

Article

# Characteristics and Sources of Heavy Metals in PM<sub>2.5</sub> during a Typical Haze Episode in Rural and Urban Areas in Taiyuan, China

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Received: 16 November 2017; Accepted: 19 December 2017; Published: 21 December 2017

**Abstract:** PM<sub>2.5</sub> samples were collected in the rural and urban areas of Taiyuan, China during a typical haze episode and the heavy metals (Cr, Mn, Ni, Cu, Zn, As, Cd and Pb) in PM<sub>2.5</sub> were analyzed. The haze was characterized by start-up stage with a daily mean PM<sub>2.5</sub> of 149.34 ± 52.33 and 146.73 ± 18.96 μg m<sup>-3</sup> in the rural and urban sites, respectively, a peak stage (288.20 ± 12.43 and 323.44 ± 5.23 μg m<sup>-3</sup>), and a weakening stage (226.59 ± 12.43 and 195.60 ± 2.93 μg m<sup>-3</sup>). The concentrations of PM<sub>2.5</sub> in the rural and urban sites in the peak stage were 5.9 and 5.5 times higher than those in the normal stage, respectively. The order of concentrations of heavy metals in PM<sub>2.5</sub> at the rural and urban sites were the same and are listed as follows: Zn > Pb > Mn > Cr > Cu > Ni > Cd > As. Pb at the rural site, As at the urban site, and Cd at the both sites failed to meet the air quality standard. The concentrations of Pb and Zn were higher at the rural site than those at the urban site. Principal component analysis indicated that the main sources of heavy metals for the rural area were raw coal combustion and soil/road dust, and for the urban area were coal combustion/industrial emissions, road/soil dust, and vehicle emissions/oil combustion.

**Keywords:** heavy metals; haze; PM<sub>2.5</sub>; rural area; Taiyuan

## 1. Introduction

Currently, large-scale regions in North China are suffering from severe haze episodes [1–5], which are essentially caused by fine particles with aerodynamic diameter less than 2.5 μm (PM<sub>2.5</sub>) [3,6,7]. This has attracted great scientific and public attention [8–13]. PM<sub>2.5</sub> can directly reduce atmospheric visibility by scattering and absorbing the sunlight [14,15]. In addition, PM<sub>2.5</sub> can affect the amount of solar ultraviolet radiation reaching the Earth's surface [16–18], the nutrient balance and acidity of soil [19,20], and climate change [21]. Chronic exposure to PM<sub>2.5</sub> is associated with a wide range of diseases [4,10], mainly since atmospheric PM<sub>2.5</sub> can carry varieties of pathogenic components, such as heavy metals, viruses, and bacteria. Previous studies have demonstrated the adverse health effect relationship between particulate matter exposure and toxic heavy metal elements [22–25]. Epidemiological studies carried out over the last decades have also demonstrated a strong relationship between exposure to elevated toxic heavy metal elements in ambient air and increased mortality and

morbidity [23,26–28]. Heavy metals, such as class I carcinogenic contaminants (such as As (regarded as metal here), Ni, Cd, and Cr), class II(B) carcinogenic (such as Pb inorganics), and non-carcinogenic contaminants (such as Cu, Zn, and Mn) [29], in atmospheric PM<sub>2.5</sub> can be enriched in the human body via inhalation, dermal absorption, and ingestion [30–32]. For example, to evaluate the exposure to heavy metals in the industrial area of Taranto, Southern Italy, Vimercati et al. [33,34] found high urinary concentrations of heavy metals (As, Cr, Mn, Hg, and Pb), which could be related to the presence of industrial plants. Subsequently, heavy metals in the human body would result in serious allergic respiratory disease and have a higher excess cancer risk to the exposed population [35,36]. For example, Pb, As, and Ni are considered to play etiological roles in some diseases, especially heart disease, stroke, kidney disease, and several blood diseases [37]. As and Cd may play a role in the development of dysglycemia (elevated fasting plasma glucose, impaired fasting glucose, and diabetes) [23].

Many studies on PM<sub>2.5</sub> have been conducted, and most studies in China have focused on emissions from traffic and industry in megacities and developed economic areas (e.g., the Beijing-Tianjin-Hebei community, the delta region of Yangtze River and Pearl River, etc.) [3,7,8,12,15,22,38–47]. However, the pollution by heavy metals in PM<sub>2.5</sub> in China's rural areas have rarely been reported. Liu et al. [3] recently reported that the periodic emissions from farmers' activities in the North China Plain possibly contributed to atmospheric water-soluble ions in Beijing. With the enhancement of farmers' incomes in recent years, coal (especially raw coal) consumption by farmers in the north China is rapidly increasing to keep their houses warm in cold seasons [48], and hence the serious air pollution in rural areas of the north China should be paid great attention.

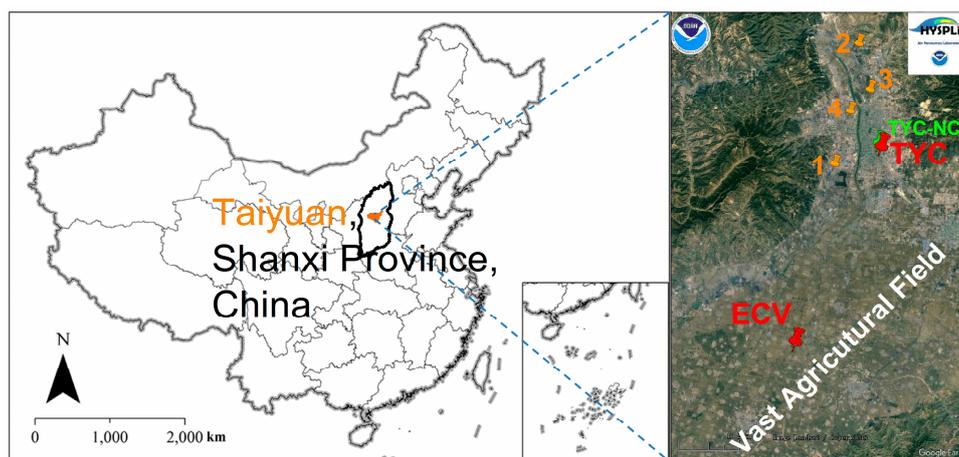
In the present study, filter samples of PM<sub>2.5</sub> were collected simultaneously during a typical haze episode in both rural and urban areas in Taiyuan, which was ranked as the most air-polluted city in China. The concentrations of Cr, Mn, Ni, Cu, Zn, As, Cd, and Pb in PM<sub>2.5</sub> were quantified and source-identified. This study will be helpful for future control measures in reducing pollutant emissions from rural areas.

## 2. Experiments

### 2.1. Description of the Study Area

Taiyuan, a typical city of heavy reliance on coal as a primary source of heat and power, is located in North China with a population of approximately 4.2 million. Its rural and urban land areas are respectively ~5500 km<sup>2</sup> and ~1500 km<sup>2</sup>, and the terrain resembles a bowl surrounded by hills in its west, north, and east directions [49,50]. The southern plain of Taiyuan is a vast agricultural field where corn and wheat are mainly planted. The heating period in the urban region is usually five months (from November to March), while the raw-coal (even peat) burning in rural areas routinely lasts more than six months for heating and almost the whole year for cooking.

The rural sampling site in this work was selected on the rooftop of a field station (~5 m above ground) which is located in the vast agricultural field of Echi village (here referred to as ECV, 37.48° N, 112.41° E) in the south of Taiyuan. The ECV site is far away from industries, traffic, and other commercial emissions and represents the agricultural countryside. The urban sampling site in Taiyuan city (here referred to as TYC, 37.81° N 112.575° E) was on the rooftop (~27 m above ground) of a residential building, which stands in central urban area of Taiyuan and is about 0.9 km away from a national controlling air sampling site (TYC-NC, 37.8195° N, 112.57° E, ~25 m). There are two power plants and two large industrial facilities upstream of the urban site (TYC), such as the First and Second Power Stations, Taiyuan Iron and Steel Factory, and Taiyuan Heavy Machinery Factory, respectively. The detailed locations are presented in Figure 1.



**Figure 1.** Locations of the sampling sites and the main industries (ECV and TYC represent the sampling sites at Echi village and Taiyuan city, respectively; TYC-NC is a national controlling air sampling site near TYC; 1, 2, 3, and 4 are the First and Second Power Stations, Taiyuan Iron and Steel Factory, and Taiyuan Heavy Machinery Factory, respectively).

## 2.2. Sampling

PM<sub>2.5</sub> samples were collected on prebaked (600 °C, 4 h) quartz microfiber filters (90 mm, Munktell Ahlstrom, Falun, Sweden) for 24 h (starting at approximately 07:00 a.m.) by a PM<sub>2.5</sub> sampler (KC-6120) at a flow rate of 100 L min<sup>-1</sup>. Before and after sampling, the filters were equilibrated for 24 h in a chamber with a constant temperature (~23 °C) and relative humidity (~50%) and then weighed by electronic microbalance (FA1004, Lichen, China). Each sample was finally stored in a dedicated filter container (100 mm) and preserved in a refrigerator (−10 °C) until analysis. Samples were collected from 7 to 22 November 2016. The wind speed and direction data were monitored at the TYC site by an anemometer (ZCF-5, Sipeik, China). The hourly PM<sub>2.5</sub> concentrations at TYC-NC were obtained from <https://data.epmap.org/>.

## 2.3. Analysis of Heavy Metals

A quarter of the sample filter was cut into pieces and then dissolved in the mixtures of HNO<sub>3</sub> (9 mL, 65%) and HClO<sub>4</sub> (3 mL, 70%). After standing for 24 h, the solution was heated with a hotplate to a boil and the temperature was maintained until the solution had nearly evaporated. After cooling to room temperature, the residue was dissolved with 10 mL of 6.5% HNO<sub>3</sub> and diluted in a 25-mL flask with ultrapure water. Then, the diluted solutions were transferred to numbered polyethylene vials and stored in a refrigerator (4 °C) for later analysis. Cr, Mn, Ni, Cu, Zn, As, Cd, and Pb were measured using an inductively coupled plasma optical emission spectrometer (ICP-OES, Agilent 5100). One blank filter (unexposed) was analyzed similarly for nine filter samples; the blank level was below the detection limits and therefore ignored.

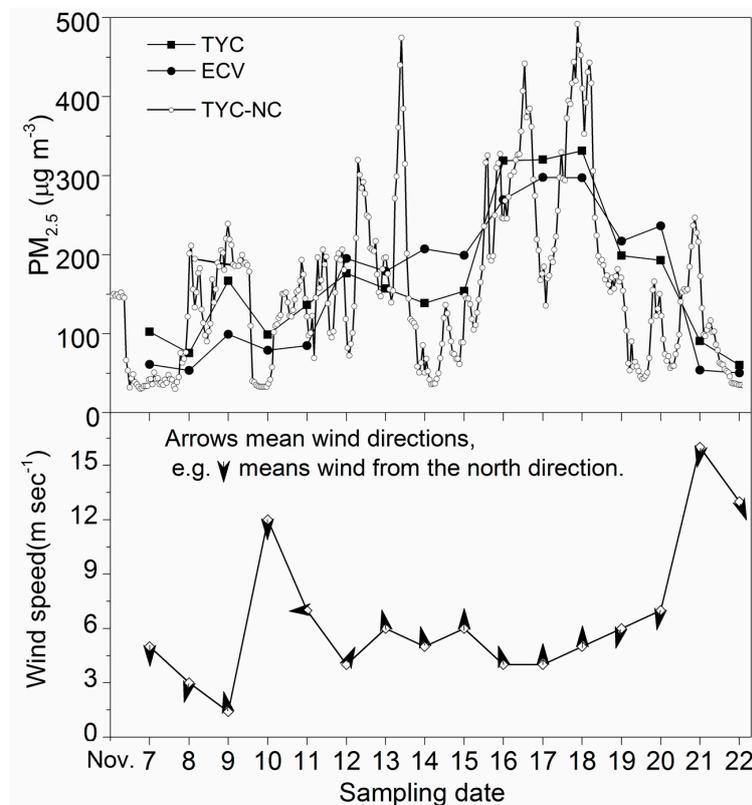
## 3. Results and Discussion

### 3.1. Levels of PM<sub>2.5</sub>

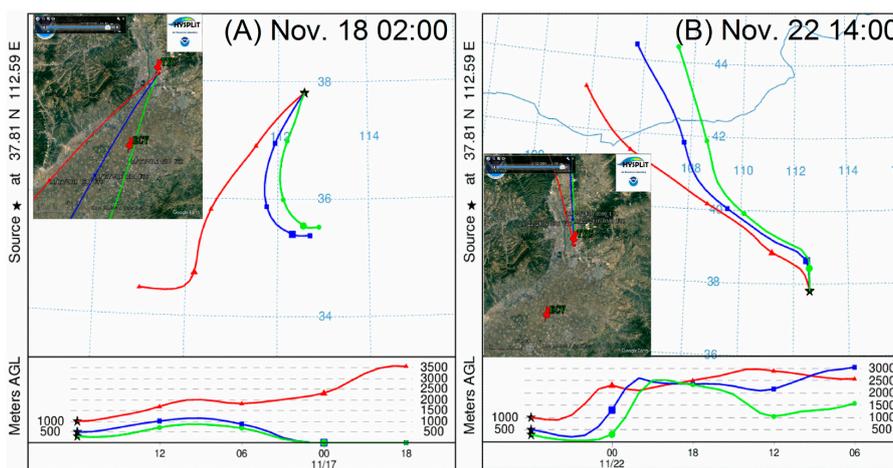
The levels of PM<sub>2.5</sub> concentrations at the two sampling sites (ECV and TYC) and the hourly PM<sub>2.5</sub> concentrations at TYC-NC were integrated into Figure 2 with wind speeds and directions. It is evident that the daily variation of PM<sub>2.5</sub> at the rural and urban sites exhibited similar trends, but was roughly opposite to wind speed. Although some dissimilar values existed, such as those recorded on 14, 19, and 20 November, the temporal variation of PM<sub>2.5</sub> at TYC was consistent with TYC-NC. All of the daily PM<sub>2.5</sub> levels both in the rural and urban sites failed to meet the air quality

limits ( $35 \mu\text{g m}^{-3}$  in standard I) set by the Ministry of Environmental Protection of China (MEPC). The haze episode can be divided into the normal stage and three haze stages (i.e., start-up stage, peak stage, and weakening stage). In the normal stage (i.e., from 7 to 8 November), the daily concentration ranges were  $53.23\sim 61.17 \mu\text{g m}^{-3}$  and  $75.55\sim 102.43 \mu\text{g m}^{-3}$  at the rural and urban sites, respectively. The concentrations of  $\text{PM}_{2.5}$  at the urban site were markedly higher than those at the rural site. In the start-up stage (i.e., from 9 to 15 November), the concentration ranges were  $78.81\sim 207.01 \mu\text{g m}^{-3}$  and  $98.79\sim 176.01 \mu\text{g m}^{-3}$  at the rural and urban sites with the mean staged concentrations of  $149.34 \pm 52.33 \mu\text{g m}^{-3}$  and  $146.73 \pm 18.96 \mu\text{g m}^{-3}$ , respectively. Comparing the mean concentrations at the two sites during the start-up stage, there is no significant difference. However, as shown in Figure 2, the concentrations of  $\text{PM}_{2.5}$  were lower at the rural site than at the urban site from 9 to 11 November, and the opposite phenomena were observed from 12 to 15 November, which suggested that local emissions of  $\text{PM}_{2.5}$  were the main sources. Wind with a speed of  $5.9 \pm 2.0 \text{ m sec}^{-1}$  was frequently from the south. The peak stage was from 16 to 18 November and the daily  $\text{PM}_{2.5}$  concentrations were  $269.55\sim 297.71 \mu\text{g m}^{-3}$  and  $318.66\sim 331.29 \mu\text{g m}^{-3}$  at the rural and the urban sites, respectively. Especially from the data monitored at the national controlling air sampling site of China, the hourly  $\text{PM}_{2.5}$  concentration at the urban area was as high as  $492 \mu\text{g m}^{-3}$  in 18 November when Taiyuan was ranked as the most air-polluted city in China. Wind with a speed of  $4.3 \pm 0.4 \text{ m sec}^{-1}$  was still from south. In the weakening stage (i.e., from 19 to 20 November), the daily concentrations were  $217.02\sim 236.15 \mu\text{g m}^{-3}$  and  $192.67\sim 198.53 \mu\text{g m}^{-3}$  at the rural and urban sites, respectively. Then, the  $\text{PM}_{2.5}$  concentrations returned to the normal levels from 21 to 22 November, and the daily concentrations were in the ranges of  $50.39\sim 53.97 \mu\text{g m}^{-3}$  and  $60.31\sim 90.58 \mu\text{g m}^{-3}$  at the rural and urban sites. The concentrations of  $\text{PM}_{2.5}$  at the rural and urban sites in the peak stage were about 5.9 and 5.5 times higher than those in the normal stages. The  $\text{PM}_{2.5}$  levels at the rural area were lower than those at the urban area in the normal stages and the peak stage, while the former were higher than the latter in the start-up stage and weakening stage, which suggested that, except for the contribution of industrial and traffic emissions, the emissions of  $\text{PM}_{2.5}$  from the rural area were also suspected to contribute to the atmospheric  $\text{PM}_{2.5}$  in Taiyuan city when wind came from the south. To reveal the influence of the air mass transport on  $\text{PM}_{2.5}$  in Taiyuan city, 24-h backward trajectories for clusters in the peak stage (18 November) and the normal stage (22 November) in Taiyuan were analyzed, and the results are illustrated in Figure 3.  $\text{PM}_{2.5}$  concentrations were accumulated when winds were from the south with a low speed, while wind from north diluted the pollution. As shown in Figure 3, in the start-up and the peak stages, winds from south swept through the southern plain and blew pollutants to the north, where the mountains in its west, north, and east directions slowed down their spreads, causing pollutants to descend upon the urban area. In the normal stage (e.g., 22 November), the concentration of  $\text{PM}_{2.5}$  declined sharply due to the winds from the north. Except for the weather and geographical conditions, the extremely high levels of  $\text{PM}_{2.5}$  during the haze episodes revealed strong sources of the pollutants around Taiyuan. According to Figure 2, in the start-up and peak stages, the daily concentrations of  $\text{PM}_{2.5}$  varied greatly while the wind direction with a low speed did not change rapidly during 13 to 18 November, which suggested that the local emissions of  $\text{PM}_{2.5}$  at the two sites played a pivotal role. In addition, there is no lag between ECV and TYC sites in terms of  $\text{PM}_{2.5}$ , suggesting that the local emissions of  $\text{PM}_{2.5}$  at both sites are main sources. The residents in the rural area frequently and randomly increased coal burning for heating due to colder weather, while the burning amount of coal for central-heating and industries in the urban area remained stable during the short period, which was suspected to be the main reason for the significant differences between the two sites during the start-up and peak stages. Although residential coal consumption in the rural area only accounts for a small fraction of the total (e.g., ~4 tons raw coal per year used for cooking and heating in each household according to our previous survey), the emission factor of  $\text{PM}_{2.5}$  from farmers' coal stoves (about  $1054\sim 12,910 \text{ mg kg}^{-1}$ ) are usually about 1~3 orders of magnitude greater than those from coal power plants or industry boilers (about  $16\sim 100 \text{ mg kg}^{-1}$ ) [3]. The activities of humans, such as coal burning for heating and cooking, were also suspected to make a

significant contribution to the deterioration of the regional air quality in winter in other rural areas in the North China Plain [3]. The most serious pollution events (or haze days) in the North China Plain have usually been coincident with the activities of farmers in recent years [3,39,41].



**Figure 2.** The concentration variation of  $PM_{2.5}$  and wind speeds and directions during the sampling period.



**Figure 3.** The 24-h backward air-mass trajectories in the peak haze stage (A) and in the normal stage (B).

### 3.2. Levels of Heavy Metals in $PM_{2.5}$

The daily variation of heavy metals in  $PM_{2.5}$  at the ECV and TYC during the sampling period is illustrated in Figure 4. It is evident that the concentrations of the heavy metals in  $PM_{2.5}$  also varied greatly during the sampling period. Comparing Figure 4 to Figure 2, the temporal variation of heavy

metals was similar to that of PM<sub>2.5</sub> with correlation coefficients ( $R^2$ ) of 0.925 at the rural site and 0.885 at the urban site (Figure 5), respectively, indicating that the concentration of heavy metals in PM<sub>2.5</sub> measured could be used as an indicator of the pollution level in the rural and urban areas.

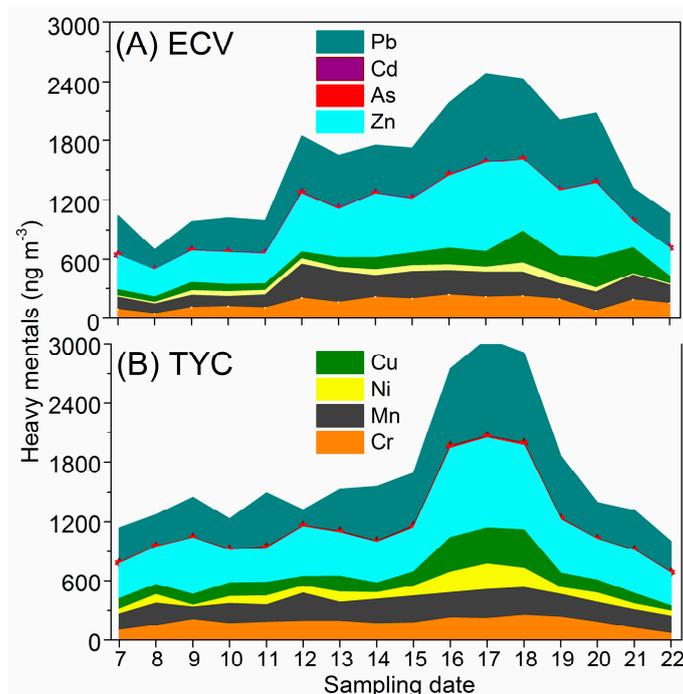


Figure 4. Variation of heavy metals in PM<sub>2.5</sub> at ECV and TYC.

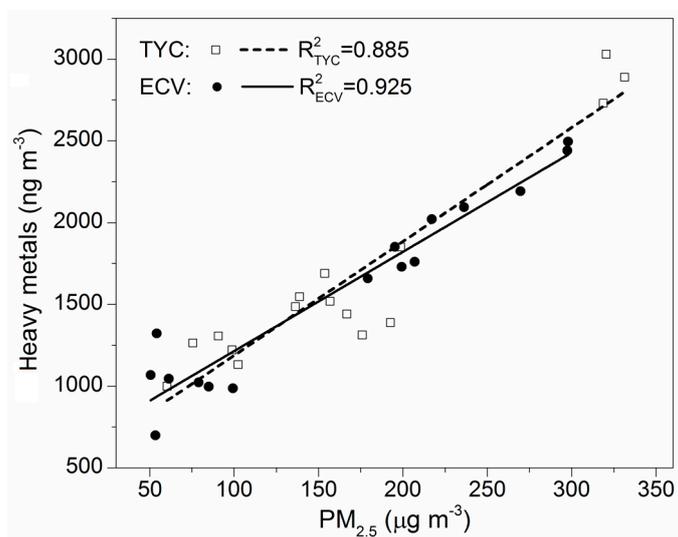


Figure 5. The correlation between PM<sub>2.5</sub> and the sum of metal mass measured at ECV and TYC.

The mean concentrations of heavy metals in different stages determined in this study and those reported in other cities in China are summarized in Table 1. Although some essential directives mitigating the effects of traffic and industrial pollutants were announced and implemented in megacities and industrial areas in the past decade [10], heavy metals pollution in PM<sub>2.5</sub> in this work was still found to be very serious during the sampling days. As shown in Table 1, it is evident that the levels of most heavy metals in PM<sub>2.5</sub> in the present study were consistent with those reported in

Taiyuan by other researchers [51,52], but much higher than most of those reported in other cities of China [38,42,45,53–56]. In particular, the concentrations of some carcinogenic contaminants were much higher than those reported in other cities. For example, the concentrations of Cr in this work were 1–3 orders of magnitude greater than those reported in other cities. However, the concentrations of Zn, As, and Cd in PM<sub>2.5</sub> in the present study were lower than those in Guangzhou city [46] and the concentration of each heavy metal in this study was significantly lower than that in Foshan city [47].

In this study, Pb in the rural area, As in the urban area, and Cd in both the rural and urban areas failed to meet the air quality standard (500 ng m<sup>-3</sup>, 5 ng m<sup>-3</sup>, and 6 ng m<sup>-3</sup>, respectively) set by MEPC. As shown in Table 1, Zn and Pb were the most abundant metals in PM<sub>2.5</sub> in both the rural and urban areas. The orders of concentrations of heavy metals in PM<sub>2.5</sub> at the rural and urban sites were the same and are listed as follows: Zn > Pb > Mn > Cr > Cu > Ni > Cd > As. The concentration of Ni in the rural area was less than half of that in the urban region. Zn and Pb were slightly higher in the rural area than those in the urban region and the others in the rural area were slightly less than those in the urban area. These may have been ascribed to several factors, including topography and meteorological conditions as well as pollution sources. The sources of heavy metals will be discussed in the next section.

**Table 1.** Concentrations (ng m<sup>-3</sup>) of heavy metals (mean concentrations and standard deviations) in PM<sub>2.5</sub> in different haze stages at ECV and TYC and other cities of China.

Sites		Cr	Mn	Ni	Cu	Zn	As	Cd	Pb	Ref.	
Taiyuan	Normal stage	ECV	116.17 ± 50.87	165.05 ± 52.71	13.21 ± 1.09	114.96 ± 75.72	301.93 ± 29.40	1.64 ± 0.51	1.80 ± 0.65	319.64 ± 58.56	This study
		TYC	107.31 ± 26.06	188.77 ± 18.19	58.69 ± 11.02	87.93 ± 18.61	378.97 ± 33.06	6.21 ± 1.76	4.17 ± 1.51	343.88 ± 28.79	This study
	Start-up stage	ECV	157.04 ± 40.73	215.31 ± 82.99	50.61 ± 6.34	93.66 ± 23.12	466.82 ± 121.92	4.58 ± 1.25	6.89 ± 1.48	435.19 ± 98.36	This study
		TYC	181.11 ± 12.22	213.47 ± 44.91	71.78 ± 19.47	124.31 ± 22.52	438.74 ± 61.53	5.57 ± 1.51	9.34 ± 4.11	415.22 ± 108.13	This study
	Peak stage	ECV	223.01 ± 7.54	248.67 ± 2.34	68.53 ± 16.32	219.14 ± 71.71	789.96 ± 76.55	7.52 ± 0.62	5.98 ± 2.84	813.91 ± 57.08	This study
		TYC	231.52 ± 13.51	272.95 ± 14.46	216.59 ± 25.78	357.98 ± 12.77	884.71 ± 21.82	12.15 ± 1.21	11.10 ± 1.17	896.68 ± 73.08	This study
	Weakening stage	ECV	132.70 ± 58.11	170.61 ± 17.73	60.52 ± 16.08	259.19 ± 48.56	715.47 ± 45.81	5.34 ± 0.38	5.94 ± 2.13	707.60 ± 2.96	This study
		TYC	206.48 ± 25.82	213.64 ± 13.85	76.25 ± 11.86	137.88 ± 8.28	480.39 ± 61.80	6.46 ± 3.24	2.93 ± 0.73	495.73 ± 129.86	This study
	All stages	ECV	156.15 ± 51.26	203.41 ± 65.28	45.85 ± 17.38	143.21 ± 72.88	517.27 ± 182.65	4.49 ± 1.92	5.33 ± 2.65	511.37 ± 182.18	This study
		TYC	175.28 ± 37.02	218.47 ± 40.13	96.22 ± 45.82	160.72 ± 73.97	512.62 ± 151.22	7.07 ± 2.73	7.60 ± 4.01	497.72 ± 181.17	This study
	Taiyuan city		90.4	313.3	28.3	38.3	/	/	3.4	489.1	[51]
	Taiyuan city		185.05 ± 45.99	/	91.45 ± 15.87	142.77 ± 9.87	/	/	11.58 ± 8.91	472.17 ± 156.46	[52]
Beijing city (Winter)		30	70	40	200	310	10	0	150	[45]	
Shanghai city (Winter)	Industrial district	49	134	39	43	775	/	6	263	[38]	
	Commercial district	7	62	6	13	235	/	1	64	[38]	
Taichung city		33.5 ± 48	19.1 ± 20.5	11.8 ± 29.9	11.5 ± 19.9	177.8 ± 103.00	/	4.3 ± 8.5	283.1 ± 252.9	[53]	
Jinan city		10	60 ± 30	10	30 ± 20	350 ± 200	40 ± 40	/	160 ± 80	[54]	
Hangzhou city		13	726	7	76	651	/	/	288	[55]	
Hong Kong		1	10	4	5	200	/	/	/	[56]	
Nanjing city (Gulou, September)		22.5 ± 7.8	48.3 ± 6.4	6.7 ± 3.7	65.7 ± 32.1	230.2 ± 69.3	4.3 ± 2.3	1.3 ± 0.1	81.5 ± 12.0	[42]	
Guangzhou city (Urban center)		51 ± 11	39 ± 8	33 ± 7	109 ± 23	680 ± 146	81 ± 17	14 ± 3	373 ± 80	[46]	
Foshan city (Winter)		/	200.6 ± 129.4	/	283.8 ± 174.7	2214 ± 1438	76.6 ± 49.1	42.6 ± 45.2	675.7 ± 378.5	[47]	

### 3.3. Source Identification

Principal component analysis (PCA) has been widely used to identify the major sources of air pollutant emissions [1,42,43,57,58]. PCA applies projection dimension reduction methods to convert several indicators into significant representative indicators without the damage of the original data [57,58]. In this study, source identification of heavy metals in PM<sub>2.5</sub> was performed by using PCA (IBM SPSS, version 22). Only components with an eigenvalue greater than 1 were retained. The PCA results of heavy metals in daily PM<sub>2.5</sub> during the sampling period are presented in Table 2.

**Table 2.** Principal component analysis (PCA) results of heavy metals in PM<sub>2.5</sub> at ECV and TYC.

Species	ECV		TYC		
	PC1	PC2	PC1	PC2	PC3
Cr	0.794	0.357		0.827	
Mn		0.893		0.815	
Ni	0.711	0.525	0.871		0.265
Cu	0.857	0.301	0.329		0.794
Zn	0.888		0.369		0.721
As	0.679	0.606	0.607	0.521	
Cd		0.953	0.819	0.258	
Pb	0.917		0.905		
Variance (%)	68.38	19.75	60.66	17.32	13.41

Only factor loading values >0.2 are shown; Extraction Method: Principal Component Analysis; Rotation Method: Varimax with Kaiser Normalization.

At the ECV site, two principal components accounted for 88.13% of the total variance in the dataset. The first factor with an eigenvalue of 5.47 can explain 68.38% of the total variance of the data, with high loadings of Pb, Zn, Cu, Cr, Ni, and As. Zhang et al. [59] reported that coal combustion was the major emission source of Pb in airborne particulate matters. In this study, the concentration of Pb was slightly higher in the rural site than in the urban site (Table 1), suggesting that local sources in rural area emitted certain amounts of Pb into the local ambient atmosphere. Considering that leaded gasoline was phased out in China in 1997, vehicular emissions should no longer be one of the main sources of Pb. In the rural area, approximately 4 tons raw coal per household has been burned in a year, according to our previous survey, and heavy smoke from chimneys of the farmers' coal stoves has been discharged directly into the atmosphere. According to the results reported by Zhang [44], the average content of Pb in coal produced in Shanxi province ( $62.56 \pm 93.44 \mu\text{g g}^{-1}$ ) is far higher than the national average value ( $15.55 \mu\text{g g}^{-1}$  [60]) in China. Similarly, the others (As, Zn, Cu, Cr, and Ni) could be also related to raw coal combustion [30,43,61–64]. In addition, the ECV site is far away from industries, main traffic, and commercial emissions. Therefore, this factor should be identified as raw coal combustion. The second factor accounted for 19.75% of the total variance with an eigenvalue of 1.58 and had high loadings of Cd, Mn, As, Ni, Cr, and Cu. Natural soil dust such as Asian dust has high loadings of Mn and Cr [63,65,66]. Previous studies have shown that Mn, Ni, and Cr were the main pollutants in soil and street dust [63,67]. Lim et al. [68] reported that Mn is a marker for soil and re-suspension dust. Manoli et al. [69] also reported that road dust had high loadings of Mn and Cr. Therefore, the second factor can be identified as mixed sources of soil and road dust.

At the TYC site, the PCA results of heavy metals in PM<sub>2.5</sub> showed three factors, accounting for 91.39% of the total variance. Moreover, 60.66% of the total variance with an eigenvalue of 4.85 was dominated by the first factor, which had high loadings of Pb, Cd, Ni, As, Zn, and Cu. This factor should be well associated with coal combustion (Pb, As, Zn) for industry, central-heating, as well as domestic coal-stove-heating and cooking and industrial emissions (As, Ni, Cd, Cu) [30,62–64,67,69–71]. Metallurgical industry, boiler factories, thermal power plants, etc. could generate huge amounts of

hazardous waste and have the potential of releasing potentially toxic trace metals, such as Ni, Cd, Mn, As, Pb, Zn, and Cu, into the atmosphere [30,72]. The second component was mostly dependent upon Mn, Cr, As, and Cd, and accounted for 17.32% of the total variance with an eigenvalue of 1.39. This factor is associated with road and soil dust (Mn, Cr) [30,63,68,69]. The third factor, which explains 13.41% of the total variance with an eigenvalue of 1.07, was strongly correlated with the elements Cu, Zn, and Ni. This factor originated from vehicle emissions/oil combustion [65,70–73].

#### 4. Conclusions

Based on the analysis of PM<sub>2.5</sub> during the haze episode collected from rural and urban areas of Taiyuan in China, PM<sub>2.5</sub> concentrations in both the rural and urban areas were extremely high. All of the daily PM<sub>2.5</sub> levels both in the rural and urban sites failed to meet the air quality limits (35 µg m<sup>-3</sup> in standard I) set by MEPC. During the haze episode, the concentrations of PM<sub>2.5</sub> at the rural and urban sites in the peak stage were about 5.9 and 5.5 times higher than those in the normal stages. Wind from the south with a low speed showed an obvious accumulative effect on PM<sub>2.5</sub>, while wind from the north diluted the pollutants.

Pollution of heavy metals in PM<sub>2.5</sub> during the haze episode was also very serious, e.g., Pb in the rural area, As in the urban area, and Cd in both the rural and urban areas failed to meet the air quality standard. The orders of concentrations of heavy metals in PM<sub>2.5</sub> at the rural and urban sites were the same and are listed as follows: Zn > Pb > Mn > Cr > Cu > Ni > Cd > As.

According to the results of PCA, the major sources of heavy metals in PM<sub>2.5</sub> at the rural site were raw coal combustion (Pb, Zn, Cu, Ni, and Cr) and soil and road dust (Mn and Cd). At the urban site, the major contributions to the heavy metals were from coal combustion (Pb, Zn, and As), industrial emissions (As, Ni, Cd, Cu), road and soil dust (Mn and Cr), and traffic emissions (Cu and Zn).

**Acknowledgments:** This work was financially supported by the Natural Science Foundation of Shanxi (201601D021135) and the Chinese National Natural Science Foundation (21707125).

**Author Contributions:** Kankan Liu, Qingmin Shang, and Liyuan Cao conceived and designed the experiments. Kankan Liu and Qingmin Shang wrote the paper; Changyuan Wan and Ping Song performed the experiments; Chanyuan Ma analyzed the data.

**Conflicts of Interest:** The authors declare no conflict of interest.

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