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Original Research

Analysis of Sources and Concentrations of Heavy Metal Contents in PM_{10} over a Four-Season Cycle in a Heavily Industrialised City in China

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Abstract

In order to study the sources and concentrations of heavy metals in atmospheric PM_{10}, a total of 144 samples were collected in two areas of Handan, China. The metal contents of Mn, Co, Ni, Cu, Cd, and Pb in the sample were analysed by inductively coupled plasma mass spectrometry. Contamination characteristics and sources were analysed by the geo-accumulation index and the enrichment factor method. Results indicated that heavy metal elements Mn, Cu, and Pb accounted for 96.9% of the total concentration. In addition, the seasonal levels of concentrations of the six elements from high to low were in the order of winter > fall > spring > summer. In both areas, the metal contents of Cd, Pb, and Cu were at levels 6, 4 and 2, respectively. Co and Ni were not considered polluting elements. The element Cd was mainly from anthropogenic sources, while the element Mn was mainly from natural sources. The enrichment factor values of Cu and Pb were between 10 and 100. The study revealed that pollution by heavy metals in industrialised cities is astounding, and leads to serious consequences for human health. Urgent measures are required to reinforce prevention and control of this pollution.

Keywords: PM_{10}, heavy industry, heavy metal, pollution assessment

Introduction

With the rapid development of China's economy, there has simultaneously been a marked increase in the level of PM_{10}, a major atmospheric pollutant [1-4]. A 2013 study conducted in the Beijing-Tianjin-Hebei region indicated that pollution and haze events occurred on approximately 60% of the days in the year, and the extent of pollution progressively worsened from north to south [5]. Handan City is located at the southernmost tip of the Beijing-Tianjin-Hebei region, bordering Henan province, and Handan was the fourth worst-polluted city in China in 2013 [6]. Various coal and steel industries

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are located in the city. These coal mining and energy industries, and iron and steel manufacturers contribute extensively to the heavy pollution in the city [7-8]. Haze events affect visibility and hinder economic development, but more importantly they lead to adverse consequences on human health [9]. A full understanding of the sources and concentrations of the pollutants as well as the extent of the associated harms are of critical importance in determining and implementing urgently needed prevention and control measures.

Particulate matter, such as PM_{10}, may contain heavy metal particles, which can be deposited in the body through respiration. Although the number of metal particles in the aerosol particles is very small (only approximately 1%) [10], they can cause bioenrichment and persistent toxicity in the environment [11], where particles slowly accumulate and persist invisibly for a long period of time [12]. PM_{10} can trigger diseases such as hyperthyroidism, mental disorders, hypertension, cardiovascular disease, and even cancer [13]. It can also damage the plasmid DNA structure [14].

Heavy metal pollutants in atmospheric particulate matter have been extensively studied. Huang et al. [15] self-developed a set of online monitoring systems to estimate the enrichment of the water-soluble elements Zn, Cd, Pb, and Cu and detect their concentration levels. In addition, Zhao et al. (2015) used Atomic Fluorescence Spectrometry (AFS) and inductively coupled plasma mass spectrometry (ICP-MS) to study the dry and wet deposition fluxes of atmospheric heavy metals in Jinan [16]. Wang et al. (2016) studied factors influencing heavy metals in $PM_{2.5}$ in certain Chinese provincial capitals [17]. Furthermore, Abuduwailil et al. (2015) studied atmospheric dust pollution caused by heavy metals and evaluated their effects on human health [18]. Chen et al. (2017) used the particle-excited X-ray emission technique to test the mass concentration of heavy metals in atmospheric particulates and analysed their pollution characteristics and sources [19]. However, studies on the pollution characteristics of heavy metals in heavily industrialised areas with excessive haze pollution are currently limited. Therefore, this study evaluated the pollution level of heavy metal elements in PM_{10} in Handan, a heavily industrialised city, with the aim of providing a reference to establish control measures for atmospheric pollution, particularly heavy metals.

Material and Methods

Sampling Techniques and Instruments

Measurements were performed at Hebei University of Engineering (the university campus) and in Baijia village (an industrial-residential area) in Handan, Hebei, China. Baijia village is located in the west of Handan City. Handan Iron and Steel Group is located 2 km southeast Baijia village – a typical industrial-residential area. Observational data, therefore, reflect the impact of heavy industrial pollution in the local environment. Hebei University of Engineering is southeast of Handan. Samples were taken on the roof of the university library in the main campus. There are no other high buildings located in the vicinity of the campus. The university canteen and the roads are certain distances away from the sampling spot. The map of sampling site and Handan Iron and Steel Group are provided in Fig. 1. Sampling was conducted over a four-season cycle (spring, summer, fall, and winter), and a total number of 144 samples were taken. The sampling periods within each season are listed in Table 1.

A KB-120E sampler was used to sample atmospheric particulate matter (PM_{10}). The samples were collected with Φ90-mm glass filter membrane, with a sampling

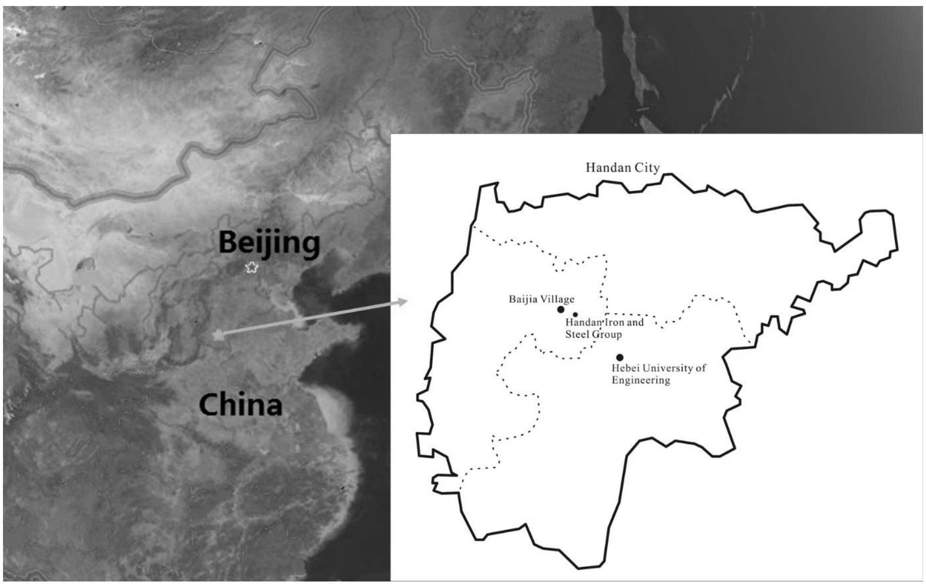

Fig.1. Map of sampling site and the Handan Iron and Steel Group.

Table 1. Sampling periods in each season.

Sampling location	Sampling season	Sampling period	Sampling height	Notes
University campus	Spring	2013.5.15-5.20	24 m	
	Summer	2013.7.2-7.8		
	Fall	2012.10.17-10.25		
	Winter	2013.1.7-1.9		
Industrial-residential area	Spring	2013.5.4-5.6	20 m	
	Summer	2013.8.14-8.18		
	Fall	2012.10.30-11.2		
	Winter	2013.1.17-1.18		After snow

flux of 120 L/min, and the sampler also recorded the sampling time and weather conditions. The filter membrane was replaced every 12 h.

Method

PM_{10} samples were weighed using an AB204-S-type 1/10000 precision balance. Prior to determining the heavy metal content, samples were processed by digestion methods. The sample filter was weighed, sheared into strips, and placed in a 100-mL beaker, and HNO_3 of 1% dilute was added prior to soaking overnight. A hot plate was then used to heat the strips until they were nearly dry. A mixture containing 20 mL of 70% HNO_3 and 10 mL of 72% $HClO_4$ was then added to the 100-mL beaker, and the samples were heated at a low temperature until they were completely dry. They were then cooled, and HNO_3 of 1% dilute filtered to a 50-mL polypropylene pipe was thoroughly shaken for testing.

Heavy metal elements were measured using an ICP-MS (X-series II) [20]. In this study, concentrations of six heavy metals (Mn, Co, Ni, Cu, Cd, and Pb) in PM_{10} were determined according to the general formula DZ/T0223-2001.

Results and Discussion

Analysis of PM_{10} Mass Concentrations

The PM_{10} mass concentrations in Handan ranged from 79.86 to 871.53 μg/m^3, with an average concentration of 254.04 μg/m^3. The average concentration in the university campus was 286.68 μg/m^3, and that in the industrial-residential area was 221.40 μg/m^3. According to the Ambient Air Quality Standard (GB 3095-2012) [21], the PM_{10} 24-h average concentration limit should be no higher than 150 μg/m^3, with an annual mean concentration limit of 70 μg/m^3 as the secondary standard. Therefore, during the sampling period, the percentage of the days exceeding the average mass PM_{10} 24-h average concentration limit was 77%. The highest measurement was 5.8 times that of Chinese National Ambient Air Quality Standards (grade-II), and the average annual concentration was 3.5 times that of grade-II.

It is suspected that these high levels of pollution are due to the location of the Beijing-Tianjin-Hebei region, which lies to the east of the Taihang Mountains. In this area, it lacks air movement conducive to the diffusion or dilution of pollutants. In addition, nearby Handan Iron and Steel Works and the coal mining industry aggravate the pollution levels.

PM_{10} concentrations were analysed by comparing those recorded in the university campus with those in the industrial-residential area (Fig. 2). The PM_{10} mass concentration seasonal levels at the two sampling points from high to low were in the order of winter > fall > spring > summer. PM_{10} concentrations in the industrial-residential area were higher than those in the university campus in spring, summer, and fall, and the PM_{10} concentration in the industrial-residential area was abnormal in winter and was only 41% of that in the university campus. Winter weather conditions are unfavourable to the diffusion and dilution of pollutants [22-23]. However, long-rang transport, local emissions, secondary formation, meteorological conditions,

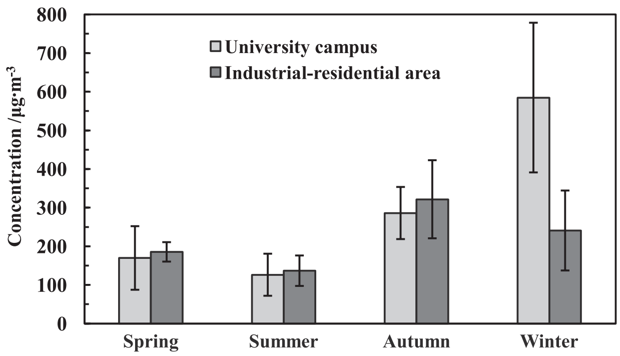

Fig. 2. Concentration distribution of PM_{10} in the university campus and the industrial-residential area.

Table 2. Comparison of mass concentrations of PM_{10} in Handan with those in other cities (μg/m³).

City		University campus	Industrial-residential area	Changsha	Dalian	Zhengzhou	China standard (grade-II)
24h average value	Min	56	57	-	24	31	150
	Max	871	458	249	308	584	
Average annual value		287	214	93	85	170	70

and other factors may all play a role affecting the pollution level. Therefore, the PM_{10} concentration is not always highest in winter. Samples from the industrial-residential area were taken immediately after snowfall. Concentrations in the village were lower in winter than in fall, as the air was somewhat purified [24]. Results indicate that pollution levels are generally higher in the industrial-residential area because of the presence of the Handan Iron and Steel Group.

A comparison of PM_{10} pollution levels was then carried out between the results from this current study and the data obtained from Changsha City (a city dominated by commercial activities), Dalian (a city with heavy shipbuilding industry), and from the serious pollution that occurred in Zhengzhou City in 2013 [25-27] (Table 2). Results showed that both the 24-h average level and the average annual level of PM_{10} in Handan were higher than those in all these other cities, and that the average annual level were more than twice those of Dalian. This finding shows the impact of heavily industrialised pollution in a city and indicates that, for improvements of air quality, pollution levels in such areas need to be reduced first.

Characteristics of Heavy Metal Pollutants

The concentrations of heavy metals in PM_{10} in Handan were (from high to low) in the order of Pb, Mn, Cu, Cd, Ni, and Co. The total concentration of all six elements was 6347.47 ng/m³. Concentrations of element Pb, Mn, and Cu accounted for 96.9% of the total concentration. Concentrations of Pb, Mn, Cu, Cd, Ni, and Co ranged from 236.11 to 825.89 ng/m³, 110.24 to 494.56 ng/m³, 62.76 to 267.62 ng/m³, 3.15 to 30.07 ng/m³, 1.91 to 26.15 ng/m³ and 0.65 to 3.57 ng/m³, respectively. In addition, the annual average concentrations of elements Pb and Cd in PM_{10} were 411.21 and 11.68 ng/m³, respectively. By the Ambient Air Quality Standard, the concentration limits of Pb and Cd are 500 and 5 ng/m³, respectively. Therefore, the annual average concentration of Pb in the sample was lower than the standard value, but that of Cd was 1.3 times higher. Pb and Cd are carcinogenic heavy metals, and Cd, when it exceeds advisable limits also causes pulmonary fibrosis and acute and chronic renal disease. It is an invisible killer of humans [28]. WHO (World Health Organization) stipulates that the limits of Ni and Mn in the atmosphere should be 25 and 150 ng/m³, respectively. In this respect, the Mn content in PM_{10} in Handan reaches 67% of the limit, and Ni level is below the limits.

The concentrations of heavy metals in PM_{10} were distributed differently throughout the various seasons. Fig. 3 represents the average mass concentration distribution characteristics of Mn, Co, Ni, Cu, Cd, and Pb in PM_{10} in Handan.

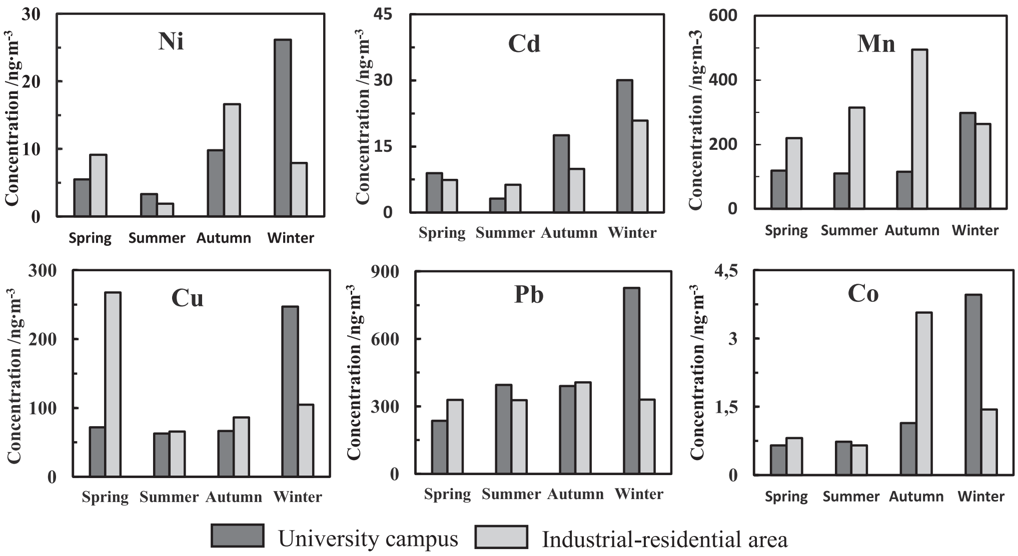

Fig. 3. Distribution of mass concentrations of heavy metals in PM_{10} over four seasons.

Table 3. Comparison of heavy metal content in PM_{10} between Handan and other cities (ng/m^3).

Element	This study	Tianjin	Shenyang	Baotou	Taiyuan	Jinan
Cd	13.02	10.9	1.87	0.29	3.8	-
Cu	121.63	254	56.4	115.1	72.3	40
Pb	405.19	504	346	104.6	559.9	200
Mn	241.94	-	-	33.99	257.8	110

The figure shows that in spring and fall, Mn, Co, Ni, Cu, and Pb over higher in the industrial-residential area than in the university campus. Ni, Cu, and Mn concentrations were higher in spring, and the Cu concentration was 2.7 times higher. Mn, Ni, and Co concentrations were higher in fall, and Mn and Co concentrations were 2.1 times higher than those within the university campus. In addition, in summer, Mn, Cd, and Cu concentrations were higher in the industrial-residential area than at the university campus. However, in winter, concentrations of all elements were lower in the industrial-residential areas than in the university campus due to the occurrence of rain and snow during the sampling period. A comparison of heavy metal concentrations between the university campus and the industrial-residential area showed that concentrations of Ni and Cd elements follow the general rule of high in winter and low in summer (winter > fall > spring > summer), and the maximum concentration was close to 30 ng/m^3. Mn and Cu concentrations were lower in the university campus than in the industrial-residential area in the spring, summer, and fall. Mn concentrations in the industrial–residential area were low in fall and high in spring, and Cu concentrations were low in spring and summer. Pb concentration in the university campus was as high as 825.89 ng/m^3 in winter but was stable at 300 ng/m^3 in the other three seasons. The Co concentration was the lowest. It increased in the industrial-residential area in fall and increased in the university campus only in winter.

In summary, the industrial-residential area was more polluted than the university campus. Intense industrial production was a major contributor to heavy metal pollution, and the seasonal levels of the six heavy metals change from high to low in the order of winter > fall > spring > summer.

Table 3 shows a comparison of heavy metal element concentrations in several major cities in China [29-32]. Cd concentration was found to be the highest in Handan (45.5 times the concentration in Baotou). Cu and Pb concentrations were lower in Handan than in Tianjin. And Cu concentration was higher in Handan than in any other city except Tianjin. In Tianjin, Shenyang, Taiyuan, and Jinan, Pb concentration was higher than concentrations of other elements, consistent with the study results. Pb and Mn concentrations in Taiyuan were higher than those in other cities, and Handan had the second-highest concentrations. It is evident that heavy metal pollution (Mn, Cu, Cd, and Pb) in PM_{10} is more serious in Handan than in other cities in China.

Evaluation of Heavy Metal Pollution by Geo-Accumulation Index

The geo-accumulation index (I_{geo}) was used to evaluate the level of heavy metal pollution in PM_{10} in the two study areas, namely the university area and the industrial-residential area in Handan City [33-35]. The formula for the I_{geo} is

$$I_{geo} = \log_2 (C_n/1.5B_n) \tag{1}$$

…where C_n is the mass concentration of the n^{th} element in the sample; B_n is the background concentration of n^{th} element [36], and 1.5 is the correction index.

The results of the geo-accumulation index method can be divided into seven grades, with levels

Table 4. I_{geo} index of heavy metals in PM_{10} over the four seasons in Handan.

	University areas				Average & Level	Industrial living areas				Average & Level
	Spring	Summer	Fall	Winter		Spring	Summer	Fall	Winter	
Ni	-2.88	-3.61	-2.04	-0.63	-2.29&0	-2.14	-4.40	-1.28	-2.35	-2.54&0
Cd	5.94	4.44	6.91	7.96	6.24&6	5.67	5.44	6.09	7.16	6.09&6
Mn	-2.88	-2.99	-2.92	-1.55	-2.58&0	-1.99	-1.48	-0.82	-1.73	-1.50&0
Cu	1.09	0.89	0.97	2.87	1.45&2	2.98	0.96	1.35	1.63	1.73&2
Pb	2.60	3.34	3.32	4.40	3.42&4	3.08	3.07	3.38	3.08	3.15&4
Co	-4.87	-4.71	-4.06	-2.49	-4.03&0	-4.56	-4.87	-2.41	-3.73	-3.89&0

Table 5. Enrichment factors of heavy metals in PM_{10} over the four seasons in Handan.

Element	Industrial living areas				University areas			
	Spring	Summer	Fall	Winter	Spring	Summer	Fall	Winter
Mn	0.8	5.5	1.0	1.1	0.7	1.1	0.4	0.4
Cu	23.4	27.5	4.2	10.6	10.5	15.2	5.4	7.6
Cd	165.4	674.2	122.1	537.0	332.6	194.2	365.2	234.7
Pb	36.0	171.7	24.5	41.7	43.1	119.5	39.8	31.6

from 0 to 6 to indicate a pollution degree from zero to strong. The results of calculations are shown in Table 4. According to the calculations, the I_{geo} values of the elements Ni and Co in PM_{10} in Handan were less than 0, which is equivalent to zero pollution. The I_{geo} value of the element Cd was higher than 5 in fall and winter (with a maximum of 7.96), which is equivalent to extremely strong pollution level 6. The I_{geo} value of the Cu element ranged between 1 and 2, where the value of 2 indicates moderate pollution. However, the I_{geo} value of Cu in the winter in the university campus was close to three times that of the other three seasons, thereby indicating that winter industrial emissions and coal-burning heating are significant contributors.

The mean I_{geo} values of Mn and Pb were -2.04 and 3.29. Mn and Pb are contaminated according to the high standard used. A comparison between the university campus and the industrial-residential area showed that the I_{geo} value of heavy metals in the university campus was slightly higher. This is due to the low I_{geo} value of various elements in winter in the industrial-residential areas as a consequence of the rain and snow effects.

Analysis of Heavy Metal Pollution Sources

To analyze and evaluate the contributions of anthropogenic and natural sources to atmospheric pollution, the element enrichment factor (EF) was used [37-38], where the formula for the EF is

$$EF_i = (C_i/C_n)_{sample}/(C_i/C_n)_{background} \quad (2)$$

…where C_i is the concentration of the studied element I, and C_n is the concentration of the reference element (this study selected Ni as the reference element).

To obtain the background element concentration, the Chinese soil element background value was consulted. A concentration factor of heavy metals that is less than 10 indicates that the element is obtained mainly from a natural source; a value between 10 and 100 indicates the element is enriched at a different degree, and an EF higher than 100 indicates the element is mainly related to an anthropogenic source [39]. Table 5 shows the EFs of four types of heavy metal elements (Mn, Cd, Cu, and Pb) in PM_{10} in Handan.

It is evident from Table 5 that the EF values of Cd, Cu, and Pb elements in the PM_{10} in this heavily industrial city were all more than 10, and, therefore, enrichment is obvious. The EF value of Cd element was more than 100, indicating that anthropogenic sources are the main contributors. The EF value of the Cu element was less than 10 in the fall, whereas it was higher than 10 in the other three seasons, thereby indicating that it may be related to anthropogenic sources. The EF value of the Pb element ranged from 10 to 100, and it is undoubtedly related to iron and steel smelting, motor vehicles, coal-fire emissions, and other sources in Handan City. The EF value of the Mn element was less than 10, which implies it is obtained mainly from a natural source and relates to crustal material. In summer and winter, the element EF values in the industrial–residential area were higher than those in the university campus, and EF values of Cu were also higher than that in the university area in spring. This is also true for the Mn element in spring and fall. This result indicates that an anthropogenic heavy metal source contributes to heavy metal enrichment, particularly for Cu.

Conclusions

The observations and analyses conducted in this study indicated that the mass concentration of PM_{10} over a four-season cycle in Handan ranged from 79.86 to 871.53 μg/m^3, and PM_{10} concentration levels exceeded the national average 24-h standard values on 77% of the days. Heavy metal elements were mainly Mn, Cu, and Pb, which accounted for 96.9% of the total element contribution. The Pb concentration was the highest; the Co concentration was the lowest; and the overall seasonal from high to low was winter > fall> spring> summer. For both daily and annual average values, the PM_{10} concentration in Handan City was higher than those in other major cities in China. The heavy metal pollution from elements Cd, Cu, and Pb is extremely serious, particularly in Handan City, and needs urgent attention. A calculation of the I_{geo} value of heavy metals showed that Cu and Cd contamination in the PM_{10} in heavily industrialized cities is serious. The element Cd in Handan reached Level 6, indicating extremely strong pollution, and the element Cu reached Level 2, indicating medium pollution. Levels of the elements Mn and Pb also indicated atmospheric contamination, while Co and Ni were considered non-polluting.

The results of EF analyses showed that EF values of heavy metals elements Cd, Cu, and Pb over the four seasons in Handan were higher than 10. In addition, the EF value of Cd was more than 100, which implies that its presence is mainly due to human activities and, therefore, it is the heavy industries which make a contribution to the heavy metal pollution in this urban atmosphere. However, the EF value of the Mn element was less than 10, indicating that it is from mainly a natural source (crustal material).

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Conflict of Interest

The authors declare no conflict of interest.

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