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Original Research

Assessment of Heavy Metals Contamination in Near-Surface Dust

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Abstract

As a result of the continuous urbanization and industrialization in China over the last few decades, heavy metals have been continuously emitted into urban environments and now pose serious threats to human health. In the present study, an extensive urban near-surface dust survey was conducted in Changchun, China, to evaluate the current status of heavy metal contamination in urban dust and to identify the potential sources. A total of 232 samples were collected and the levels of Cu, Pb, Zn, Cd, Hg, As, Cr, and the major elements were then determined. The results indicated that the urban dusts were enriched with metals, particularly Cu, Cd, Zn, Pb, and Hg. Principal component analysis (PCA) was employed to identify sources of heavy metals, and the results revealed distinctly different associations among the trace metals and the major elements in the urban dusts. The concentrations of Cr and As appeared to be controlled by natural factors. Cu and Pb were mainly from vehicle emissions. Zn primarily come from traffic sources, especially vehicle tires. Additionally, Hg primarily originated from coal combustion, while Cd was mainly associated with industrial sources. A revised pollution index for each metal and a Nemerow integrated pollution index (NIPI) of the seven metals were attributed to each sampling site to assess the degree of metal contamination. The results showed that the pollution indexes (PI) of each metal are not especially high. However, a significant degree of metal pollution exists in some urban dusts in Changchun, particularly for Cu, Cd, Zn, and As. The mean value of the Nemerow integrated pollution index (NIPI) of the seven metals also indicated that urban dusts in Changchun city were classified as having moderate levels of pollution. However, it should be noted that about 11% of all samples had a NIPI value above 3, suggesting a high level of contamination by metals. These findings indicate that more attention should be paid to metal pollution of the urban dusts in Changchun in the future.

Keywords: urban dust, heavy metals, Changchun city, pollution assessment

Introduction

The continuous urbanization and industrialization that has occurred in many countries over the last few decades has been accompanied by unprecedented environmental changes. Several categories of contaminates, such as heavy metals [1, 2] and polycyclic aromatic hydrocarbons (PAHs) [3], etc., are continuously emitted into the terrestrial environment, thus posing a great threat to human health [3-5]. Nowadays, more and more people dwell in urban areas. Therefore, the changes of urban environment are crucially important for city dwellers. Heavy metal pollution of urban

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dust and soil is one of the fastest growing types of environmental pollution and is raising serious concerns of researchers because of the pollution accumulation and health risks [4, 6-11].

Urban soil and dust are important reservoirs of trace metal contaminants in urban environments [12, 13]. Meanwhile, compared with urban soils, urban dusts, as a special type of environmental medium, have been reported in many countries to contain toxic organic and inorganic pollutants, especially high levels of trace metals [1-4, 6-11]. These metals can remain in urban environments for a long time or be re-suspended into the atmosphere and thus pose a potential threat to local ecosystems and public health. The results of total metal loadings and Pb isotope ratios reveal that dust is much more sensitive than bulk surface soil to anthropogenic contamination in urban areas [14]. Therefore, urban dusts can reflect pollutants from multimedia, and were considered as an important environmental indicator of urban environmental pollution [7, 15].

Many previous studies have shown that heavy metal contents in street dusts are generally much higher than those in urban soils [1, 16]. Long-term exposure to an urban dust environment that contains elevated concentrations of heavy metals would cause chronic damage through ingestion via the hand-mouth pathway, inhalation, and dermal contact [1, 3, 5]. It has been estimated that adults could ingest up to 100 mg dust per day in dusty environments [17]. Children are usually exposed to greater amounts of dust than adults as a result of "pica" (the mouthing of non-food objects) and play behavior. Meanwhile, research indicates that dust transported and stored in the urban environment also has the potential to provide considerable loadings of heavy metals to receiving water and water bodies [18]. Therefore, several authors have stated that there is a growing need for increased knowledge, investigation, and assessment of trace metal contamination in urban dust [9, 10].

Numerous studies have tried to ascertain various aspects of heavy metal contamination in urban dust. These studies focused on element contents, fractionation and contamination assessment, particle-size and spatial distribution, and source identification. Although many studies have concentrated on the total levels of heavy elements in street dust, environmental and health effects of heavy metals in dust are dependent, at least initially, on the mobility and availability of the elements, and mobility and availability is a function of their chemical speciation and partitioning within, or on, dust matrices [10, 19]. Previous studies also showed that the chemical fractionation patterns of heavy metals in urban dusts in different locations vary. Meanwhile, the high exchangeable phases of heavy metals in urban dust were found [10, 19].

Behavior and other properties of urban dust strongly depend on particle size distribution. Generally, toxic heavy metals were found to be mainly associated with fine particulates compared to coarse ones [20, 21]. Based on spatial analysis, it was found that areas with highly elevated metal concentrations were generally located in industrial and residential areas, and roadside and crowded commercial districts. In contrast to heavy metals in urban soils, those in urban dust have many more possible sources, especially derived from anthropogenic sources. For urban dusts, the anthropogenic sources of heavy metals include traffic emissions such as vehicle exhaust particles [15, 22], tire wear particles [22, 23], weathered street surface particles, and brake-lining wear particles [22, 23], plus industrial emissions (power plants, coal combustion, metallurgical industry, auto repair shops, chemical plants, etc.) [8, 9, 15], domestic emissions, weathering of building and pavement surfaces, and other activities such as waste incineration. Multivariate statistical methods have commonly been applied to assess the sources of heavy metals in urban soils [7, 19]. The HRTEM/EDX approach, magnetic, and lead isotopes were also used to identify the source of dust particles in urban dust [13, 15, 24, 25]. Most recently, research also has focused on heavy metal pollution of the dust deposited on foliage [25, 26].

Despite numerous studies of heavy metal contamination in urban dust that have been conducted in developed countries and in some Chinese metropolitan areas, there is little information available regarding heavy metals contamination of urban dust in Changchun City, China. Little attention has been given to trace elements in the near-surface dust (1.5 m height above ground surface, which is considered the average breath height, was defined in the present study) which is the accumulative mixture of atmospheric particulate and surface dust for a relatively long time, and is much more easily touched and re-suspended [27, 28]. Meanwhile, research on near-surface dust has supplemented the research on air pollution of 0~5m, and has important significance in urban administration, urban planning, and the study of element transformative mechanisms in urban ecological environments. As the first heavy industrial base since the establishment of the P.R.C., Changchun has made great contributions to the economic development of China. The rapid economic development in Changchun over the last two decades, however, has led to a significant release of waste into the urban environment and placed great pressure on the local environment, including heavy metal contamination of urban soils and dust, such as Cd, Cu, Pb, and Zn [27-30]. The environmental quality of Changchun is vital for future sustainable development. Therefore, a more detailed study is required to elucidate the environmental changes associated with the rapid economic growth that is occurring, as well as to find appropriate methods for sensible and responsible development to maintain a sustainable environment.

The present study was conducted to:

1) determine the concentrations of heavy metals (As, Cd, Cr, Cu, Hg, Pb, and Zn) in near-surface urban dust collected from Changchun,
2) identify their natural or anthropogenic sources by principal component analysis (PCA),
3) assess the level of heavy metal contamination in the near-surface dust based on pollution index values and the Nemerow integrated pollution index (NIPI).

This information will be helpful to urban planners and environmental risk managers who seek to encourage responsible, environmentally friendly economic development strategies.

Materials and Methods

Study Site

The study site is located in the urban area of Changchun, the capital of Jilin Province and an important social-economic center of northeastern China located in the hinterland of the Northeast Plain. The main urban area of this city consists of five administrative districts: Chaoyang, Lvyuan, Kuancheng, Erdao, and Nanguan (Fig. 1). The city spans 4,789 km^2 and had an urban population of 4,876,500 in 2011, growing from approximately 1,643,000 in 1980 and 2,110,000 in 1990. The population density is about 3,295.6 per km in urban areas. The area is dominated by relatively flat topography with an altitude of 200 to 250 m, and the area falls within the North Temperate Zone, which is characterized by a sub-humid climate with a continental monsoon. The annual average temperature of the region is 4.8ºC and the average annual rainfall is 569.6 mm. The prevailing wind direction is from the southwest to northeast throughout the year, and there is an annual average wind speed of 4.3 m/s. Urban land uses are primarily industrial, residential, and commercial. There were more than 0.617 million motor vehicles in the city in 2011, and this number has been increasing by 230 per day. In 2012, approximately 24.7 million tons of coal were consumed in the city. The major industries are the automotive industry, coal-fired power plants, metallurgy, iron and steel mills, machine manufacturing, electric and electronics manufacturing, and medicine. Most of the automotive industry is located in the northwestern and southwestern portions of the city, while the northern and eastern areas are dominated by metallurgy, coal-fired power plants, iron and steel mills, and some cement plants and other building material production facilities (Fig. 1). This study area has experienced substantial uncontrolled development over the past 30 years. The central part of the study area comprises the historic center district and is characterized by high residential and commercial activities and traffic levels. The southern region is the new district and comprises an important residential and educational area, with only a few industrial activities.

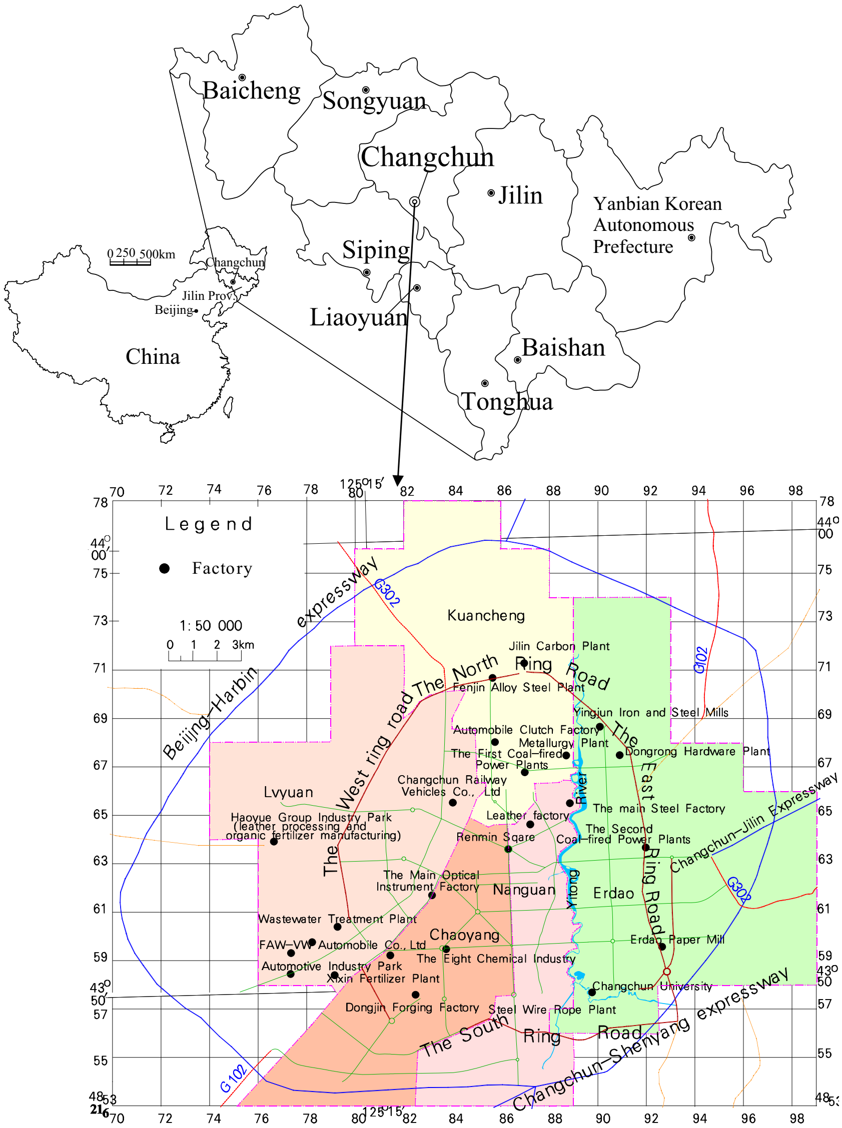

Fig. 1. Locations of Changchun are shown together with the communication network, the administrative division, and the main industries (only a few factories are shown because of the limited page space).

Sampling and Analysis

In the present study, the scope of the sampling area focused on the urban area of Changchun, which has an area of 215 km^2. According to the Specifications of the Multi-purpose Regional Geochemical Survey carried out by the China Geological Survey (CGS) in 2005 and methods documented by [31], a systematic sampling strategy was adopted to provide a sampling scheme over the entire study area. The total area was divided into 215 cells of 1 km×1 km in size, within which the near-surface urban dust samples (about 1.5 m high above ground surface) were collected. A sampling density of one sample per km^2 was adopted wherever possible in urban areas. Each of the dust samples consisted of about 3-5 sub-samples collected by sweeping all kinds of platform surfaces, mainly on nonmetallic windowsills using a clean plastic dustpan and brushes for each sampling site. The sampling data were chosen in April, the end of the dry spring season, and there was no rain when the samples were collected. Meanwhile, the sampling site was far from significant pollution sources such as main streets, construction sites, industrial factories, etc. Because the samples were collected from a nonmetallic platform and did not touch any metal tools during sampling and subsequent sample preparation and analysis, the impacts of platform and sampling tools on the content of heavy metals in dust were neglected. For each cell, a total of 30 g of dust was taken from the mixed samples using a quartile method. Overall, 232 samples were collected. The collected dust samples were stored in paper bags for transport and storage. The exact location (longitudes and latitudes) of each sample point was measured by a GPS instrument, and environmental observations were described during fieldwork (Figs. 1 and 2).

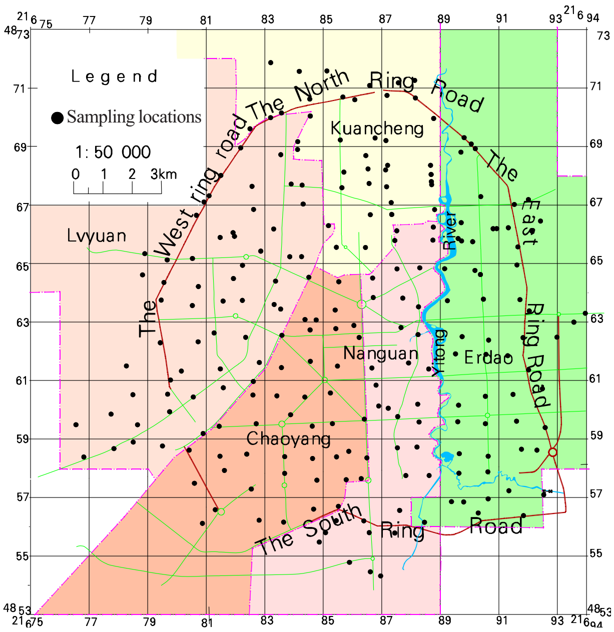

Fig. 2. Locations of sample sites in Changchun City.

The samples were air-dried naturally in a laboratory for about two weeks, after which they were sieved through a 20-mesh polyethylene sieve (<0.8 mm) to remove stones, coarse materials, and other debris. A total of 20 g of dust was collected from the sieved samples using the quartile method and then stored in polyethylene bottles in a desiccator.

All analyses were completed in the lab of the Changchun Inspection and Testing Center of Geology and Mineral Resources, a subordinate research institute of the Ministry of Land and Resources of China. The prepared urban dust samples were ground in an agate mortar to pass through a 200-mesh sieve prior to analysis. For each sample, approximately 0.2 g of urban dust was digested using HF, HNO_3, and $HClO_4$ for measuring the heavy metals and major elements concentrations. The total concentrations of As and Hg were analyzed by atomic fluorescence spectrometry (AFS, AFS-230E, Beijing, China), whereas Cd was measured by Graphite Furnace Atomic Absorption Spectrometry (GF-AAS, M6, thermo Elemental, USA), and the total concentrations of Cu, Pb, Zn, Cr, and the major elements (Mn, Al_2O_3, CaO, Fe_2O_3, MgO, SiO_2, K_2O, and NaO) in soil samples were measured by x-ray fluorescence spectrometry (XRF, ADVANT'XP$^+$, ARL, Switzerland).

For each sample, approximately 10 g of a 20-mesh sieved urban dust sample was weighed for measuring the pH of the dust. Values of pH were measured in H_2O with a soil/solution ratio of 1:2.5 (m/v), using the glass electrode method (GL, pHS-3C, REX, Shanghai, China) according to the agricultural sector standard (NY/T1377-2007) of the People's Republic of China.

To ensure quality assurance and quality control (QA/QC), the precision and accuracy of all the methods were determined by analyzing the four standard reference materials (GSS-1, GSS-2, GSS-10, and GSS-11 soils) obtained from the Center of National Standard Reference Material of China for each batch of 12 samples analyzed. The analytical precision of all the methods, measured as the relative standard deviation (RSD), was generally below 0.05, while the analytical accuracy, measured as the logarithmic deviation (ΔlogC) according to the Specification of Multi-purpose Regional Geochemical Survey recommended by CGS, was also generally below 0.05. Pre-cleaned and acid-washed glassware were used for all analyses. All reagents and acids used were of analytical grade and were used without further purification. Distilled water was used to prepare the reagents. Reagent blanks and duplicate samples (about 16% of samples) were also used in the analytical program to detect contamination and assess the precision and accuracy of the analysis process. The recovery rates for the heavy metal and major elements in the standard reference materials (GSS-1, GSS-2, GSS-10 and GSS-11 soils) obtained from the Center of National Standard Reference Material of China were between 96.1 and 105%. The analytical results showed no signs of contamination and revealed that the precision and accuracy of the analysis process, measured as logarithmic standard deviation and logarithmic deviation according to the Specification of

Multi-purpose Regional Geochemical Survey recommended by CGS (2005), respectively, were both generally below 5%. The detection limits for As, Hg, Cu, Pb, Zn, Cr, MN, and Cd were 0.6, 0.0004, 1, 2, 2.5, 10, and 0.01 mg/kg, respectively, while those for Al_2O_3, CaO, Fe_2O_3, MgO, SiO_2, K_2O, and NaO were 0.03%, 0.03%, 0.05%, 0.03%,0.1%, 0.05%, and 0.1%, respectively.

Methodology

Descriptive Analysis

Descriptive statistics, including the mean, range, standard deviation (SD), and coefficient of variations (CVs), were performed after analysis. The SD and CVs were incorporated to reflect the degree of dispersion distribution of different metals to indirectly indicate the activity of the selected elements in the examined environment [6]. In addition, the normal distribution of each variable was evaluated using the Kolmogorov-Smirnov test based on the raw data.

Multivariate Statistical Analysis

Pearson correlation coefficients were calculated to determine the relationships among metals and major elements to investigate elemental associations among the heavy metals and major elements in the soils.

Principal component analysis (PCA) is the most common multivariate statistical method used in environmental studies and is employed to extract a small number of latent factors for analyzing relationships among the observed variables [7, 19]. Like other multivariate statistical methods, PCA is useful in reducing data dimension while retaining important information and representing variables in a form that can be easily interpreted [6, 24]. In this method, PCA results will vary considerably depending on whether the covariance or correlation matrix is used. In this study, PCA are calculated based on the correlation matrix because the concentrations of the heavy metals evaluated in this study vary by different orders of magnitude [6]. The first few Eigenvectors (or PCs) account for most of the data set variability [6]. To make the results of the present study more easily interpretable, varimax with Kaiser normalization was used as the rotation method to maximize the variances of the factor loadings across variables for each factor [6, 7]. Moreover, prior to conducting PCA, Kaiser-Meyer-Olkin (KMO) and Bartlett's sphericity tests were used to examine its validity [32]. The KMO and Bartlett's results for this study were 0.642 and 1432.871 ($p<0.05$), respectively, indicating that PCA may be useful for dimensionality reductions. In this study, all principal factors extracted from the variables were retained with the percentage of accumulated contribution of variances being larger than 80%.

The raw data were standardized according to the Z score (whose mean and variance were set to zero and one, respectively) to minimize the effects of differences in measurement units or variance, and to render the data dimensionless [6].

Table 1. Threshold concentrations of each level of pollution for each metal in urban dust (mg/kg).

	X_a	X_c	X_p	X_{bp}
As	15.00	25.00	30.00	50.00
Cd	0.15	0.30	1.00	3.00
Cr	90.00	200.00	300.00	600.00
Cu	30.00	50.00	400.00	1000.00
Hg	0.15	0.30	1.50	3.00
Pb	35.00	250.00	500.00	1000.00
Zn	100.00	200.00	500.00	1000.00

Methods of Heavy Metal Pollution Assessment

To assess the degree of metal contamination, a revised pollution index for each metal and NIPI of the seven metals were attributed to each sampling site as proposed by [33]. The PI was defined as follows:

$$\begin{cases} PI = C_i / X_a & C_i \le X_a \\ PI = 1 + \left(C_i - X_a\right) / \left(X_c - X_a\right) & X_a < C_i \le X_c \\ PI = 2 + \left(C_i - X_c\right) / \left(X_p - X_c\right) & X_c < C_i \le X_p \\ PI = 3 + \left(C_i - X_p\right) / \left(X_{bp} - X_p\right) & X_c < C_i \le X_p \\ PI = 4 + \left(C_i - X_{bp}\right) / \left(X_{bp} - X_p\right) & C_i > X_{bp} \end{cases} \tag{1}$$

...where C_i is the measured concentration of each metal (As, Cu, Cr, Cd, Hg, Pb, and Zn) in this study, X_a is the threshold concentration of the heavy metal enrichment, X_c is the threshold concentration of the low level of pollution, X_p is the threshold concentration of the high level of pollution, and X_{bp} is the threshold concentration of serious pollution. The values of X_a, X_c, X_p, and X_{bp}, defined in Table 1, were the integration results of the document published by [6], the Environmental Quality Standard for Soils published by the National Environmental Protection Agency of China, and the Environmental Quality Standard for urban dust in Changchun suggested by [34].

The PI of each metal was classified as: non-pollution ($PI<1$), indicating that the level of metals was below the threshold concentration, but does not necessarily mean there was no pollution from anthropogenic sources or other enrichment over the background, low-level pollution ($1\le PI<2$), moderate-level pollution ($2\le PI<3$), high-level pollution ($3\le PI<4$), and extremely high-level pollution ($PI\ge 4$). The NIPI of the seven metals for each sampling site was defined as follows [33]:

$$NIPI = \sqrt{\frac{PI^2{}_{i\max} + PI^2{}_{iave}}{2}} \tag{2}$$

...where PI_{imax} is the maximum PI value of each heavy metal and PI_{iave} is the average PI value of each heavy metal.

Table 2. Heavy metal (mg/kg) and main element (%) concentrations of urban dusts in Changchun.

	As	Cd	Cr	Cu	Hg	Pb	Zn	Mn
Minimum	8.95	0.212	66.11	35.71	0.038	40.3	164	547
Maximum	114.4	7.379	175.54	636.95	6.656	523.6	2533	1326
Mean	23.26 [a]	0.624 [a]	95.98 [b]	68.41 [a]	0.239 [a]	93.6 [a]	465.35 [b]	692 [a]
Standard deviation	10.37	0.84	18.19	45.05	0.54	54.43	295.96	75.22
Variation coefficient	0.45	1.35	0.19	0.66 [c]	2.26 [c]	0.58 [c]	0.64	0.11 [c]
Skewness	1.09 [c]	1.43 [c]	1.95	1.94 [c]	0.96 [c]	1.24 [c]	3.84	1.85
Kurtosis	5.696 [c]	3.127 [c]	4.865	8.981	1.880	3.679	18.59	9.316 [c]
TD [d]	LgN	LgN	Skewed	LgN	LgN	LgN	Skewed	LgN
Changchun topsoil MV [e]	12.5	0.132	66.0	29.4	0.118	35.4	90.0	743
Atmospheric dry and wet deposition [f]	38.9	2.241	92.3	76.17	0.27	115.2	462	—
Changchun BK[g]	10.590	0.09	54.17	18.9	0.04	19.06	59.860	—
Jilin BK [h]	11.60	0.09	42.40	14.8	0.03	14.96	45.950	605
Jilin MV [i]	8.82	0.11	51.4	18.0	0.03	24.4	57.22	674
Residential/recreational intervention limits	20	2	150	120	1	100	150	—
Industrial/commercial intervention limits	50	15	800	600	5	1000	1500	—
	Al_2O_3	CaO	Fe_2O_3	K_2O	MgO	Na_2O	SiO_2	S
Min	9.56	2.82	3.71	1.38	1.26	0.98	36.61	0.06
Max	16.01	19.60	8.46	3.51	4.27	3.47	60.95	1.92
Mean	13.17 [b]	5.06 [a]	4.53 [a]	2.31 [b]	1.76	1.85	54.35 [b]	0.26
Standard deviation	0.64	1.59	0.55	0.19	0.30	0.25	2.83	0.22
Variation coefficient	0.05	0.31	0.12	0.08	0.17	0.14	0.05	0.85
Skewness	-0.63	1.66 [c]	1.80 [c]	1.11	1.43 [c]	0.15 [c]	-1.85	0.33 [c]
Kurtosis	5.924	7.784 [c]	6.668 [c]	11.693	5.882 [c]	6.840 [c]	8.126	0.912 [c]
TD [d]	Normal	LgN	LgN	Skewed	LgN	LgN	Normal	LgN
Changchun topsoil MV	14.05 [b]	1.74 [b]	4.56 [a]	2.61 [b]	1.27 [b]	1.77 [b]	64.52 [b]	—
Jilin BK [h]	ND	1.010	ND	1.940	ND	1.540	ND	—
Jilin MV [i]	12.83	1.28	3.92	2.59	1.18	1.91	66.58	—

[a] Geometrical mean, [b] Arithmetical mean, [c] Values were obtained using a lognormal transformation, [d] Type of distribution, [e] Average values of the topsoil in Changchun, A layer (0~20 cm), total 352 samples [30], [f] Average values of atmospheric dry and wet deposition in Changchun [27], [g] Background values of the soil in Changchun [36], [h] Background values of the soil in Jilin Province [36], [i] Average values of the soil in Jilin Province, A layer (0~20 cm), about 8,000 samples,
ND – No data, Normal – normal distribution, LgN – lognormal distribution

The NIPI was classified as: non-pollution (NIPI≤0.7), warning line of pollution (0.7<NIPI≤1), low level of pollution (1<NIPI≤2), moderate level of pollution (2<NIPI≤3), and high level of pollution (NIPI>3) [33].

Results and Discussion

Concentrations of Heavy Metals and Major Elements

The concentrations of heavy metals and major elements in the urban dusts of Changchun and their descriptive statistical results are listed in Table 2. The results of the Kolmogorov-Smirnov test ($p<0.01$) showed that Al_2O_3 and SiO_2 concentrations fit a normal distribution (K-S probability were 0.32 and 0.04, respectively), while the As, Cd, Cr, Cu, Hg, Pb, Zn, Mn, CaO, MgO, Na_2O, S, Fe_2O_3, and K_2O were skewed. However, the As, Cd, Cu, Hg, Pb, Mn, CaO, MgO, Na_2O, S, and Fe_2O_3 levels were found to almost fit a normal distribution after the data were naturally logarithmically transformed (K-S probability were 0.48, 0.02, 0.11, 0.15, 0.03, 0.08, 0.14, 0.09, 0.04, 0.58, and 0.5, respectively). There is an extremely significant difference for Cr, Zn, and K_2O, which is observed to fit a skewed distribution

Table 3. Comparison of mean concentrations (mg/kg) of metal in urban dusts from different cities (mg/kg).

City	As	Cd	Cr	Cu	Hg	Pb	Zn	height	References
Changchun (N=232)	23.26	0.62	95.98	68.41	0.24	93.6	465.35	Urban dusts	This study
Nanjing (N=35)	13.40	1.10	126.00	123.00	0.12	103.0	394.00	Street dusts	[4]
Xi'an (N=65)	10.62	—	167.28	94.98	0.64	230.5	401.46	Street dusts	[6]
Zhuzhou (N=55)	89.00	41.40	125.00	139.00	0.92	956.0	2379.00	Street dusts	[9]
Greater Toronto (N=42)	—	0.51	197.90	162.20	—	182.8	232.80	Street dusts	[11]
Shanghai (N=273)	—	1.23	159.30	196.80	—	294.9	733.80	Street dusts	[16]
Xi'an (N=92)	28.50	—	—	102.70	—	266.3	798.00	Street dusts	[20]
Chengdu (N=318)	41.90	4.33	112.00	240.00	0.54	372.0	1078.00	1.5 m height	[40]
Birmingham (N=100)	—	1.62	—	466.90	—	47.1	385.70	Street dusts	[41]
Beijing (N=50)	—	0.64	69.33	72.13	—	201.8	219.20	Street dusts	[42]
Baoji (N=38)	19.80	—	126.70	123.20	1.10	433.2	715.30	Street dusts	[43]
Urmqi (N=169)	—	1.17	54.28	94.54	—	53.5	294.47	Street dusts	[44]

"—" – No data, N – Number of samples.

even though the data were naturally logarithmically transformed. The mean values were adopted for analyses when the heavy metal concentrations had normal distributions. The geometric mean values were used when the heavy metal concentrations had lognormal distributions. Each heavy metal showed a wide variety of concentration, which is typical in urban dusts and may imply that there are some hot-spots in Changchun that have been polluted by heavy metal pollution in urban dust [11]. However, most of the major elements showed a relatively narrow interval, except for CaO and S. The relatively wide range of CaO may have been caused by the excessive input of building materials into urban dusts, such as cement and calcareousness [29]. When compared with the background values of the topsoil in the Changchun region, the mean concentrations of all elements were significantly elevated, and these values were also significantly higher than the background values of Jilin [35, 36]. In addition, it should be noted that the heavy metal and CaO concentrations in urban dusts of Changchun are generally higher than those of the mean concentrations in soils from Jilin Province. The mean concentrations of Al_2O_3, Fe_2O_3, K_2O, MgO, Na_2O, and SiO_2 were comparable with the mean concentrations in soils from Jilin Province and in topsoil from Changchun. Compared with the mean concentrations of heavy metals in atmospheric dry and wet depositions, the mean concentration of heavy metals in urban dust was generally low, except for Cr. However, it is much higher than those of the mean concentrations in topsoil in Changchun [30]. Taken together, these findings may imply that anthropogenic input is responsible for the presence of these elements in urban dust.

Based on the variation coefficients (VCs), the examined elements can be classified into three groups: Cr, Mn, Al_2O_3, K_2O, Fe_2O_3, MgO, Na_2O, and SiO_2, which had VC values lower than 0.2; Cd and Hg, which had VC values higher than 1.0; and all other elements with VC values between 0.2 and 1.0. We would expect those elements dominated by a natural source to have low VCs, while the VCs of elements influenced by anthropogenic sources would be expected to be quite high. This is the case for urban dusts, for they have undergone erosion and aeolian transport before ultimate deposition, and have therefore been fully mixed [6].

The mean values of most heavy metals in the analyzed urban dusts in Changchun are generally much lower than those reported for samples from some large and/or industrialized cities (Chengdu, Nanjing, Shanghai, Xi'an, Hong Kong, Zhuzhou, Baoij, Toronto, Birmingham, etc.), but are generally higher than those measured in smaller, undeveloped cities (i.e. Urmqi), particularly Cr, Pb, and Zn (Table 3). Although Changchun is one of the oldest industrial cities in China, the industrial history of Changchun is shorter than that of other industrialized cities, and pollution occurred at a later stage in these areas. Therefore, although near-surface urban dust from a height of 1.5 m was collected in this study, and samples were collected from street surfaces and from areas with different background environments in other studies (Table 3), the relatively lower concentrations of heavy metals in the urban dust in Changchun could be the result of a shorter accumulation time and lower rate. Certainly, the relatively high sampling height of this study may also influence the concentration levels. Particularly, it should also be noted that the concentrations of Pb in the analyzed samples are also significantly lower than those reported in studies of metropolitan areas in cities such as Xi'an, Chengdu, Beijing, London, Shanghai, Zhuzhou, and Baoji; these samples are comparable to those measured in some other parts of the world (i.e. Nanjing and Hong Kong). Meanwhile, the concentrations of Zn in the analyzed samples are also significantly lower than those in Chengdu, Hong Kong, and Zhuzhou, and are comparable to

Table 4. Pearson's correlation matrix for metal concentrations.

	As	Cd	Cr	Cu	Hg	Pb	Zn
As	1.000						
Cd	0.081	1.000					
Cr	-0.094	0.085	1.000				
Cu	0.012	0.065	0.205**	1.000			
Hg	0.020	-0.063	-0.053	0.054	1.000		
Pb	0.205**	0.137*	0.281**	0.427**	-0.005	1.000	
Zn	0.072	0.068	0.066	0.170**	-0.027	0.130*	1.000

*Correlation is significant at the 0.05 level (2-tailed).
**Correlation is significant at the 0.01 level (2-tailed).

Table 5. Matrix of the principal component analysis loadings of metals and major elements of urban dusts in Changchun city[a].

Variable	Principal loading factors							Communalities
	PC1 (18.213%)	PC2 (14.39%)	PC3 (11.20%)	PC4 (10.01%)	PC5 (9.44%)	PC6 (9.17%)	PC7 (9.12%)	
As	-0.180	0.171	**0.849**	-0.177	-0.058	-0.025	0.074	0.823
Cd	0.088	0.064	0.042	0.007	0.034	-0.026	**0.988**	0.992
Cr	**0.688**	0.402	-0.030	0.119	-0.075	-0.031	-0.020	0.657
Cu	0.116	**0.752**	-0.075	0.125	0.195	0.106	-0.016	0.650
Pb	-0.025	**0.867**	0.125	-0.136	-0.023	-0.062	0.097	0.800
Zn	0.016	0.130	0.058	-0.021	**0.960**	-0.026	0.034	0.945
Hg	-0.080	0.033	0.012	0.005	-0.024	**0.988**	-0.026	0.984
CaO	-0.021	0.014	-0.006	**0.946**	-0.023	0.002	0.008	0.896
Fe_2O_3	**0.788**	0.035	0.083	-0.171	0.141	-0.106	0.051	0.691
MgO	0.293	-0.191	**0.688**	0.301	0.213	0.059	-0.033	0.737
Mn	**0.873**	-0.121	-0.062	0.079	-0.041	0.021	0.075	0.795

[a] Extraction Method: Principal Component Analysis. Rotation Method: Varimax with Kaiser Normalization. Rotation converged in 7 iterations.
The percentage in brackets is the explained contribution of principal components to the total variance.
The boldfaced numbers are the dominant elements in different PCs.

those measured in London, Birmingham, and Xi'an. Because each city has its own characteristic combination of elemental compositions, the observed similarities, as well as variations, may not reflect actual natural and anthropogenic diversities among the different urban settings. Therefore, there is an immediate need to establish a standard procedure to represent and analyze urban samples.

Correlation Coefficient Analysis

To a certain extent, Pearson's correlation coefficient can be used to measure the degree of correlation between the metal data and can provide suggestive information regarding heavy metal sources and pathways [6, 18]. The results of Pearson's correlation coefficients of heavy metals in urban dusts in Changchun are summarized in Table 4. Pb were significantly positively correlated to As, Cd, Cr, Cu, and Zn. Meanwhile, Cu also displayed a significant positive correlation with Cr and Zn, indicating that the general contamination sources for these metals were primarily traffic and industrial activities [18]. Furthermore, it should be noted that Hg was not significantly correlated with all other heavy metals considered, indicating that there are some unique sources of Hg that are currently unknown.

Multivariate Statistical Analysis

In the present study, PCA was applied to assist in the identification of the sources of pollutants [7, 22]. The results of PCA of the metal concentrations in the urban soils are shown in Table 5. The first seven principal components were considered during the PCA, accounting for over 81.54% of the total variance, reflecting the majority of the data [7].

Table 6. The average concentrations of metals in gasoline, diesel oil, and tire and vehicle exhaust in Changchun.

	Gasoline (g/L)	Diesel oil (mg/kg)	Tire dust (mg/kg)	Vehicle exhaust (gasoline) (mg/kg)	Vehicle exhaust (diesel oil) (mg/kg)
Cu	0.002	2.430	16.480	255.0	109
Pb	2.11	0.130	12.7	440.0	45.75
Zn	No data	No data	No data	1900	1898
References	[29]	[29]	[29]	[30]	[37]

PC1 was dominated by Cr, Fe_2O_3, and Mn, accounting for 18.21% of the total variance. In general, Fe_2O_3 and Mn represented a natural geochemical association of major rock-forming elements in soils, which may reflect a natural origin from weathered soil dust [7, 19]. The first principal component accounted for most of the variability in the data, and each succeeding component accounted for a reduced percentage of the remaining variability. Hence, fugitive dust from soil dust was shown to have a dominant influence on urban dusts in Changchun, and PC1 may be summarized as a ‘natural’ factor. Even some previous studies reported that Cr might originate from industrial discharges or traffic sources [7, 19]. The relatively low mean concentration of Cr (95.98 mg/kg) can be compared with the mean concentrations in Changchun topsoil (66.0 mg/kg) and may further support the conclusion that Cr may still be controlled by natural sources. However, it should be noted that Cr was positively correlated with Cu and Pb upon correlation coefficient and PCA analysis (PC2), suggesting less significant anthropogenic origins such as traffic emissions.

PC2 was dominated by Cu and Pb. The strong relationship between these two elements may reflect anthropogenic contamination of urban dusts. In fact, this factor source may usually be explained by contributions mainly from vehicle emissions, especially cars that consume lead gasoline [19]. Over the past 50 years, vehicle emissions have been considered to be the principal source of Pb in soil and urban dusts. This has been verified by many related studies [7, 19]. Previous studies showed that even though the use of lead petroleum has been banned in Changchun since 1998, and the content of Pb in the troposphere has decreased [37]. The concentration of Pb in urban soils still reflects the significant degree of historical Pb contamination and the long half-life of Pb in soils, which is one of the sources of urban dust [37]. The mean concentration of Pb in urban dusts was 93.6 mg/kg in the present study, while it was found to be 255.0 mg/kg in the car emission dust of 2005 [29]. In addition, Cu was also present in high concentrations in the automotive exhaust in Changchun (Table 6). Copper alloy is a material used in mechanical parts due to its desirable qualities such as corrosive resistance and strength. Copper is also used in Cu-brass automotive radiators and brakes due to its high corrosive resistance and high thermal conductivity. However, the deterioration of the mechanical parts in vehicles over time will result in Cu being emitted into the surrounding environment (Adachi and Tainosho documents that the Cu content in brake dust is 1.2%) [23].

PC3 is loaded primarily by As and MgO and accounts for 11.20% of the total variance. The source for this factor is soil dust. MgO generally represents a natural geochemical association of major rock-forming elements in soils and may reflect a natural origin from parent rocks. The strong relationship between As and MgO indicates that As in urban dusts may primarily originate from natural sources, and the effect of some other anthropogenic sources, such as traffic emissions, industrial discharges, coal combustion, etc., could be negligible. Based on these findings, PC3 may be labeled as a ‘natural’ factor.

CaO, Zn, Hg, and Cd were unequivocally isolated in the fourth, fifth, sixth, and seventh components (PC4, PC5, PC6, and PC7), respectively. The PCA analysis showed that CaO displayed a relatively weak association with most of the other elements; but it did show a relatively significant correlation with MgO. These features may indicate that CaO has some unique sources. In fact, excessive input of building materials into the urban environment may be the reason following the analysis mentioned above. Meanwhile, the content of Ca in building dust was 12.15% in Changchun [30]. However, the origin of CaO in urban dust in Changchun should be confirmed by further studies. Zn also displayed a relatively weak association with most of the other elements. Some studies imply that traffic activities may be a significant contribution to Zn [6, 19], especially the mechanical abrasion of vehicles. Zinc, added as an additive (mainly as antioxidants, e.g., Zinc carboxylate complexes and zinc sulphonates) during the vulcanizing process, comprises from 0.4% to 4.3% of the resulting tire tread, and is a vital and significant source of Zn pollution in dusts [23]. Therefore, the origin of Zn may mainly result from anthropogenic sources, especially traffic dust. In addition, spatial distribution of high values of Zn are always near heavy traffic routes, and do show some relationship to traffic activities [34]; this feature is in agreement with the literature published by Han et al. [6].

The sixth group of elements only consists of Hg (PC6). Elements such as Hg are commonly considered to be indicators of coal combustion and waste incineration [38]. The analysis of six common coal combustion fly ash samples collected from Changchun revealed that the mean concentrations of Hg in the coal combustion fly ash were 3.492 mg/kg. In 2012, approximately 24.7 million tons of coal were consumed in this city. Meanwhile, it should be pointed out that, besides the coal combustion emission, Hg can also be emitted from the smelting of sulfide mineral.

Table 7. Statistical results of pollution index (PI) of heavy metals in Changchun urban dusts.

	PI			Number of samples				
	Min	Max	Mean	Non-pollution	Low	Middle	High	Extremely high
As	0.59	8.22	2.03	13	131	52	32	4
Hg	0.25	7.44	1.53	64	92	70	5	1
Cr	0.73	1.78	1.05	99	133	0	0	0
Cu	1.29	3.39	2.04	0	26	205	1	0
Pb	1.02	3.05	1.31	0	226	5	1	0
Zn	1.64	8.07	2.82	0	4	169	49	10
Cd	1.41	7.19	2.56	0	7	196	22	7

The atmospheric depositions of Hg were 0.030 mg/m^2/a, and the contribution of coal burning was significant [27]. In addition, spatial distribution of high values of Hg do show some relationship to industrial activities such as waste incineration, metal refining and manufacturing, papermaking, coal-fired power plants, and the chemical industry [34]; this feature is in agreement with the literature [39].

Cd was unequivocally isolated in the seventh component (PC7) of Changchun urban dust and showed a weak association with other elements. This may indicate that it had some unique sources. In fact, industrial discharge may contribute significantly to the accumulation of Cd in urban dusts [19]. The spatial distribution of Cd also confirmed these inferences, as documented previously by literature [34] in more detail.

Although the results of the present study provide preliminary conclusions regarding the origin of each metal, further studies are necessary to gain a better understanding of pollution sources based on the spatial distribution of each metal.

Metal Pollution Assessment

The minimum, maximum, and mean values of PI for each element are shown in Table 7. The mean PI values were generally low, indicating that there was no serious dust pollution. However, the PI values varied greatly among metals and among different sample locations. The PI values for Cr in urban dust ranged from 0.73 to 1.78, indicating that urban dust in Changchun was uncontaminated to low-level contaminated. Indeed, most of the samples (133 samples) had low PI values for Cr, and the other 99 samples had PI values indicating non-pollution. These findings suggest that the concentrations of Cr in urban dusts were comparable to the first grade threshold value established by the Environmental Quality Standard for Soils and suggested by literature [34]; also, there was no obvious pollution of Cr in the urban dust. This finding may further support the conclusion that Cr may still be controlled by natural sources. The PI values for Cu and Pb in urban dust ranged from 1.29 to 3.39 and 1.02 to 3.05, respectively. Most of the samples (231 samples) had low or moderate PI values for Cu and Pb. However, only 1 sample had PI values indicating heavy pollution. Thus, the dust quality of Changchun has deteriorated relative to natural conditions, and it is likely that many of the urban dusts in Changchun have been polluted with low or moderate levels of Cu and Pb. Meanwhile, the mean value of the PI value for Cu was 2.04, displaying a moderate level of pollution; the Pb was a low level of pollution (mean PI value for Pb was 1.31). Nearly 76.93% of the study area displayed a moderate level of pollution for Cu.

The PI values of Cd, Zn, As, and Hg in urban dust varied greatly, ranging from 1.41 to 7.19, 1.64 to 8.07, 0.59 to 8.22, and 0.25 to 7.44, respectively, implying that some sample locations were seriously contaminated. Furthermore, high PI values (higher than 2) were observed in 97.83% of the samples for Cd, 98.76.3% for Zn, 37.93% for As, and 32.76% for Hg. These data indicate that Cd, Zn, As, and Hg pollution, particularly Cd, Zn and As pollution, is relatively serious in Changchun topsoil when compared with other elements. The mean PI values of Cd, Zn and As were 2.56, 2.82, and 2.03, respectively.

There was no significant difference for PI values among different districts (Fig. 3). However, the mean PI values for As and Hg in urban dust were generally higher in Nanguan and Chaoyang. Nanguan is an old urban area with a dense population, heavy traffic, and many industrial activities when compared with other districts, indicating the presence of relatively serious heavy metal pollution in Nanguan District. Chaoyang District is the downtown area with a dense population and has the highest traffic density. The reason for the relatively high PI values in Nanguan and Chaoyang districts should be considered and further studies should be conducted in the future.

The NIPIs of all samples varied from 1.76 to 6.12, with an average of 2.63, indicating a moderate level of pollution of local urban dusts (Table 8). Assessment of the data shows that there were only 19 samples (about 8.19% of all samples) with an NIPI value below 2, while approximately 80.60% of all samples (187 samples) had a NIPI value between 2 and 3, indicating high levels of pollution of local urban dusts, only 1.72% of all samples (4 samples) had a NIPI value between 3 and 4 and 9.48% of all samples

Table 8. Statistical results of Nemerow integrated pollution index (NIPI) of heavy metals in urban dusts of Changchun.

NIPI			Number of samples				
Min	Max	Mean	Non-pollution (NIPI≤0.7)	Warning line pollution (0.7<NIPI≤1)	Low level pollution (1<NIPI≤2)	Middle level pollution (2<NIPI≤3)	High level pollution NIPI≥3
1.76	6.12	2.63	0	0	19	187	26

(22 samples) had an NIPI>4, indicating a high level of pollution of local urban dusts. Overall, these findings suggest that the dust of Changchun has been polluted by anthropogenic emissions. Fig. 4 shows the spatial distributions of NIPIs in Changchun. Dust sample sites with extremely high levels of pollution (NIPI>3) were primarily located in areas close to facilities for smelting, the chemical production, tanneries, etc.. Meanwhile, the hot-spot areas are distributed relatively separately. Therefore, metals present in the emissions from industrial facilities may significantly affect urban dust in these sites. Although most of these facilities have been relocated in recent years, the historical accumulation of metals should not be ignored.

Conclusions

The present study examined the content of metals in the urban dusts in Changchun in northeast China. Major elements in urban dusts were also analyzed, including Mn, Al_2O_3, CaO, Fe_2O_3, MgO, SiO_2, K_2O, and NaO. Urban dusts in Changchun were found to have elevated concentrations of a number of heavy metals, including Cu, Pb, Hg, As, Cd, Cr, and Zn. The mean concentrations of metals and CaO were significantly higher than the background values of topsoil in the Changchun region, and were also higher than the mean concentrations in soils from Jilin Province, while the mean concentrations of K_2O, MgO, Na_2O, and SiO_2 were comparable to the mean concentrations in soils from Jilin Province and in topsoil from Changchun. These findings may reflect the influence of urbanization and industrialization on the areas considered. However, the concentrations of heavy metals in urban dusts from Changchun were generally lower than those recorded in other large and/or industrialized cities, particularly for Cr, Pb, and Zn. This may indicate that the accumulation time and rate have an important influence on the content of heavy metals. Pb was significantly positively correlated to As, Cd, Cr, Cu, and Zn. Meanwhile, Cu also displayed a significant positive correlation with Cr and Zn. Furthermore, it should be noted that Hg was not significantly correlated with all other heavy metals considered.

Based on total metal concentrations and VC analysis, coupled with correlation coefficient analysis, two metal groups originating from natural and anthropogenic sources may be distinguished. Specifically, the concentration of Cr appears to be controlled by natural sources, whereas the levels of As, Hg, Cd, Cu, Pb, and Zn in Changchun urban dusts mainly originate from anthropogenic sources. Furthermore, PCA results revealed that Cu and Pb primarily came from vehicle emissions, while Zn in the urban dusts came from traffic sources – especially vehicle tires. Meanwhile, Hg primarily originated from coal combustion, and Cd was found to be mainly associated with industrial sources inferred from the PCA, concentration level, and analogous studies in other areas. Finally, Cr and As were found to be mainly controlled by natural factors originating from soil dust; the influence of anthropogenic activities on the Cr levels was generally low. However, it should be noted that the Cr was positively correlated with Cu and Pb upon correlation coefficient and PCA analysis (PC2), which may imply less significant anthropogenic origins such as traffic emissions.

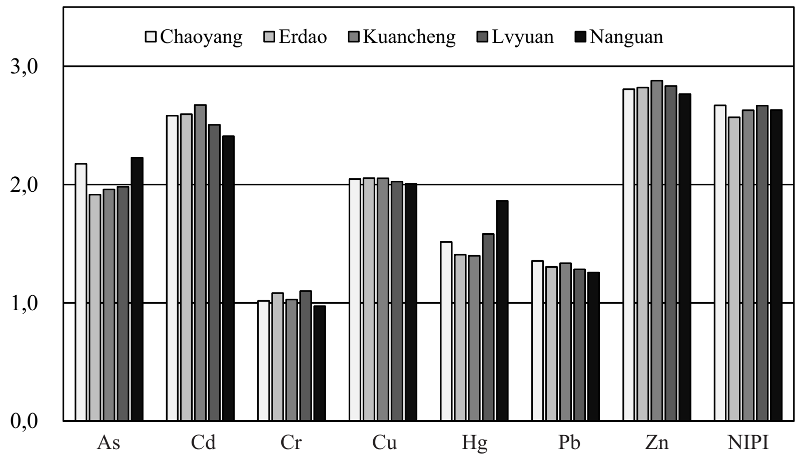

Fig. 3. The mean pollution index (PI) and Nemerow integrated pollution index (NIPI) of heavy metals in the urban dusts in different districts of Changchun City.

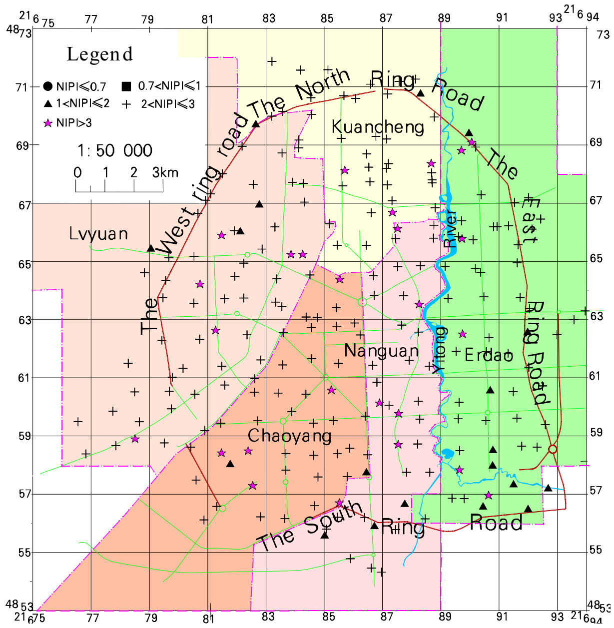

Fig. 4. Spatial distribution of the Nemerow integrated pollution index (NIPI) in the studied area.

The PI values of the metals measured in the urban dusts indicate that a significant degree of metal pollution exists in some urban dusts within the urban areas of Changchun, particularly for Cu, Cd, Zn, and As. The levels are not especially high as a whole, but there are clearly contaminated hot-spot areas distributed separately in the studied region. The mean NIPI values also indicate that urban dusts in Changchun have reached the moderate level of pollution by metals. However, it should be noted that about 11% of all samples had a NIPI value above 3, suggesting high levels of contamination by metals, particularly inside the loop road. These findings indicate that more attention should be given to metal pollution of the urban dusts in Changchun. The ecological and health implications of these findings should be considered in future investigations.

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