

Characteristics, Sources and Health Risk Assessment of Trace Metals in PM10 in Panzhihua, China

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Abstract Ambient PM₁₀ air samples were collected at two industrial sites and one urban residential site in the mining city of Panzhihua, China, from April, 2014, to January, 2015. Mass concentrations of ten trace metals (As, Cd, Cr, Ni, Co, V, Mn, Cu, Pb, and Zn) in PM_{10} were determined by inductively coupled plasma-mass spectrometry. The results showed Zn, Pb, Cu, Mn and V were the most abundant elements from the industrial sites. Concentrations for Cd, Cr, Co, Ni, Mn and Cu at industrial sites greatly exceeded the air quality standards of the World Health Organization and the Chinese Ministry of Environmental Protection. Principal component analysis indicated that the main sources of the trace metals were steel smelting, fuel combustion, geological and mineral dust. Four different clusters of particles (i.e., mineral, calcium-containing, soot and aluminosilicate) were identified by scanning electron microscopy coupled with energy dispersive X-ray spectrometry. Chromium (Cr) was found to present the highest excess cancer risk, implying the potential for carcinogenic health effects in local inhabitants. Manganese (Mn) presented a non-carcinogenic health risk to children and adults, while the other metals were within acceptable limits.

Keywords Mining City \cdot PM₁₀ \cdot Trace metals \cdot SEM-EDX · Health risk assessment

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1 Applied Nuclear Technology in Geosciences Key Laboratory of Sichuan Province, Chengdu University of Technology, Chengdu 610059, Sichuan, China

2 College of Earth Science, Chengdu University of Technology, Chengdu 610059, Sichuan, China Rapid economic growth and associated increase in energy demands, mining activities and industrialization in China have resulted in high levels of atmospheric particulate matters (PM) with the exposure of many populations to high levels of particulate pollution (Kong et al. [2011](#page-7-0); Zhou et al. [2014](#page-7-1)). In particular, these particles are commonly associated with significantly elevated levels of contaminants, such as As, Cd, Cr, and Pb. These may be inhaled or ingested, causing serious impacts upon health (Zheng et al. [2010](#page-7-2)). Epidemiological studies have provided solid evidence for the relationship between concentrations of fine particulate matter (with aerodynamic diameter $\langle 10 \mu m,$ PM_{10}) and adverse respiratory health effects (Taus et al. [2008](#page-7-3)). The World Health Organization (WHO) has estimated that 4–8% of the deaths occurring annually in the world are related to air pollution (WHO [2002\)](#page-7-4). In fact, high concentrations of airborne particulate trace metals in PM_{10} have been reported in many cities in China (Duan and Tan [2013](#page-6-0)). In an effort to improve air quality and to protect human health, legislation (Ambient Air Quality Standard of China GB3095-2012) has been issued in China, for the first time establishing the limits for allowable concentrations of Cd, Hg, As and Cr (VI), as well as to reduce the allowable concentration of Pb.

Numerous natural and anthropogenic activities can increase aerosol emissions and contaminant levels, among which, mining and metallurgical industrial activities are commonly known to dominate over natural sources of particulate matter and associated heavy metals (Allen et al. [2001](#page-6-1); Csavina et al. [2012\)](#page-6-2).

Panzhihua located in the dry-hot-valley mining region of southwest China. Here, V–Ti magnetite mining, coal mining, iron and steel smelting, and chemical and coal-fired power plants are major industries. Highly intense industrial activities have emitted high concentrations of metal-laden

particulates into the air (Xu et al. [1984;](#page-7-5) Xue et al. [2010](#page-7-6)), making this one of the most polluted areas in Sichuan Province (Tan [2005\)](#page-7-7). Wang et al. [\(1985](#page-7-8)) as early as 1985 documented the potential detrimental health effects of atmospheric particulates and toxic pollutants in this region. In spite of numerous efforts by local government authorities to reduce the air pollution, PM_{10} particles have continued to be a primary air pollutant in this region. In recent years, rapid urbanization and industrialization have aggravated particulate pollution in this region. In spite of risks to the health of population, few studies have been carried out in this region to assess the metal levels and to evaluate the health risks of exposure to airborne trace metals.

The aims of this work were to assess the metals (As, Cd, Cr, Ni,Co, V, Mn, Cu, Pb and Zn) in two industrial districts and one urban residential district of Panzhihua, to investigate the possible sources of trace metals, and for the first time estimate the health risk linked to the exposure to airborne trace metals in this region.

Materials and Methods

Panzhihua (26°58′N, 101°72′E) is located in a denuded mountainous area of the southern part of the Pan-Xi Rift valley in southwestern China. It has a typical subtropical monsoon dry-hot-valley climate, which is characterized by indistinct seasons except for a distinct rainy (from May to September) and dry season (during the rest of the year), abrupt daily temperatures change, concentrated rainfall, and diverse microclimates. The annual precipitation is 760–1200 mm, with 90% of the total rainfall occurring during the rainy season. The annual average temperature is 20.2°C. Jinsha River (upper stream of the Yangtze River) traverses the city from west to east, and the industrial enterprises are located on both sides of the river and surrounded by high mountain ranges. Strong thermal inversions occur frequently in this region, which seriously restricts the dispersion of air pollutants.

Three sampling sites were selected based on their different district functions and levels of metal pollution (Fig. [1](#page-1-0); Table [1\)](#page-2-0). PM_{10} samples were collected using moderate-volume air samplers (Wuhan Tianhong Co.TH-150C, Wuhan, China). Samples were collected at a flow rate of 0.1 m^3 min−1 on pre-weighted quartz filters (Φ: 90 mm; Whatman-QMA, Buckinghamshire, UK). A total of 80 PM_{10} samples along with 4 blank samples were collected at each sampling site from April 2014 to January 2015 during the following periods: 15 April–5 May (representative of spring), 15 July–5 August (summer), 15 October–5 November (autumn), and 10–30 January (winter). Collection duration of each sample was 24 h; three field blanks were collected during every seasonal campaign, which were then analyzed

Fig. 1 Location of atmospheric particulate sampling sites in Panzhihua

together with the samples. Particle concentrations were determined gravimetrically.

For trace metal analysis, PM samples were extracted from filter using a mixed acid [ultra-pure HF and $HNO₃$] (3:2)] microwave digestion method and analyzed for ten trace metals using a PE 6000 inductively coupled plasmamass spectrometry (ICP-MS) (Perkin Elmer corp., Norwalk, CT, USA) at Applied Nuclear Technology in Geosciences Key Laboratory of Sichuan Province (Chengdu University of Technology). The reagent and filter blanks were analyzed using the same procedure; all data were blank corrected by subtracting blanks from measured values. A certified reference standard, NIST, SRM-1648, was used to ensure accuracy and precision. Resulting recoveries fell within $\pm 10\%$ of certified values for most elements.

Obtained data were processed using principal component analysis (PCA) with a Varimax rotated factor matrix method (Heidam [1982\)](#page-6-3), to identify sources of airborne trace metals in PM_{10} . Principal components having an eigenvalue higher than 1 in the component data set were generally retained.

Scanning electron microscopy coupled with energy dispersive X-ray spectrometry (SEM-EDX) may be used to obtain information on the morphology and chemical composition of particles, and insight into their origin (Samara et al. [2016\)](#page-7-9). This approach was used in our study, with the analysis performed at the State Key Laboratory of Oil and Gas Reservoir Geology and Exploitation of Chengdu University of Technology, China, using a FEI Quanta 250 FEG (FEI, Bragg, Czech) operated at 20 kV and equipped with an X-ray energy dispersive spectrometer (EDX-Oxford IN-CAx-max20, Oxford, UK).

Human health risk assessment including non-carcinogenic and carcinogenic risks was carried out according to standard EPA methods (US EPA [2011\)](#page-7-10). The adults and children (1–15 years) living in this area are potential receptors. The trace elements of concern involved in risk

Sampling sites	District function	Ambient environment
XSG (26°29'0.42"N, 101°45'10.63"E)	Commercial and residential district	It is characterized by traffic, residential and construction activities
HMK (26°36'38.62"N, 101°35'49.06"E)	Mining and industrial complex and residential district	It is characterized by detonations in rocks, a crushing plant, a blast furnace slag pool, coal-fired power plants and building materials industrial as well as heavy traffic
NNP (26°33'58.86''N, 101°39'59.39''E)	Industrial complex district	It is characterized by iron and steel smelling, vanadium and titanium industries, and heavy traffic, especially the heavy-loaded trucks

Table 1 Locations and ambient environment of sampling sites

assessment were As, Cd, Cr, Co, Ni, Cu, Mn, V, Pb, and Zn, which are also the typical contaminants associated with local industrial activities. Among these, As, Cd, Cr, Co and Ni are carcinogenic metal (loid) s, while Cu, Pb, Zn, V and Mn are known as non-carcinogenic. Risk assessment was performed by using mean concentrations of elements in PM_{10} . Inhalation was assumed to be the only exposure pathway to airborne trace elements.

The non-carcinogenic elements average daily dose (ADD, in mg kg⁻¹ d⁻¹) and carcinogenic elements lifetime average daily dose (LADD, in mg kg⁻¹ d⁻¹) were calculated as follows:

$$
ADD (or LADD) = C \times IR \times ED/BW \times AT \tag{1}
$$

where C is the concentration of pollutants, mg m⁻³; IR is the inhalation rate (15.2 m³ d⁻¹ for male, 11.3 m³ d⁻¹ for female, and 8.7 m³ d⁻¹ for children); BW is the body weight (62.7 kg for male, 54.4 kg for female, and 36 kg for children); ED is the duration of expose (30 years for adults, 18 years for children); AT is the average time for residents (for carcinogenic, $AT = 70 \times 365 = 25{,}550$ days, for noncarcinogenic, $AT = 30 \times 365 = 10,950$ days for adults, and 6570 days for children).

The cancer risk for individual is calculated as in the following equation:

$$
R = [1 - exp(-LADD \times SF)]/70 \tag{2}
$$

where R is the average annual excess risk of cancer for an individual (the acceptable or tolerable risk is 1×10^{-6} to 10−4), dimensionless, SF is the slope factor (reference values are shown in Table [2](#page-2-1)), which is used to estimate the risk of cancer associated with exposure to carcinogenic, or potentially carcinogenic chemicals, mg kg⁻¹ d⁻¹, 70 is number of individual' s average lifetime, years.

For non-carcinogenic risks, it is calculated by:

$$
R = (ADD \times 10^{-6})/(RfD \times 70) \tag{3}
$$

where R is the probability of non-cancer occurring in the exposed population over a 70-year lifetime (if $\langle 1 \rangle$ is not considered a health concern, for $R \geq 1$, adverse health effects are possible and more attention should be paid); RfD is the reference dose (reference values are shown in

Table 2 Reaction parameters for heavy metals entering the human body through respiratory system

Elements	Nature	SF(mg) $kg^{-1} d^{-1}$	RfD (mg $kg^{-1} d^{-1}$)		
As	Carcinogenic	15.1			
Cr	Carcinogenic	42.0			
C _d	Carcinogenic	8.4			
Ni	Carcinogenic	0.8			
Co	Non-carcinogenic	9.8			
V	Non-carcinogenic		7.0×10^{-3}		
Mn	Non-carcinogenic		1.43×10^{-5}		
Cu	Non-carcinogenic		2.0×10^{-3}		
Ph	Non-carcinogenic		4.3×10^{-4}		
Zn	Non-carcinogenic		1.0×10^{-2}		

Table [2\)](#page-2-1), mg kg⁻¹ d⁻¹, 10⁻⁶ is the level of risk acceptance for the RfD.

Results and Discussion

The measured levels of PM_{10} and trace metals are sum-marized in Table [3.](#page-3-0) Mean PM_{10} levels obtained for the sampling periods were 127.7, 165.5, and 187.5 μ g m⁻³ in XSG, HMK and NNP, respectively; with an overall mean of 160.0 μ g m⁻³. These values all exceeded the Chinese annual PM₁₀ guideline of 70.0 μg m⁻³. The two industrial stations showed significantly higher levels of PM_{10} particles, underscoring the importance of industrial emission sources.

Table [3](#page-3-0) shows the maximum, minimum, and average concentration values found for metals in PM_{10} at three sites during the sampling periods. The most abundant heavy metal in PM_{10} was Zn, followed by $Pb > Cu > M$ $n > V > Cr > Ni > Cd > As > Co$, at the majority of sampling sites. Levels of trace metals in the industrial districts (NNP and HMK) were higher than in the residential area (XSG). This may have been attributable to several factors, including pollution sources (distance from the sources and relative importance of local sources), topography, and

meteorological conditions (such as wind speed and direction). Laborers and residents living around the industrial complex are assumed to be exposed to elevated levels of the airborne trace metals compared to those living further distances from the industrial area.

Table [4](#page-3-1) provides a comparison between the results of our study with trace metals in PM_{10} from other industrial areas in China and other countries, as well as with guidelines for specific metals. The concentrations for As, Cd, Cr, Co, Ni, Mn, and Cu at the two industrial sites greatly exceeded the limit values. Even in urban residential sites, As, Cr, Ni, Co and Cu exceeded their respective threshold. Concentration of Pb and V were below the limit values. However, In comparison with other industrial area, the levels of V in our study were high, exceeding the concentrations in Wuhan, Beijing, Shanghai Banwol and Jeddah by approximately 2.6–15.0 times. At the NNP station, the V concentration also exceeded the Chinese average concentration in the atmosphere of 18.7 ng m⁻³ (Tan and Duan [2013](#page-7-11)) by approximately 5–7 times. The weathering of the rock, fuel combustion and the mining of V–Ti magnetite and smelting are the major sources of V in ambient air particles. Vanadium compounds are toxic to humans, causing damage to the heart and kidneys following uptake via the respiratory tract (Zou and He [1993](#page-7-12)). Research on the toxicity of V has indicated a correlation between V levels in the environment and mortality due to certain types of cancer (Wu and Lan [2004](#page-7-13)). Japan has promoted the monitoring of V as an atmospheric particle pollutant (Kitani et al. [1998](#page-7-14)). Therefore, special attention should be accorded to V due to its high concentrations in the ambient air particles of Panzhihua. The levels of Zn (586.8 and 625.5 ng m⁻³ at NNP and HMK, respectively) were similar to those reported for Wuhan, Beijing, Sihwa and Dhanbad, higher than those reported for Shanghai, Tarragona and Jeddah, and lower than those in Banwol areas. Based on the results, the present study area can be considered as a highly polluted area with respect to metal species.

Table 4 Comparison of metal concentrations (ng m⁻³) determined during the present study and those measured by other researchers

"/" no data, *PZH* Panzhihua, a WHO ([2000\)](#page-7-15), b GB3095-2012 ([2012\)](#page-6-4)

Table 5 Factor loading from principal component analysis

Only factor loading values >0.2 were shown

Fac factor, *Var* variance, *Acc* accumulation variance

Table [5](#page-4-0) presents the principal component (PC) loading for the metal date of the study period with corresponding variances for each site. Only components with eigenvalue >1 were retained. Four main source factors were identified: iron and steel smelting industries, fuel combustion (coal combustion and traffic emissions), geological sources (soil or road dust), and mineral dust.

At the NNP site, 29.68% of total variance was dominated by the main factor which had high loadings for Cd, Cr, Cu, Ni and V. Its sources were identified as steel smelting industry, mineral and fuel combustion particles. Nickel, Cd, Cr and Cu were generally related with iron/steel manufacturing and vehicle emissions (Mansha et al. [2012](#page-7-21)). Vanadium can serve as a marker for local mining and industrial dust and vehicle emission. Factor 2 accounted for 25.05% of the data variance, and was characterized by Co, V and Mn. This indicated sources of traffic emission, as well as road re-suspension dust. Cobalt, V and Mn are regarded as indicators of emissions from fuel combustion and vehicles (Manoli et al. [2002](#page-7-22); Canepari et al. [2008\)](#page-6-7). Manganese is a marker for soil and re-suspension dust, and it is also released from construction sources (Lim et al. [2010](#page-7-19)). The third factor contributed 11.28% of the data variance with high loadings of As, Pb and Zn, indicating coal combustion and smelting industry as the sources (Allen et al. [2001](#page-6-1)).

At the HMK site, PC1 accounted for 32.69% of the variance and had high loadings for Cr, Ni, Co, V, Mn and Cu, which may represent mine wastes, combustion and traffic. The mining, mineral processing and smelting activities have generated huge amounts of hazardous mine wastes (ore stockpiles, slag deposits, spoil heaps, mine tailings and waste rock piles) that have been deposited over extensive areas. These deposits are subject to water runoff, leaching and wind erosion. Therefore, these materials have the potential for releasing potentially toxic trace elements, such as Cr, V, Mn, Cu, Pb and Zn into the environment (Sonia et al. [2013](#page-7-23)). The second factor explained up to 18.59% of the variance and was defined by As, Co, Pb and V, the sources of which were considered to be fuel combustion (Tian et al. [2012\)](#page-7-24). As the third factor, Cd, Mn, and Cr had high factor loading values, which may be associated with industrial and fuel combustion (Thomaidis et al. [2003](#page-7-25)). The fourth factor, with high loadings for Zn and V may attribute to industries and vehicle emission.

At the XSG site, Cr, Co, Cu, Mn and V were represented in factor 1, accounting for 27.66% of the variance. This was well associated with soil and road dust. The second factor explained 20.49% of date variance, and was characterized by Cr, Ni, Pb, and Zn, indicating traffic discharges as the source. The third factor (As and Zn) may have had coal combustion as the source for these elements. The results of the present study demonstrated the abundance of Zn, Cu, Pb, Mn, and V in PM_{10} . This implies that the pollution in the region is primarily industrial. Similarly, previously studies have shown that the heavy metals Mn, Zn, Cr, Cu, V, Co, Ni, and Pb are the main pollutants in urban street dust and soil, and are distributed primarily in areas within and surrounding mining and industrial activities.

Based on SEM-EDX analysis of the particles for morphology and elemental composition, four different cluster types were identified (i.e., mineral, Calcium-containing, soot and aluminosilicate; Fig. [2](#page-5-0)). Mineral particles (Fig. [2](#page-5-0)a) had a lump structure, and possessed high levels of V, Cr, Mn, Ti and Fe, suggesting a mining activity origin (Slezakova et al. [2008\)](#page-7-26). Particles contained a high concentration of Ca (Fig. [2](#page-5-0)b) and minor concentrations of Si, indicating these particles had a crustal source, and were derived mainly from limestone mining in this region. Soot particles

Fig. 2 SEM micrographs and EDX spectra of particles in PM10, **a** mineral particles, **b** calcium-containing particles, **c** soot particles, **d** aluminosilicate particles

(Fig. [2c](#page-5-0)) had a fluffy and amorphous structure, clearly demonstrated the presence of carbonaceous spherules, possibly originated from fuel combustion associated with traffic emission and biomass burning (Slezakova et al. [2008\)](#page-7-26). Aluminosilicate particles (Fig. [2d](#page-5-0)) were composed primarily of Si, Fe, and AI, and had a spheroidal morphology, indicating an origin related to a high temperature combustion process (Slezakova et al. [2008](#page-7-26)).

Based on the average annual concentrations of airborne trace metals determined during the study periods, a health risk assessment was performed to obtain an understanding of the health hazard. As indicated in Table [6,](#page-6-8)

Elements	NNP			HMK			XSG		
	Male	Female	Children	Male	Female	Children	Male	Female	Children
As	1.82E-05	l.56E-05	$3.63E - 06$	$1.91E - 0.5$	$1.64E - 05$	$3.81E - 06$	$.56E - 05$	$1.34E - 05$	$3.12E - 06$
Cr	$2.16E - 04$	1.85E-04	$1.29E - 04$	$2.05E - 04$	$1.76E - 04$	$1.23E - 04$	$1.41E - 04$	$1.23E - 04$	$8.43E - 05$
C _d	$1.21E - 0.5$	1.03E-05	$7.21E - 06$	$5.70E - 06$	$4.88E - 06$	$3.40E - 06$	$3.35E - 06$	$2.87E - 06$	$2.00E - 06$
Ni	$1.62E - 06$	1.38E-06	$9.66E - 07$	$3.06E - 06$	$2.62E - 06$	1.83E-06	$1.18E - 06$	$1.01E - 06$	$7.06E - 07$
Co	$7.71E - 06$	$6.61E - 06$	$4.61E - 06$	$5.37E - 06$	$4.60E - 06$	$3.21E - 06$	$2.25E - 06$	$1.92E - 06$	$1.34E - 06$

Table 6 Carcinogenic risks via inhalation exposure

Table 7 Non-carcinogenic risks via inhalation exposure

Elements	NNP			HMK			XSG		
	Male	Female	Children	Male	Female	Children	Male	Female	Children
Cu	$5.19E - 02$	$4.44E - 02$	$1.55E - 01$	$3.64E - 02$	$3.12E - 02$	1.09E-01	$3.89E - 03$	$3.33E - 03$	$1.16E - 01$
Mn	$4.70E + 00$	$3.17E + 00$	$11.11E + 01$	$3.26E + 00$	$2.79E + 00$	$9.74E + 00$	1.26E+00	$1.08E + 00$	$3.76E + 00$
V	$3.79E - 03$	$3.24E - 03$	$1.74E - 02$	$3.39E - 03$	$2.91E - 03$	$1.02E - 02$	$9.07E - 04$	$7.77E - 04$	$2.71E - 03$
Pb	$2.13E - 01$	$1.83E - 01$	$6.37E - 01$	$8.29E - 02$	$7.10E - 02$	$2.48E - 01$	$5.63E - 02$	$4.82E - 02$	$1.68E - 01$
Zn	$1.36E - 02$	$1.17E - 02$	4.08-E02	$1.45E - 02$	$1.25E - 02$	$4.25E - 02$	$4.14E - 03$	3.55E-03	$1.24E - 02$
sum	$4.98E + 00$	$3.41E + 00$	$11.91E + 00$	$3.39E + 00$	$2.90E + 00$	$10.15E + 00$	$1.32E + 00$	$1.13E + 00$	$4.01E + 00$

the risk levels of the carcinogenic trace metals for exposure through the respiratory system were between from 1.18×10^{-6} and 2.16×10^{-4} for male, 1.01×10^{-6} and 1.85×10^{-4} for female, and 7.06×10^{-7} and 1.29×10^{-4} for children. The risk level for Cr was higher than the level of average risk acceptance of 10−6/year (EPA [1989\)](#page-6-9) at all sites, which should cause concern. The risk levels for the carcinogenic heavy metals occurred in the following order: $Cr > As > Cd > Co > Ni$ in three sampling sites. In addition, carcinogenic metals posed the greatest cancer risk to men, then to women and lastly to children.

As indicated in Table [7](#page-6-10), the risk levels of the noncarcinogenic toxic metals for exposure through the respiratory system were between 9.07×10^{-4} and 4.70×10^{-0} for male, from 7.77×10^{-4} and 3.17×10^{-0} for female, and 2.71×10^{-3} and 11.11×10^{1} for children. Manganese posed non-carcinogenic risks to children and adults, while others were within acceptable levels.

Compared to residents living further away from the industrial area, laborers and residents near industrial complexes are assumed to be exposed to elevated levels of the toxic metals associated with PM_{10} . As a result of individual differences in body weight, respiration rate, and outdoor exposure, the carcinogenic and non-carcinogenic risks to men, women, and children are different. This study has shown that carcinogenic risks posed by metals were considerably higher than non-carcinogenic risks, indicating a concern for carcinogenic health effects in local inhabitants.

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