A Study on Characteristics of Heavy Metal Elements in Atmospheric PM2.5 During Winter in Chengdu

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Abstract. This paper conducted an in-depth analysis on the characteristics of components of heavy metals in PM\(_{2.5}\) by studying the atmospheric heavy metal elements during winter in Chengdu. On the basis of the limiting value of heavy metal element concentration in PM\(_{2.5}\) in major cities at home and abroad and the national standard limiting value on heavy metal element concentration, this paper compared and evaluated the pollution level of heavy metal elements in PM\(_{2.5}\) during winter in Chengdu and analyzed the typical characteristics of heavy metal elements in haze days and non-haze days as well as the potential sources. The following conclusions were drawn: 1) in PM\(_{2.5}\) during winter in Chengdu, the major component was K, minor components were Fe, Ca, Zn, Cu, Pb, Ba, Mn, As, Cr and Cd, trace components were Se, Ag, Hg, Ni, V, Au and Ga and ultra-trace components were Co, Sb, Ti and Sn, and the proportion of the aforementioned components was 2.10706\%, 2.14783\%, 0.01933\% and 0.00009\% respectively; 2) the major component, minor component, trace component and ultra-trace component of heavy metal elements respectively accounted for 1.621\%, 1.641\%, 0.016\% and 0.00004\% of PM\(_{2.5}\); 3) the proportion of all the heavy metal components in PM\(_{2.5}\) in haze days was lower than that in non-haze days, which indicates that the relatively high concentration of PM\(_{2.5}\) might be caused by non-metallic salts; 4) in PM\(_{2.5}\), the proportion of Pb, Cd, Mn and Cu was lower than both the domestic average level and the international average level; the proportion of Zn and Ni was at the domestic average level but slightly higher than the international average level; and the proportion of As was at both the domestic average level and the international average level; 5) the mean value of Pb concentration during winter was 0.08717\(\mu\)g/m\(^{3}\), which was lower than the limiting value of the national standard GB 3095-2012; and 6) there was a rather high level of consistency between such elements as were K, Ca, Fe, Zn and Ag as well as PM\(_{2.5}\) on the typical haze day, which shows that obvious colinearity existed in the emission source of the aforesaid elements and such elements might come from road dust sources; on the typical non-haze day, random-peak daily variation occurred in heavy metal components, which might result from unorganized emission from peripheral pollution sources; however, the proportion of ultra-trace components of heavy metals on the typical non-haze day was slightly higher than that on the typical haze day, which might be closely related to the local background transmission.
1. Introduction
Recently, China has been shrouded by widespread haze, especially Beijing, Tianjin, Hebei and West Sichuan Plain where haze has constantly occurred. In the light of the existing studies, it can be found that PM$_{2.5}$, fine atmospheric particles, is a major cause of haze [1]. As aerosol with extremely small particle diameter and relatively large specific surface area, PM$_{2.5}$ can cause visibility to drop suddenly. Besides, it is easy for poisonous and hazardous substances in ambient air including trace components of heavy metals such as Cd, Pb, Hg and As to gather in PM$_{2.5}$ and enter human bodies in the biological process including respiration, even to enter the pulmonary alveoli and blood, which will obviously increase the probability of developing cancer, malformation and mutation and threaten human health and environment [2]. Heavy metal elements generally refer to the metal elements with densities more than 4.5g/cm$^3$, such as Cr, Mn, V, Fe, Cu, Zn, Ag, Ni, Cd, Sn, Ba, Hg and Pb, etc.. As is metalloid with great toxicity, thus it is often classified as heavy metal [3]. China has lagged behind in promulgating atmospheric environmental protection standard on heavy metal elements in ambient air. The Ambient Air Quality Standards in effect (GB 3095-2012) only specifies that the annual average limiting value and quarterly average limiting value of Pb are 0.5µg/m$^3$ and 1µg/m$^3$ respectively and that the annual average limiting value of Cd, Hg, As and Cr$^{6+}$ is 0.005µg/m$^3$, 0.05µg/m$^3$, 0.006µg/m$^3$ and 0.000025µg/m$^3$ respectively. However, the daily average limiting value and the annual average limiting value of other metal elements are not covered therein. This paper studied the characteristics of heavy metal elements in PM$_{2.5}$ of ambient air in Chengdu during winter, the period of high incidence of haze, with the heavy metal elements of which minute quantity can be severely harmful to human health as the object of study [4].

2. Research Area and Monitoring Method
The city of Chengdu is located at the western edge of Sichuan Basin, bordered by Longmen Mountain and Qionglai Mountain, sitting on the transition zone between Chengdu Plain and Hilly Area of Central Sichuan Basin, neighboring on Deyang, Ziyang, Meishan, Ya’an and Ngawa Tibetan and Qiang Autononomous Prefecture and covering an area of 12121 km$^2$. As shown in Figure 1, from the west to the east, the topography of Chengdu can be classified as mountainous region, plain and hilly area. To the west lie the high and steep Longmen Mountain and Qionglai Mountain, with an elevation of over 3000m; in the middle lies the vast Chengdu Plain that slightly slopes down from the northwest to the southeast, with an elevation that varies from 450m to 720m and a relative altitude difference that is no more than 20m in general; to the east lie the low Longquan Mountain and the west edge of Hilly Area of Central Sichuan Basin, the elevation of most of which is less than 1000m.

Chengdu has a monsoon-influenced humid subtropical climate with rare snow and frost, low wind speed, high humidity and low atmospheric pressure year-round. Most of the days are overcast without
much sunshine. And temperature inversion frequently occurs. In accordance with statistical yearbooks [5], in Chengdu, the annual average temperature is 16.3 °C, the percentage of annual average sunshine ranges from 23% to 30%, the annual average amount of solar radiation ranges from 80.0 kcal/cm² to 93.5 kcal/cm² and the annual average wind speed is 1.1 m/s.

The monitoring equipment was installed at the roof of the seven-storey Huanbao Building (30.66°N, 104.04°E). The monitoring on particulate matters was carried out strictly in accordance with Technical Specifications and Test Procedures of Continuous Automated Monitoring System for Ambient Air Particulate Matters (PM₁₀ and PM₂.₅) (HJ 653-2013) and Ambient Air Quality Standards (GB3095-2012) issued by Ministry of Environmental Protection. METONE BAM 1020, Thermo Fisher TEOM 1405 and Xact™ 625 heavy metal elements online monitor were used. In this study, 1891 groups of hourly valid data on PM₂.₅ and heavy metal elements (23) were acquired through data quality control. The aforementioned data was measured from 0:00 on November 1, 2016 to 23:00 on January 31, 2017.

3. Result and Discussion

3.1. Characteristics of Components

This study classified the heavy metal elements in ambient air into the major component, minor components, trace components and ultra-trace components on the basis of the amount of the components to be tested in the analytical samples in analytical chemistry. The heavy metal element whose proportion in PM₂.₅ was ≥1% was classified as major component; the heavy metal element whose proportion in PM₂.₅ ranged from 0.01% to 1% was classified as minor component; the heavy metal element whose proportion in PM₂.₅ ranged from 0.0001% to 0.01% was classified as trace component; and the heavy metal element whose proportion in PM₂.₅ was ≤0.0001% was classified as ultra-trace component. The online monitor on heavy metal elements Xact™ 625 monitored 23 elements. In general, it is believed that ambient air does not contain Pd, so Pd was regarded as hour-by-hour correction element, not included in the statistics in Table 1.
Table 1. Statistical Table on Heavy Metal Element Components in PM$_{2.5}$ During Winter in Chengdu

| Time average µg/m$^3$ | Component ratio | Component division | Compon | Minor components |
|---------------------|-----------------|-------------------|-------|-----------------|
| PM$_{2.5}$ 93.94235 | Proportion of heavy metal elements 4.27432% | **As** 0.029 35% | **Cr** 0.011 17% | **Cd** 0.010 14% | **Se** 0.004 89% | **Ag** 0.004 65% | **Hg** 0.003 34% | **Ni** 0.003 06% | **V** 0.001 24% | **Au** 0.000 66% | **Ga** 0.000 33% | **Co** 0.000 05% | **Sb** 0.000 03% | **Ti** 0.000 01% | **Sn** 0.000 00% |
| K 1.97942 2.10706% | Major component 2.10706% | **Mn** 0.06343 0.06752% |
| Fe 0.76888 0.81846% | Minor components 2.14783% |
| Ca 0.51729 0.55065% | **Cu** 0.08880 0.09453% |
| Zn 0.35706 0.38008% | **Pb** 0.08717 0.09280% |
| Ba 0.08443 0.08987% | **Ba** 0.08443 0.08987% |
| Mn 0.06343 0.06752% |

From Table 1, it can be found that the average value of concentration of PM$_{2.5}$ in ambient air during winter in Chengdu was 93.94235µg/m$^3$, which was much higher than 35µg/m$^3$, the Level-2 standard for annual average limiting value of PM$_{2.5}$ specified in the existing Ambient Air Quality Standards (GB 3095-2012). It even exceeded 75µg/m$^3$, the Level-2 standard for daily average limiting value of PM$_{2.5}$. In PM$_{2.5}$ in ambient air during winter in Chengdu, the average value of concentration of 22 heavy metal elements including Cr, Mn and Pb was 4.01539µg/m$^3$, accounting for 4.27432%; K was the major component, accounting for 2.10706%; Fe, Ca, Zn, Cu, Pb, Ba, Mn, As, Cr and Cd were minor components, accounting for 2.14783%; Se, Ag, Hg, Ni, V, Au and Ga were trace components, accounting for 0.01933%; and Co, Sb, Ti and Sn were ultra-trace components, accounting for 0.00009%.

Ambient Air Quality Standards (GB 3095-2012) in effect prescribes that the daily average concentration of PM$_{2.5}$ shall be ≤75µg/m$^3$. To research the characteristics of heavy metal elements in PM$_{2.5}$ in ambient air during winter in Chengdu, this study defined the time of sampling in which the daily average concentration of PM$_{2.5}$ was more than 75µg/m$^3$ as haze day and the other valid data as non-haze day. According to the statistics, the valid data of the winter in Chengdu were 84 days in total,
accounting for 91.3% of the winter. In accordance with the definition of haze day, the daily average concentration of PM$_{2.5}$ of 53 days was more than 75µg/m$^3$, which means that the proportion of haze days was 63.1%.

In this study, 3 days with maximum hourly concentration of PM$_{2.5}$ each month were chosen as typical haze days, and 9 days were chosen as typical haze days all through the winter accordingly. 9 typical non-haze days were chosen in the same way. The statistical result is shown in Table 2:

**Table 2.** Characteristics of Heavy Metal Elements in PM$_{2.5}$ in Haze Days and Non-haze Days During Winter in Chengdu (Daily Average, Unit: µg/m$^3$)

| Classification | PM$_{2.5}$ Daily average concentration | Major component | Minor components | Trace components | Ultra trace components |
|----------------|----------------------------------------|-----------------|-----------------|-----------------|-----------------------|
| Haze day       | 163.97                                  | 2.658537        | 2.690972        | 0.026838        | 0.000058              |
| Proportion     | 1.621%                                  | 1.641%          | 0.016%          | 0.00004%        |                       |
| NO-haze day    | 36.881                                  | 0.878246        | 1.142617        | 0.011722        | 0.000122              |
| Proportion     | 2.381%                                  | 3.098%          | 0.032%          | 0.00033%        |                       |

As shown in Table 2, the daily average concentration of PM$_{2.5}$ in haze days during winter in Chengdu was 163.97µg/m$^3$, 4.5 times of that in non-haze days. The major component, minor components, trace components and ultra-trace components of heavy metal elements in PM$_{2.5}$ in haze days accounted for 1.621%, 1.641%, 0.016% and 0.00004% of PM$_{2.5}$ respectively. The proportion of all the components in haze days was lower than that in non-haze days. That the heavy metal elements in non-haze days were always closer to the local background value indicates that during winter in Chengdu, the more increase in concentration of PM$_{2.5}$ in haze days was caused by non-metallic salts such as organic aerosol.

3.2. Characteristics of Concentration Level

Table 3 shows the concentration of heavy metal elements in PM$_{2.5}$ in ambient air of some cities at home and abroad.
Table 3. Concentration of Heavy Metal Elements in PM$_{2.5}$ in Major Cities at Home and Abroad (Unit: µg/m$^3$)

| City            | Pb   | Mn   | Zn   | Ni   | Cr   | Cd   | Cu   | As   |
|-----------------|------|------|------|------|------|------|------|------|
| Beijing         | 0.11 | 0.03 | 0.32 | 0.06 | 0.02 | 0.0034 | 0.04 | 0.01 |
| Tianjin         | 0.29 | 0.22 | 0.025 | 0.352 |
| Shanghai        | 0.109 | 0.0603 | 0.01 | 0.0273 | 0.003 | 0.0308 |
| Guangzhou       | 0.216 | 0.033 | 0.432 | 0.0065 | 0.00696 | 0.061 | 0.032 |
| Shenzhen        | 0.291 | 0.0983 | 0.0241 | 0.013 | 0.0286 |
| Wuhan           | 0.416 | 0.1556 | 0.0065 | 0.014 | 0.009 | 0.0469 |
| Changsha        | 0.0925 | 0.0333 | 0.0389 |
| Nanjing         | 0.19 | 0.225 | 0.085 |
| Shenyang        | 0.346 | 0.0401 | 0.0269 | 0.0355 | 0.002 | 0.0302 |
| Jinan           | 0.0765 | 0.0856 | 0.0473 | 0.0579 | 0.0199 |
| Taiyuan         | 0.107 | 0.1053 | 0.0399 | 0.0699 | 0.0384 |
| Lanzhou         | 0.1027 | 0.291 | 0.0097 | 0.0297 | 0.0003 | 0.084 |
| Hangzhou        | 0.13 | 0.05 | 0.65 | 0.08 | 0.04 |
| Yinchuan        | 0.0671 | 0.0769 | 0.214 | 0.003 |
| Urumqi          | 0.037 | 0.027 | 0.0072 | 0.019 | 0.001 | 0.0018 |
| Lhasa           | 0.07 | 0.08 | 0.28 | 0.01 | 0.03 |
| Chongqing       | 0.182 | 0.0871 | 0.0037 | 0.0113 | 0.007 | 0.0059 |
| Chengdu         | 0.08717 | 0.06343 | 0.35706 | 0.00306 | 0.01117 | 0.01014 | 0.0888 | 0.02935 |
| Xiamen          | 0.119 | 0.057 | 0.009 | 0.09 | 0.009 | 0.007 |
| Hong Kong       | 0.0718 | 0.012 | 0.174 | 0.0054 | 0.0235 | 0.0009 | 0.0009 | 0.0009 |
| Taiwan          | 0.0665 | 0.0155 | 0.328 |
| Islamabad       | 0.144 | 0.079 | 0.024 | 0.01 | 0.004 |
| Ankara          | 0.071 | 0.0049 | 0.0031 | 0.0032 | 0.00011 | 0.0015 |
| Bern            | 0.049 | 0.025 | 0.003 | 0.00026 | 0.0008 |
| Budapest        | 0.024 | 0.03 | 0.0032 | 0.0089 | 0.0014 |
| Toulouse        | 0.0097 | 0.0075 | 0.03522 | 0.03597 | 0.00025 |
| Lahore          | 4.4 | 0.3 | 0.018 | 0.03 | 0.077 | 0.022 |
| Mexico          | 0.1 | 0.02 | 0.26 | 0 | 0 | 0.02 |
| Toronto         | 0.0034 | 0.0038 | 0.016 | 0.0008 | 0.0003 | 0.0025 |
| New Jersey      | 0.0079 | 0.029 | 0.01 | 0.0004 | 0.017 |
| Los Angeles     | 0.011 | 0.106 | 0.035 | 0.0044 | 0.013 |
| Auckland        | 0.008 | 0.003 | 0.036 | 0.005 | 0.005 |
| Seoul           | 0.0964 | 0.039 | 0.0196 | 0.0137 |
| Tokyo           | 0.125 | 0.0401 | 0.00563 | 0.00609 |
| New York        | 0.0079 | 0.01 | 0.0027 | 0.00034 |
| California      | 0.039 | 0.017 | 0.012 | 0.015 | 0.043 | 0.006 |
| Average of China b) | 0.16000 | 0.09410 | 0.36258 | 0.03310 | 0.05578 | 0.00745 | 0.06969 | 0.02906 |
| Average of the World c) | 0.22985 | 0.07326 | 0.23754 | 0.02153 | 0.05143 | 0.00937 | 0.04263 | 0.02338 |

a) This Study; b) Table 2  Average Value of Domestic Cities (Except for Hong Kong and Taiwan); c) Table 2  Average Value of All Cities

As shown in Table 2, it can be found that the amount of Pb and Cd in PM$_{2.5}$ of ambient air in Chengdu was not only less than that in cities including Beijing, Shanghai and Guangzhou, but also lower than the domestic average level and the international average level; the amount of Mn was more...
than that in cities including Beijing and Guangzhou, the same as that in Shanghai, and lower than the
domestic average level and the international average level; the amount of Zn and Ni was at the
domestic average level but slightly higher than the international average level; the amount of Cu was
more than that in cities including Beijing and Guangzhou, but lower than the domestic average level
and the international average level; and the amount of As was at the domestic average level and the
international average level, higher than that in cities such as Beijing and slightly higher than that in
cities including Shanghai and Guangzhou.

Table 4. Major Standards for Evaluation of Heavy Metal Elements in Ambient Air at Home and
Abroad (Annual Average Value, Unit: µg/m$^3$)

| Country / organization | Release time | Pb   | Cd   | Hg   | As   | Cr$^{6+}$ | V     | Mn   | Ni   |
|------------------------|--------------|------|------|------|------|-----------|-------|------|------|
| WHO[44]                | 2000         | 0.5  | 0.005| 1    | 0.0066| 0.00025   | 1.05  | 0.025|
| The European Union[45] | 2004         | 0.005| 0.006| 0.02  |       |           |       |      |      |
| China[46]              | 2012         | 0.5  | 0.005| 0.05 | 0.006 | 0.000025  |       |      |      |

The average value of concentration of Pb in PM$_{2.5}$ during winter in Chengdu was 0.08717µg/m$^3$,
lower than 1µg/m$^3$, the limiting value standard of quarterly average value of Pb in the existing
Ambient Air Quality Standards (GB 3095-2012). In this study, there was only the quarterly average
value of other heavy metal elements in PM$_{2.5}$ in Chengdu, and the standard limiting value of quarterly
concentration of the heavy metal elements except for Pb was not covered in the national standards,
thus this study did not evaluate whether other heavy metal elements met the corresponding standard.
Special attention shall be paid to that the annual average limiting value of Cr$^{6+}$ rather than Cr is
specified in GB 3095-2012. On the basis of the relation between $\rho$(Cr$^{6+}$) and $\rho$(Cr), the ratio is 0.13 [47].
Accordingly, the conversion result can be obtained and the evaluation on Cr$^{6+}$ can be completed.

3.3. Characteristics of Variation in Typical Days
The typical days were chosen respectively from the 9 haze days and 9 non-haze days mentioned in 3.1.
The date on which the maximum hourly concentration value of PM$_{2.5}$ was the median value of that of
the 9 haze days was chosen as typical haze day. The typical non-haze day was chosen in the same way.
Characteristics of variation in typical days are shown in the following graphs (Figure 2 - Figure 5).

Figure 2. Daily Variation of Major Component of Heavy Metal Elements in PM$_{2.5}$ in Typical Days
During Winter in Chengdu
As shown in Figure 2 to Figure 4, on the typical haze day, there was a high level of consistency between such elements as were K, Ca, Fe, Zn and Ag as well as PM$_{2.5}$, which shows that obvious colinearity existed in the emission source of the aforesaid elements. Ca and Fe generally come from diastrophism. Some of K comes from the soil source of the earth’s crust (including soil, road dust and building dust, etc.), and the other is generally used as the indicative element of biomass burning and fuel oil, etc.. Zn is a kind of impurity in lubricating oil, usually emitted into ambient air by vehicles. Therefore, the strong colinearity of emission sources indicates that the elements such as K, Ca, Fe, Zn and Ag, etc. may come from road dust. On the typical non-haze day, there was no consistency between the daily variation of the components of heavy metal elements and that of PM$_{2.5}$. Random-peak daily variation appeared instead. Besides, obvious characteristics of pollution did not exist, which might result from unorganized emission from peripheral pollution sources.
Figure 5 shows that the proportion of ultra-trace components of heavy metal elements in PM$_{2.5}$ on the typical non-haze day was slightly higher than that on the typical haze day, which may be closely related to the local background transmission.

4. Conclusion
This paper conducted an in-depth analysis on the characteristics of components of heavy metals in PM$_{2.5}$ by studying the heavy metal elements in ambient air during winter in Chengdu. The following conclusions can be drawn.

1) In PM$_{2.5}$ in ambient air during winter in Chengdu, the average value of concentration of heavy metal elements was 4.01539µg/m$^3$, accounting for 4.27432%; the major component was K, minor components were Fe, Ca, Zn, Cu, Pb, Ba, Mn, As, Cr and Cd, trace components were Se, Ag, Hg, Ni, V, Au and Ga and ultra-trace components were Co, Sb, Ti and Sn, and the proportion of the aforementioned components was 2.10706%, 2.14783%, 0.01933% and 0.00009% respectively.

2) During winter in Chengdu, the proportion of haze days was 63.1%. In haze days, the major component, minor components, trace components and ultra-trace components of heavy metal elements respectively accounted for 1.621%, 1.641%, 0.016% and 0.00004% of PM$_{2.5}$.

3) The proportion of all the heavy metal components in PM$_{2.5}$ in haze days was lower than that in non-haze days, which indicates that the increase in concentration of PM$_{2.5}$ in haze days during winter in Chengdu may be caused by non-metallic salts.

4) The amount of Pb and Cd in PM$_{2.5}$ of ambient air in Chengdu was not only less than that in cities including Beijing, Shanghai and Guangzhou, but also lower than the domestic average level and the international average level; the amount of Mn was more than that in cities including Beijing and Guangzhou, the same as that in Shanghai, and lower than the domestic average level and the international average level; the amount of Zn and Ni was at the domestic average level but slightly higher than the international average level; the amount of Cu was more than that in cities including Beijing and Guangzhou, but lower than the domestic average level and the international average level; and the amount of As was at the domestic average level and the international average level, higher than that in cities such as Beijing and slightly higher than that in cities including Shanghai and Guangzhou.

5) The mean value of concentration of Pb in PM$_{2.5}$ during winter in Chengdu was 0.08717µg/m$^3$, which was lower than the limiting value of the national standard GB 3095-2012. The quarterly standard limiting value of other heavy metal elements was not covered in the national standards, thus this study did not evaluate whether other heavy metal elements met the corresponding standard.

6) There was a high level of consistency between such elements as were K, Ca, Fe, Zn and Ag as well as PM$_{2.5}$ on the typical haze day, which shows that obvious colinearity existed in the emission source of the aforesaid elements, and that such elements might come from the sources of road dust. On typical non-haze day, there was random-peak daily variation instead of consistency between the daily variation of components of heavy metal elements and that of PM$_{2.5}$, which might result from unorganized emission from peripheral pollution sources. The proportion of ultra-trace components of heavy metal elements in PM$_{2.5}$ on the typical non-haze day was slightly higher than that on the typical haze day, which might be closely related to the local background transmission.

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