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Soil Heavy Metal(loid) Pollution Evaluation, Risk Assessment, and Source Analysis of a Mineral Processing Plant

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Abstract: Yunnan Province is rich in mineral resources. Early mining, processing, metallurgy, and other mining activities produce three industrial wastes (waste water, waste gas, and waste residue) causing environmental pollution. Considering the legacy site of a mineral processing plant in Yunnan as the research object, 21 sampling points in the study area and 12 control sampling points in the periphery were set up to determine the contents of the heavy metal(loid)s As, Hg, Cd, Cu, Ni, Pb, and Cr in the soil. The spatial distribution of heavy metal(loid)s was interpolated and analyzed using Arcmap10.8, and combined with the single-factor index, Nemero Comprehensive Pollution Index, and the health risk assessment method for the heavy metal(loid) pollution status and health risk of the soil were evaluated. The soil in the study area was acidic, with the largest average value of elemental As and the largest percentages of control and screening values. The results of the single-factor and Nemero composite pollution index showed the following trend: As > Pb > Cd > Cu > Ni > Hg. Cd, Cu, and Pb mainly originate from mining and metallurgy and Hg from the combustion of fossil fuels, while soil-forming substrates are the main sources of Ni. Pollution by As was the most prominent element, whereas pollution by Cd, Cu, and Pb in some areas also cannot be ignored to prevent negative impacts on residents. It is recommended to remediate and treat the soil on site for public events; therefore, this study fills the gap in studying potential ecological risks, human health risk assessments, and sources of exposure (oral ingestion, respiratory ingestion, dermal contact).

Keywords: pollution assessment; health risk assessment; soil heavy metal(loid)s; mineral processing plant legacy sites; Yunnan

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1. Introduction

Heavy metal(loid)s have received widespread attention owing to their cumulative, non-degradable, persistent, and toxic characteristics [1,2], As, Pb, Hg, Cr, and Cd are included in China's List of Priority Controlled Chemicals (First Batch) and China's List of Toxic and Hazardous Air Pollutants (2018) [3–5].

Heavy metal(loid) pollution is characterized by regional concentrations that accumulate during long-term mineral extraction, processing, and industrialization [6–8]. At the same time, mining activities are considered one of the most dangerous sources of environmental pollution. High concentrations of heavy metal(loid)s are manifested in river sediments around mines [9], soil, and the atmosphere, and the sources are mainly the weathering of nearby bedrock and ore-rich zones [10], leachate discharges from the processing of ores [11], mining dust emissions [12], and emissions from industrial operations and vehicles [13].

Heavy metal(loid)s pose the greatest threat among inorganic pollutants, and their enrichment poses a serious threat to human health, as well as the environment [14]. Site contamination has been described as a "chemical time bomb" [15], and contaminated sites are also known as "brownfield sites". Identifying the extent of contamination and the sources of heavy metal(loid)s at a contaminated site is a prerequisite for initiating any remediation work. Therefore, it is important to highlight the sources of contamination and identify the exposure threats to humans and their associated ecology. The (Igeo), Contamination factor (CF), Contamination Degree (CD), Enrichment Factor (EF) Pollution Load Index (PLI), and PERI are valuable tools for assessing the levels of HM contamination and ecological risk in the soil. The methods of heavy metal(loid) source resolution have also gradually increased with the continuous improvements in research, and soil pollutant traceability is mainly categorized into qualitative source identification (principal component analysis and factor analysis) and quantitative source resolution (isotope tracer method and chemical mass balance method) [16].

Various approaches have been explored for soil remediation. Among them, the treatment of tailings, as one of the most hazardous solid wastes, is getting more and more attention, and the previous generations have managed the tailings through effective methods with obvious results. The use of Portland cement increased the geotechnical stability of treated Chromite Ore Processing Residue [17], cement and bentonite [18], rice husk ashes (RHAs) and highly active pozzolanic solid waste [19], mechanical activation of tailings [20,21], iron/biochar composites [22], modified amorphous calcium phosphate prepared with phosphogypsum waste [23], and micro and vermicompost-assisted phytoextractors [24].

Since the 12th–14th Five-Year Plan, China has been committed to preventing and treating heavy metal(loid) pollution and has issued a series of policy documents related to this initiative to maintain the safety of the ecological environment and protect human health [25–28]. Although the prevention and control of heavy metal(loid) pollution in China has been effectively controlled, the historical legacy of heavy metal(loid) pollution is still increasingly prominent, posing a potential threat to ecological and environmental safety, as well as human health; therefore, heavy metal(loid) pollution treatment and remediation has a long way to go.

The Yunnan Province is known as the "Kingdom of Nonferrous Metals" because of its abundant mineral resources [29]. However, while driving regional economic development, the surrounding soil ecological environment is adversely affected by wastewater, exhaust gas, and slag produced by mining activities, such as extraction and smelting [30]. Scholars have studied soil and groundwater pollution in and around the mining areas in Yunnan Province [31–34], while a few of them have also studied heavy metal(loid) pollution caused by smelting factories [35–37], and found that there are different degrees of pollution in the region. However, information on the pollution of mineral processing plants is still very limited, and the long-term accumulation of heavy metal(loid)s exceeding the environmental carrying capacity will have adverse effects on human beings. Therefore, it is particularly important to determine the characteristics of heavy metal(loid) pollution in the soil, its ecological risk, and its source identification [38,39].

At the same time, the status of soil heavy metal(loid) contamination at the historical legacy site affected by the mineral processing plant and the health risks of the contaminated soil to the residents of the study area are unclear. The study area is relatively sensitive and has a high environmental risk. However, research on the spatial distribution of soil heavy metal(loid) contamination and the associated health risks is limited. The objectives of this study were (1) to identify the spatial distribution characteristics and possible sources of heavy metal(loid)s, (2) assess the carcinogenic and non-carcinogenic risks of heavy metal(loid)s through multiple exposure pathways in soils, and (3) provide mathematical support for the restoration of historical sites, which is of great significance for the sustainable development of legacy sites.

2. Materials and Methods

2.1. Overview of the Study Area

The study area was located in the southeastern part of the Wenshan Prefecture and belongs to the peak landscape of the karst plateau in southeastern Yunnan. The study area was the legacy site of a mineral processing plant in Yunnan (Figure 1), covering an area of approximately 24,738.4 m^2, with an overall arrangement along the northeast-southwest direction. The beneficiation plant was built in 1998 and shut down in 2013. The production process was a comprehensive heavy flotation process that mainly focused on washing the tin, zinc, and copper ores. Village 1 was located directly east of the site, with a straight-line distance of 760 m; Village 2 was located southwest of the site, with a straight-line distance of 805 m, and a small number of residents lived around the site. The site's former historical production area included a residue deposit, raw ore yard, workshop, staff dormitory, dewatering tank, sedimentation tank, and analysis laboratory. The southwest corner of the factory area was originally part of the Workshop and Residue deposit and is now occupied by the Timber mill. The strata in the area are Quaternary fill layer (Q_4^{ml}), Quaternary alluvial layer (Q_4^{al+pl}), Middle Cambrian Tianpeng Formation (Є$_2$t) [40], and the lithology is mainly moderately weathered graywacke, strongly weathered muddy siltstone, and Quaternary sediments. The land-use types around the site were mainly woodland and farmland, and the soil types were mainly red, yellow, and yellow-red. No treatment measures were taken at the site, and waste residues were randomly accumulated in the plant. The plant had not been dismantled, and the residual waste and waste slag had seriously contaminated the soil and groundwater in and around the site through rainfall washout, groundwater migration, soil exposure, and air migration.

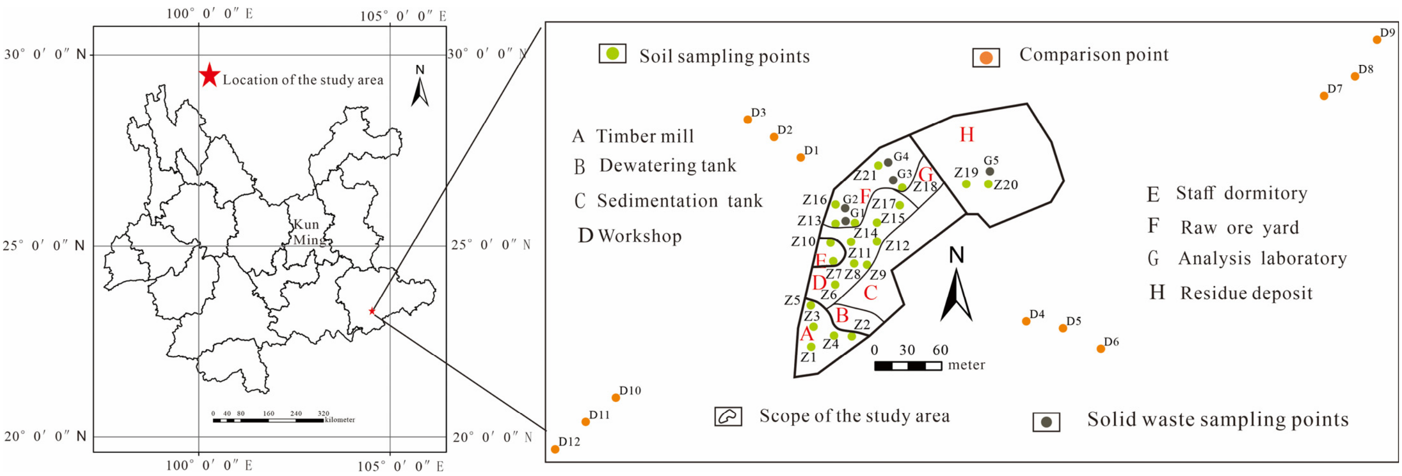

Figure 1. Map of study area.

2.2. Sample Collection and Analysis

Thirty-three soil monitoring points (12 control samples) and 199 samples (12 control samples) were collected from the study area. Heavy metal(loid) samples were analyzed and tested at the Testing Center of the Kunming Geological Survey Institute of the General Administration of Metallurgical Geology of China GAMC, which has CMA and CNAS qualification certifications.

According to China's "Technical Specification for Soil Environmental Monitoring" (HJ/T166-2004) [41], each batch of samples is measured with a quality-controlled parallel double sample, with the use of national-level standards for the accuracy control of soil sample testing and analysis and quality control. The blank standard addition recovery rate was set within 90–110%, the sample standard addition recovery rate was 70–130%, the relative deviation of parallel samples in the laboratory was controlled at 20%, the absolute deviation of pH is 10% (0.1 pH), and all analytical test results fell within the permissible range, qualified, and were reliable. Specific detection and analysis methods were used, like the following: As and Hg, atomic fluorescence spectrometry; Cr, atomic absorption spectrometry; and Cd, Cu, Ni, and Pb, inductively coupled plasma mass spectrometry.

(1) Location program

This investigation adopted the principle of strictness based on China's Technical Guidelines for Soil Environment Investigation and Assessment of Construction Land [42], combined with a detailed investigation stage of pollution identification and preliminary screening of suspected contaminated areas. The systematic distribution method combined with the professional judgment method of distribution, according to a grid of not more than 40 m × 40 m, divided the site into several monitoring units that were sampled in the center of each monitoring unit, and the number of soil sampling points was not less than one for every 400 m^2 for key areas and not less than one for every 1600 m^2 for other areas, with a total of 21 soil sampling boreholes arranged. Twenty-one soil sampling boreholes were used, five in the raw ore yard, seven in the workshop, five in the timber mill, two in the staff dormitory, and two in the Residue Deposit. Owing to the thin soil layer, the sampling depth of each sampling point was based on actual geological conditions to determine the maximum sampling depth. The sampling depth range was selected to be 1–10.5 m, and the number of samples for each monitoring point was 6–13.

(2) Soil control point layout

According to China's Technical Guidelines for Site Environmental Investigation (HJ 25.1-2014) [43], three soil control monitoring points were established at equal intervals along the four vertical axes in the external area of the site (undisturbed by humans), and 12 soil control monitoring points were established to collect samples from 0 to 20 cm. GPS was used to locate the samples in the field, and a Sampling Record Sheet was filled out to record the environmental conditions around the sampling points in detail.

(3) Solid waste samples

Following China's General Rules for Sampling Solid Chemical Products (GB/T6679-2003) [44] Technical Specification for Sampling and Sampling of Industrial Solid Wastes (HJ/T20-1998) [45], using the seriously polluted building (structure) area for zoning sampling, workshop, analysis laboratory, staff dormitory, sedimentation tank, raw ore yard, and timber mill (random sampling of the top 2 cm of buildings), one sample was taken from each places, and a total of six samples were collected.

The waste residues stockpiled at the site were mainly in the raw ore yard, and waste residue deposits were not being protected. The surrounding environment contained varying degrees of potential pollution. Five samples were collected, four from the raw ore yard and one from the residual deposit.

2.3. Human Health Risk Assessment Methods

According to the health risk assessment model published by the USEPA, three routes of oral intake, dermal contact, and respiratory intake of soil particulate matter were selected [46] to evaluate the health risks to adults and children, and specific reference values were derived from the standard values of the China's Technical Guidelines for the Risk Assessment of Construction Land Use (HJ25.3-2019) [47] and related domestic and international studies [48–50]. The average daily exposure of the human body to soil heavy metal(loid)s was calculated as follows:

$$ADD_{iing} = C_i \times (R_{ing} \times EF \times ED/BW \times AT) \times 10^{-6} \quad (1)$$

$$ADD_{iinh} = C_i \times (R_{inh} \times EF \times ED/PEF \times BW \times AT) \quad (2)$$

$$ADD_{iderm} = C_i \times (SA \times SL \times ABF \times EF \times ED/BW \times AT) \times 10^{-6} \quad (3)$$

where C_i is the concentration of heavy metal(loid)s in soil (mg·kg^{-1}), ADD_{iing} is oral ingestion, ADD_{iinh} is respiratory inhalation, ADD_{iderm} is dermal contact, R_{ing} is daily soil ingestion (mg·d^{-1}), R_{inh} is daily soil inhalation (m^3·d^{-1}), EF is exposure frequency (d·a^{-1}), ED is exposure duration (a), BW is mean body weight (kg), AT is mean exposure time (d), SA is skin exposure area (cm^2), SL is skin adhesion factor (mg· (cm^2·d)$^{-1}$), ABF is skin adsorption factor (unitless), and PEF is particulate emission factor (m^3·kg^{-1}). Specific reference values are shown in Table 1 below.

Table 1. Values of relevant parameters in human health risk assessment.

Parameters	Reference Value	
	Child	Adult
R_{ing} (mg·d^{-1})	200	100
EF(d·a^{-1})	350	350
ED (a)	6	24
BW (kg)	15.9	56.8
AT (d)	25,550 (carcinogenic) 2190 (non-carcinogenic)	25,550 (carcinogenic) 8760 (non-carcinogenic)
Rinh (m^3·d^{-1})	7.5	14.5
PEF (m^3·kg^{-1})	1.36×10^9	1.36×10^9
SL (mg·(cm^2·d) $^{-1}$)	0.2	0.07
SA (cm^2)	2800	5700
ABF (zero dimension)	0.01 (carcinogenic) 0.001 (non-carcinogenic)	0.01 (carcinogenic) 0.001 (non-carcinogenic)

The hazard quotient (HQ) and cancer risk (CR) represent the non-carcinogenic and carcinogenic risks associated with exposure to individual heavy metal(loid)s, respectively, where HQ < 1 indicates no non-carcinogenic health risk, and vice versa [51]. Referring to the U.S. Superfund Risk Evaluation Guidelines, a total carcinogenic risk (TCR) < 10^{-6} does not pose a carcinogenic risk to humans, and when the TCR > 10^{-4}, it is considered to pose an unacceptable carcinogenic risk, and the range of acceptable carcinogenic risk is $10^{-6} \leq \text{TCR} \leq 10^{-4}$ [52].

$$HI = \sum HQ_i = \sum\left(\frac{ADD_{iing}}{RfD_{iing}} + \frac{ADD_{iinh}}{RfD_{iinh}} + \frac{ADD_{iderm}}{RfD_{iderm}}\right) \quad (4)$$

$$CR_n = ADD_{iing} \times SF_{iing} + ADD_{iinh} \times SF_{iinh} + ADD_{iderm} \times SF_{iderm} \quad (5)$$

where: SF_{ij} and RfD_{ij} are the carcinogenicity slope factor and the reference dose of heavy metal(loid) element i corresponding to the j pathway, as shown in Table 2 [53].

Table 2. Reference dose and carcinogenicity slope factor for heavy metal(loid)s in different exposure pathways.

Heavy Metal(loid)	RfD/(mg·(kg·d)$^{-1}$)			SF/(kg·d)·mg^{-1})		
	Oral Intake	Inhalation	Skin Exposure	Oral Intake	Inhalation	Skin Exposure
Cu	4.00×10^{-2}	4.02×10^{-2}	1.20×10^{-2}	-	-	-
Zn	3.00×10^{-1}	3.00×10^{-1}	6.00×10^{-2}	-	-	-
Cr	3.00×10^{-3}	2.86×10^{-5}	6.00×10^{-5}	0.5	42.00	20.00
Ni	2.00×10^{-2}	2.06×10^{-2}	5.40×10^{-3}	1.70	0.84	42.50
Pb	3.50×10^{-3}	3.52×10^{-3}	5.25×10^{-4}	8.5×10^{-3}	4.2×10^{-2}	1.7×10^{-2}
Cd	1.00×10^{-3}	1.00×10^{-5}	1.00×10^{-5}	6.10	6.30	6.10
As	3.00×10^{-4}	1.23×10^{-4}	1.23×10^{-4}	1.50	15.10	3.66
Hg	3.00×10^{-4}	8.57×10^{-5}	2.10×10^{-5}	0.0003	0.0003	3×10^{-7}

2.4. Ecological Risk Evaluation Methods and Standards

In this soil pollution evaluation, the screening value in China's Soil Environmental Quality Soil Pollution Risk Control Standard for Construction Land (GB 36600-2018) [54] was selected as the evaluation standard, which was combined with the single-factor index and Nemero Comprehensive Pollution Index to evaluate the soil heavy metal(loid) contamination status and the comprehensive degree of contamination in the study area [45].

(1) Single-factor indices

The calculation formula is as follows:

$$P_i = C_i / S_i \quad (6)$$

where C_i is the measured value of heavy metal(loid) content in soil, S_i is the limited standard of heavy metal(loid) pollution in soil(screening value), and the standard relationship of heavy metal(loid)s pollution level division is as follows: $P \leq 1$, no pollution; $1 < P \leq 2$, light pollution; $2 < P \leq 3$, moderate pollution; $P > 3$, heavy pollution.

(2) Nemero Composite Pollution Index Method

This refers to the combined effect of a variety of pollutants and more serious pollutants, focusing on the pollutants that cause serious environmental pollution and is calculated using the following formula [45]:

$$P_N = \sqrt{\frac{P_{iave}^2 + P_{imax}^2}{2}} \quad (7)$$

P_{iave} is the average value of the single-factor pollution index, P_{imax} is the maximum value of all single-factor pollution indices, and the heavy metal(loid) pollution level is divided according to the following ranges: $P_N \leq 0.7$, no pollution; $0.7 < P_N \leq 1$, not yet polluted (cordon sanitaire); $1 < P_N \leq 2$, mildly polluted; $2 < P_N \leq 3$, moderately polluted; $P_N > 3$, heavily polluted.

3. Results and Discussion

3.1. Characteristics of Soil Heavy Metal(loid) Content

The heavy metal(loid) content in the study area is shown in Figure 2 and Table 3 (the data were logarithmically (ln) processed to maintain the aesthetics of the graph, considering the large difference in the data while making the graph. The distribution range and characteristics of the soil content of the six heavy metal(loid)s in the legacy site of the processing plant were compared and analyzed with respect to the screening value of the risk of soil contamination of China's construction land and the control value (GB 36600-2018). As far as the average value is concerned, the average value of the As element was the largest, and the main exceeding heavy metal(loid)s were As, Cd, and Pb. The distribution of Hg, Cu, Ni, and Pb content was relatively concentrated, while the distribution of As and Cd was relatively scattered. An anomaly appeared in the box plot corresponding to the content of Pb, which was the object of focus in the later stage, and the biotoxicity of As and Pb was higher; certain control measures need to be taken to reduce the harm of As and Pb to the environment and human health based on the study's results.

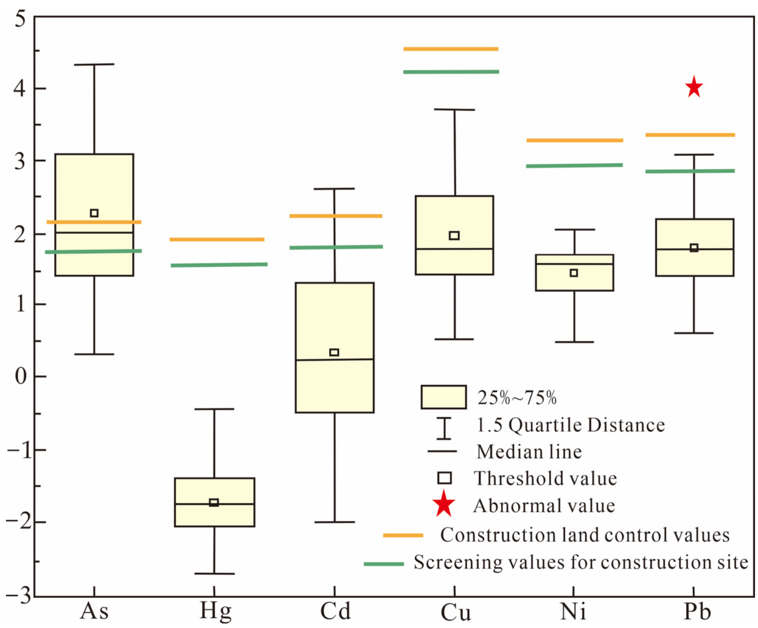

Figure 2. Box plots of heavy metal(loid) contents in the soil of construction land in the study area.

Table 3. Concentration distribution of heavy metal(loid)s in the study area(mg·kg^{-1}).

	Parameterization	pH	As	Hg	Cd	Cu	Ni	Pb
Sample point of a study area	Min	2.700	2.090	0.002	0.010	3.500	3.230	4.240
	Max	8.900	21,764.000	0.364	414.000	5101.000	113.000	15,007.000
	Ave	6.545	2210.248	0.041	21.264	318.038	34.267	286.881
	Median	7.29	105.000	0.019	2.090	64.800	37.700	63.400
Comparison point	Min	5.490	1.280	0.006	0.010	10.700	43.200	5.910
	Max	7.880	52.200	0.088	0.880	192.000	63.200	45.800
	Ave	6.833	23.282	0.037	0.177	52.983	51.675	28.131
	Median	6.95	19.15	0.028	0.12	39.55	49.9	27.8
	SD		4931.624	0.063	48.047	558.125	19.434	1389.251
	CV		2.373	1.521	2.403	1.848	0.550	5.121
	Background value for Yunnan Province		18.400	0.058	0.218	46.300	42.500	40.600
	K1		120.122	0.707	97.541	6.869	0.806	7.066
	Soil background value for China		9.600	0.038	0.079	20.700	24.900	23.500
	K2		230.234	1.079	269.165	15.364	1.376	12.208
	Screening value (% exceedance)		13.37	0	9.63	0	0	2.14
	Control value (% exceedance)		47.59	0	1.07	0	0	1.07

Note: K1 and K2 represent the ratio of the average value of the study area to the background value of Yunnan Province and the ratio of the average value to the background value of the whole country, respectively; the exceedance rate is calculated by comparing the control value of the screening value of the risk of soil contamination of construction land and is expressed as a percentage.

The characteristics of the heavy metal(loid) content of the soil in the study area have been shown in Table 3 (Cr was below the detection limit (0.01); therefore it is not listed in the table). Sampling and analysis of the surrounding undisturbed soil showed that the pH of the study area was 6.6, which was lower than that of the control point, indicating that the soil in the study area was acidic because of the long-term impact of mineral-processing operations. Except for Hg and Ni, the contents of the rest of the heavy metal(loid) elements in the study area were much higher than the contents of the control points, indicating that they were greatly affected by anthropogenic activities. The mean values of the soil heavy metal(loid) As, Hg, Cd, Cu, Ni, and Pb contents were 120.122, 0.707, 97.541, 6.869, 0.806, and 7.066 times higher than the background values in Yunnan Province [55] and 230.234, 1.079, 269.164, 15.364, 1.376, and 12.208 times higher than the background values of the soil in the whole country [56]; in general, As and Cd showed significant enrichment, as they had the highest exceedance rate of screening and control values. The spatial coefficients of variation of the six heavy metal(loid) elements were in the following order from the largest to the smallest: Pb > Cd > As> Cu > Hg > Ni. The magnitude of the coefficients of variation were related to the source of soil heavy metal(loid)s. The smaller the coefficient of variation, the more the dominant natural sources and the larger the coefficient of variation, the greater the anthropogenic influence [57].

3.2. Characterization of Heavy Metal(loid) Content in Solid Waste

3.2.1. Characterization of Building (Structure) Content

The main contaminant in the buildings (structures) at the site was As, with the highest concentration of 683 mg/kg (sedimentation tank G-1) (Table 4). Areas, such as analysis laboratories, workshops, sedimentation tanks, and dewatering tanks, have relatively high As contents because of long-term direct contact during the production process.

Acid leaching toxicity analysis of the buildings (structures) (Table 4) and building (structures) samples did not exceed the concentration limits listed in China's Hazardous Waste Identification Standards Leaching Toxicity Identification (GB 5085.3-2007) [58]. It was determined that the buildings (structures) within the site did not contain hazardous wastes. The on-site samples from the buildings (structures) met the standards for Class I general industrial solid waste.

Table 4. Solid waste content characteristics.

Item			As	Hg	Cr	Cd	Cu	Ni	Pb
Buildings	$mg\cdot kg^{-1}$	Staff dormitory	78.8	0.007	<2.00	1.08	34.7	30.6	38.9
		Timber mill	89.2	0.006	<2.00	0.37	43.6	54.7	50.7
		Analysis laboratory	634	0.021	<2.00	2.73	85.7	33.0	42.2
		Workshop	344	0.013	<2.00	3.72	140	40.2	103
		Residue deposit	47.5	0.010	<2.00	0.81	17.2	2.73	15.9
		Sedimentation tank	683	0.017	<2.00	12.2	188	12.4	50.9
	Acid leaching toxicity ($mg\cdot L^{-1}$)	Standard limit value	5	0.1	5	1	100	5	5
		Staff dormitory	0.0925	<0.00002	<0.004	<0.0012	0.0125	<0.0038	<0.0042
		Timber mill	1.339	<0.00002	<0.004	<0.0012	<0.0025	<0.0038	<0.0042
		Analysis laboratory	0.0947	<0.00002	0.005	<0.0012	<0.0025	<0.0038	<0.0042
		Workshop	0.0205	<0.00002	0.005	<0.0012	0.0089	<0.0038	<0.0042
		Residue deposit	0.0019	<0.00002	0.006	<0.0012	<0.0025	<0.0038	<0.0042
Slag	$mg\cdot kg^{-1}$	G1	9685	0.016	<2	16.9	426	6.84	326
		G2	26,803	0.015	<2	72.9	1750	17	357
		G3	17,995	0.012	<2	36.1	928	12	294
		G4	23,010	0.011	<2	71	1518	17.7	325
		G5	8809	0.006	<2	1.68	117	1.37	230
	Acid leaching toxicity ($\mu g\cdot L^{-1}$)	Standard limit value	5000	100	5000	1000	100,000	5000	5000
		G1	35,584	<0.02	<0.004	832	20,053	259	10.6
		G2	21,946	<0.02	<0.004	3494	57,736	732	7.5
		G3	16,425	<0.02	<0.004	1826	36,853	511	17.6
		G4	9450	<0.02	<0.004	2608	26,779	546	32.1
		G5	6172	<0.02	<0.004	129	2481	21.8	<4.2

3.2.2. Characteristics of Waste Slag Content

The heavy metal(loid) elements As, Cu, and Pb were higher in the waste slag samples, and those in the original ore dumps (G1, G2, G3, and G4) were higher than those in the waste slag dumps (G5).

When acid leaching toxicity tests were performed on waste slag, the heavy metal(loid) contents of the samples from both the raw ore dump and waste slag dump were higher than the standard limit value of China's Hazardous Waste Identification Standards for Leaching Toxicity Identification (GB 5085.3-2007). The main factors that exceeded the raw ore dump were As (max. 7.12 times) and Cd (max. 3.49 times), and the exceeding factor for the waste slag dump was As (1.23 times).

3.3. Spatial Distribution Characteristics of Soil Heavy Metal(loid)s

Using ArcMAP10.8 statistical analysis, the inverse distance-weighted interpolation analysis of the six elements was used to interpolate the spatial distribution of the six soil heavy metal(loid) pollution characteristics (Figure 3); the six heavy metal(loid) distribution patterns were obvious, and the distribution was more concentrated. As, Cd, and Cu had similar distribution patterns, with the high-value points located in the southeast and northeast (timber mill, workshop, and residue deposit), and the high value of Pb was located in the workshop, Hg was mainly located in the southwest (timber mill), and Ni was mainly located in the workshop. The index of the red points in the southeast was significantly higher than that of the other points, indicating that beneficiation activities may influence the content. There is a risk of leakage of raw materials, waste residue during the workshop's production process, and residue deposits. The timber mill was part of the workshop, and the residue was deposited in the early days. In contrast, the heavy metal(loid)s in the waste residue and wastewater directly entered the soil environment through soil infiltration and rainwater washing, resulting in a high heavy metal(loid) content [59]. This is consistent with previous findings, mainly related to ore processing, such as leachate discharge and tailings accumulation [60–62].

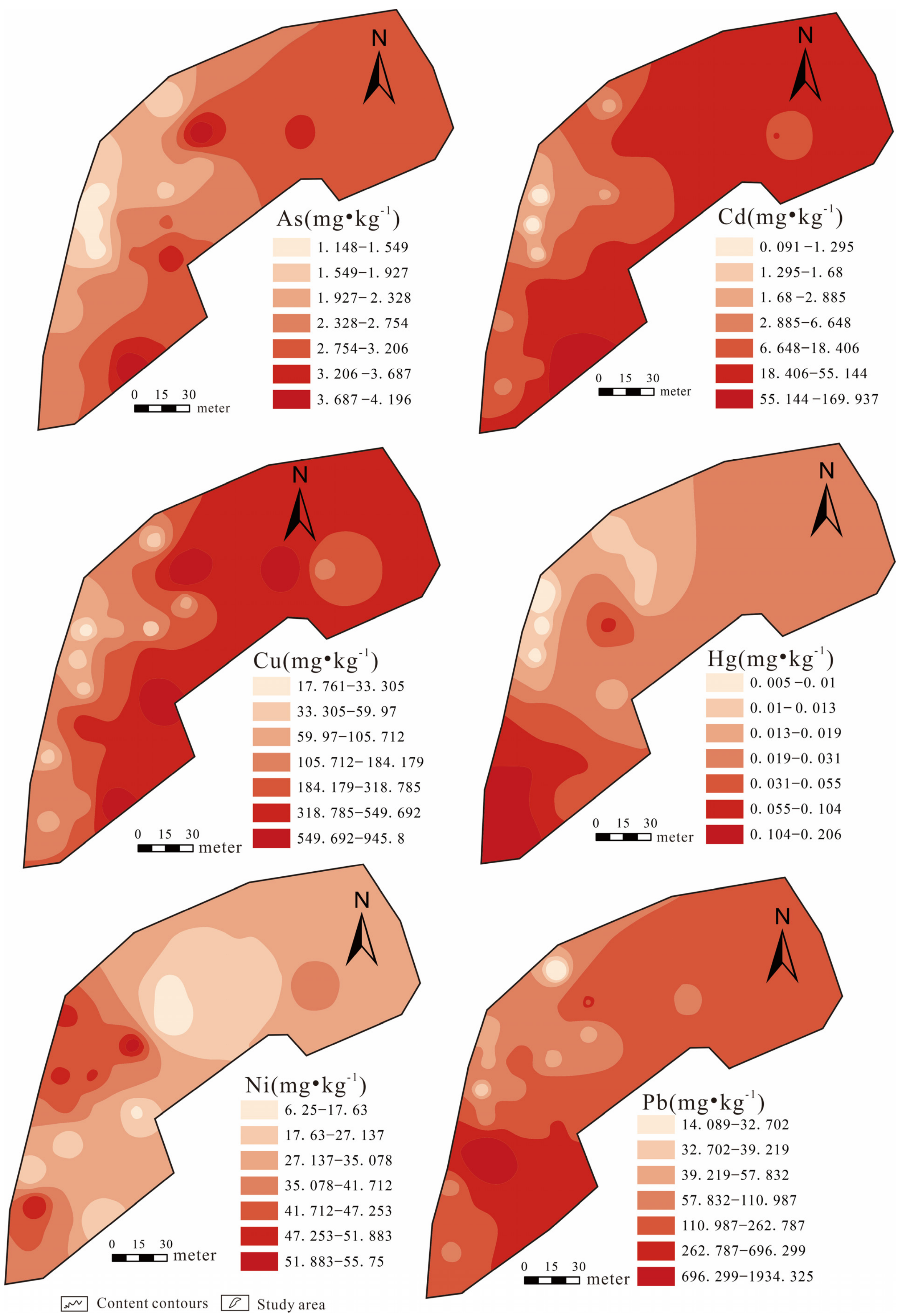

Figure 3. Spatial distribution of soil heavy metal(loid)s in the study area.

3.4. Soil Pollution Risk Evaluation

(1) Soil heavy metal(loid) pollution soil heavy metal(loid) single factor evaluation results and Nemero comprehensive pollution index results are shown in Table 5, which shows that the pollution degree of the As element sampling points in the study area from non-pollution to heavy pollution were distributed, of which non-pollution (39.04%) and heavy pollution (44.39%) accounted for the ratio of equal strength and, at the same time, indicates that the As element should be the object of key concern. Cd and Pb were also distributed in the light and heavy pollution ranges, indicating that Cd and Pb cannot be ignored. Mercury, copper, and nickel were not found to be polluting. Cd and Pb were also distributed in mild-to-severe pollution, indicating that Cd and Pb should not be neglected, whereas Hg, Cu, and Ni were not found to be polluting. The Nemero Composite Pollution Index also showed that the order of heavy metal(loid) pollution in the region was As > Pb > Cd > Cu > Ni > Hg, which was consistent with the single-factor index.

Table 5. Comprehensive pollution index evaluation of heavy metal(loid)s in soil.

Item	Parameterization	As	Hg	Cd	Cu	Ni	Pb
Single-factor index (percentage of each sample, %)	Uncontaminated	39.037	100	89.305	100	100	96.791
	Light pollution	12.299	0	9.091	0	0	2.139
	medium pollution	4.278	0	0.535	0	0	0
	High pollution	44.385	0	1.070	0	0	1.070
Single-factor exponential distribution values	Max	362.733	0.010	6.369	0.283	0.126	18.759
	Min	0.035	5.3×10^{-5}	0.0002	0.0002	0.004	0.005
	Avg	36.837	0.001	0.327	0.018	0.038	0.359
P_N		257.810	0.007	4.510	0.201	0.090	13.327
Pollution degree		High pollution	Uncontaminated	High pollution	Uncontaminated	Uncontaminated	High pollution

(2) Soil heavy metal(loid) health risk.①Through exposure assessment analysis (Table 6), non-carcinogenic and carcinogenic three pathways average daily intake of different heavy metal(loid)s had the order of As > Cu > Pb > Ni > Cd > Hg, in addition to the element Hg; the daily intake and total daily intake of non-carcinogenic oral intake, dermal contact, carcinogenicity of a single pathway of all the heavy metal(loid) elements are manifested in the children higher than in the adults. Non-carcinogenic adult respiratory intake of all the heavy metal(loid)s was higher than that of children. The average daily intake of non-carcinogenic and carcinogenic adults and children by different routes was ranked as $ADD_{inh} < ADD_{derm} < ADD_{ing}$, and the oral intake was much higher than the respiratory intake.

Health risk assessment: The health risk assessment combines human health and environmental pollution and the odds of harmful factors adversely affecting human health to assess the risk of changes in human health from exposure factors [63], health risk assessments, and a quantitative study of the magnitude of the risk of pollutants in the human body to produce effects.

Health risk evaluation Indices (Table 7). The order of carcinogenic and non-carcinogenic risks by different routes were $CR_{ing} > CR_{derm} > CR_{inh}$ and $HQ_{ing} > HQ_{derm} > HQ_{inh}$ for adults and children, respectively, suggesting that oral intake was the main route. The order of carcinogenic risk in adults and children was As > Cd > Ni > Pb > Hg. The order of non-carcinogenic risk was As > Pb > Cd > Cu > Ni > Hg. In terms of the mean value, the oral route of As posed a carcinogenic risk, while the oral route of Cd and Ni and the dermal route of As and Ni were in the acceptable range, and the dermal route of Cd posed no carcinogenic risk; the oral route of As, Cd, and Ni posed no carcinogenic risk, the dermal route of Cd posed no carcinogenic risk, and the dermal route of Cd posed no carcinogenic risk. As, Cd, and Ni respiratory inhalation did not pose a carcinogenic risk to the human body; in addition, we can also find in the table that the As and Cd elemental adults and children oral intake pathway was part of the sampling point of the carcinogenic risk; for Hg and Pb, all exposure pathways were less than 10^{-6}, and they do not pose a carcinogenic risk to the human body, and the oral intake pathway and dermal exposure pathway of the maximum value are located in the range of 10^{-6}–10^{-4}, belonging to the carcinogenicity risk of the human body. The maximum values for the oral and dermal routes of exposure were 10^{-6}–10^{-4}, which were within the acceptable ranges.

Table 6. Average daily exposure to non-carcinogenic and carcinogenic heavy metal(loid)s in soil.

Heavy Metal(loid)		ADD_{ing}		ADD_{inh}		ADD_{derm}		ADD	
		Adult	**Child**	**Adult**	**Child**	**Adult**	**Child**	**Adult**	**Child**
Non-carcinogenic	As	0.0037	0.0267	3.98×10^{-7}	7.35×10^{-7}	1.49×10^{-5}	7.46×10^{-5}	3.75×10^{-3}	2.67×10^{-2}
	Hg	6.99×10^{-8}	4.99×10^{-7}	7.45×10^{-12}	1.38×10^{-11}	2.79×10^{-10}	1.40×10^{-9}	7.02×10^{-8}	5.01×10^{-7}
	Cd	3.59×10^{-5}	0.0003	3.83×10^{-9}	7.07×10^{-9}	1.43×10^{-7}	7.18×10^{-7}	3.60×10^{-5}	2.57×10^{-4}
	Cu	0.0005	0.0038	5.72×10^{-8}	1.06×10^{-7}	2.14×10^{-6}	1.07×10^{-5}	5.39×10^{-4}	3.85×10^{-3}
	Ni	5.96×10^{-5}	0.0004	6.17×10^{-9}	1.14×10^{-8}	2.31×10^{-7}	1.16×10^{-6}	5.99×10^{-5}	4.14×10^{-4}
	Pb	0.0005	0.0035	5.16×10^{-8}	9.54×10^{-8}	1.93×10^{-6}	9.69×10^{-6}	4.86×10^{-4}	3.47×10^{-3}
	ADD	0.0048	0.0346	5.17×10^{-7}	9.55×10^{-7}	1.93×10^{-5}	9.70×10^{-5}	4.87×10^{-3}	3.47×10^{-2}
Carcinogenic	As	0.0013	0.0023	1.36×10^{-7}	6.30×10^{-8}	5.10×10^{-5}	6.40×10^{-5}	1.33×10^{-3}	2.35×10^{-3}
	Hg	2.40×10^{-8}	4.28×10^{-8}	2.55×10^{-12}	1.18×10^{-12}	9.56×10^{-10}	1.20×10^{-9}	2.49×10^{-8}	4.40×10^{-8}
	Cd	1.23×10^{-5}	2.20×10^{-5}	1.31×10^{-9}	6.06×10^{-10}	4.91×10^{-7}	6.16×10^{-7}	1.28×10^{-5}	2.26×10^{-5}
	Cu	0.0002	0.0003	1.96×10^{-8}	9.07×10^{-9}	7.34×10^{-6}	9.21×10^{-6}	1.91×10^{-4}	3.38×10^{-4}
	Ni	1.98×10^{-5}	3.54×10^{-5}	2.11×10^{-9}	9.77×10^{-10}	7.91×10^{-7}	9.92×10^{-7}	2.06×10^{-5}	3.64×10^{-5}
	Pb	0.0002	0.0003	1.77×10^{-8}	8.18×10^{-9}	6.63×10^{-6}	8.30×10^{-6}	1.73×10^{-4}	3.05×10^{-4}
	ADD	0.0017	0.0030	1.77×10^{-7}	8.18×10^{-8}	6.63×10^{-5}	8.31×10^{-5}	1.73×10^{-3}	3.05×10^{-3}

Table 7. Carcinogenic health risk index of heavy metal(loid)s in the soil.

Heavy Metal(loid)			CR_{ing}		CR_{inh}		CR_{derm}		TCR	
			Adult	**Child**	**Adult**	**Child**	**Adult**	**Child**	**Adult**	**Child**
Carcinogenic	As	Min	1.81×10^{-6}	3.24×10^{-6}	1.95×10^{-9}	9.00×10^{-10}	1.77×10^{-7}	2.21×10^{-7}	1.99×10^{-6}	3.46×10^{-6}
		Max	0.0189	0.0338	2.03×10^{-5}	9.37×10^{-6}	0.0018	0.0023	0.0208	0.0361
		Avg	0.0019	0.0034	2.06×10^{-6}	9.51×10^{-7}	0.0002	0.0002	0.0021	0.0037
	Hg	Min	3.47×10^{-13}	6.20×10^{-13}	3.70×10^{-17}	1.71×10^{-17}	1.39×10^{-17}	1.74×10^{-17}	3.47×10^{-13}	6.20×10^{-13}
		Max	6.32×10^{-11}	1.13×10^{-10}	6.74×10^{-15}	3.11×10^{-15}	2.52×10^{-15}	3.16×10^{-15}	6.32×10^{-11}	1.13×10^{-10}
		Avg	7.19×10^{-12}	1.28×10^{-11}	7.66×10^{-16}	3.54×10^{-16}	2.87×10^{-16}	3.59×10^{-16}	7.19×10^{-12}	1.28×10^{-11}
	Cd	Min	3.53×10^{-8}	6.31×10^{-8}	3.89×10^{-12}	1.80×10^{-12}	1.41×10^{-9}	1.77×10^{-9}	3.67×10^{-8}	6.48×10^{-8}
		Max	0.0015	0.0026	1.61×10^{-7}	7.44×10^{-8}	5.83×10^{-5}	7.31×10^{-5}	0.0015	0.0027
		Avg	7.51×10^{-5}	0.0001	8.27×10^{-9}	3.82×10^{-9}	3.00×10^{-6}	3.75×10^{-6}	7.80×10^{-5}	0.0001
	Cu	Min	-	-	-	-	-	-		
		Max	-	-	-	-	-	-		
		Avg	-	-	-	-	-	-		
	Ni	Min	3.18×10^{-6}	5.68×10^{-6}	1.67×10^{-10}	7.73×10^{-11}	3.17×10^{-6}	3.97×10^{-6}	3.97×10^{-6}	9.65×10^{-6}
		Max	0.0001	0.0002	5.86×10^{-9}	2.71×10^{-9}	0.0001	0.0001	0.0001	0.0003
		Avg	3.37×10^{-5}	6.02×10^{-5}	1.78×10^{-9}	8.21×10^{-10}	3.36×10^{-5}	4.35×10^{-5}	4.21×10^{-5}	0.0001
	Pb	Min	2.09×10^{-8}	3.73×10^{-8}	1.10×10^{-11}	5.08×10^{-12}	1.66×10^{-9}	2.09×10^{-9}	2.25×10^{-8}	3.94×10^{-8}
		Max	7.38×10^{-5}	0.0001	3.89×10^{-8}	1.80×10^{-8}	5.89×10^{-6}	7.39×10^{-6}	7.98×10^{-5}	0.0001
		Avg	1.41×10^{-6}	2.52×10^{-6}	7.44×10^{-10}	3.43×10^{-10}	1.13×10^{-7}	1.41×10^{-7}	1.52×10^{-6}	2.66×10^{-6}
Heavy metal(loid)			HQ_{ing}		HQ_{inh}		$HQ_{d \times 10rm}$		HI	
			Adult	Child	Adult	Child	Adult	Child	Adult	Child
Non-carcinogenic	As	Min	0.0118	0.0840	1.14×10^{-4}	5.65×10^{-6}	7.01×10^{-5}	0.0006	0.0119	0.0846
		Max	122.4741	875.0352	1.1919	0.0588	1.1919	5.9759	124.8579	881.0699
		Avg	12.4379	88.8644	0.1210	0.0060	0.1138	0.6069	12.6728	89.4773
	Hg	Min	1.13×10^{-5}	8.04×10^{-5}	4.20×10^{-9}	7.76×10^{-9}	6.42×10^{-7}	3.22×10^{-6}	1.19×10^{-5}	8.36×10^{-5}
		Max	0.0020	0.0146	7.64×10^{-7}	1.41×10^{-6}	0.0001	0.0006	0.0022	0.0152
		Avg	0.0002	0.0017	8.69×10^{-8}	1.61×10^{-7}	1.33×10^{-5}	6.66×10^{-5}	0.0002	0.0017
	Cd	Min	1.69×10^{-5}	0.0001	1.80×10^{-7}	3.33×10^{-7}	6.74×10^{-6}	3.38×10^{-5}	2.38×10^{-5}	0.0002
		Max	0.6989	4.9935	0.0075	0.0138	0.2789	1.3982	0.9852	6.4055
		Avg	0.0359	0.2565	0.0004	0.0007	0.0143	0.0718	0.0506	0.3290
	Cu	Min	0.0001	0.0011	1.57×10^{-8}	2.90×10^{-8}	1.96×10^{-6}	9.85×10^{-6}	0.0001	0.0011
		Max	0.2153	1.5382	2.28×10^{-5}	4.22×10^{-5}	0.0029	0.0144	0.2182	1.5526
		Avg	0.0134	0.0959	1.42×10^{-6}	2.63×10^{-6}	0.0002	0.0009	0.0136	0.0968
	Ni	Min	0.0003	0.0019	2.82×10^{-8}	5.21×10^{-8}	4.03×10^{-6}	2.02×10^{-5}	0.0003	0.0020
		Max	0.0095	0.0681	9.87×10^{-7}	1.82×10^{-6}	0.0001	0.0007	0.0097	0.0689
		Avg	0.0030	0.0207	2.99×10^{-7}	5.53×10^{-7}	4.27×10^{-5}	0.0002	0.0030	0.0209
	Pb	Min	0.0020	0.0146	2.17×10^{-7}	4.01×10^{-7}	5.44×10^{-5}	0.0003	0.0021	0.0149
		Max	7.2386	51.7171	0.0008	0.0014	0.1925	0.9654	7.4319	52.6839
		Avg	0.1384	0.9886	1.47×10^{-5}	2.71×10^{-5}	0.0037	0.0185	0.1421	1.0071

Note: "-" indicates that these data are not available.

As is a high-risk element, the mean values of the oral route for children is approximately seven times higher than that for adults. These values were greater than 1, indicating that the effects of As on children via the oral route were more pronounced. The maximum values of Cd, Cu, and Pb in the oral route for children, Cd in the dermal route, and Pb in the oral routes for adults and children were all greater than 1, indicating that they pose a non-carcinogenic risk to the human body. The maximum values of Hg and Ni in the different exposure routes were all less than 1, and that of Ni was less than 1 for the different exposure routes, indicating that they did not pose a significant risk to human health.

3.5. Analysis of Heavy Metal Sources

Heavy metal(loid)s exist in the soil during natural formation and are usually harmless to the soil. However, under the influence of human activities, heavy metal(loid)s continue to accumulate in the soil such that the heavy metal(loid) content is much higher than the natural background value [64], posing a serious threat to the ecosystem and human health [65]. A Comprehensive characterization of the heavy metal(loid) content in the soil and the solid waste, single factor index, Nemero pollution index, and health risk assessment revealed that the main pollution factors in the study area were As, Cd, Pb, and Cu, which were greatly affected by human influence (Table 3). Simultaneously, identifying the sources of soil heavy metal(loid)s in the study area is particularly important for the subsequent management of the study area. Correlation analysis is an important means of identifying the sources of soil heavy metal(loid)s [66,67]; Pearson correlation coefficients of heavy metal(loid) elements in the soil are shown in Table 8, where $p < 0.01$. As was highly significantly and positively correlated with Cd, Cu, and Pb, Cd was with Cu, and Pb, Cu, Pb, Hg, Ni, Cu, and Cu was with Pb, whereas Hg and Ni did not significantly correlate with the remaining elements, indicating other sources of Hg and Ni.

Table 8. Pearson correlation coefficients.

	As	Hg	Cd	Cu	Ni	Pb
As	1					
Hg	0.122	1				
Cd	0.893 **	0.153 *	1			
Cu	0.902 **	0.101	0.899 **	1		
Ni	−0.471 **	0.214 **	−0.493 **	−0.367 **	1	
Pb	0.759 **	0.376 **	0.760 **	0.777 **	−0.309 **	1

Note: * indicates a significant correlation in the two-sided test (0.05); ** indicates a significant correlation in the 0.01 level (two-sided test).

Principal component analysis is an important means of discriminating heavy metal(loid)s in soil [68,69]; using the software IBM SPASS 25 to carry out the Kaiser Meyer Olkin (KMO) test for the elemental content, the KMO value was 0.779, indicating that the test data have the conditions for factor analysis. Simultaneously, the maximum variance method was used to rotate the factor-loading matrix (Table 9). The cumulative contribution rate of the three principal component factors was 93.19%, with the first principal component factor (PC1) contributing approximately 56.37%, on which As, Cd, Cu, and Pb had the highest scores, and the second principal component factor (PC2) contributed approximately 18.61%, with the highest score on PC2.The third principal component factor, PC3, contributed approximately 18.22%, and Ni scored the highest on PC3. The distribution of the elements in Figure 4 is highly similar to that of the elemental components, with high scores for PC1, PC2, and PC3 indicating that they have similar origins or characteristics. The results of the cluster analysis show that at a distance of 5–10, it can also be categorized into three classes (Figure 4-(2)).

Table 9. Principal component analysis results of soil heavy metal(loid) element contents.

Element	Component		
	PC1	**PC2**	**PC3**
Eigenvalue	3.382	1.116	1.093
Variance (%)	56.372	18.605	18.216
As	0.925	0.038	−0.243
Hg	0.106	0.977	0.123
Cd	0.911	0.078	−0.280
Cu	0.968	−0.004	−0.103
Ni	−0.268	0.129	0.954
Pb	0.822	0.372	−0.142

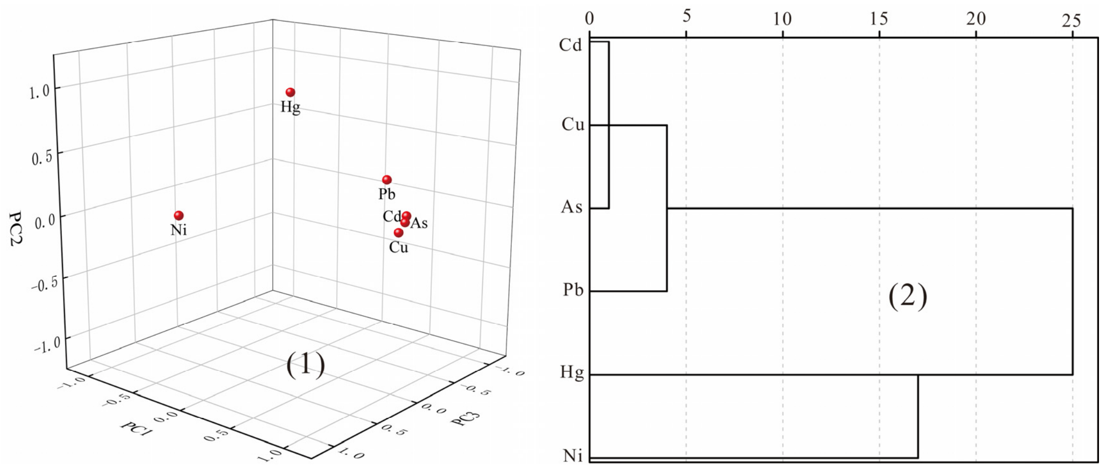

Figure 4. Spatial distribution plots of heavy metal(loid)s in soils based on principal components analysis—(1), cluster analysis of soil heavy metals—(2).

Comprehensive correlation analysis, principal component analysis, and cluster analysis showed that the heavy metal(loid)s in the study area could be divided into three categories: (PC1) As, Cd, Cu, Pb, As, Cd, Cu, and Pb contents in the soils of the beneficiation plant were much larger than the control point and the background value of the soils in Yunnan Province. The coefficients of variation were all greater than 1, indicating that it is subject to anthropogenic factors, and the study area belongs to the typical nonferrous metal area with a long history of mining and smelting. As, Cd, Cu, and Pb pollution is related to the abandoned ore, slag, and tailings pile in the mining area [70–72]; at the same time, the deposits in the study area are polymetallic deposits of tin, zinc, and copper, etc. [73], and As, Cd, Cu, Pb, and so on are the mineralizing elements of the deposits in the area [74–76]; for the second category (PC2) Hg, the Hg content in the study area is higher than that in the control point, close to the Yunnan Province background value. The coefficient of variation was greater than 1, indicating that the anthropogenic factors had a significant influence. Previous studies have found that the combustion of fossil fuels, such as coal, causes the accumulation of Hg in the soil [77,78], and it is hypothesized that PC2 originates from fossil fuels. In the third category (PC3) Ni, the content of Ni in the study area is lower than the content of the unperturbed content of the surrounding area, as well as the background value of the soils in Yunnan Province, and the coefficient of variation is less than 1 (Table 3), indicating that anthropogenic factors have not significantly influenced Ni; this indicates that Ni was not affected by anthropogenic factors, which is consistent with previous studies [79,80], and PC3 represents the source of the soil-forming parent material.

3.6. Risk Prevention and Control

According to the future planning of the site as a construction site, which belongs to a class of land, the exposure pathway of pollutants needs to be considered for its impact on human health and environmental health risks [81], while the site has complex hydrogeological conditions, the source of heavy metal(loid)s in the soil is mainly caused by waste gas and waste water discharges, waste residue stockpiling, or improper disposal. On the one hand, the remediation needs to be combined with the actual situation of the site, such as the site's hydrogeological conditions, the degree of contamination, and the multi-metal synchronous curing/stabilizing materials, and soil-groundwater synergistic remediation techniques can be used to remediate the site [82]; on the other hand, there are still residents in the vicinity of the site, and the remediation process is a long-term process. The public also needs to be encouraged to participate and to understand the site in depth in order to avoid the risk, so as to protect the public's health.

4. Conclusions

(1) The average value of soil pH in the sample points of the study area is 6.56, and the whole is acidic; the average value of Hg and Ni elements is lower than the content of the control points and the background value of the soil in Yunnan Province and higher than the national background value of the soil; the average content of As, Cd, Pb, and Cu is significantly higher than the background value of the soil in Yunnan Province, which indicates that there is a certain degree of enrichment of them in the soil of the study area; the coefficients of variation for As, Cd, Pb, and Cu are all greater than 1, which may be affected by human activities; the main exceeding heavy metal(loid)s are As, Cd, and Pb, and at the same time, the main pollution factors in solid waste are As, Cd, Pb, and Cu.

(2) Pollution in the study area was mainly distributed in the southeast, and the spatial distribution of most heavy metal(loid)s was concentrated in lumber mills, factories, and waste dumps in the study area.

(3) The evaluation results of the single-factor and Nemero composite pollution indices were consistent as follows: As > Pb > Cd > Cu > Ni > Hg. The proportion of the heavy pollution degree of the single-factor index was As > Cd > Pb > Hg = Cu = Ni.

(4) For the exposure assessment analysis, non-carcinogenic, and carcinogenic average daily intake of different heavy metal(loid)s, it is in the order of As > Cu > Pb > Ni > Cd > Hg, with the different pathways for ADDinh < ADDderm < ADDing, and for the health risk evaluation, adults and children in the order of different pathways for cancer risk are CRing > CRderm > CRinh. The order of non-cancer risk is HQing > HQderm > HQinh. The carcinogenic risk of element As is the largest, and the carcinogenic risk of Cd, Ni, and Pb in some sampling points is within the acceptable range. Hg does not pose a carcinogenic risk to humans.

(5) The sources of soil heavy metal(loid)s in the study area can be divided into three categories: As, Cd, Cu, and Pb, which are mainly derived from mineral extraction and metallurgy; Hg is related to the combustion of fossil fuels, and soil-forming matrices are the main sources of Ni.

In summary, As elemental pollution is the most prominent, while the pollution status of Cd, Cu, and Pb in some areas should not be ignored, and the overall pollution level was relatively low; however, timely measures are still needed to prevent the further accumulation of heavy metal(loid)s. Plots with high heavy metal(loid) contents need to be monitored for a long time, and waste treatment needs to be scientific.

However, this study has potential limitations. Only a basic pollution evaluation, health risk evaluation, and source analysis were performed, and there is a lack of content prediction.

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