



Assessment of toxic elements in road dust from Hutou Village, China: implications for the highest incidence of lung cancer

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Abstract

We attempt to understand the pollution characteristics and carcinogenic risk of toxic elements around Hutou Village, Xuanwei City, Yunnan Province, China. For this propose, 48 road dust samples were collected systematically, and the concentrations of Cr, Ni, Cu, Zn, As, Cd, Pb, Co, and Cr(IV) were analyzed and compared; the spatial distribution was obtained. The I_{geo} and EF indices and multivariate statistical analysis (CA, PCA, HACA) were carried out for source investigation, and human health risk assessment was also adopted to evaluate local non-carcinogenic and carcinogenic risks. The result showed that Cr, Ni, Cu, Cd and Co contaminations were quite serious; Zn, As, Cd, and Pb had similar distribution pattern, and Cr and Ni also shared similar distribution characteristics; Cd, Pb, Zn, and As ascribed to anthropogenic sources, while Cr and Ni originated from either anthropogenic activity or natural sources; Co and Cu originated from natural sources; the non-carcinogenic risk of Co cannot be ignored. The carcinogenic risk of Ni was considered unacceptable. Finally, an indoor coal-burning pattern was established that the high Cd and Ni inhalation and ingestion model was associated with lung cancer.

Keywords Toxic elements · Road dust · Spatial distribution · Pollution characteristic · Risk assessment · Hutou Village

Highlights

1. The pollution in the southeast of Hutou Village was more serious than that in the northeast.
2. The Cr, Ni, Cu, Cd, and Co contaminations were quite serious.
3. The source of toxic elements originated from both anthropogenic activity and natural sources.
4. The carcinogenic hazard of Ni and carcinogenic hazard of Co in Hutou Village were obvious.
5. The implications of the indoor coal-burning pattern could lead to lung cancer.

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Introduction

Road dust is particulate, liquid and gaseous substances (Wang et al. 2019a, b) that accumulate short-term surrounding materials from not only natural sources (e.g., soil weathering, volcanic ash, forest fires, plant remains, and pollen) (Žibret 2018) but also various kinds of human activities (e.g., weathering of construction materials, vehicular emissions (exhaust, tire, and brake wear), industrial activity, fuel combustion) (Li et al. 2013; Tang et al. 2013; Liu et al. 2014; Škrbić et al. 2018). The accumulation of toxic elements would cause long-term damage through inhalation (respiratory system), ingestion (digestive system), and dermal contact (skin) on people (Zhang et al. 2013). Furthermore, dust that contains toxic elements can access the soil environment by wind and aquatic systems via runoff (Al-Awadhi and AlShuaibi 2013). Therefore, the pollution of toxic elements in road dust has attracted extensive concerns and a large number of studies in the world (Ordóñez et al. 2015; Soliman et al. 2015; Tang et al. 2017).

The quantification of the contamination level in road dust could be determined by various pollution indices (Jafarabadi et al. 2017). The basic indices mainly include (1) geoaccumulation index (I_{geo}) (Müller 1969) and (2) enrichment factor (EF) index (Zhuang et al. 2018). The basic multivariate statistical analysis (Pan et al. 2017) mainly the following contents: (1) Pearson correlation analysis (CA); (2) principal component analysis (PCA) (Alahabadi and Malvandi 2018); (3) hierarchical agglomerative cluster analysis (HACA) (Kynčlová et al. 2017; Jafarabadi et al. 2017). Human health risk assessment of toxic elements could quantify the potentially harmful effects on human (US EPA 1992; Baghaie and Aghili 2019; Saher and Kanwal 2019), included indicators such as hazard quotient (HQ_{ij}), hazard index (HI_i), $Risk_i$, and TRisk (Xu et al. 2014).

Xuanwei City has one of the highest rates of lung cancer incidence and mortality (Lan et al. 2008), which is about five times of the Chinese national average (Downward et al. 2014; Chen et al. 2015a) among both males and females, although almost all women are non-smokers (Chuang et al. 1992), especially in Hutou Village. Through literature, we found that toxic elements (Downward et al. 2017) in the local environment are relatively high which were associated with not only natural sources owing to geological origin of the margin of Southwest China Emeishan large igneous province (ELIP) (YGMRB 1982; Xie et al. 2008) but also anthropogenic activity of coal mine development (Downward et al. 2014; Dai et al. 2014).

Many scholars believe that the highest incidence and mortality of local lung cancer was related to the products of indoor coal-burning (Tian 2005; Xiao et al. 2012; Barone-Adesi et al. 2012; Kim et al. 2014), such as PAHs (Downward et al. 2014; Chuang et al. 1992), toxic elements (Lu et al. 2013; Tan et al. 2018; Dai et al. 2008), and nano-quartz particles (Lu et al.

2016; Downward et al. 2017). These materials have greatly enriched the understanding of local environmental problems and achieved certain results. In this case, the development of Yantang coal mine (near Hutou Village) had been stopped, and the transformation of local stoves (Li 2016) was completed in 1980; indoor coal-burning has been scarce; however, the local incidence and mortality of lung cancer (Chen et al. 2015a) did not decline as expected. At the same time, many scientific achievements have some limitations in explaining the high incidence of lung cancer in local women.

The most important objectives were (1) to obtain the spatial distribution of Cr, Ni, Cu, Zn, As, Cd, Pb, Co, and Cr(VI); (2) to quantify the pollution level of toxic elements; (3) to explore the origins; (4) to evaluate the potential health effects on human; and (5) to obtain some implications between toxic elements and high incidence of lung cancer in this region.

Methodology

Study area

Hutou Village belonging to Laibin Town, Xuanwei City, Yunnan Province, China, is located in the southwest side of Laibin Town, 4 km away from Laibin Town and 12 km away from the urban area (26° 17' 55.10" N, 104° 7' 58.99" E), in the vicinity of the Xuanwei Changzheng middle school and Xuan-Ke road with heavy vehicle traffic (Fig. 1) and neighboring the Yantang coal mine with a distance less than 2 km in the north. Moreover, Hutou Village is the area with the highest incidence and mortality of lung cancer in Xuanwei, especially in women.

Sample collection and analysis

Sample collection

This work was accomplished in October 2017, when the weather was cool and dry. Each sample was collected by sweeping about a 5–20-m² area near the curb (within 0.5–2 m of the road) with new clean plastic tools (broom and dustpan). Forty-eight samples were collected (2 duplicate samples per sampling point) around Hutou Village (the sampling area was 2.5 km²); each sample weighed about 1000 g, stored in plastic bags. The locations (Fig. 1) were recorded by GPS (Magellan Xplorer 510). The samples were taken back to the laboratory and dried at room temperature for 1 month; then, the leaves, large sand grains, and other impurities were removed and thereafter were sieved by a 200-mesh (about 74.2 μm) nylon sieve. They were then stored in plastic bags to be analyzed for concentrations of toxic elements.

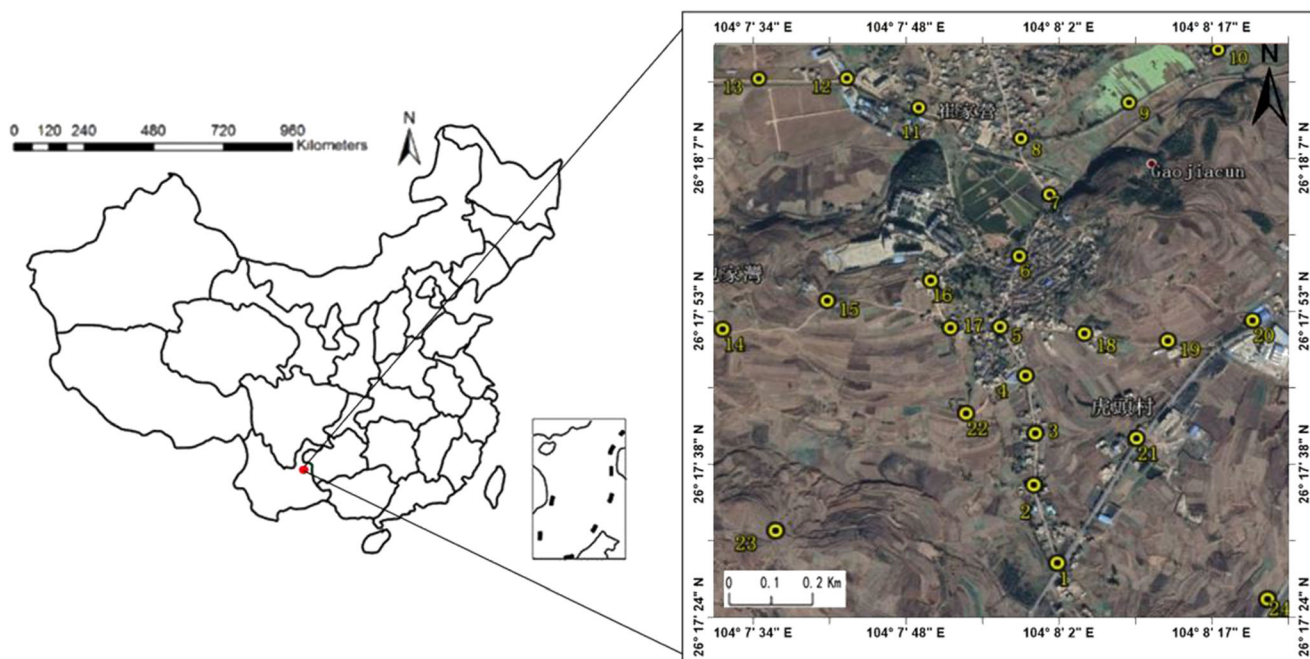


Fig. 1 Sampling point location

Sample digestion

All the chemicals were processed in a laboratory where all the equipment was ultra-clean. Meanwhile, the guaranteed reagent of chemical reagents, nitric acid and hydrofluoric acid, were used in all experimental procedures.

0.1000 g sample powder, 2.00 ml HNO_3 , and 1.00 ml HF were added in a 25.00-ml PTFE-lined stainless-steel bomb (Hu et al. 2010) (high-pressure digestion tank). The sealed vessel was put in an electric oven (WGL-230B), heated to 100 °C for 1 h, and then heated to 180 °C for 29 h. After cooling down, the vessel was opened and dried on a 130 °C hot plate, then added 1.00 ml of HNO_3 , evaporated to dryness, and repeated this procedure another time. Then, 5 ml ultra-pure water, 1 ml HNO_3 , and 1 ml Rh internal standard to digestion liquid were added. Next, the sealed vessel was screwed and heated for 4 h at 140 °C in an electric oven. Finally, the solution was composed of 10 ml liquid by adding ultra-pure water and then pipetted 1 ml solution to the next 10-ml centrifugal tube by the addition of ultra-pure water.

In this paper, the digestion of Cr(VI) from road dust was processed with the alkaline digestion method referring to the US EPA standard 3060A (Zhou et al. 2017).

A reagent blank, procedural blank solution, and Two GSS-4 standard solutions were prepared in the same way. Besides, all the chemical containers used were dipped in diluted HNO_3 (20% v/v) for 24 h, then rinsed with deionized water three times before the experiment.

Data acquisition

The concentrations of Cr, Ni, Cu, Zn, As, Cd, Pb, and Co in the sample solution obtained were analyzed by ICP-MS (Perkin Elmer SCIEX). The concentration of Cr(VI) was determined with the 1,5-diphenyl-carbohydrazide spectrophotometric method (722S) (referring to China national standard GB/T15555.4-1995).

Quality control

To obtain reliable quality data, we analyzed duplicate samples, two blank samples (a procedural blank, a reagent blank), and two standard samples (GBW07404: GSS-4). All standard samples were free of pollution. The accuracy of the repeated analysis is less than 5% RSD, and the analytical accuracy calculated with duplicate and standard samples ranged between 94 and 108%. Quality control enables all samples to have good accuracy ($\text{SD} < 5\%$). The range of analyzed concentrations was from $n \cdot 10^{-2}$ mg/kg to $n \cdot 10^3$ mg/kg by ICP-MS (Timofeev et al. 2019). The detection limit of Cr(VI) was 2 mg/kg.

Evaluation of road dust contamination

The pollution level of road toxic dust samples was quantitatively analyzed by geoaccumulation index (I_{geo}) and enrichment factor (EF).

Geoaccumulation index

Müller (1969) introduced the I_{geo} , which was described as the following short equation, and which is a very useful standard for quantifying the pollution intensity of toxic elements in the local environment:

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

where C_n is represented the concentration of the n th element, B_n is the geochemical background value of the n th element in a particular area (Yunnan soil), and 1.5 is the correction factor. The basis of the pollution division is shown in Table S1.

Enrichment factor

The enrichment factor (EF) is applied for assessing toxic elements' contamination through anthropogenic sources and deriving the origin of elements (Bourennane et al. 2010). The method standardizes the measured element content relative to the reference sample, such as iron or aluminum (in this study iron), which is calculated as follows:

$$EF = (C_n/Fe)_{sample}/(C_n/Fe)_{background} \tag{2}$$

where $(C_n/Fe)_{sample}$ is the sample toxic elements' value to the iron content ratio in this study, while $(C_n/Fe)_{background}$ is the background toxic elements' value to the iron content ratio (Malvandi 2017).

The results are shown in Table S2, EF numerical value < 1.5 deeming natural origins, EF numerical value > 1.5 deeming anthropogenic origins (Ghrefat et al. 2010).

Human health risk assessment

Human exposure assessment

The Human Exposure Model has been widely applied to evaluate the exposure risk of humankind to dust toxic elements based on equations provided by (US EPA 1989, 2004) concerning their toxicities as well as accumulative characteristics. Generally, exposure is expressed by daily dose and computed individually for each element as well as every exposure pathway (inhalation (respiratory system), ingestion (digestive system), and dermal contact (skin)). For toxic elements in road dust, inhalation is considered the main pathway.

Specifically, the doses contacted through inhalation, ingestion, and dermal absorption have been computed using Eqs. (3)–(8) (US EPA 1989, 2004; Kaur et al. 2018):

$$ADD_{ing} = C \times \frac{R_{ing} \times EF \times ED}{BW \times AT} \times 10^{-6} \tag{3}$$

$$ADD_{inh} = C \times \frac{R_{inh} \times EF \times ED}{PEF \times BW \times AT} \tag{4}$$

$$ADD_{derm} = C \times \frac{SA \times SL \times ABS \times ED \times EF}{BW \times AT} \times 10^{-6} \tag{5}$$

$$LADD_{ing} = \frac{C \times EF}{AT(\text{carcinogenic})} \times \left(\frac{R_{ing}(\text{child}) \times ED(\text{child})}{BW(\text{child})} + \frac{R_{ing}(\text{adult}) \times ED(\text{adult})}{BW(\text{adult})} \right) \times 10^{-6} \tag{6}$$

$$LADD_{inh} = \frac{C \times EF}{PEF \times AT(\text{carcinogenic})} \times \left(\frac{R_{inh}(\text{child}) \times ED(\text{child})}{BW(\text{child})} + \frac{R_{inh}(\text{adult}) \times ED(\text{adult})}{BW(\text{adult})} \right) \tag{7}$$

$$LADD_{derm} = \frac{C \times SL \times EF \times ABS}{AT(\text{carcinogenic})} \times \left(\frac{SA(\text{child}) \times ED(\text{child})}{BW(\text{child})} + \frac{SA(\text{adult}) \times ED(\text{adult})}{BW(\text{adult})} \right) \times 10^{-6} \tag{8}$$

where ADD_{ing} (mg/kg) is the everyday dose through ingestion (digestive system) of road dust; ADD_{inh} (mg/kg) is the everyday dose through inhalation (respiratory system) of road dust; ADD_{derm} (mg/kg) is the everyday dose through dermal contact (skin) with road dust; $LADD_{ing}$ is lifetime average daily dose of carcinogenic toxic elements through ingestion; $LADD_{inh}$ is lifetime average daily dose of carcinogenic toxic elements via inhalation; and $LADD_{dermal}$ is lifetime average daily dose of carcinogenic toxic elements via dermal contact. The additional parameters' values and meanings are defined in Table S3.

Health risk assessment model

Non-carcinogenic risk The HQ_{ij} (hazard quotient) was used to estimate the potential non-carcinogenic risk of each toxic element in road dust, which could be calculated with a daily dose of each toxic element divided by corresponding reference dose (RfD_{ij}) (Table S4).

$$HQ_{ij}(\text{mg kg}^{-1} \text{day}^{-1}) = \frac{ADD_{ij}(\text{mg kg}^{-1} \text{day}^{-1})}{RfD_{ij}(\text{mg kg}^{-1} \text{day}^{-1})} \tag{9}$$

The HI_i (hazard index) was used to estimate the non-carcinogenic risk of the mixed pollutions of each toxic element by summing each HQ_{ij} under the following equation:

$$HI_i = \sum_{j=1}^3 HQ_{ij} \tag{10}$$

There is no significant non-carcinogenic risk effect HQ or HI value is less than or equal to 1. If HQ or HI is greater than 1, it is more likely to have a non-carcinogenic risk.

Table 1 Statistics results of toxic elements' concentrations of Hutou Village dust in Xuanwei (mg/kg)

	Cr	Ni	Cu	Zn	As	Cd	Pb	Co	Cr(VI)
Max	952.64	181.2	180.35	349.85	10.36	3.8	54.87	52.26	18.26
Min	371.25	81.04	98.39	136.31	5.45	0.62	16.82	31.27	5.34
Median	503.97	99.03	154.23	235.55	8.45	1.72	37.39	41.84	10.94
Mean	558.44	108.4	150.97	233.69	8.06	2.02	37.9	41.17	11.37
SD	162.21	28.65	20.8	70.46	1.54	1.17	9.26	6.27	3.21
Kurtosis	0.96	1.32	0.63	-1.45	-1.09	-1.58	-0.35	-1.44	-0.34
CV (%)	29	26	14	30	19	58	24	15	28
Skewness	1.4	1.51	-0.89	0.11	-0.48	0.25	-0.24	-0.04	0.25
Yunnan background	65.2	42.5	46.3	89.7	18.4	0.2	40.6	17.5	/

Max, maximum; SD, standard deviation; CV (%), coefficient of variance

Carcinogenic risk Carcinogenic risk refers to the increased risk of cancer when people are exposed to carcinogens in their lifetime, which can be calculated with the following equations:

$$\text{Risk}_i = \text{SF}_i (\text{kg day mg}^{-1}) \times \text{LADD}_i (\text{mg kg}^{-1} \text{day}^{-1}) \quad (11)$$

where SF_i (kg day mg^{-1}) is the carcinogenic slope factor through each exposure pathway as shown in Table S4, representing the maximum probability of carcinogenic effects on humans exposed to certain toxic elements.

The total risk (TRisk) was calculated (Cocârță et al. 2016; Timofeev et al. 2019) by adding each of the individual risks with the following equation:

$$\text{TRisk} = \sum \text{Risk}_{\text{ing}} + \sum \text{Risk}_{\text{inh}} + \sum \text{Risk}_{\text{dermal}} \quad (12)$$

TRisk or Risk_i surpassing 1×10^{-4} is considered unacceptable, risks below 1×10^{-6} are considered negligible or no noticeable effect, and risks between 1×10^{-4} and 1×10^{-6} are commonly considered acceptable, depending on the exposure pattern and environment.

Statistical analysis

The range of the elements, median, coefficient of variation (CV), standard deviation (SD), Skewness, Kurtosis, mean values, and $\log_{10}(x + 1)$ function (Kynčlová et al. 2017; Ranjbaret al. 2017) were calculated by Microsoft Excel. All statistical tests were accomplished by using the SPSS (Windows version 25). Spatial distribution pattern maps of eight toxic elements were carried out by Surfer 14 and ArcGis 10.2 with Kriging interpolation; data analyses for I_{geo} and EF were calculated with the Microsoft Excel. The multivariate statistical analysis of toxic elements' concentration was done by the SPSS (Window version 25) which included PCA and CA. The biplot and HACA were

implemented using R version 3.6.3 (R Core Team (2020)). The health risk assessment was done by Excel.

Results and discussion

Potential toxic element concentration

The concentrations of these elements are listed in Table S5, and the descriptive statistics of each concentration are shown in Table 1. The numerical value of each toxic element ranged widely; the mean concentrations of toxic elements were 558.44 ± 162.21 mg/kg for Cr, 108.4 ± 28.65 mg/kg for Ni, 150.97 ± 420.8 mg/kg for Cu, 233.69 ± 70.46 mg/kg for Zn, 8.06 ± 1.54 mg/kg for As, 2.02 ± 1.17 mg/kg for Cd, 37.90 ± 9.26 mg/kg for Pb, 41.17 ± 6.27 mg/kg for Co, and 11.37 ± 3.21 mg/kg for Cr(VI), respectively.

Considering the limitation of dust information, the soil background value of Yunnan Province was used to evaluate the elements' contamination. Therefore, the concentrations of Cr, Ni, Cu, Zn, As, Cd, Pb, and Co were approximately 8.6, 2.6, 3.3, 2.6, 0.4, 10.1, 0.9, and 2.4 times of the corresponding background values of Yunnan soil (Zhang and Li 2015) except Cr(VI) for its soil background values is unknown.

The results revealed that the source of these toxic elements was clear; at the same time, the contents of these toxic elements were higher in Xuanwei City according to the book of *Seventy-Six Element Geochemical Atlas of Southwest China* (Xie et al. 2008).

To obtain the variability of the road dust properties, the variability coefficient (CV) was used and classified as four types (Phil-Eze 2010), more than 20% is considered to be low variability, 21 to 50% is considered to be mild variability, 51 to 100% is considered to be high variability, and CV above 100% is supposed to be high variable. In this study, the CV of Cd showed maximum variability. CV of Cr, Ni, Pb, Zn, and

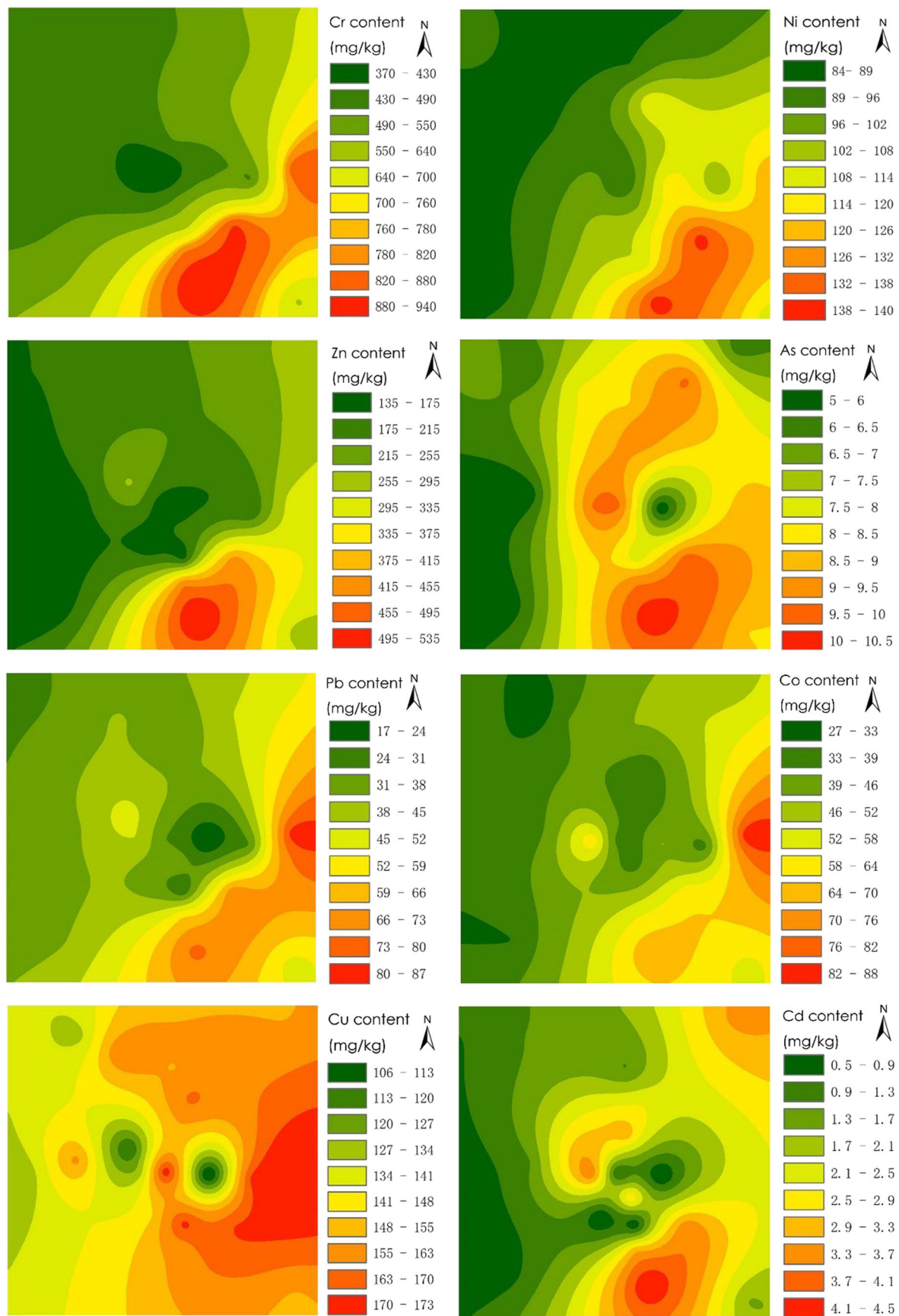


Fig. 2 Contour map of the toxic elements' content in road dust of Hutou Village in October 2017

Cr(VI) showed medium variability, and variability for Cu, As, and Co was the low (Pan et al. 2017). The high CV numerical value of Cd indicates the anthropogenic sources (Karim et al. 2014).

Kurtosis and skewness numerical values are useful tools to obtain data distribution (Zhang et al. 2008). The skewness numerical value manifested that Cu, As, Pb, and Co are negatively distribution.

Road dust comparison

The concentrations of toxic elements in Hutou Village have been compared with some other areas of the world in Table S6, using the Chinese soil background values (Chen et al. 1991) as the reference values. The average concentrations of all toxic elements surpassed the relevant Chinese soil background values except As. What's more, Cr, Ni, Cu, Zn, Cd, and Co concentrations were higher than the soil background values in Yunnan Province (Zhang and Li 2015). Carcinogenic elements (Cr, Ni, As, Cd, Co) were making a comparison to those reported of other cities. The Cr, Ni, Cd, and Co concentrations in Hutou Village were much higher than other cities, considering the high Cr background (Xie et al. 2008) and comparably immobile Cr(III) (Stefánsson et al. 2015).

Cr(VI) is a proven human respiratory system carcinogen (ATSDR 2000; Beaver et al. 2009), an ingestion carcinogen (Venter et al. 2016), and a contact allergen (NTP 2008). However, Cr(III) is a necessary trace element for the human body. Meanwhile, the higher concentration of Cr comes from not only the road dust but also the soil together with basalt rock; hence, distinguishing the forms of chromium in road dust is urgent; the comparison of Cr(VI) of some other locations all over the world is meaningful.

In this study, the concentration of Cr(VI) is listed in Table S7. We found Cr(VI) appears to be a little high compared with some other places.

Spatial distribution of road dust

The spatial distribution contour map of toxic elements in road dust is an effective method to identify hot spots, classify safe zones and unsafe zones, and evaluate potential pollution sources. The contour maps of Cr, Ni, Cu, Zn, As, Cd, Pb, and Co are presented in Fig. 2, and Cr(VI) is presented in Fig. S2. A similar distribution pattern of Zn, As, Cd, and Pb around Hutou has been found, which might relate to the anthropogenic activities such as atmospheric deposition of coal combustion, especially Cd for it is highly dispersed and it has diffused characteristic (McLaughlin and Singh 1999).

Meanwhile, the distribution patterns of Cr, Ni, and Cr(VI) also shared similar characteristics (south-eastern area), which may be related to either anthropogenic activity for the

unknown cause (we also inadvertently noted a high concentration of chromium slags (Fig. S1-1) near 1 site (Cr = 41,289 mg/kg, Ni = 239 mg/kg, Co = 25 mg/kg; data are not shown in this study)) or natural origin (tholeiite Cr = 6253 mg/kg, Ni = 1644 mg/kg, Co = 97 mg/kg; data are not shown in this study) (Oze et al. 2007) for the established cause of ELIP (Xiao et al. 2004; Xie et al. 2008). Moreover, the heavily polluted hotspots are located in the south-eastern parts and the vicinity of the Xuan-Ke road; this is a strong indication of the effect of anthropogenic activities on the road in these areas, which may increase air pollution in downwind Hutou Village. Cu and Co contour maps were different from those of the above elements and may indicate the natural source. To sum up, Cd, Cr, Ni, and Cr(VI) were the pollutants with a high concentration in the road dust in Hutou Village.

Estimation of road dust quality

Geoaccumulation index

The results of geoaccumulation index (I_{geo}) are drawn in Fig. 3 and listed in Table S8. According to the classification (Table S1), the calculated indices indicate that some elements studied (i.e., As, Pb) at all sites belonged to class No. 0 (uncontaminated). The elements such as Ni, Zn, and Co belonged to class No. 1 (uncontaminated to moderately contaminated) at all sites except for Co at sites 1#, 2#, 17#, 20#, 21#, and 24# and Zn at sites 1#, 2#, 20#, 21#, and 24# where they belonged to class No. 2 (moderately contaminated). Cu belonged to class No. 2 (moderately contaminated) except for those at sites 12#, 13#, 14#, 16#, 17#, and 18#.

Cr and Cd had higher I_{geo} values than other toxic elements, and the pollution level ranged from No. 2 class (moderately contaminated) to No. 4 class (strongly contaminated).

On the whole, the quality of road dust throughout Hutou Village was quite serious concerning Cr, Cu, and Cd, especially Cd based on the I_{geo} .

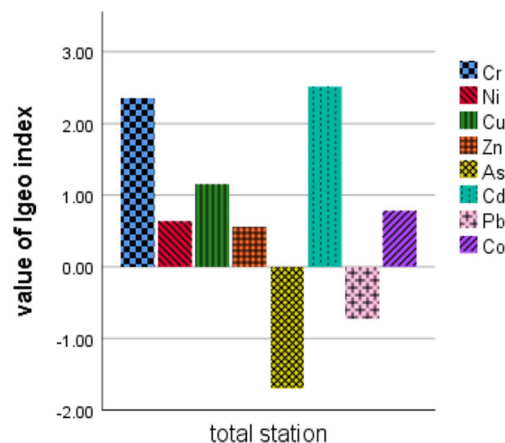


Fig. 3 The mean values of I_{geo} in road dust

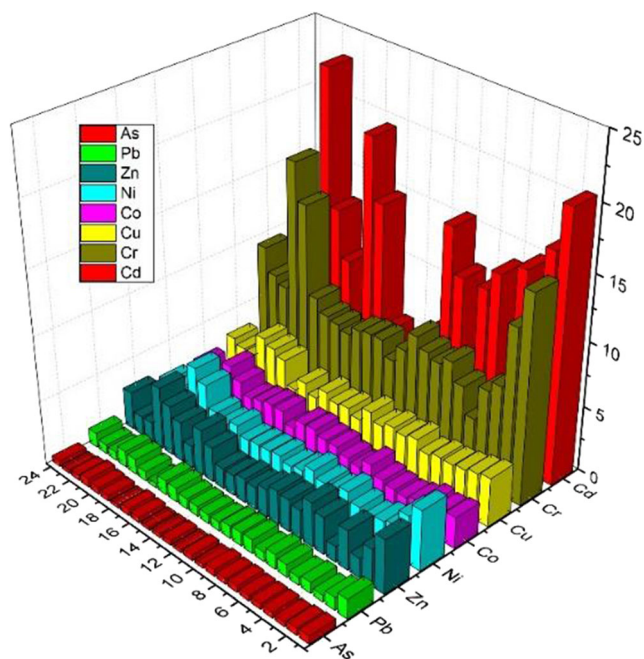


Fig. 4 Three-dimensional plots of enrichment factor (EF)

Enrichment factor

The values of the EF index are presented in Fig. 4 and in Table S9. According to the EF pollution classification, Hutou road dust toxic elements showed a wide range of enrichment at each sampling location. It is worth noting that Cd enrichment above class No. 3 (moderate severe enrichment) was higher than other elements at most sites except for sites 3#, 5#, 13#, 14#, 18#, 22#, and 23# due to the high dispersion nature of cadmium (McLaughlin and Singh 1999). Cr was the second higher degree of enrichment belonging to class No. 3 (moderate severe enrichment) at every site. The lower enrichment degree was As and Pb belongs to class No. 1 (no enrichment). Briefly, the order of the average values of the EF index was Cd > Cr > Cu > Co > Ni > Zn > Pb > As.

Although the enrichment factor (EF) index could deduce the origin of elements between anthropogenic and natural sources. The geological background, types of rock assemblages, and distribution of mineral resources should also be noticed. In this study, all elements except Pb and As represented anthropogenic origins based on enrichment factor (EF values > 1.5); nevertheless, when the geological reasons were taken into consideration, only Cd represented anthropogenic origins.

The higher EF values were Cd and Cr, which were consistent with the results of the I_{geo} . The EF values for Cd and Cr ranged from 2.54 to 23.1 and 4.65 to 17.19, respectively. A literature study (Xie et al. 2008; Chen et al. 2015b) and some other analytical tests indicate that tholeiite and chromium slags may be responsible for Cr pollution, and coal combustion (Cd = 16.13 mg/kg in Yantang coal (Fig. S1-11); data not shown in this study) for Cd pollution.

Sources apportionment of elements in road dust

Various statistical methods were used for statistics, with 95% confidence interval ($p < 0.05$) and 99% confidence interval ($p < 0.01$). Principal component analysis (PCA) and hierarchical agglomerative cluster analysis (HACA) can be used to identify possible sources (natural or anthropogenic) of toxic element pollution in road dust. The Pearson correlation analysis (CA) is the best way to describe the possible relationships between the analyzed elements.

Pearson correlation analysis

CA is widely used not only to understand the dimensions of similarity but also to estimate the interrelationships between toxic elements and to identify the sources listed in Table 2.

The correlation matrix of this study (Table 2) observed that Cd and Pb had a positive correlation ($P < 0.01, r = 0.797$), Cd and Zn had the best positive correlation ($P < 0.01, r = 0.854$), and Pb and Zn had some correlation ($P < 0.01, r = 0.688$),

Table 2 Pearson correlation analysis of toxic elements in the Hutou Village

	Cr	Ni	Cu	Zn	As	Cd	Pb	Co
Cr	1.000							
Ni	.884**	1.000						
Cu	.415*	0.389	1.000					
Zn	.465*	.524**	0.233	1.000				
As	0.271	0.310	0.396	.575**	1.000			
Cd	0.341	.405*	0.063	.854**	.564**	1.000		
Pb	0.333	0.255	0.044	.688**	.524**	.797**	1.000	
Co	-0.019	0.039	-0.173	0.079	-0.065	-0.059	-0.163	1.000

*Significant at $p < 0.05$ levels

**Significant at $p < 0.01$ levels

indicating similar origins and identical behavior. Besides, the correlation between Cr and Ni is better ($P < 0.01$, $r = 0.884$), which confirmed that Cr and Ni were homologous.

Principal component analysis

PCA could explore the sources of toxic elements and clarify potential sources. To support the above conclusion, PCA of 8 toxic elements in road dust was carried out. All factors were extracted and rotated using the varimax method based on Kaiser normalization, when the eigenvalue is greater than 1, in five iterations, the rotation converges. Three factors were obtained, included eigenvalues, percentage of the variance, and cumulative percentage presented in Table 3.

The first HCA explained 50.507% (PC1) of the total variance of toxic element concentration (Fig. 5) and 4.041 of the eigenvalue consisted of Cd, Zn, Pb, and As. All these sulphophile elements are considered, and Xuanwei has many coal mines (Downward et al. 2014) in Yunnan Province, China. Therefore, high Cd levels are more likely attributed to anthropogenic activity such as coal combustion. Although indoor air pollutions were decreased for the transformation of local stoves (Li 2016), the local custom of burning coal has not changed; the number of large trucks transporting coal has no reduction in Xuan-Ke road. At the front, the contour map around Hutou Village characteristics of Cd was obtained which would establish some relationship between lung cancer and Cd pollution.

Meanwhile, Li (2017) obtained the average cadmium concentration of chimney ash and indoor dust of three Hutou

Table 3 Varimax rotated component matrix loading for analyzed elements in sediment samples. Rotated component matrix

Element	Component		
	PC1	PC2	PC3
Cd	<i>0.941</i>	0.189	
Zn	<i>0.881</i>	0.331	
Pb	<i>0.857</i>		0.232
As	<i>0.65</i>	0.275	0.269
Cr	0.262	<i>0.927</i>	
Ni	0.31	<i>0.913</i>	
Co	-0.116	0.266	-0.833
Cu		0.51	<i>0.764</i>
Eigenvalue	4.041	1.416	1.210
% of variance	50.507	17.701	15.130
Cumulative %	50.507	68.207	83.337

Extraction method: principal component analysis

Rotation method: varimax with Kaiser normalization

The italicized values indicate loading coefficients > 0.5

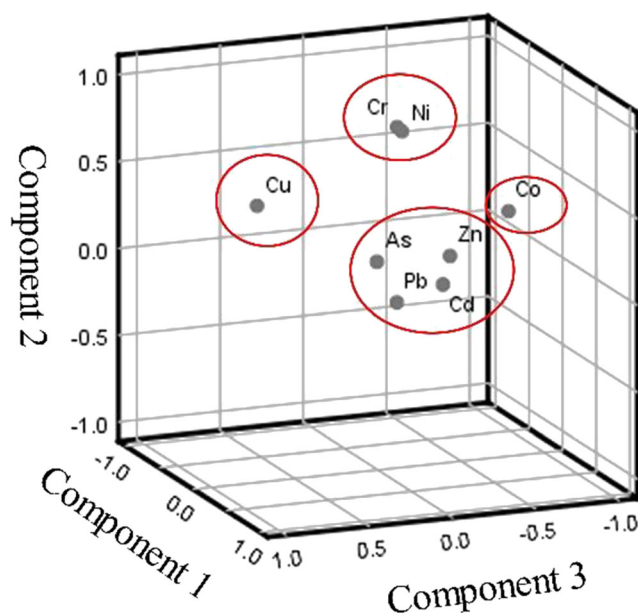


Fig. 5 Loading plots of the three components of toxic elements

farmer's houses as 203.41 mg/kg and 24.72 mg/kg respectively; hence, cadmium contamination was very serious. Friberg et al. (1971) believed that cadmium was more likely to accumulate in women and affect their health. In addition, there is a phenomenon that women have a stronger correlation with Itai-Itai disease (Vahter et al. 2002), which reflects the severity of cadmium pollution. Then, is there a correlation between the highest incidence and mortality of lung cancer (Person et al. 2013; Wang et al. 2019a, b) in women and cadmium pollution in Xuanwei area? Zn, Pb, and As may originate geological weathering of sulfide minerals partly. The elements in PC1 were mostly from the anthropogenic activity and geological weathering.

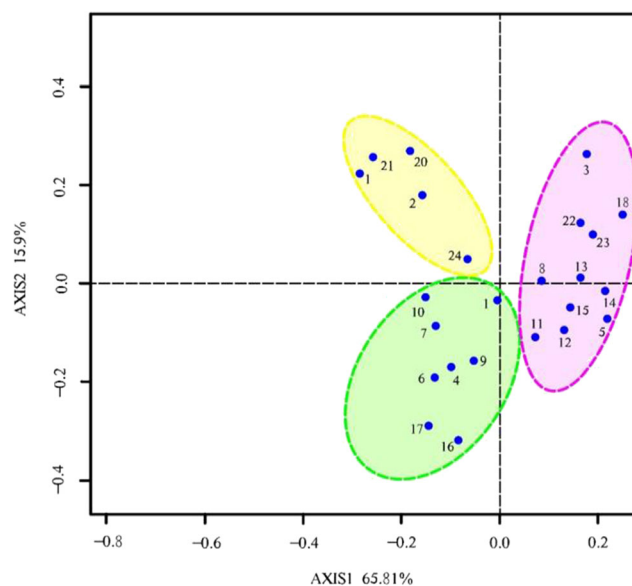


Fig. 6 Biplot showing the results of PCA (sampling site)

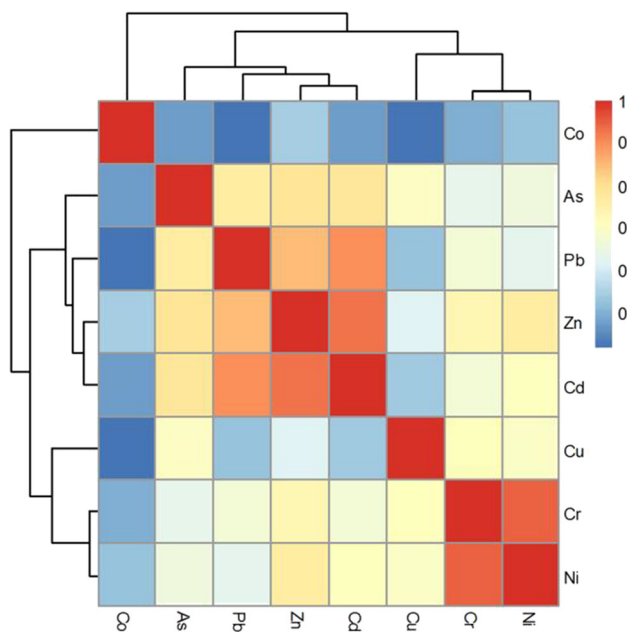


Fig. 7 Dendrogram obtained based on hierarchical agglomerative cluster analysis (HACA)

The second HCA explained 17.701% (PC2) of the total variance of toxic elements concentration (Fig. 5) and 1.416 of the eigenvalue consisted of Cr and Ni. These elements are siderophile elements, and the higher Cr and Ni levels were probably attributed to both anthropogenic activity such as coal combustion and unknown chromium slags (Fig. S1-1) and geological weathering of rocks of the ELIP (Xiao et al. 2004; Xie et al. 2008).

The third HCA explained 15.130% (PC3) of the total variance of toxic element concentration (Fig. 5) and 1.210 of the eigenvalue consisted of Cu. Cu levels were more likely attributed to geological weathering of rocks.

To establish the relationship between sampling points, the biplot (Fig. 6) based on PCA was also carried out. The first group consists of three sampling sites (1#, 2#, 20#, 21#, 24#) and may have some relationship with Xuan-Ke road; the second group consists of four sampling sites (4#, 6#, 7#, 9#, 10#, 16#, 17#) that may have some relationship with coal combustion; the third group consists of the rest of the sampling sites which may have some relationship with a natural source.

Hierarchical agglomerative cluster analysis

In this paper, the results of the HACA were consistent with CA and PCA. Based on the HACA (Fig. 7), we obtained four groups of toxic elements. The first group of toxic elements, represented by Cd, Pb, As, and Zn, maybe the same sources. The second group is represented by Cr and Ni. The third group is represented by Cu. The fourth group is the rest of the Co.

Health risk assessment

The parameter values of ADD are shown in Table S10, and HQ and HI are shown in Table 4. The trends of exposure dose of toxic elements of HQ_{ij} and HI_i were the same for children and adults. The HQ values of the three pathways decreased in the sequence of ingestion > dermal contact > inhalation. HQ_{ing} has a higher contribution rate to HI, of more than 80%. It can be seen that the ingestion of toxic elements is the main way to harm human health, followed by skin contact and inhalation.

Table 4 Exposure dose of toxic elements

	HQ						HI	
	HQ_{ing}		HQ_{inh}		HQ_{dermal}		Children	Adult
	Children	Adult	Children	Adult	Children	Adult		
Cr	4.76E-03	6.39E-04	9.66E-05	1.04E-04	3.30E-03	2.46E-03	8.15E-03	3.20E-03
Ni	6.93E-02	9.30E-03	1.02E-03	1.10E-03	1.56E-02	1.16E-02	8.59E-02	2.20E-02
Cu	4.83E-02	6.47E-03	9.14E-07	9.81E-07	4.34E-04	3.24E-04	4.87E-02	6.80E-03
Zn	9.96E-03	1.34E-03	1.89E-07	2.02E-07	8.96E-05	6.68E-05	1.00E-02	1.40E-03
As	3.44E-01	4.61E-02	4.55E-04	4.88E-04	3.09E-03	2.30E-03	3.47E-01	4.89E-02
Cd	2.59E-02	3.47E-03	1.71E-04	1.84E-04	9.32E-03	6.95E-03	3.54E-02	1.06E-02
Pb	3.46E+00	4.64E-01	2.61E-06	2.80E-06	3.12E-02	2.32E-02	3.49E+00	4.88E-01
Co	1.75E+00	2.35E-01	5.80E-03	6.22E-03	1.58E+00	1.18E+00	3.34E+00	1.42E+00
Cr(VI)	4.85E-02	6.50E-03	9.63E-05	1.03E-04	4.36E-02	3.25E-02	9.22E-02	3.91E-02

Table 5 Carcinogenic risk of toxic elements

Heavy mental	Risk			Group (IARC 2019)
	Risk _{ing}	Risk _{inh}	Risk _{dermal}	
B24Ni	1.63E-04	4.62E-08	9.17E-06	1 (IARC 2019)
As	2.00E-05	4.53E-08	4.49E-08	1 (IARC 2019)
Cd		1.42E-08		1 (IARC 2019)
Pb	5.32E-07	7.45E-10	1.20E-09	2A (IARC 2019)
Co		6.16E-07		2B (IARC 2019)
Cr(VI)	9.39E-06	2.71E-06	8.43E-07	1 (IARC 2019)

The conclusion is consistent with the previous research results. Among all the toxic elements, HQs and HI values show that children are more vulnerable than adults.

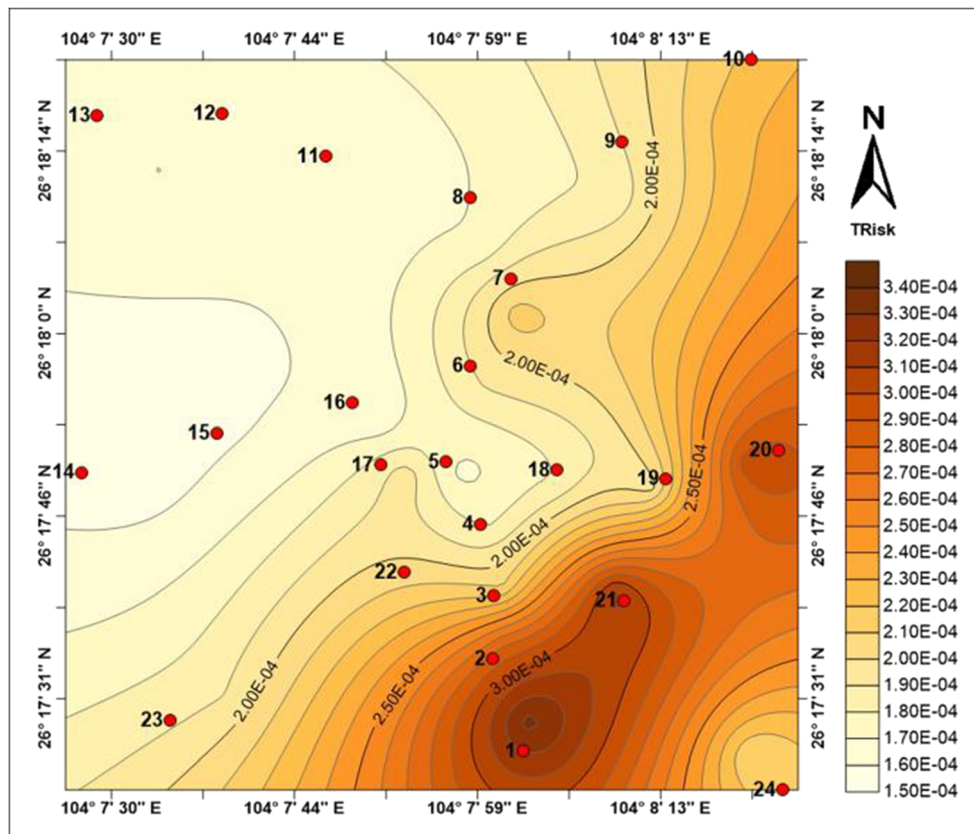
Pb and Co were at certain risk for children based on the results of HQ_{ing}, HQ_{dermal}, and HI. In general, the HI value decreased in the order of Co > Pb > As > Ni > Cr(VI) > Cu > Cd > Zn > Cr. On account, As and Pb concentrations were lower than the Yunnan Province background soil and children have been recognized to be the most vulnerable to lead exposure risk (Lidsky and Schneider 2003; CDC 2005; Paulson and Brown 2019); therefore, Co and Pb were the most concerned in non-carcinogenic risks for children. For adults, HQ_{ing}, HQ_{dermal}, and HI value of Co is greater than 1; hence,

we need to pay attention to the non-carcinogenic exposure risk of Co.

In summary, the non-carcinogenic health effects of Co concentration on children and adults in the local environment cannot be ignored because of its low reference dose (Timofeev et al. 2019).

The carcinogenic risk values of Ni, As, Cd, Pb, Co, and Cr(VI) in ingestion, inhalation and skin contact in Hutou Village are shown in Table 5. Higher risk values for ingesting nickel (Ni) than 1×10^{-4} indicate high levels of this carcinogen, which is considered unacceptable due to its low reference dose (Timofeev et al. 2019). The results showed that the order of decreasing the overall carcinogenic health risk was Ni > As > Cr(VI) > Pb; The order of the reduction of carcinogenic health risk in inhalation was Cr(VI) > Co > As > Ni > Cd > Pb; The order of carcinogenic health risk reduction by skin contact was Ni > Cr(VI) > As > Pb. However, as the concentration is lower than the Yunnan background soil, we should pay close attention to the carcinogenic risk of nickel pollution in the local environment.

The total carcinogenic risk of the whole toxic elements (Trisk) contour map is shown in Fig. 8). The relatively high Trisk value (3.34×10^{-4} at site 1#) in the south-eastern of Hutou Village together with the relatively low Trisk value (1.55×10^{-4} at site 14#) indicated that carcinogenic risk is unacceptable.

Fig. 8 Total carcinogenic risk patterns in Hutou Village

Implications from the assessment to lung cancer

Through the assessment of road dust, we found that the carcinogenic risk of Ni was considerable, the non-carcinogenic risk of Co was higher, and the I_{geo} and EF of Cd and Cr were higher. This is an important conclusion. Are Ni, Co, Cd, and Cr the same as other substances? What about the toxic elements in coal, other dust, PM₁₀. We need a wide range of comparisons to get some implications.

Through data comparison and analysis, the Ni, Co, and Cd concentrations in Yangtang coal seams (B1, B2, B3, C1) are generally high especially C1 coal seam for its higher Cd concentrations. In this case, the indoor and outdoor coal-burning processes could cause high Ni and Cd pollution in the dust, particulate matters, and water. In the previous discussion, the carcinogenic effect of Ni mainly comes from ingestion, and the carcinogenic effect

of Cd mainly comes from inhalation; hence, the Ni in the whole fraction and soluble fraction of PM₁₀ may increase the probability of respiratory diseases; the indoor concentrations of Cd in the dust, whole fraction, and soluble fraction of PM₁₀ may also increase the accumulation in lung tissue especially in women (Table 6).

Based on the above data and analysis, a possible pattern map was established which showed Cd and Ni may enter the human respiratory system through an indoor coal-burning process. High Ni and Cd product would be enriched around the chimney, the bottom of the pot, and around stove by the indoor burning of high Ni and Cd coal through fire pot and stove. The other part would be enriched in the dust and particles in the room and enter the human body through inhalation. Because of the high water solubility of Ni and Cd, a small part of them would be taken orally through the meal. Long-term exposure to such bad indoor fumes is the root of pulmonary damage (Fig. 9).

Table 6 The concentrations of toxic elements between different substances

Type	Site	Ni	Co	Cd	Cr	Ref
Coal (mg/kg)	Yangtang B3	47.08	24.84	0.43	19.85	(Li 2016)
	Yangtang	30.64	27.66	0.26	17.93	(Wang 2015)
	Yangtang B1	41	31	2.9	17	(Dai et al. 2008)
	Yangtang B2	40	28	2.5	36	(Dai et al. 2008)
	Yangtang B3	27	24	1.4	9	(Dai et al. 2008)
	Yangtang C1	18.7	22.45	0.96	13.06	(Li 2015)
	This study	49.82	31.14	16.13	24.05	Unpublished data
	Chinese coal	13.72	7.07	0.25	15.35	(Dai et al. 2003)
Dust (mg/kg)	Laibin road dust	117.52	62.92	4.91	546.35	(Zhang and Li 2015)
	Hutou chimney ash	102.61	129.88	203.41	86.26	(Li 2017)
	Hutou indoor ash	92.64	78.92	24.72	122.52	(Li 2017)
	Hutou chimney ash	87.29	73.57	102.75	54.79	(Li 2016)
	Hutou indoor ash	84.35	72.14	58.64	72.98	(Li 2016)
	Bottom of pot ash	45.84	37.10	118.63	32.48	(Li 2016)
	Stove ash	79.03	74.14	35.17	51.37	(Li 2016)
	Hutou road dust	108.4	41.17	2.02	558.44	This study
Hutou toxic elements of PM ₁₀ (ng/m ³)	Indoor PM ₁₀ (2007.2)	10.1	0.87	347.2	25.8	(Zhou 2010)
	Outdoor PM ₁₀ (2007.2)	4.4	0.59	29.3	16.8	(Zhou 2010)
	Indoor PM ₁₀ (2009.1)	9.16	0.8	489.14	22.93	(Zhou 2010)
	Indoor PM ₁₀ (2011.1)	278.1	6.9	217.9	11.25	(Fan 2013)
	Indoor PM ₁₀ (2013.11)	42.72	0.83	2.91	15.26	(Hu 2016)
	Indoor PM ₁₀ (2014.6)	34.86	0.59	7.57	3.92	(Hu 2016)
	Inhalation Unit Risk	0.26	9	18	/	(US EPA 2004)
	Hutou soluble fraction of PM ₁₀ (ng/m ³)	Indoor PM ₁₀ (2007.2)	5.1	0.23	234.4	0.43
Outdoor PM ₁₀ (2007.2)		1.5	0.15	16.6	0.21	(Zhou 2010)
Indoor PM ₁₀ (2009.1)		2.19	0.24	210.52	0.49	(Zhou 2010)
Lung tissue (mg/kg)	Cancer tissue	2.23	0.68	1.99	6.09	(Li 2016)
	Paracancerous tissue	1.55	0.83	4.14	4.30	(Li 2016)
	Normal tissue	1.04	0.56	2.64	2.79	(Li 2016)

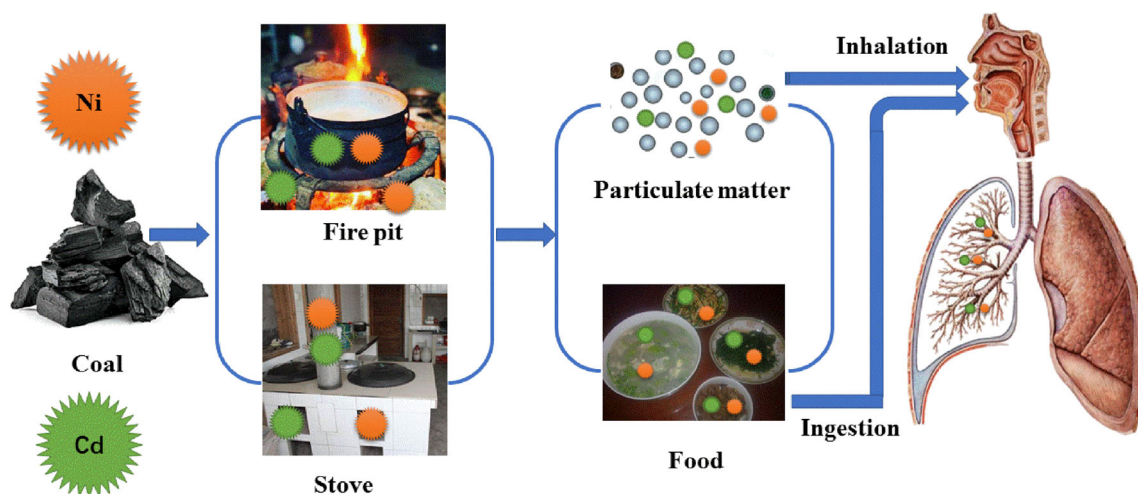


Fig. 9 The pattern of indoor Ni and Cd entering the human respiratory tract in Hutou Village

Conclusions

This study is performed to analyze the concentrations of toxic elements in road dust around Hutou Village, Xuanwei City, Yunnan Province, China. The spatial distribution, pollution characteristics, sources, and human health risks were obtained.

The mean concentrations of Cr, Ni, Cu, Zn, As, Cd, Pb, and Co were 8.6, 2.6, 3.3, 2.6, 0.4, 10.1, 0.9, and 2.4 times of the soil background values of Yunnan Province, respectively. The spatial distribution patterns of the Zn, As, Cd, and Pb were the same (around Hutou Village), which may relate to anthropogenic activity. The distribution patterns of Cr, Ni, and Cr(VI) also share similar characteristics (south-eastern direction), which may relate to either anthropogenic activity or natural origin. According to I_{geo} and EF indices, the pollution degree of Cr, Cu, Cd, Co, and Ni is serious. The multivariate statistical analysis showed that Cd, Pb, Zn, and As originated from anthropogenic sources, and Cr and Ni originated from either anthropogenic activity or natural origin while Co and Cu originated from natural sources. The high value of HI of Co in the local environment indicated that the non-carcinogenic risk of Co cannot be overlooked. The results of carcinogenic health risk clearly show that the entire ingestion risk decreased in the sequence: Ni > As > Cr(VI) > Pb, and the inhalation risk decreased in the sequence: Cr(VI) > Co > As > Ni > Cd > Pb while the dermal contact risk decreased in the sequence: Ni > Cr(VI) > As > Pb. Moreover, the TRisk of road dust decreased in the sequence: Ni > As > Cr(VI) > Co > Pb > Cd.

The implications from the indoor pattern map manifested that high pollution of Cd and the carcinogenic risk of Ni would cause lung cancer.

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Compliance with ethical standards

Conflict of interest The authors declare that they have no conflict of interest.

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