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Original Research

Ecological Risk Assessment and Spatial Distribution of Heavy Metal in an Abandoned Mining Area, a Case Study of Liaoyang Pyrite Mining Area in China: Implications for the Environmental Remediation of Mine Sites

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Abstract

An important hidden source of soil heavy metal pollution was the discharge of wastewater and waste rock during mining production activities. Numerous processes could result in the buildup of heavy metals in soil, and exposure to these metals could irreversibly harm a person's health. The source, distribution and quantity of historical waste residue in a pyrite mining area were described, and the nature of the waste residue was determined based on the results of toxic leaching identification in this study. The study showed that the pH value of the leaching solution of the waste residues was less than 6. Therefore, these waste residues was the second category of general industrial solid waste. The soil around the waste residues was basically acidic. Also, the content of arsenic, cadmium and copper in farmland soil around the waste residues exceeded the screening value in the Soil Environmental Quality Standard Risk Control of Soil Pollution in Farmland (GB15618-2018). Therefore, the arsenic, cadmium and copper in the waste residue could migrate with rainwater and cause some pollution to the neighboring farmland soil. The surface water was acidic and the arsenic concentration in the surface water exceeded the standard limit. Therefore, surface water was affected by acidic wastewater. However, the impact of project wastewater on groundwater was not significant. Through the analysis of the pollution status, comprehensive treatment countermeasures and secondary pollution prevention measures for the waste residue were proposed, as well as the issues

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that need attention when formulating specific treatment plans, which could provide a reference for similar projects.

Keywords: historic waste residue, pyrite, leachate treatment, ecological restoration

Introduction

Mineral resources were crucial for human development, reproduction, and survival. However, human activities including coal burning, metal smelting, mining for minerals, and sewage irrigation were significant sources of heavy metal pollution [1]. Pollution and environmental issues could arise when mining mineral resources. The continual expansion of scale brought about by the quick development of mineral resource mining had resulted in a significant accumulation in tailings [2]. The majority of mine tailings were kept in natural stacks and ponds, which might take up a lot of space and could seriously pollute the environment. The main causes of heavy metal migration during the extraction of heavy mineral resources were natural hydrological and atmospheric processes. The primary elements that could provide possible risks to the mining environment were the weathering and leaching of the ore and discharged industrial waste, which were significant sources of heavy metal pollution in soil [3].

There were various degrees of heavy metal contamination in the soil, surface water, and groundwater in the mining area as a result of the waste residues produced during mining, beneficiation, and smelting operations migrating and spreading to the surrounding areas through rainfall and dustfall processes [4]. Heavy metals are persistent, accumulate, and degrade slowly, making them some of the most prevalent soil environmental contaminants. Once heavy metals enter the soil, they could not be biodegraded due to the lack of effective export routes. Because heavy metals were present in the soil for a long time, they continue to accumulate [5]. This process, which also could reduce the yield and quality of agricultural products, could affect the nature and function of the soil [6]. Additionally, heavy metals could be accumulated in crops and infiltrate the food chain, posing a health concern to the local resident [7].

The non-degradable nature of heavy metals could result in the accumulation of pollutants in the substrate of water bodies, sites, and soil in some areas [8]. The potential pollution risk was more serious due to the prolonged duration of pollution, outdated treatment technology, and poor supervision and management [9]. Therefore, the pilot project to address historical legacy issues about ailing ponds, waste storage sites, agricultural land contaminated by heavy metals, landfill surroundings, and ecological damage in mining areas was explicitly listed as one of the key projects in the national ‘Comprehensive Prevention and Control Plan for Heavy Metal Pollution’ [10]. The state of heavy metal contamination in a pyrite mining location with historical residues was examined in this study and the remediation strategies that could serve as a model for similar operations were provided.

Material and Methods

Study Area

This sulfur iron mine, which was shut down in 1988 and is now abandoned, was mined in 1972 and was situated in Liaoyang County, Liaoyang City, Liaoning Province. A source of waste contamination upstream of the Tanghe reservoir was created by the mine’s abandoned waste residue dumps. The nearby soil environment, surface water environment, groundwater environment and the nearby rural environment were seriously polluted by the toxic and harmful substances contained in the waste residue, which also posed a threat to the Tanghe Reservoir. Over time, the mining waste rocks have been haphazardly piled up naked, producing yellow acidic wastewater. The project was surrounded by 135hm^2 of farmland. The waste residue from the contaminated site would be washed away by rainwater, which was very likely to cause the continuous spread of pollution and would continue to cause more serious pollution and damage to the neighboring farmland [11]. Also, the water quality of the Tanghe Reservoir's drinking water sources were affected by the pollutants in the waste residue. Contamination of the drinking water source water quality of the Tonghe Reservoir, as well as agricultural soils, could affect the health and safety of local residents. This was because the waste residue was located upstream of the Tanghe reservoir. Therefore, the ecological restoration of the abandoned waste residue piles was essential.

The cause of site pollution was mainly due to the accumulation of waste residue in this project area [12]. The exudate of waste residue had a high content of iron and manganese. Pyrite ions in the waste residue were oxidized by oxygen in the air to form iron hydroxide precipitation. This could lead to reddish and yellowish seepage and the formation of "sulfur water". Thus, a layer of yellowish-brown sediment had settled at the bottom of the river over time. Also, the acid water generated in the project area was in the pH range of 3-5, and the water was strongly acidic.

Site Status

The project’s scope was illustrated in the Fig. 1. The project's total solid waste treatment area was

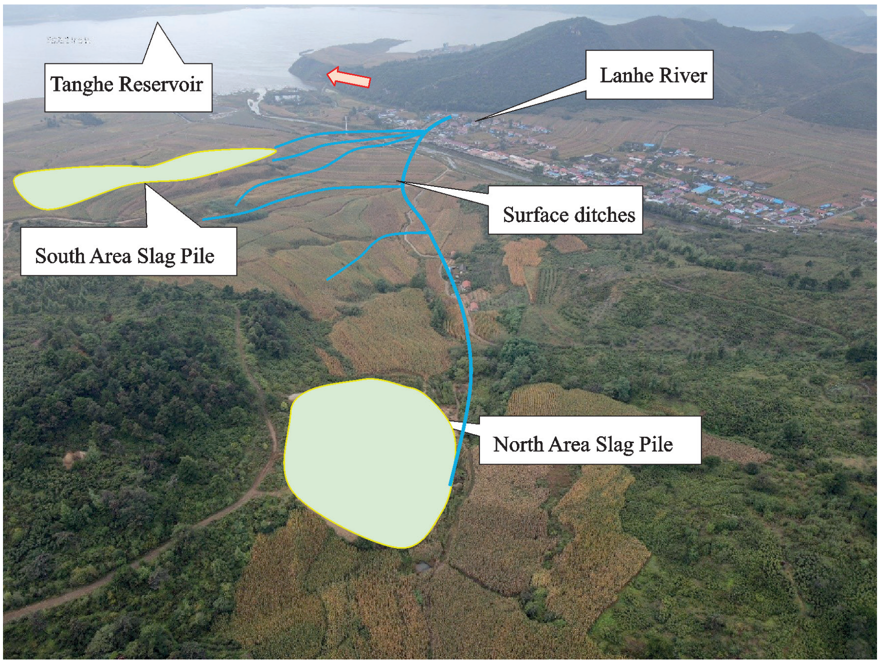

Fig. 1. General aerial view of the project site.

approximately 34,800 m^2. In the north and south portions of the iron sulfide mine during mining, there were two abandoned waste residue stockpiles. 6080.44 m^2 was the size of the north area, and 28680.4m^2 was the size of the south area. The storage capacity of waste residue was 208565.04 m^3.

The majority of the sulfur and iron waste residue was piled up in a dispersed manner due to the early mining methods, and the available land resources were occupied. Acidic wastewater was generated under the action of air oxidation, rainwater drenching, and surface runoff, and the nearby soil and water environment was polluted by the acidic wastewater. Tanghe Reservoir was located 2,000 meters downstream of the Lan River. Therefore, Tanghe Reservoir's water quality was also affected by the acid water. In addition, soil and nearby vegetation might also be polluted by the two waste residue piles in different degrees. Therefore, the two waste residue piles posed a possible threat to the surrounding environment.

(1) The present state of waste residue pile in the region of the north

There are no environmental protection facilities for waste residue piles in the north, such as waste residue retaining walls or flood drains around the waste residue accumulation area. The surface area of the waste residue pile in the north area was 6080.44 m^2, with a height of 22 m, and the estimated volume was 36,482.64 m^3. The waste residue pile in the north was piled in the open, and the exposed waste residue was blade-like after natural weathering, with a loose structure, and the overall shape of the regional slope was irregular. The surface layer of the slope body was covered with thin loose accumulation, which was primarily small-grained gravel falling blocks during the early mining process, and the slope was steeper. The subsequent mining process resulted in a more uneven shape. The pile could easily slide downstream into the river, which would have a negative impact on the surrounding area. The pile might continue to slide to the foot of the slope. One mining cave with a 1 x 1.5 m shaft size was located in the project's northern region. The pollution status was shown in Fig. 2.

(2) Waste residue pile's southern section

The surface area of the south section's waste residue pile is 28680.4 m^2. The volume of waste residue was estimated to be 172,082.4 m^3. The waste residue pile in the southern area was covered with planting soil, shrubs, and grass. Also, drainage ditch had been built. However, there was no isolation and anti-permeation measure above the waste residue pile. As a result, under rainfall conditions, acidic wastewater could seep out of the waste residue pile in the southern zone and sink into the drainage ditch in the southern zone.

(3) The condition of the drainage ditches around the project site

On the project site, there were four drainage ditches. The site survey revealed that there were exposed waste materials and loose construction in the project site ditches. With surface water infiltration and rainfall erosion, acidic wastewater from the project site could seep out of the drainage ditches and was discharged into the Lan River with the on-site ditches. Agricultural land was located downstream of the project site, and both the waste residue and the agricultural land were in the same watershed. The leachate was generated by precipitation drenching and could enter the project site ditch with surface runoff, which in turn could pose a threat to farmland.

An aerial image of the project's northern waste residue pile

The slope of the waste residue heap in the Northern area

Pollution statue in northern region

Mine cave in the northern region

The waste residue pile in the southern area

Drainage ditch in the southern area

Drainage ditch

Drainage ditch

Fig. 2. The pollution current state of the study area.

Sampling

Waste Residue Sampling

The site's waste residue samples were examined. Five profile waste residue samples (0-600 cm) were collected using a stainless-steel auger in May 2023 [1]. And two soil control was collected in the farmland area upstream of the waste site and the bottom of the hill, respectively. The location of the collected samples were shown in the Fig. 3 and recorded using Global Positioning System (GPS, Garmin 72).

All the samples were collected using a plastic shovel and put in the polyethylene bags, labeled and transported to the laboratory [13]. Samples were homogenized, air-dried, and crushed manually. Samples were ground to pass through a < 0.149 mm sieve and analyzed for their heavy metal contents and tested for leaching toxicity (water leaching test) [14].

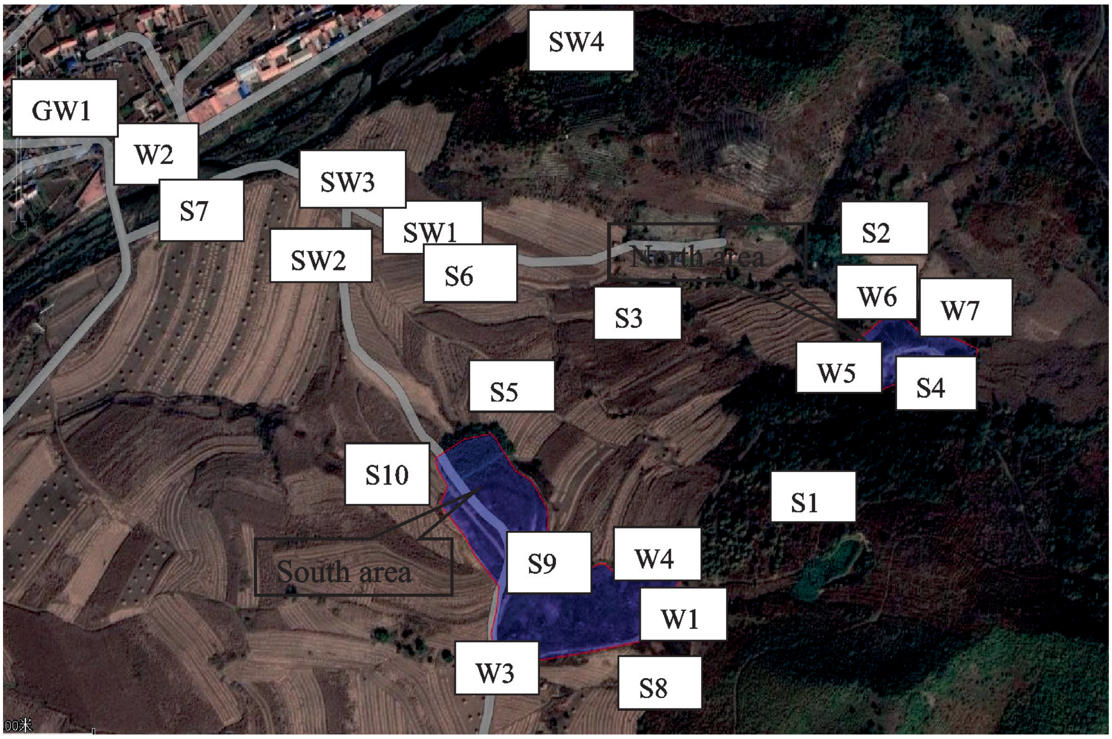

Fig. 3. Schematic diagram of waste residue,soi, surface water and underground water sampling points.

Soil Sampling

Ten profile soil samples (0-100 cm) were collected using a stainless-steel auger in May 2023 [15]. The sample point map was displayed in Fig. 3, and sampling depth and details about the sampling points was listed in the following table.

All the samples were collected using a plastic shovel and put in the polyethylene bags, labeled and transported to the laboratory [16]. Soil material was homogenized, air-dried, and crushed manually. Samples were ground to pass through a <0.149 mm sieve and analyzed for their heavy metal contents [17].

Surface Water Sampling

The Lan Rive could play a critical role in the Tang River Reservoir, which was located about 500 m downstream of the waste residue. The sampling points were shown in the table below. Four unfiltered surface water samples were collected from the center of the river for pH, arsenic, suspended particles, and sulfides analyses. The samples were then transferred into acid cleaned 100 mL polypropylene bottles. One mL of ultrapure nitric acid was added in each polypropylene bottle to achieve a pH of 1 [18].

Groundwater Sampling

One groundwater monitoring site was established in a groundwater well in the community downriver of the waste residue site to examine the effect of waste residue on groundwater pollution. The Groundwater Quality Standard (GB/T 14848-2017) III standard limits were used to analyze the groundwater test findings.

Sample Analysis

Waste Residue Analysis

All the chemicals used in our study were of analytical grade and aqueous solutions were prepared with deionized water [1]. The contents of the heavy metal for waste residue was determined according to the Solid Waste-Determination of metals - Inductively coupled plasma mass spectrometry (ICP-MS, HJ766-2015) [19].

The water leaching test for waste residue was performed according to the Solid waste-Extraction procedure for leaching toxicity-Horizontal vibration method (HJ557-2009) [20]. Distilled water was used to extract samples. The samples were extracted at a liquid to solid (L/S) ratio of 10 in a capped polypropylene bottle on a rotary tumbler at 110 rpm for different time at room temperature. After the extraction, the final pH of the leachate was measured. The liquid was separated by filtration through a 15 μm paper filter and the concentrations of the Cd, Cu, Ni, Hg, As, Zn, Cr and Pb was analyzed. The test results of water leaching were calculated using the maximum allowable emission limits for pollutants of Class I standards of the Integrated Sewage Discharge Standards (GB8978-1996) [8]. The results could allow researchers to better understand the properties of heavy metal scrap in the area.

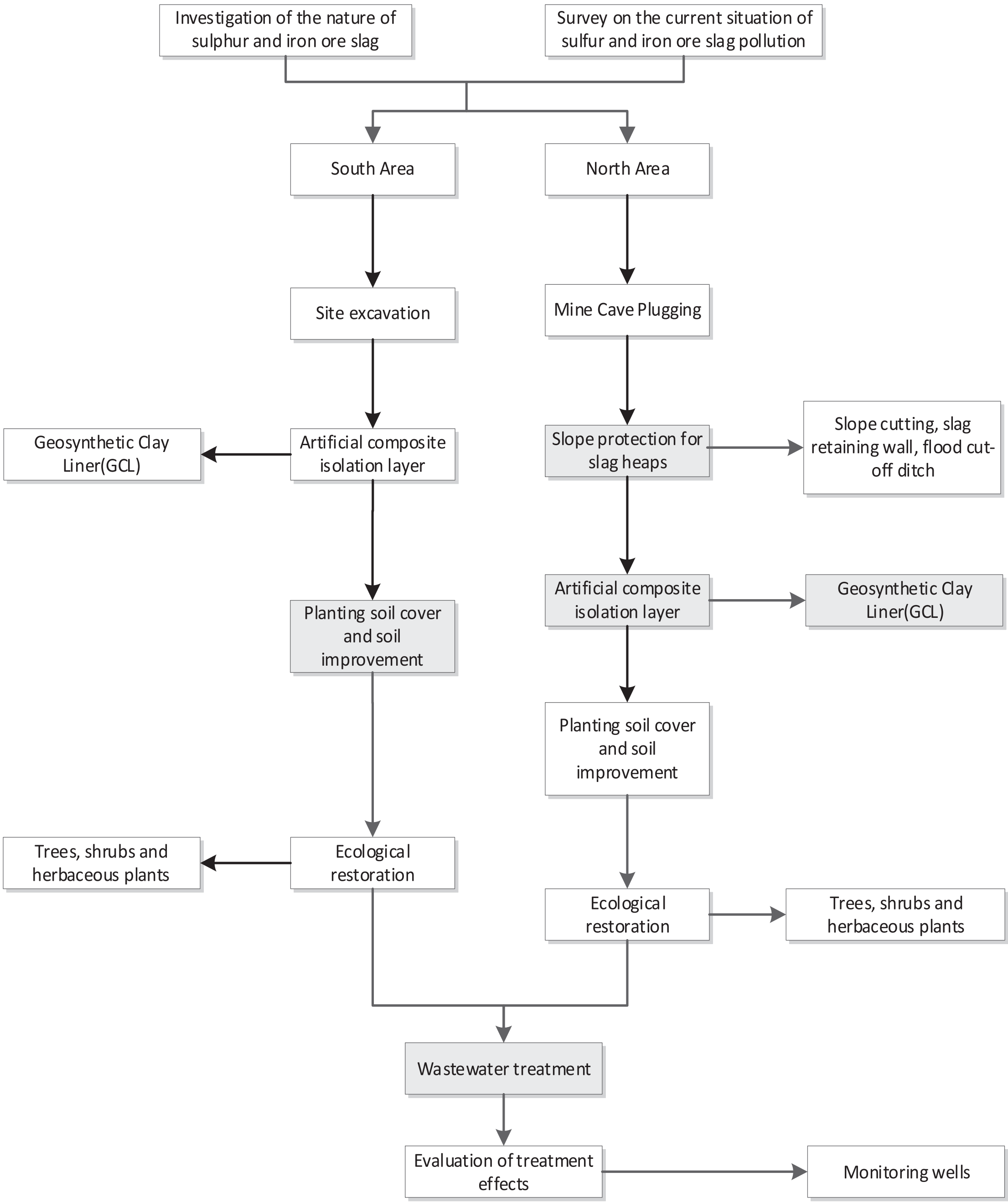

Fig. 4. Technology route.

Soil Sample Analysis

The soil samples were digested according to USEPA Method 3051A [21] for total heavy metal concentrations analysis with minor modifications by digesting 0.200 g soil sample with 15 mL of tri-acidic mixture (HNO_3, HCl, $HClO_4$) in a volume ratio of 1:3:1.The concentrations of Cd, Cu, Ni, Pb and Zn in digested samples were determined using atomic absorption spectrophotometry [22]. Detection limits for Cd, Cu, Ni, Pb, and Zn were 1.8, 2.3, 6.8.27.3, and 1.6 g L^{-1}, respectively.

The concentrations of Cd, Pb, mercury (Hg), arsenic (As), Zn, copper (Cu), nickel (Ni), and chromium (Cr) were determined [23]. Briefly, soil samples (0.5 g) were digested with a mixture of HNO_3, HCl, HF, and $HClO_4$ using an Automated Sample Digestion System (Thomas Cain Inc., Mequon, WI, USA). The total concentration of Hg was determined using a Hydra-C atomic absorption spectrophotometer (Leeman, USA). The total concentration of As was determined using an atomic fluorescence spectrometer (JiTian, Bejing, China). An inductively coupled plasma atomic emission spectrometer (Leeman, USA) was used to detect the total concentrations of Pb and Zn. The total concentrations of Cd, Cu, Ni, and Cr were measured using inductively coupled plasma-mass spectrometer (Agilent, Japan) [24].

Soil pH were measured in this study [23]. A digital pH meter (ORION5 STAR, thermo scientific) was used for measuring the pH of the extraction solution (1:10, w/v sample: distilled water) after extraction by shaking for 2 h at 150 rpm [25].

Water Analysis

The surface water and underground water sampling bottles were rinsed with distilled water in order to eliminate any primary contamination [26]. The pH of samples was set to lower two by the nitric acids (65% Merch, Germany) to reduce HMs absorption [27]. The samples were sampled in 2-L polypropylene bottles and transported to the lab at 4ºC. Based on the guidelines, the time between collecting the samples and delivering them to the laboratory was about 5-6 h. In all the analysis, double-distilled water was used. The standard solution of all metals was produced by diluting 1000 mg/L predetermined amount of standard solutions. Inductively coupled plasma mass spectrometry is the method for measuring the contents of heavy metal of surface water and underground water [28].

Results and Discussion

Nature of Waste Residue

After the solid waste was subjected to toxic leaching experiments, the concentration of any one pollutant in the leachate obtained from the solid waste did not exceed the maximum allowable discharge concentration specified in the "Comprehensive Sewage Discharge Standard" (GB8978-1996), and the pH value of the leachate was within the range of 6~9 [29]. This industrial solid waste was the first category of general industrial solid waste according to the "General Industrial Solid Waste Storage and Disposal Site Pollution Control Standards" (GB18599-2001). Leachate obtained from industrial solid waste on the toxic leaching experiments had one or more pollutant concentrations exceeding the maximum allowable discharge concentration specified in the “Integrated Sewage Discharge Standard” (GB8978-1996), or the pH value of the leachate was outside the range of 6 to 9 [30]. This industrial solid waste was the second category of general industrial solid waste.

Although the number of pollutants in the waste sludge in the north and south areas did not exceed the maximum allowable discharge concentration specified in the “Integrated Sewage Discharge Standards (GB8978-1996)”, the pH of the leaching solution for the waste sludge was less than 6. Therefore, the wastes in the north and south districts were classified as Class II general industrial solid wastes.

Soil Contamination Results

Their test results were shown in Table 8, and the Soil Soil Environmental Quality Risk Control Standards for Soil Contamination on Agricultural Land (GB15618-2018)were used for assessment (Table 4).

The soil in the project area was basically acidic. This result could be due to acidic soil caused by the acidic water generated at the project site. The arsenic contents of soil in the exceeded the screening value of the "Soil Environmental Quality Risk Control Standard for Soil Contamination on Agricultural Land" (GB15618-2018). The contents of cadmium and copper in the farmland soil exceeded the screening value of "Soil Environmental Quality Standards for Soil Contamination Risk Control on Agricultural Land" (GB15618-2018). The soil exceeded the standard to some extent. Therefore, it was determined that the migration of arsenic, cadmium and copper in the waste residue with rainwater caused some pollution to the downstream soil.

Surface Water Pollution Results

The result of the surface water was listed in Table 9. The surface water on the south, west and east side of the drainage canal and the entrance to Lanhe River was acidic, and the pH was lower than the standard limit of category III of the "Surface Water Environmental Quality Standard" (GB3838-2002). The surface water at the mouth of the Lanhe River and the east, south, and west drains all contained more arsenic than was permitted. The arsenic concentration in the west drains reached 1.31 mg/L, which could exceed

Table 1. Statistical table of waste residue sampling points.

Test category	Sampling number	Sampling points	Sampling coordinates	
Soil	W1	The top of the hill in the south area	123.414599°	41.041010°
	W2	Control point at the bottom of the hill	123.409423°	41.045920°
	W3	Farmland in the south area	123.412118°	41.040376°
	W4	South slope	123.414324°	41.041304°
	W5	North slope foot	123.416820°	41.043612°
	W6	North slope	123.416835°	41.043619°
	W7	Farmland in the north area	123.416889°	41.043631°

Table 2 Evaluation criteria for waste residues.

Number	Category	Pollution factor	Concentration limit value (mg/L)	Implementation standard
1	Water leaching	pH	6-9	Integrated Sewage Discharge Standards (GB 8978-1996)
2		Pb	1.0	
3		Cd	0.1	
4		As	0.5	
5		Zn	2.0	

Table 3 Statistical table of soil sampling points.

Test category	Sampling number	Sampling points	Sampling coordinates	
Soil	S1	Farmland	123.416504742°	41.042210966°
	S2	Farmland	123.417953135°	41.043557435°
	S3	Farmland	123.414069297°	41.043702275°
	S4	Farmland	123.417061301°	41.043301284°
	S5	Farmland	123.413563700°	41.042764843°
	S6	Farmland	123.411521198°	41.044142157°
	S7	Farmland	123.409847500°	41.044753701°
	S8	Farmland	123.413789006°	41.040447414°
	S9	Farmland	123.412608834°	41.041101873°
	S10	Farmland	123.411257001°	41.042593181°

Table 4. Soil evaluation criteria.

Number	Pollution factor		Risk Screening Value			
			pH<5.5	5.5<pH≤6.5	6.5<pH≤7.5	pH>7.5
1	Cd	Water Field	0.3	0.4	0.6	0.8
		Other soil	0.3	0.3	0.3	0.6
2	Hg	Water Field	0.5	0.5	0.6	1.0
		Other soil	1.3	1.8	2.4	3.4
3	As	Water Field	30	30	25	20
		Other soil	40	40	30	25
4	Pb	Water Field	80	100	140	240
		Other soil	70	90	120	170
5	Cr	Water Field	250	250	300	350
		Other soil	150	150	200	250
6	Cu	Water Field	150	150	200	200
		Other soil	50	50	100	100
7	Ni		60	70	100	190
8	Zn		200	200	250	300

Table 5. Surface water sampling point statistics.

Test Category	Sampling Number	Sampling points	Sampling coordinates	
Surface water	SW1	East side drainage canal	123.410431°	41.044373°
	SW2	South side drainage canal	123.410280°	41.044335°
	SW3	West side drainage canal	123.410233°	41.044070°
	SW4	Into the Lan River mouth	123.413498°	41.046587°

Table 6. Groundwater sampling points.

Test category	Sampling number	Sampling points	Sampling coordinates	
Groundwater	GW1	Groundwater wells in the village downstream of the waste residue	123.405631°	41.044632°

the standard limit of category III of the "Surface Water Environmental Quality Standard" (0.05 mg/L) and exceed the standard by 25.2 times.

The arsenic content of the south side drain was 0.137 mg/L, which was 1.74 times higher than the 0.05 mg/L limit set by the standard limit of category III of Surface Water Environmental Quality Standard (GB3838-2002). The pH of surface water decreased significantly after the waste residue, while the arsenic content in water increased significantly. This demonstrated the arsenic in the waste was continuously migrating to the surrounding soil and downstream farmland with rainwater. As a result, the surface water was affected by the acidic effluent from this operation.

Groundwater Contamination Results

The groundwater of the project was not polluted and the concentration of the groundwater could meet the standard of "Groundwater Environmental Quality Standard" (GB/T14848-2017) for Class III. It was obvious that the waste residue in the project might pose little effect on the groundwater.

Conclusion and Discussion

Waste Residue Risk Assessment

Based on the result, the waste residue could be determined as the second general industrial solid waste. The exposed pile of waste residue layer was vulnerable to rainfall erosion or drenching. Meanwhile, the site was located in the higher mountainous terrain. During spring and summer rainy seasons, heavy rainfall directly washed the waste residue pile in the site, which could cause waste migration [31]. In addition, rainwater leaching could cause the heavy metals in the waste residue to infiltrate the lower layer and the surrounding area, leading to the spread of heavy metal pollution in the soil below and around the site.

Risk Assessment for Soil Pollution

The total amount of heavy metals arsenic, cadmium and copper of the waste residue within the site area could exceed the standard, and the plants within the site area were sparse, with soil erosion and bare ground state. The waste residue has caused serious pollution to the soil in this area. The waste residue and its contaminated soil distributed within the site had become an important source of pollution [32]. According to the site investigation results, the site residue was distributed along the mountain road. Therefore the waste residue was washed by rainwater, which was very susceptible to the continuous spread of pollution. The waste residue would continue to cause more serious pollution and damage to the surrounding soil and ecological environment [33].

Risk Assessment for Agricultural Pollution

According to the site study, agricultural land was located downstream of the project site. And both the waste residue and the agricultural land were located within the same watershed. Leachate was generated from the waste residue under the effect of precipitation leaching, which might enter the ditch of the project site with surface runoff, and then could threaten the farmland [34]. As shown in the table below, the pH of the waste leachate in the south and north areas does not meet the irrigation water quality standard (GB 5084-2021). Therefore, the acidic wastewater could have an impact on farmland.

Pollution of Surface Waters Risk Assessment

About 500 m downstream of the contaminated site was surface water of Lanhe River was about 500 m downstream of the waste residue. There was acidic wastewater with a yellow color and a certain degree of pollution at the project site according to the survey. As shown in the result of the surface water, the arsenic

Table 7. Leaching test results of waste residue (mg/L) (water leaching).

Detection factor	W1	W2	W3	W4				W5				W6	W7	Concentration limit value
Depth	(0-0.5 m)	(0-0.5 m)	(0-0.5 m)	(0-0.5 m)	(0.5-1.5 m)	(1.5-3 m)	(3-6 m)	(0-0.5 m)	(0.5-1.5 m)	(1.5-3 m)	(3-6 m)	(0-0.5 m)	(0-0.5 m)	
As	0.7×10^{-3}	0.6×10^{-3}	2.7×10^{-3}	1.1×10^{-3}	1.0×10^{-3}	0.9×10^{-3}	1.2×10^{-3}	1.4×10^{-3}	1.3×10^{-3}	1.0×10^{-3}	1.0×10^{-3}	0.5×10^{-3}	1.3×10^{-3}	0.5
Cd	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	<0.05	0.1
Cr	<0.03	<0.03	<0.03	<0.03	<0.03	<0.03	<0.03	<0.03	<0.03	<0.03	<0.03	<0.03	<0.03	1.5
Cu	<0.04	0.06	<0.04	<0.04	<0.04	<0.04	<0.04	0.45	0.45	0.45	0.44	0.12	<0.04	0.5
Pb	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1	1.0
Hg	0.08×10^{-3}	0.26×10^{-3}	0.67×10^{-3}	0.48×10^{-3}	0.43×10^{-3}	0.47×10^{-3}	0.40×10^{-3}	0.93×10^{-3}	0.94×10^{-3}	0.89×10^{-3}	0.92×10^{-3}	0.85×10^{-3}	0.88×10^{-3}	0.05
Ni	<0.007	<0.007	<0.007	<0.007	<0.007	<0.007	<0.007	0.017	0.016	0.016	0.016	<0.007	<0.007	1.0
Be	<0.008	<0.008	<0.008	<0.008	<0.008	<0.008	<0.008	<0.008	<0.008	<0.008	<0.008	<0.008	<0.008	0.005
pH	7.3	7.0	7.1	**5.1**	**5.3**	**5.2**	**5.1**	**4.3**	**4.1**	**4.2**	**4.0**	**4.4**	7.2	6~9
Se	0.9×10^{-3}	0.9×10^{-3}	1.0×10^{-3}	0.9×10^{-3}	1.0×10^{-3}	0.9×10^{-3}	0.9×10^{-3}	1.1×10^{-3}	0.9×10^{-3}	0.8×10^{-3}	0.9×10^{-3}	1.0×10^{-3}	1.0×10^{-3}	0.1
Zn	0.013	0.039	0.210	0.018	0.018	0.019	0.018	0.033	0.032	0.028	0.026	0.014	0.184	2.0
Mn	0.04	0.05	<0.01	0.03	0.03	0.03	0.03	0.34	0.34	0.26	0.24	0.10	<0.01	2.0
Cr^{6+}	<0.004	<0.004	<0.004	<0.004	<0.004	<0.004	<0.004	<0.004	<0.004	<0.004	<0.004	<0.004	<0.004	0.5
Fluoride	0.359	0.644	0.272	0.300	0.701	0.365	0.198	0.287	0.157	0.381	0.335	0.160	0.212	10
Sulfide	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	<0.01	1.0
COD	4	<4	25	4	5	4	4	6	6	5	6	<4	9	100
SS	12	17	15	14	14	17	16	12	15	11	15	16	16	70

Bold numbers represent the exceedance

Table 8. Heavy metal content in the waste residue (mg/kg).

Sampling points	Sampling depth	Category	pH	As	Cd	Cr	Cu	Pb	Hg	Ni	Zn
W1	(0-0.5 m)	Concentration	7.24	18.5	0.2	48	50	19	0.176	58	41
		Exceeding times	/	/	/	/	/	/	/	/	/
W4	(0-0.5 m)	Concentration	5.31	**91.5**	**0.4**	88	**73**	29	0.208	49	97
		Exceeding times	/	**1.29**	**0.33**	/	**0.46**	/	/	/	/
W4	(0.5-1.5 m)	Concentration	5.29	72.7	**0.44**	72	**68**	26	0.173	48	93
		Exceeding times	/	**0.82**	**0.47**	/	**0.36**	/	/	/	/
W4	(1.5-3 m)	Concentration	5.88	**44.4**	**0.42**	62	**63**	23	0.16	46	89
		Exceeding times	/	**0.11**	**0.40**	/	**0.26**	/	/	/	/
W4	(3-6 m)	Concentration	6.07	**42.8**	**0.41**	73	**59**	22	0.175	41	80
		Exceeding times	/	**0.07**	**0.37**	/	**0.18**	/	/	/	/
W5	(0-0.5 m)	Concentration	3.34	**98.4**	**0.4**	45	**94**	31	0.183	53	48
		Exceeding times	/	**1.46**	**0.33**	/	**0.88**	/	/	/	/
W5	(0.5-1.5 m)	Concentration	3.28	**70.4**	**0.39**	41	**90**	29	0.187	50	44
		Exceeding times	/	**0.76**	**0.30**	/	**0.80**	/	/	/	/
W5	(1.5-3 m)	Concentration	5.54	**48.1**	**0.37**	38	**86**	27	0.179	48	39
		Exceeding times	/	**0.20**	**0.23**	/	**0.72**	/	/	/	/
W5	(3-6 m)	Concentration	5.83	**43.6**	**0.36**	36	78	23	0.158	45	35
		Exceeding times	/	**0.09**	**0.20**	/	**0.56**	/	/	/	/
W6	(0-0.5 m)	Concentration	3.28	**95.8**	0.18	38	49	22	0.165	52	48
		Exceeding times	/	**1.40**	/	/	/	/	/	/	/
GB15618-2018 Screening Value			/	40	0.3	150	50	70	1.3	60	200
GB15618-2018 Control value			/	200	1.5	800	/	400	20	/	/

Bold numbers represent the exceedance

and pH content of the drainage canal and the estuary into Lanhe River could exceed the water quality standard of Class III of the Surface Water Environmental Quality Standard (GB3838-2002). And the acidic wastewater produced by the waste residue could flow into the river, polluting the surface water quality [35]. Therefore, the acidic wastewater produced by the waste residue flowed into the river and caused pollution to the surface water quality.

Groundwater Pollution Risk Assessment

The main source of groundwater recharge was precipitation in the atmosphere [36]. Due to the large slope of the surface topography, most of the precipitation gathered on the surface of the mountainous area was rapidly collected in the low-lying valley areas, thus forming surface streams [37]. Additionally, the project site was located in a mountainous area with high terrain, and thus the groundwater of the site was buried deep. Therefore, the impact of waste residue on groundwater is minimal, as shown by the monitoring data from groundwater wells.

Risk from the Geological Environment

A large amount of waste residue was piled up in the project area. The piling of waste residue along the slope was susceptible to geological hazards such as landslides and mudslides [38]. No landslide and mudslide geological hazards were found during the investigation. However, the waste residue in the project area was exposed and no protective measures had been laid, which was prone to landslides and mudslides under special circumstances such as heavy precipitation [39].

Comprehensive Management Strategies

Solution Comparison

The treatment technology of this project mainly consisted of off-site landfill and *in-situ* storage.

Table 9. Heavy metal contents in the farmland soil (mg/kg).

Sampling points	Sampling depth	pH	Pb	Cu	Cr	Ni	Cd	Zn	Hg	As
S1	(0-0.2 m)	6.43	20	29	96	50	0.18	71	0.055	10.1
	(0.2-0.5 m)	6.47	15	28	86	46	0.18	65	0.031	9.38
	(0.5-1.0 m)	6.45	11	26	75	43	0.17	63	0.043	8.38
S2	(0-0.2 m)	**5.44**	63	**76**	121	56	**0.41**	79	0.090	87.2
	(0.2-0.5 m)	**5.47**	54	**73**	105	53	**0.41**	78	0.052	86.4
	(0.5-1.0 m)	**5.47**	51	**67**	94	51	**0.39**	76	0.059	86.0
S3	(0-0.2 m)	**5.41**	37	**72**	126	52	**0.42**	68	0.144	68.1
	(0.2-0.5 m)	**5.43**	30	**64**	120	50	**0.37**	65	0.048	64.3
	(0.5-1.0 m)	**5.40**	17	**65**	91	46	**0.38**	64	0.054	65.7
S4	(0-0.2 m)	**3.37**	31	**92**	164	56	**0.43**	70	0.151	96.5
	(0.2-0.5 m)	**3.41**	25	**90**	137	52	**0.43**	69	0.071	93.7
	(0.5-1.0 m)	**3.45**	15	**88**	103	47	**0.41**	68	0.051	87.9
S5	(0-0.2 m)	**5.32**	31	**73**	68	47	**0.41**	63	0.125	77.1
	(0.2-0.5 m)	**5.34**	26	**70**	48	45	**0.41**	62	0.070	75.4
	(0.5-1.0 m)	**5.40**	15	**66**	29	40	**0.40**	61	0.054	75.3
S6	(0-0.2 m)	6.94	32	**58**	129	51	**0.40**	68	0.097	65.3
	(0.2-0.5 m)	6.91	22	**56**	104	48	**0.36**	65	0.057	60.7
	(0.5-1.0 m)	6.88	13	**52**	87	46	**0.31**	65	0.046	58.9
S7	(0-0.2 m)	6.45	31	**53**	89	50	**0.35**	54	0.083	44.7
	(0.2-0.5 m)	6.41	21	**52**	78	48	**0.33**	51	0.078	43.5
	(0.5-1.0 m)	6.44	14	**52**	69	45	**0.31**	49	0.036	37.2
S8	(0-0.2 m)	**5.31**	27	**79**	108	57	**0.41**	83	0.118	73.2
	(0.2-0.5 m)	**5.36**	20	**78**	89	55	**0.41**	83	0.068	68.9
	(0.5-1.0 m)	**5.44**	11	**75**	63	52	**0.37**	78	0.044	69.2
S9	(0-0.2 m)	**3.34**	50	**94**	76	51	**0.44**	67	0.182	98.2
	(0.2-0.5 m)	**3.36**	42	**90**	78	52	**0.42**	68	0.064	97.3
	(0.5-1.0 m)	**3.41**	40	**86**	86	52	**0.40**	69	0.030	72.7
S10	(0-0.2 m)	**5.07**	29	**76**	110	55	**0.43**	81	0.104	84.5
	(0.2-0.5 m)	**5.32**	21	**73**	94	52	**0.42**	78	0.070	84.0
	(0.5-1.0 m)	**5.41**	13	**71**	64	49	**0.40**	76	0.043	76.3
GB15618-2018 Screening Value		/	70	50	150	60	0.3	200	1.3	40
GB15618-2018 Control value		/	400	/	800	/	1.5	/	20	200

Bold numbers represent the exceedance

(l) *In-situ* storage

By disposing of the waste residue *in-situ*, the contaminated soil was sealed off from the surrounding area, preventing pollutants from migrating with rain or groundwater and contaminating the neighborhood, and harming human health [40]. *In-situ* storage of waste residue could be sealed based on not destroying the original structure of the waste residue piles. The area of the waste residue pile could be effectively protected by constructing a retaining wall under the waste residue pile and interception and diversion channels around the waste residue pile. Through the upper sealing and ecological restoration measures, the cost of excavation and transportation of waste residue pile could be effectively reduced.

Table 10. Surface water monitoring results.

Testing factor	SW1	SW2	SW3	SW4	Environmental Quality Standard for Surface Water (GB3838-2002) III implementation standards
pH	**4.2**	**3.6**	**3.2**	**5.8**	6~9
SS (mg/L)	30	15	15	17	/
Sulfide (mg/L)	<0.01	<0.01	<0.01	<0.01	0.2
As (mg/L)	**0.117**	**0.137**	**1.31**	**0.06**	0.05

Bold numbers represent the exceedance

Table 11. Groundwater monitoring results.

Sampling number	Testing factor	Concentration	Groundwater Environmental Quality Standard (GB/T14848-2017) for Class III water bodies
GW1	pH	7.2	6.5~8.5
	Sulfide (mg/L)	<0.003	0.02
	As (mg/L)	0.001	0.01

(2) Off-site landfill

Off-site landfill disposal was to select a special waste residue disposal site within the perimeter of the waste residue area, and then transport waste residue to the disposal site for landfill disposal. After the completion of landfilling, the disposal site was sealed and greened. The disposal of waste residue by landfill could isolate the contact between waste residue and human and animals, to ensure the safety of personal property and the health of regional residents. Off-site landfill disposal technology was a relatively more thorough treatment technology. However, it could require relocation and disturbance of the waste residue, the waste residue transfer process might bring risks, and the vegetation restoration area was large, and the corresponding investment cost could be increased [41].

Although off-site landfill disposal technology could have a better treatment effect, the volume of waste residue in this project was large. The relocation of large disturbed areas of waste materials was not only expensive but also difficult to find another suitable site for a new landfill [42]. In the process of waste residue relocation and transfer, the waste residue was exposed to the air, which might cause serious pollution [43]. Also the construction period was risky, and the amount of handling work was enormous, and thus the consequences were numerous. The *in-situ* disposal project was small in volume, short in duration, and the cost of *in-situ* disposal was small. *In-situ* disposal could also effectively solve the pollution problem of waste residue [44]. Therefore, *in-situ* sequestration for waste residue was chosen in this study. The desired purpose could be achieved by retaining surface water and controlling the leaching effect of surface water on the waste residue, thus reducing the generation of acidic wastewater.

General Technical Route

There were two waste residue pile regions in the project. The north area was currently exposed and was a steep slope platform. Due to the previously built interception and drainage ditches, the southern section was covered in bushes and grasses. Different restoration technology alternatives were therefore presented in accordance with the divergent conditions of the north and south areas.

The mine cave in the north area should be closed off. By constructing a retaining wall, the waste residue pile could be prevented from producing landslides. And the stability of the waste residue pile could be guaranteed by cutting and shaping the slope of the pile. The construction of interception and drainage facilities on the periphery of the remediated waste residue pile to reduce surface water infiltration and surface runoff into the waste residue pile. The surface impermeability of the north area was carried out with artificial composite materials to achieve the purpose of blocking the transmission pathway and controlling the environmental risk. The treated waste residue pile area was covered with planting soil and soil fertilization to improve the soil structure and reach the purpose of controlling the acidification of the slag pile area from the source. Vegetation was also planted to restore the ecological environment of the pile-up area.

Artificial composite surface impermeability was employed in the south to achieve the purpose of blocking the transmission pathway and controlling the environmental risk [45]. To restore the biological environment of the waste residue pile area, the waste pile area was covered with planting soil, and vegetation was also planted.

Three primary methods for treating acidic wastewater were source control, process interception, and final treatment [46]. Source control is mainly to establish interception drains to collect acidic wastewater in the waste residue pile area to reduce surface runoff. Process interception was mainly to establish ecological ditches to adsorb acidic wastewater pollution by improving the existing drainage channels with *in-situ* substrate and adding microbial bacterial agents and carbon sources. The final treatment was mainly to treat the acidic wastewater generated by using mobile wastewater treatment equipment [47].

Specific Treatment Programs

(1) North area.

In the northern area of the waste residue area, the first step was to block the mine cave. One mine cave was sealed to reduce the generation of acidic wastewater at the source. The second step was to protect the slope of the waste residue pile. The intercepting ditch was set at the bottom of the waste residue retaining wall and at the top of the slope. Meanwhile, drainage ditches were set up on each side of the waste residue pile to reduce the infiltration of surface water on the periphery and reduce surface runoff into the waste residue pile. The third step was to build the physical isolation layer. A physical isolation layer was used to limit environmental threats and to create an impermeable surface. This layer was made of artificial composite material. The fourth step was to perform soil fertilization. Soil fertilization could improve the soil's structure and increase the soil's capacity to store water and retain fertilizer. Finally, ecological restoration was performed in this waste residue pile by planting.

(2) South area

The first step was to clear vegetation and topsoil in the southern area. And then artificial composite materials were utilized to create an impermeable surface, to block the course of transmission and to limit environmental concerns. Through soil fertilization, the capacity of soil to store water and retain nutrients could be improved. Lastly, vegetation was planted in the waste residue pile area to restore the ecological environment of the pile area.

(3) Acidic wastewater treatment

Acidic wastewater treatment was performed by source control, process interception, and final treatment. The major method of source control was to install intercepting drains in the vicinity of the waste residue stockpile to collect acidic wastewater and lessen surface runoff. The major goal of process interception was to create ecological ditches and absorb acidic wastewater pollution by enhancing the current drainage channels with *in-situ* substrate and microbial bacterium. mobile sewage treatment technology was used in the final treatment to treat the produced acidic wastewater.

Conclusion

Due to the primitive mining techniques used in the early days, the majority of the sulfur and iron waste residue was piled up in a dispersed manner, taking up some of the available land resources. The waste residue produced acidic wastewater under the action of air oxidation, rainwater drenching, and surface runoff, polluting the surrounding soil and water environment. The acidic wastewater directly entered the river channel of Lanhe River through surface runoff and the reservoir of Tanghe River. The water quality of Tanghe Reservoir was affected. Furthermore, to varying degrees, the waste residue in the north area and southern areas of this study could pollute the soil and harm the nearby vegetation and biological landscape, which would pose a possible threat to the environment.

Controlling the migration of heavy metals to the outside world and reducing the heavy metal pollution in the mining area of the watershed through remediation would significantly enhance the ecological environment of the region and safeguard the health of downstream inhabitants. The local economy, community, and environment might all develop sustainably at the same time. The management precautions and strategies suggested in this study could serve as technical guidelines for projects of a similar nature. The management strategies put forward in this paper could serve as technical references for projects of a similar nature.

Conflict of Interest

The authors declare no conflicts of interest.

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