10^{-6}	8.8×10^{-5}	3.2×10^{-4}			5.0×10^{-3}	4.6×10^{-2}		
Ni	NA	2.7×10^{-3}	2.5×10^{-2}	2.4×10^{-7}	6.8×10^{-7}	5.3×10^{-5}	1.9×10^{-4}			2.7×10^{-3}	2.5×10^{-2}	1.5×10^{-9}	1.0×10^{-9}
	IA	2.8×10^{-3}	2.7×10^{-2}	2.6×10^{-7}	7.2×10^{-7}	5.7×10^{-5}	2.0×10^{-4}			2.9×10^{-3}	2.7×10^{-2}	1.5×10^{-9}	1.1×10^{-9}
Pb	NA	5.0×10^{-2}	4.7×10^{-1}	4.7×10^{-6}	1.3×10^{-5}	1.8×10^{-3}	6.4×10^{-3}			5.2×10^{-2}	4.7×10^{-1}		
	IA	1.3×10^{-1}	1.2	1.2×10^{-5}	3.3×10^{-5}	4.6×10^{-3}	1.6×10^{-2}			1.3×10^{-1}	1.2		
Zn	NA	2.1×10^{-3}	1.9×10^{-2}	1.9×10^{-7}	5.4×10^{-7}	5.5×10^{-5}	2.0×10^{-4}			2.1×10^{-3}	1.9×10^{-2}		
	IA	5.4×10^{-3}	5.0×10^{-2}	5.1×10^{-7}	1.4×10^{-6}	1.5×10^{-4}	5.2×10^{-4}			5.5×10^{-3}	5.1×10^{-2}		
HI_T/$Risk_T$	NA									2.1×10^{-1}	1.9	2.0×10^{-7}	1.4×10^{-7}
	IA									4.4×10^{-1}	3.9	2.7×10^{-7}	1.8×10^{-7}

non-industrial and industrial areas. Cr had the highest exposure dose amongst them. The exposure doses of Cr for adults and children were 4.65×10^{-9} mg/(kg·day) and 3.23×10^{-9} mg/(kg·day) in non-industrial areas and 6.17×10^{-9} mg/(kg·day) and 4.27×10^{-9} mg/(kg·day) in industrial areas, respectively.

Human Health Risk Assessment

Different heavy metals have different toxicities, so the health effects of heavy metals on humans cannot be accurately assessed via exposure dose alone. Thus, the non-carcinogenic and carcinogenic risk indices of eight dustfall heavy metals in non-industrial and industrial areas were calculated based on the above exposure doses.

The results are shown in Table 4. The hazards of dustfall heavy metals on children and adults in non-industrial and industrial areas of China were inconsistent. In non-industrial areas, the *HI* was 1>As>Cr>Pb>Cd>Cu>Ni>Zn>Hg for children and 1>As>Cr>Pb>Cd>Cu>Ni>Zn>Hg for adults. In industrial areas, the *HI* was As>Pb>1> Cr>Cd>Zn>Cu>Hg>Ni for children and 1>As>Pb>Cr>Cd>Zn>Cu>Hg>Ni for adults. In non-industrial areas, the non-carcinogenic risk for children and adults was consistent with the exposure doses. The health risk of the three pathways for children and adults was less than 1, indicating that the risk was low and negligible. However, in industrial areas, the non-carcinogenic risk of As and Pb exceed the safety threshold of 1. As is related to diseases of the nervous system, blood and skin ulcer in both children and adults [56]. The HQ of As for children via the ingestion pathway was 1.80 in industrial areas, indicating that the potential non-carcinogenic risk of As in industrial areas was more severe than that in non-industrial areas. Moreover, the non-carcinogenic health effects for children were much more vulnerable than those for adults.

For the carcinogenic risk index, the carcinogenic risk values of As, Cd, Cr and Ni were within the range of 10^{-6}–10^{-4}, but attention should be still paid to the ecological hazards of Cr because the carcinogenic risk index is close to 10^{-6}.

Uncertainty in Risk Assessment

It can be seen from the analysis results of different methods that the sources of atmospheric dustfall in China are extensive and the composition is complex, and the maximum contents of different heavy metal elements are distributed in different areas. The data collected in this study come from different research literature. Different papers have different divisions of functional fields, and the heavy metals studied in each literature are also different. Limited by objective factors such as time and space, it is difficult to collect data of heavy metals, and the research scope of this study cannot cover the whole country. For example, Jiangxi, Tibet, Yunnan, Hainan, Hong Kong, Macau and Taiwan Province have not yet carried out relevant studies. In addition, the sampling methods, time and analysis methods of different literatures will also have an impact on the research results. At present, there is no unified and complete detection system for heavy metals in dustfall in China, so it is suggested that the above factors should be taken into account in the follow-up research.

Conclusions

In recent years, many scholars within domestic and foreign research areas have carried out a large number of studies on heavy metal pollution in atmospheric dustfall. However, most of these research areas are concentrated in a kind of specific functional area, such as urban streets [55], parks [3] and multiple functional areas [5]. Compared with other studies within domestic and foreign research areas, the study is first divided into industrial and non-industrial areas, and the spatial distribution of heavy metals is relatively more comprehensive.

The distribution characteristics and ecological and health risk assessment of dustfall heavy metals in non-industrial and industrial areas of China were evaluated in this study. Compared with relevant studies within domestic and foreign research areas, the results showed that (1) the respective concentrations of As, Hg, Cd, Cr, Cu, Ni, Pb and Zn were 1.07-333.06, 0.04-1.10, 0.06-14.77, 9.43-436.76, 22.74-696.09, 16.40-91.00, 17.81-559.85 and 80.81-3479.07 mg/kg in non-industrial areas and 4.35-3912.08, 0.07-27.60, 0.01-195.00, 39.70-1591.80, 24.46-1674.19, 6.47-248.23, 20.10-5287.00 and 89.50-15343.27 mg/kg in industrial areas, the concentration of heavy metals in industrial areas was significantly higher than that in non-industrial areas. Liu et al. [16] found in relevant studies that the concentration of heavy metals in industrial areas was significantly higher than that in surrounding areas, but the study areas were only industrial areas and surrounding areas, so it might not be possible to accurately judge the he degree of heavy metal pollutions. (2) The chronological statistics indicated that the heavy metal concentrations decreased in non-industrial areas but increased in industrial areas from 2006 to 2016, which suggested that only controlling the concentration of heavy metals was limited for heavy metal pollution. (3) The ecological risk assessment of dustfall heavy metals varied widely, the ecological risk of heavy metals in industrial areas is generally higher than that in non-industrial areas, and the ecological risk was mainly caused by As, Hg and Cd. The pollution levels were mostly moderate to considerable. In industrial areas, the ecological pollution of As, Hg, Cd, Pb and Zn mainly ranged from considerable to serious, and the ecological risk of Liaoning, Shaanxi, Beijing–Tianjin-Hebei, Hubei and the southwest and southeast coastal regions of China was serious. However, Das et al. [31] and Kim et al. [8] found that the concentrations of As and Pb in non-industrial areas in India were much higher than those in industrial areas. The study found that the concentrations of As and Pb in China's industrial areas are higher than those in India, while those in non-industrial areas are much lower than those in India. (4) The results of human health risk assessment show that among all the investigated heavy metals, the headquarters of industrial area has the highest content of As. The HI value of heavy metals indicates that the non-carcinogenic risks of children and adults in industrial areas are significantly higher than that in non-industrial areas, and the non-carcinogenic health effects of children are far more vulnerable than adults. Shabbaj etal. [57] found that the carcinogenic risk (CRA) of heavy metals in Jeddah was within the safe limits for children and adults, and the CRA value of children was higher than that of adults. In view of these results, the government should put forward measures to monitor and control heavy metal pollution.

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Author Contributions

Mingshi Wang, Chunhui Zhang participated in the design of this study, Chunhui Zhang analyzed the data and wrote the manuscript, Yuchuan He, Chun Chen, Qiao Han gave many good suggestions for data analyzing and writing the manuscript.

Conflicts of Interest

The authors declare no conflict of interest.

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Supplementary Material

Table S1. The concentration of dustfall heavy metals in non-industrial areas of China (mg/kg).

Province	Sampling site	Sampling time	Number	As	Hg	Cd	Cr	Cu	Ni	Pb	Zn	Reference
Beijing	Beijing and surrounding areas	2013.6-2014.3	66			4	134.3	158.9	46.85	154.8	741.25	Xiong et al., 2016
	Urban college	2014.11	50			0.77	85.07	82.41	34.03	54.3	308.31	Yan et al., 2016
	Subway station	2010	16	9.09	0.35	1.11	131.92	67.33	41.77	437.41		Yang et al., 2011
	Urban area[1]	2013.12-2014.3	35			2.72	133.92	249.01	62.62	114.38	567.08	Sun et al., 2016
	Urban area[2]	2009.6-2009.9	225			0.47	77.36	63.73	23.6	50.79		Tang et al., 2012
	Urban area[3]	2010.2-2010.5	154			0.72	84.7	69.9	25.20	105	222	Wei et al., 2015
	Urban Park	2010.4-2010.7	50			0.64	69.33	72.13	25.97	201.82	219.2	Du et al., 2013
Shanghai	Living area	2005-2006	48			0.97	218.91	186.41	64.91	212.94	687.25	Jing et al., 2009
	Urban area	2004.11-2007.2		8.73	0.16	1.24	157			287		Wang et al., 2009
Xinjiang	Zhundong Coalfield	2014.7-2014.12	50	35.17	0.09		436.76	69.23		20.14		Liu et al., 2017b
	Ebinur basin	2013.8-2014.7	36	1.07	0.04	0.21	95.32	22.74		17.81	106.89	Zhang et al., 2015
	Urumqi[1]	2012.3-2013.3		151.5		1.39	66.05	59.05	48.79	33.94	806	Abuduwailil et al., 2015
	Urumqi[2]	2010.1-2010.12		6.94		0.46	62.97	77.86	47.81	100.43	649.09	Zhang et al., 2014b
	Urumqi and Changji	2008.5-2009.5		17.88	0.07	1.02	40.48	44.21	21.38	53.38	669.13	Wei et al., 2013
Province	**Sampling site**	**Sampling time**	**Number**	**As**	**Hg**	**Cd**	**Cr**	**Cu**	**Ni**	**Pb**	**Zn**	**Reference**
Shanxi	Baoji	2006.2	38			5.5	126.7	123.2	48.8	408.4	715.1	Wang et al., 2011
	Xi'an[1]	2013.10-2013.11		92.69		4.12	199.31	131.98	69.98	283.82	724.25	Chen et al., 2007
	Xi'an[2]	2014.1-2014.12				7.36	148.17	55.91	41.53	407.98	446.56	Yang et al., 2017
Gansu	Lanzhou	2010.6-2011.5				4.40	84.76	83.07	38.97	129.75	367.02	Li et al., 2014
Jiangsu	Nangjingpark	2013.12	30	10.77		5.18	81.29	168.28	45.32	147.35	526.66	Wang et al., 2016
	Nanjing	2015.12		4.02		1.50	57.38	173.40	43.02	117.25	668.40	Tian et al., 2018
	Nantong	2013.12	19	33.71		5.05		113.3		175.55	335.6	Peng et al., 2015
	Suzhou[1]	2012.6	43	13.4	0.18	2.45	25.7	104.8	16.4	262.2	376.9	Ma et al., 2016
	Suzhou[2]	2014.3-2015.2	276				112.9	27.5		45.2	225.3	Lin et al., 2017
Hubei	Huangshi	2012.5	21				391.09	696.09		391.32	3479.07	Yao et al., 2016

Table S1. Continued.

Fujian	Quanzhou[1]	2007	13	17.05		2.18	107.94	119.64	65.88	242.10	647.98	Hu et al., 2013
	Quanzhou[2]	2006				1.64	109.66	116.57	71.00	290.90	1094.42	Yu et al., 2010
	Quanzhou[3]	2006.10--12				5.17	154.20	159.08	82.12	226.03	522.33	Zhang et al., 2016
	Quanzhou district	2006.10--12		22.79		2.42	108	211.88	78.04	328.21	522	Hu et al., 2010
Zhejiang	Hangzhou	2006.6--2009.6				14.77		240.82	27.90	341.27	1755.15	Li et al., 2013
Hebei	Shijiazhuang[1]	2007.11-2008.11		14.55	0.32	5.22	97.53	86.34	31.77	159.53	822.46	Kui et al., 2012
	Shijiazhuang[2]	2013.5	27			2.97	166.94	149.26	49.34	228.64	654.22	Wan et al., 2016a
	Tangshan	2009	162	12.65	0.27	3.4	132.35	69.86	40.71	121.48	815.42	Chen et al., 2011
Liaoning	Shenyang	2006.4	61			4.35		81.33		106.26	334.47	Li et al., 2010a
	Fuxin district	2015.3-2016.2	32	12.2		4.8	110.9	74.7	44.2	114.6	756.9	Zhao et al., 2017
Qinghai	Xining	2012.7		3.6				40.8	22.6	52.9	108.9	Zhao et al., 2014
Guangdong	Shunde	2007	10	48.8	0.92	0.31	420	379	91	502	899	Wu et al., 2009
Province	**Sampling site**	**Sampling time**	**Number**	**As**	**Hg**	**Cd**	**Cr**	**Cu**	**Ni**	**Pb**	**Zn**	**Reference**
Guangdong	Guangzhou	2011	18			4.22	62.2	116.3	31.9	72.6	504	Cai et al., 2013
Sichuan	Chengdu[1]	2010	335	32.8	1.1	3.75	277		39	354	634	Shi et al., 2012
	Chengdu[2]	2014.8-2015.8	75			1.68	84.8	98.9	24.5	81.3	296	Li et al., 2017
	Panzhihua	2012.11				0.84	313.78	92.07	54.56	83.09	340.56	Yang et al., 2016b
Guizhou	Guiyang	2013.7	32	12.85				197.55		83.43	571.77	Zhang et al., 2014a
	Panxian	2012.3-2013.2		53.1	0.7	7.1	62.2	376.4	64.1	304.9	555.2	Zeng et al., 2014
Shanxi	Taiyuan basin	2006	26	22.4	0.19	1.96				131		Lai et al., 2008
	Taiyuan	2008				2.27	9.43	55.85		71.76	187.55	Yang., 2010
Hunan	Changsha[1]	2009.9				2.5	403.5	126		348	1541.5	Gao et al., 2015
	Changsha[2]	2013.10.	51			9.11	80.7	43.9		66.6	215	Li et al., 2016
Anhui	Huainan	2014.9	46	6.81	0.12	0.25	59.17	37.53		43.57		Tang et al., 2017
	Culture and education area	2010				5.28	69.61	68.01		148.65	2302.84	Tong et al., 2012

Table S1. Continued.

Henan	Rural area	2013		91.6		10.00				308.00		Qiu et al., 2015
	Pingdingshan	2008.5		22.98		3.30	142.55	99.2	52.35	331.15	498.80	Liu et al., 2009
	Jiaozuo	2013.12-2014.3		37.21		7.79	72.59	69.39	41.44	159.24	1058.29	Wang et al., 2017
Chongqing	Culture and education area	2007.3--2008.2				12.58	160.73	191.98	40.58	559.85	653.18	Jiang et al., 2008
Inner Mongolia	Hohhot[1]	2006.3--2006.9		11.4				51.80		99.45		Liu., 2008
	Hohhot[2]	2006	91	9.76	0.59	0.61	76.05	55.75	30.90	77.06	247.08	Bao., 2008
	Baotou	2012.10.	121				182.1	29.1	21.2	58.2	85.9	Han et al., 2016
Jilin	Changchun[1]	2006.10-2007.2		333.60				142.77		114.68	111.70	Li., 2007
	Changchun[2]	2008.10				2.58		45.77	19.22	38.61	80.81	Zhao et al., 2009
	Changchun[3]	2013.6	28			0.31	60.30	43.56	23.16	70.89	170.80	Liu et al., 2015
Province	**Sampling site**	**Sampling time**	**Number**	**As**	**Hg**	**Cd**	**Cr**	**Cu**	**Ni**	**Pb**	**Zn**	**Reference**
Heilongjiang	Harbin	2008.4--2009.4	46	23.5	0.25	1.46	87.13	104.9	34.58	117.15	370.77	Tang et al., 2011
	Songnen plain	2006.11-2007.11	166	12.3	0.12	1.01	51.9	49	21.5	58.5	277.3	Li., 2011
Ningxia	Yinchuan	2014.5-2015.4				0.06	78.13	61.28	21.59	43.21	178.37	Liu et al., 2017
Shandong	Qingdao	2010				1.79	109.27			126.02	326.88	Qiao et al., 2011
Tianjin	Tianjin	2013				0.99	121.41	100.62	43.35	61.48		Sun et al., 2015
	Park	2011.12-2012.1	41			1.14	103.18	113.18	40.58	63.32		Guo et al., 2014

Table S2. The concentration of dustfall heavy metals in industrial areas of China (mg/kg).

Province	Sampling site	Sampling time	Number	As	Hg	Cd	Cr	Cu	Ni	Pb	Zn	Reference
Shanghai	Yangpu district	2009	11	33.10		4.10	48.60	1072.90		168.60	3014.90	Shuai et al., 2011
Xinjiang	ZhunDong coalfield	2014.5-2014.12	52	40.40	0.19		482.07	78.66		20.10	5840.19	Yang et al., 2016a
Shanxi	Industrial area	2013.10-2013.11	18	73.53		9.01	136.49	42.92	35.74	148.93	327.69	Chen et al., 2016b
	Baoji[1]	2013	21			68.40	39.70	160.20	26.80	4509.10	5859.60	Sun et al., 2015
	Baoji[2]	2013.6				62.10	49.60	154.10	27.00	4201.70	5264.10	Liu et al., 2015b
	Industrial park	2012.11-2013.3	9			79.17	48.98	127.04	36.76	2436.47	4129.27	Liang et al., 2014
	Changqing industrial area	2010.10.	38	23.30	0.24		1591.80	178.20	40.20	1586.20	1918.80	Wang et al., 2014
Gansu	Industrial area	2010.6-2011.5				3.89	143.22	87.65	52.87	122.45	461.31	Li et al., 2014
Jiangsu	Nanjing[1]	2013.9-2014.5	10	20.40			144.89	127.10	52.84	96.68	468.93	Chen et al., 2014a
	Nanjing[2]	2015.12		7.51		1.73	67.75	214.70	47.15	141.80	727.20	Tian et al., 2018
	Taicang	2007	5	8.40	0.37	2.90	111.00	155.90	44.70	261.00	716.00	Liu et al., 2009
Hubei	Huangshi[1]	2012.5	11				652.64	555.84		296.84	15343.27	Yao et al., 2016
	Huangshi[2]	2012.5	17			41.35		1674.19		949.30	2122.53	Liu et al., 2014a
Province	**Sampling site**	**Sampling time**	**Number**	**As**	**Hg**	**Cd**	**Cr**	**Cu**	**Ni**	**Pb**	**Zn**	**Reference**
Fujian	Quanzhou[1]	2006				3.47	195.70	95.73	248.23	258.84	3765.6	Hu et al., 2013
	Quanzhou[2]	2007.11-2008.1	8		1.86	3.71	97.40	106.3	126.3	255.1	2240	Zhang et al., 2016
	Industrial area	2012.11-2013.1	10				248.00	621.00	52.58	205.00	4245.00	Wen et al., 2015
Zhejiang	Near industrial and mining	2008.9			2.51	8.77	256.00	373.00	78.00	544.00	898.00	Yang et al., 2010
	Hangzhou	2006.6--2009.6				20.8		106.9	21.0	376.6	2222.6	Ma et al., 2016
Hebei	Shijiazhuang[1]	2007.11-2008.11		15.09	0.37	11.98	92.97	76.17	30.15	132.18	1357.71	Kui et al., 2012
	Shijiazhuang[2]	2013.5	16			3.58	143.2	101.2	43.0	173.2	560.0	Wan et al., 2014b
Liaoning	Anshan	2008.6-7		9.35		0.79	167.2	56.52	41.05	196.17	377.33	Xing et al., 2010
	Huludao	2008	35		1.22	72.84		264.40		533.20	5271.00	Zheng et al., 2010
Guangdong	Shaoguan	2012.8-2012.10	12			42.57		78.34		314.67	794.31	Huang et al., 2013
	Huizhou		27			8.40	237.00	1313.00		462.00	1860.00	Qiu et al., 2007
	Power plant[1]			130.00		6.66	571.00	147.00		446.00	1184.00	Liu et al., 2017a
	Power plant[2]	2013.9-2014.3	20			49.30	170.00	1340.00		1980.00	306.00	He et al., 2017

Table S2. Continued.

Sichuan	Panzhihua[1]	2007.7-2007.12	21	78.60		5.20	311.50	113.30		1929.00	924.10	Jiang et al., 2010
	Panzhihua[2]			4.35	0.07	0.27	107.50	66.50	56.20	31.50	155.90	Xu et al., 2009
Guizhou	Smelting plant			26.10	27.60	1.25	100.00	48.20	24.20	398.00	2037.00	Lin et al., 2017
Shanxi	Linfen	2008.4-2009.3				0.44	80.24	24.46	47.31	151.30	195.42	Cheng et al., 2010
Hunan	Industrial area	2011.1-2014.12	16			2.16				211.71		Zeng et al., 2014
	Loudi	2008.7	18			2.20	225.00	141.00	49.30	228.00	583.00	Zhang et al., 2012
Anhui	Huainan mining area	2014.9	18	6.95	0.21	0.21	54.80	36.60		43.70		Tang et al., 2017
	Industrial area	2014.9	24	6.67	0.29	0.27	65.40	38.70		46.80		Tang et al., 2017
Province	**Sampling site**	**Sampling time**	**Number**	**As**	**Hg**	**Cd**	**Cr**	**Cu**	**Ni**	**Pb**	**Zn**	**Reference**
Anhui	Hefei	2010.4-2010.7	4			3.62	121.90	82.33		236.04	3308.10	Tong., 2012
Henan	Smelting plant	2013.9-2014.2		1419.00		195.00				5287.00		Qiu et al., 2015
	Jiaozuo	2013.12-2014.3		26.26		8.74	61.50	60.44	35.60	140.90	4594.33	Wang et al., 2017
Chongqing	Industrial area	2007.3-2008.2				14.28	273.58	397.48	38.5	2970.43	983.65	Jiang et al., 2008
Guangxi	Nandan[1]	2014.10.		1332.81	2.63	38.00				650.88		Chen et al., 2016c
	Nandan[2]	2016.3	28	3912.08		58.67		521.36		981.40	4252.10	Wei et al., 2016
Jilin	Changchun[1]	2006.10-2007.2		73.2				89.70		123.80	149.50	Li., 2007
	Changchun[2]	2008.10.				3.50		71.38	23.49	71.38	110.89	Zhao et al., 2009
Inner Mongolia	Baotou	2012.1	83				189.60	29.40	21.60	64.90	89.50	Han et al., 2017
Ningxia	Yinchuan	2014.5-2015.4				0.01	49.26	93.14	6.47	50.65	128.43	Liu et al., 2017c
Shandong	Zibo	2008				0.30	193.00	160.00	92.90	455.50	1252.50	Li et al., 2010b
Tianjin	Industrial area	2015					64.54	226.58	44.32	4696.15	5846.24	Xing., 2017

Table S3. *RfD* and *SF* of the heavy metals in dustfall.

	As	Hg	Cd	Cr	Cu	Ni	Pb	Zn
RfD_{ing}	3.00E-04	3.00E-04	1.00E-03	3.00E-03	4.00E-02	2.00E-02	3.50E-03	3.00E-01
RfD_{inh}	3.01E-04	8.57E-05	1.00E-03	2.86E-05	4.02E-02	2.06E-02	3.52E-03	3.00E-01
RfD_{dermal}	1.23E-04	2.10E-05	1.00E-05	6.00E-05	1.20E-02	5.40E-03	5.25E-04	6.00E-02
RfD_{vapour}		8.57E-05						
SF_{inh}	1.5		6.30	42.0		0.84		