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Vertical Distribution of Cyanide and Heavy Metals in a Tailings Pond in Jilin, China

Zhehao Li [1,†], Qianfei Zhao [2,†], Hongying Yang [2,*], Qiang Liu [1,*], Yu Zhang [1], Ying Wang [1] and Linlin Tong [2]

[1] Changchun Gold Research Institute Co., Ltd., No. 6760 Nanhu Road, Changchun 130012, China
[2] School of Metallurgy, Northeastern University, Shenyang 110819, China
* Correspondence: yanghy@smm.neu.edu.cn (H.Y.); liuqiang27@163.com (Q.L.); Tel.: +86-13889803669 (H.Y.); +86-13624315723 (Q.L.)
† The first two authors contributed equally to this paper.

Abstract: After long-term accumulation, weathering and rainfall of metals and non-metals in tailings ponds, the chemical composition will change, and the surface heavy metals may gradually migrate to the bottom layer, thus showing a cumulative effect. At present, the distribution of cyanide and heavy metals in vertical space has not been reported. In this work, 12 sampling points were arranged for a cyanide tailings pond by the grid method in Jilin, China. The contents of cyanide and heavy metals were determined according to the standard method. The results show that the heavy metals in the tailings were mainly Cu, Pb, Zn and Mn (342.83 mg/kg, 571.09 mg/kg, 610.15 mg/kg and 796.63 mg/kg, respectively), and the concentration of heavy metals in the horizontal direction does not change significantly. It should be noted that the concentrations of zinc and manganese did not change much in the vertical direction. The concentration of copper and lead increased with the increase in depth. The concentration of cyanide at the sampling site far away from human activities was higher at 6.71 mg/kg, and the average concentration is 2.5 mg/kg. In addition, because the cyanide is unstable and affected by factors such as light and rainfall, the concentration change in the vertical direction fluctuates.

Keywords: cyanide; heavy metals; distribution; cyanide tailings pond

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1. Introduction

Gold ore mining produces a large amount of waste rock, and the amount of cyanide gold extraction tailings is similar to that of daily ore treatment. Compared with ferrous metal and non-ferrous metal mining, gold ore mining is a production process that produces a larger proportion of solid waste [1,2]. The cyanide-bearing tailings produced in the gold cyanidation process are generally stored in tailings ponds, which are major risk sources of environmental pollution. If not handled properly, the cyanide in them will cause serious pollution to the local water environment and soil.

The evaluation of potentially toxic elements pollution in tailings ponds has become a concern of many experts and scholars, who pay attention to the pollution degree of tailings ponds, the spatial distribution of pollution elements and the impact of pollution on the surrounding environment [3–5]. Zhang et al. [6] found that Cu, Zn, Cd and S were the major pollutants in an HTM tailings pond when they studied the vertical distribution of heavy metals in the abandoned tailings ponds of an HTM copper mine. In addition, the process of sulfide oxidation and heavy metal release has continued in the 20 years of tailings stockpiling. During their research on the feasibility of tailings as additives in the production of port cement, Celik et al. found that tailings need to be subject to two-stage chemical treatment in order to destroy free cyanide, stabilize and solidify heavy metals [5]. Zhang et al. [7] conducted spatial distribution and risk assessment of pollutants in a gold mine tailings pond in Pinggu, China, and pointed out that cyanide and potentially toxic elements in cyanide tailings would cause harm to the surrounding environment through rainwater infiltration and tailings seepage. In order to reduce the impact of tailings ponds on the environment, it is necessary to take appropriate treatment measures, which must be based on the properties of tailings, heavy metal content and spatial distribution characteristics of early research.

The Jiapigou area in Jilin, China, is rich in gold resources. In order to meet the production needs of enterprises, tailings ponds were built and put into use as early as the 1970s. The characteristics of tailings are related to the mining method and the characteristics of the original deposit [8,9]. As a result, the tailings from Jiapigou are mixed with arsenic, cyanide and other toxic heavy metals [10]. The tailings ponds in this area adopt the open-air stockpiling method. A large number of cyanide residues are piled on the ground and exposed to the environment. Cyanide is a highly toxic substance, which can volatilize under natural light to form hydrogen cyanide, especially under acidic conditions, thus causing acute or chronic damage to residents and livestock [11,12]. Pollutants in tailings can also affect the surrounding ecological environment through evaporation, rainfall seepage, tailings pond leakage and dust deposition [7]. Although the establishment of tailings ponds has improved the ecological environment, the pollution caused by tailings ponds is still a problem worthy of attention.

Therefore, the investigation and evaluation of cyanide and heavy metals in the tailing pond of a gold mine in Jiapigou can deeply understand the pollution status of the surrounding environment, which plays an important role in the treatment and ecological restoration of the tailing pond. In this paper, the spatial vertical distribution of cyanide and four heavy metals, including Cu, Pb, Zn and Mn, in the Jiapigou cyanide tailings pond, Jilin, China, were studied to determine the concentration and distribution of cyanide and heavy metals, so as to provide a scientific basis for the subsequent comprehensive treatment of tailings ponds.

2. Materials and Methods

2.1. Study Area

The cyanide tailings in the tailings pond come from the full slime cyanide process, and the cyanide concentration of the cyanide slag is 50 mg/kg. The total storage capacity is about 331.95×10^4 m^3 and the effective storage capacity is about 256.11×10^4 m^3. The tailings pond has been built and put into use since 2005. The tailings pond is located northeast of Jiapigou district, Jilin, China, which is an open-basin geological structure (Figure 1). The study area belongs to the climate of the continental cold forest mountain area in the north cold temperate zone, with an annual average temperature of 4.8 °C and rainfall of 748.4 mm [13]. The catchment area of the tailings pond is 2.38 km^2, the maximum channel length is 2.15 km and the average slope is 0.137%. The on-site exploration and analysis showed that the gold content in the tailings was higher, with an average grade of 0.83 g/t. The morphology and sample map of sampling sites in the tailings pond are shown in Figure 1.

2.2. Sample Collection and Chemical Analyses

According to the principle of systematic distribution in HJ/T 20-1998, the rectangular area in Figure 1 was selected as the distribution area, with 12 sampling points and a distance of 12 m between each point. According to the slope of the bottom of the tailings pond, the sampling depth of Points 1, 6, 7 and 8 is 30 m, the sampling depth of Points 2, 3, 4 and 5 is 25 m, and the sampling depth of Points 9, 10, 11 and 12 is 20 m. The specific layout is shown in Figure 1. The vertical samples were collected at an interval of one meter, and the temperature and humidity of samples were measured on site (Figure 2). A total of 300 tailings samples were collected.

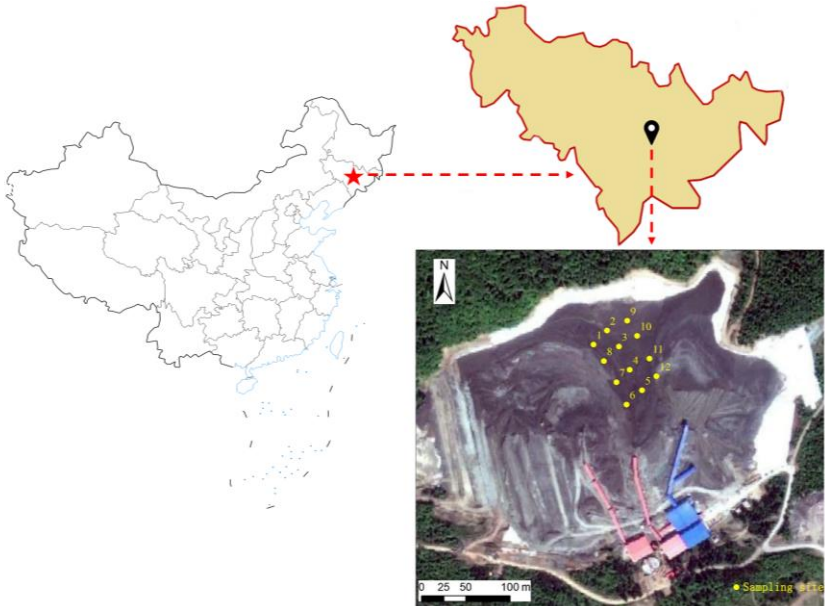

Figure 1. Location map of tailings pond and layout of sampling points.

Figure 2. Morphology and sample map of sampling sites in tailings pond.

The collected samples were transferred to a polythene bag, stored in triplicate in a portable cryostat and immediately transported to the laboratory for screening to remove large particles such as debris, stones and pebbles. The weight of each sample is 10 g, sealed at 4 °C for cyanide content analysis, and the storage time should not exceed 24 h. The remaining samples were ground and crushed with a particle size <75 μm after air-drying in the room and used for the determination of heavy metal content after digestion.

Mineralogical analysis of the samples was obtained from the average values of all samples, including the initial cyanide concentration and the mass fraction of each element. In addition, the uniformly mixed samples from each region were prepared for particle size distribution (BT−9300H, Bettersize, Dandong, China). X-ray diffraction (XRD, D 8, Saarbrucken, Germany) and microscopic (LEICA−DMLP, Wetzlar, Germany) observation, in order to analyze the main phase composition in the cyanide slag.

2.3. Sample and Data Analysis

The sample to be tested was mixed with deionized water in accordance with 1:2.5 (v/v), and phosphoric acid was added to ensure that the pH of the pulp was less than 2. Then, it was placed in an electric heating jacket and heated for distillation at 150 °C. Bivalent tin and bivalent copper were used to inhibit the interference of sulfide and catalyze the decomposition of complex cyanide. Finally, the concentration of cyanide was determined by HJ 823-2017.

Determination of heavy metal concentrations was performed in accordance with standard procedures (ISO11047, 1998). A 1 g amount of the sample to be tested was placed in the vessel of Teflon digestion with 48 mL of a HNO_3 and HCl mixture (1:3 v/v), followed by digestion on a heating plate at 150 °C for 2 h. After digestion, the samples were cooled, filtered and diluted to 100 mL. Finally, the contents of Cu, Pb, Zn and Mn were determined by an inductively coupled plasma optical emission spectrometer (ICP-OES, Avio500, Waltham, MA, USA). For the analysis of heavy metal content in cyanide tailings, the accuracy of the analytical method was checked by using duplicates, blanks and standard reference materials (GBW07405, Institute of Geophysical and Molybdenum Exploration, Langfang, China).

The presentation of the study area used Google earth 7.1.2 (Google, Mountain View, CA, USA). The descriptive statistical software SPSS 24.0 (IBM, Chicago, IL, USA) was used for multivariate statistical analysis of cyanide and heavy metals in the tailings pond, including the maximum, minimum, mean, standard deviation (SD) and coefficient of variation (CV).

Ultra-pure water was used throughout the experiment. In order to ensure the accuracy of the analysis results, the test results of all samples in the experiment were the average of three parallel samples, and the standard deviation of the analytical data from duplicate samples was less than 5%.

3. Results and Discussion

3.1. Process Mineralogy of Tailings

3.1.1. Particle Size Distribution of Cyanide Tailings

The result of the particle size analysis is shown in Figure 3. The result shows that the cyanide tailings had a fine particle size, of which the particles less than 38 μm accounted for about 90%. This is because the gold concentrate undergoes a grinding process prior to cyanide leaching so that the minerals can be separated as much as possible to expose the wrapped gold, resulting in very fine particle size of the cyanide tailings. The smaller the particle size, the greater the harm to the human body [3]. In addition, the finer the mineral particles, the more likely the minerals are to have non-selective agglomeration, which increases the difficulty of the flotation separation of valuable elements, which is not conducive to the secondary recovery and utilization of tailings [14].

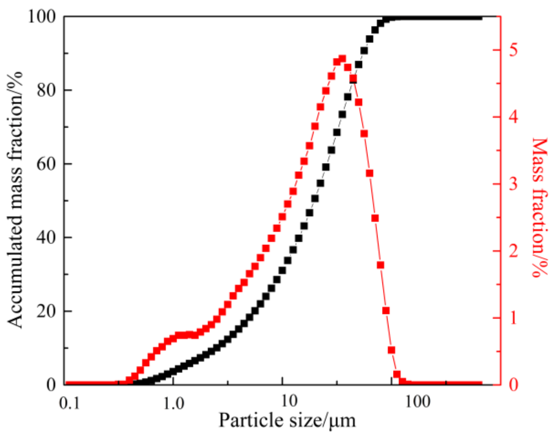

Figure 3. Particle size distribution of cyanide tailings.

3.1.2. Chemical Composition of Cyanide Tailings

The chemical composition analysis results of cyanide tailings are shown in Table 1. SiO_2 and Al_2O_3 accounted for the largest proportion, the mass fraction was 76.79% and 5.61%, respectively, indicating gangue was the main mineral. The next high content was Fe, with a mass fraction of 4.33%. The contents of Cu, Pb and Zn were more than 0.30%, which can be recycled. The contents of Au and Ag were 0.90 g/t and 19.29 g/t, respectively, but they also reached the standard of recycling due to their high value.

Table 1. Analysis results of chemical components of cyanide tailings.

Composition	Au *	Ag *	Cu	Pb	Zn	Mn	Fe	SiO_2	Al_2O_3
wt/%	0.90	19.29	0.34	0.36	0.35	0.19	4.33	76.79	5.61

* g/t.

3.1.3. Composition and Content of Minerals in Cyanide Tailings

Figure 4 shows the XRD pattern of cyanide tailings. It can be seen from the XRD pattern that the main minerals were quartz (SiO_2), pyrite (FeS_2), orthoclase ($KAlSi_3O_8$) and albite ($NaAlSi_3O_8$). The XRD pattern does not show other minerals with lower content. The XRD results show that gangue and pyrite were the main mineral compositions of cyanide tailings. This is because the cyanide tailings in the study area come from the whole slime cyanide tailings, which are directly produced after cyaniding gold concentrate by gold flotation.

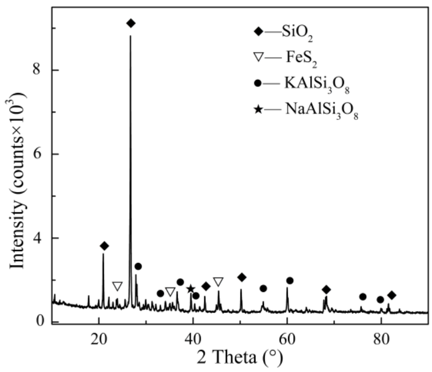

Figure 4. XRD pattern of cyanide tailings.

According to the statistical analysis of the metallographic microscope (Figure 5), sulfide is the main metal mineral in cyanide tailings, and the analysis results of each mineral component are shown in Table 2. The highest content was pyrite, with a content of 4.44%, followed by a small amount of chalcopyrite, sphalerite and galena, with a content of 1.28%. The non-metallic mineral was mainly quartz, with a content of 73.56%, followed by a small amount of silicate minerals, such as feldspar and mica, with a content of 11.98%. In addition, carbonate minerals, including calcite and ankerite, account for 8.37%.

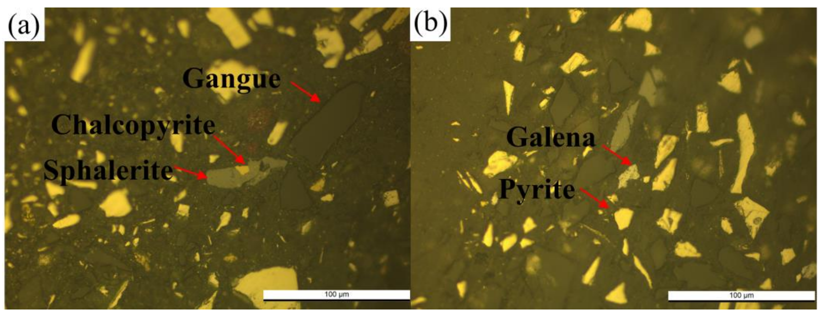

Figure 5. Occurrence state of main minerals in cyanide tailings (**a**) chalcopyrite and sphalerite intergrowth, (**b**) pyrite and galena monomer.

Table 2. Analysis results of mineral composition of cyanide tailings.

Minerals	Pyrite	Chalcopyrite	Sphalerite	Galena	Quartz	Silicate Minerals	Carbonate Minerals	Others
wt/%	4.44	0.97	0.01	0.30	73.56	11.98	8.37	0.47

3.2. Statistical Analysis of Cyanide and Heavy Metals in Tailings Pond

The descriptive statistical results of cyanide and Cu, Pb, Zn and Mn in the cyanide tailings are shown in Table 3 and Figure 6. The maximum and average values of cyanide were within the standard limits, which was far less than the maximum allowable stacking concentration of cyanide of 50 mg/kg. However, in the study area, the distribution of cyanide showed an uneven phenomenon. The reason for the above may be that during the storage of the cyanide tailings, the cyanide was migrated or degraded due to factors such as rainfall and sunlight exposure. The migration and transformation of cyanide during storage will be discussed later. In addition, the maximum toxicity of the potentially toxic elements (Mn, Cu, Pb and Zn) in the study area exceeded the standard limit, and the concentration distribution changed greatly. The coefficient of variation is the ratio of the standard deviation to the mean value, which can be used to express the degree of variation in the data of contaminated elements. It is not difficult to find that the coefficient of variation of Pb and Zn in the study area was high, and the coefficient of variation of Pb was more than 100%. These results show that the distribution of these pollutants in the tailings pond is very uneven, which is bound to affect the selection of the secondary utilization process of cyanide tailings. Therefore, it is necessary to find out the distribution of each metal in the tailings pond. In addition, the distribution of metals and non-metals and the properties of chemical components in the tailings pond will be affected by different tailings ponds due to the environment, topography, ores and deposition time, which are the key to evaluating the secondary resource utilization of tailings ponds.

Table 3. Descriptive statistics of cyanide and heavy metal concentration in tailings (mg/kg).

	N [1]	Max	Min	Mean	S.D	CV (%)
Cyanide	300	19.20	0.11	2.50	2.88	115.2
Copper	300	824.00	88.00	342.83	114.99	33.54
Lead	300	2360.00	76.90	571.09	581.75	101.87
Zinc	300	2620.00	68.30	610.15	568.29	93.14
Manganese	300	1430.00	663.00	796.63	66.61	8.36

[1] N is the total number of samples.

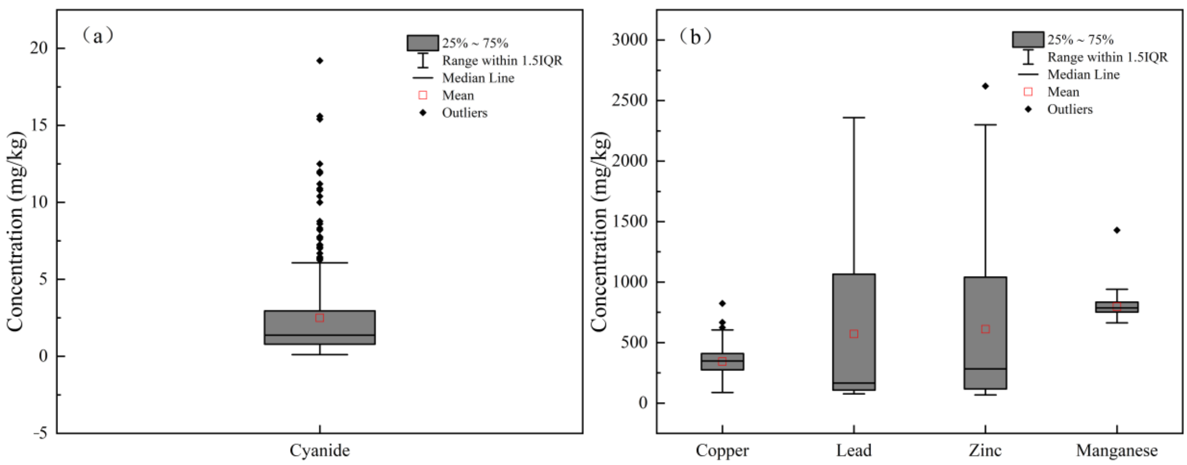

Figure 6. Box diagram of cyanide and heavy metal content analysis in tailings (**a**) cyanide, (**b**) metal ions.

3.3. Horizontal Distribution of Cyanide and Heavy Metals in Tailings Pond

The average distribution of cyanide and heavy metal content at each sampling point is shown in Figure 7. As can be seen from Figure 7a, the distribution of cyanide concentration in the tailings pond is quite different. However, due to the long accumulation time, rainfall, percolation and natural degradation, the cyanide content in the tailings was much lower than that in the fresh slag (1300 ppm), which was distributed below 10 mg/kg. The highest cyanide concentration was 6.71 mg/kg, which appeared in an area far away from human activities (Sampling Point 1), and the lowest cyanide concentration at Sampling Point 6 was 0.88 mg/kg.

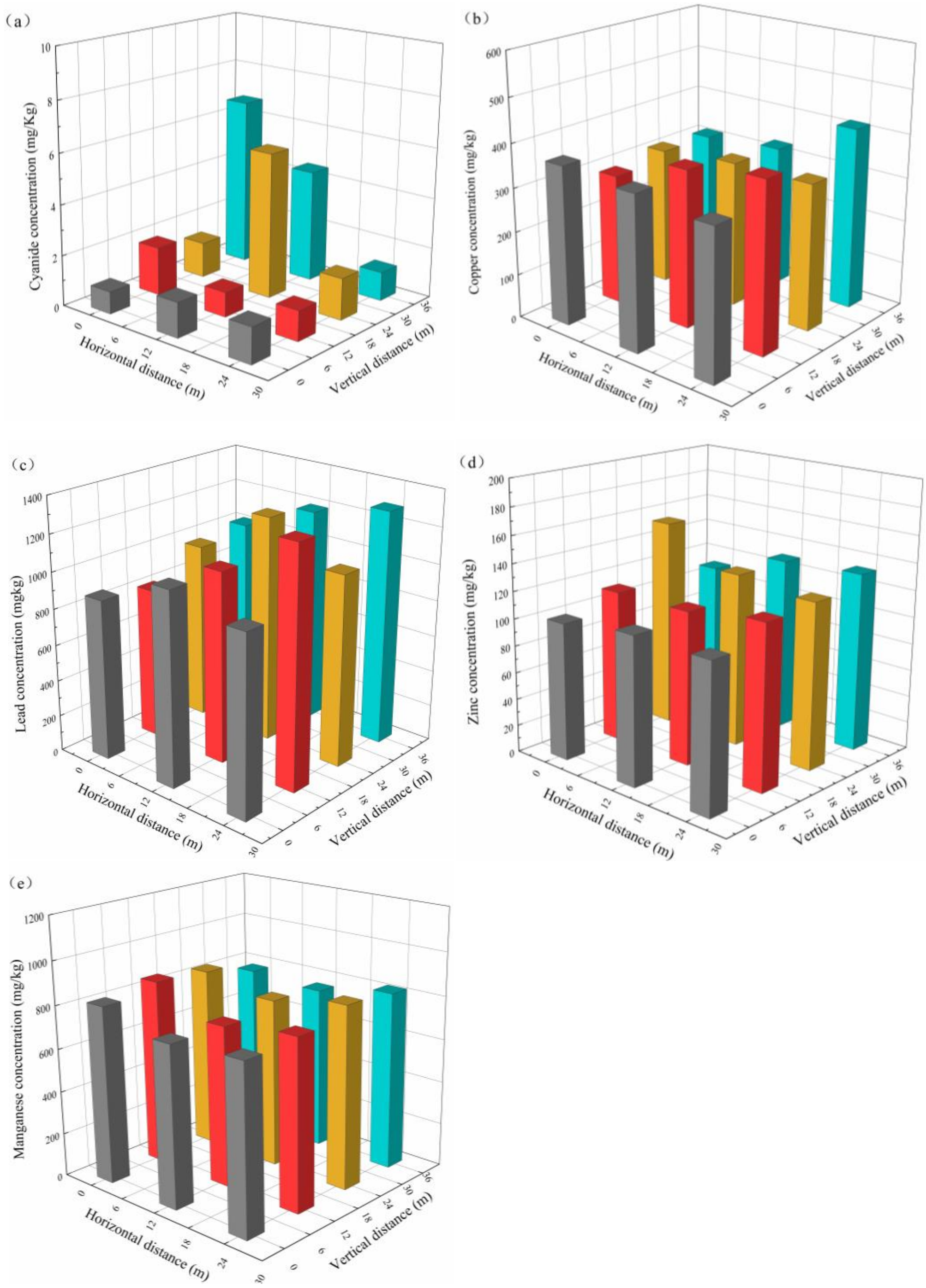

Figure 7. Distribution diagram of cyanide and heavy metal concentration in horizontal direction in tailings pond (**a**) cyanide, (**b**) Cu, (**c**) Pb, (**d**) Zn, (**e**) Mn.

It can be seen in Figure 7b–e that the concentration of heavy metals in the tailings sample was Pb > Mn > Cu > Zn. The highest concentration of Pb was 1311.25 mg/kg at Point 3. Among them, the Pb concentration at Points 1, 2, 3, 4, 9, 10 and 11 was greater than 1000 mg kg. The highest concentration of Mn was 859.87 mg/kg at Point 7. In addition, it is found in Figure 7e that the Mn content of each point varies little, with the concentration ranging from 740 to 860 mg/kg. The maximum concentrations of Cu and Zn were 416.3mg/kg and 155.07 mg/kg at Points 9 and 7, respectively. The change in Cu, Pb, Zn and Mn contents shows that the distribution of four heavy metals in the tailings pond are basically the same in the horizontal direction. The percolation performance of tailings is better, which can be verified by the water washing and leaching test of tailing slag [15].

3.4. Vertical Distribution of Cyanide and Heavy Metals in Tailings Pond

The vertical average concentration of cyanide and four heavy metals in the study area of the tailings pond are shown in Figure 8. The cyanide content of samples fluctuated irregularly with the depth. The highest value appeared at a depth of 30 m with a content of 4.72 mg/kg, and the lowest value appeared at a depth of 13 m with a content of 1.52 mg/kg. From the overall distribution trend of cyanide, the content of cyanide was distributed uniformly along the depth direction. As can be seen from Figure 8b, the contents of heavy metals Zn and Mn in the tailings did not change significantly along the direction of depth and showed a uniform distribution, while the concentrations of Cu and Pb peaked at the depth of 10 m with concentrations of 424.33 mg/kg and 1440.58 mg/kg, respectively. At the same time, the concentrations of Cu and Pb both showed a trend of decreasing with the depth increasing until the content became stable. The reason for the above phenomenon may be that in the inactive tailing pond, the tailing profile often shows the segmentation of the oxidation zone, cementation zone and non-oxidation zone with the increase in depth [6,16]. On the surface of the tailings pond, most of the sulfide is oxidized by the air and water, and then the metal concentration in the cementation zone increases, indicating that the metal released from the surface tailings moves downward and accumulates.

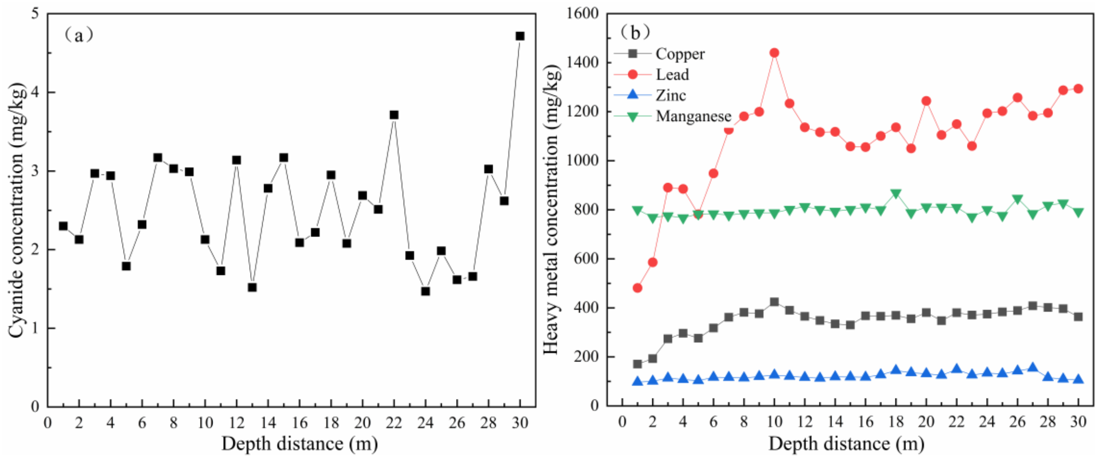

Figure 8. Vertical distribution of (**a**) cyanide and (**b**) heavy metal concentration in tailings pond.

According to the site conditions, sampling inspection and analysis, etc., the spatial distribution diagram of cyanide content was made, as shown in Figure 9. It can be seen that the cyanide content in tailings is not high in general, most of which was less than 6 mg/kg, and only a few were close to 20 mg/kg. As can be seen from Figure 9a, the cyanide content on the northwest side of the tailings pond was higher than that on the southeast side. The representative section 5-4-3-2 was selected, and it was found that with the increase in the depth from the surface, the cyanide content first increased and then decreased, especially at Point 3. The reasons for the above situation are that because the cyanide tailings were divided into regions and layers, the initial content of cyanide in the tailings was different. In addition, due to the different conditions of the tailings accumulation time and the degree of compaction, the natural degradation degree and rate of cyanide in tailings will be different [17]. At the same time, affected by the topography of the tailings pond and different surface infiltration conditions, cyanide in tailings migrated from the shallow surface to the deep and from the high to the low after precipitation and rainfall.

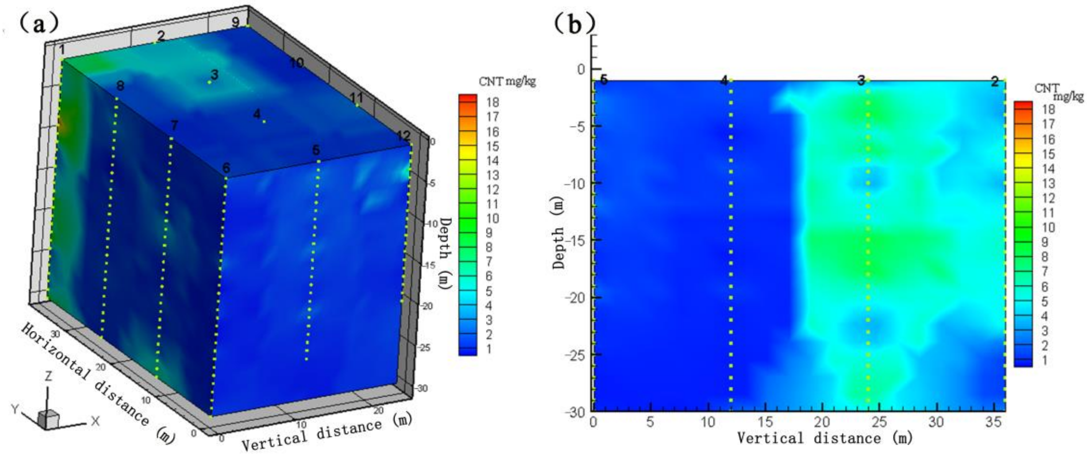

Figure 9. Profile of cyanide concentration distribution in (**a**) vertical and (**b**) horizontal direction in tailings pond.

4. Conclusions

The application of tools, methods and detection analysis revealed the distribution characteristics of cyanide and heavy metals in a cyanide tailings pond in Jilin, China. The results show that the cyanide concentration was basically kept below 10 mg/kg, and the maximum concentration value appeared in the northwest direction, which is far away from the human activity area. In addition, the accumulation of different batches of tailings and the topography of tailings ponds are the reasons for the fluctuation of cyanide concentration in the vertical direction. The oxidation of sulfide on the surface of tailings results in little change in heavy metal concentration in the horizontal direction, but with the increase in depth, some metals will accumulate in the cemented layer. The maximum concentration of heavy metals Cu and Pb appeared at the depth of 10 m, and then the concentration gradually decreased until stable. However, the concentration distribution of Zn and Mn did not change significantly with the depth. Through the process mineralogy study, it is found that the Cu, Pb and Zn in the tailings mainly exist in the form of chalcopyrite, galena and sphalerite, but the proportion of sphalerite was only 0.01%, which does not have the value of secondary utilization. In addition, the topographic and climatic conditions of the tailings pond will affect the oxidation rate of metals and non-metals, thus affecting the existence form of minerals, which is a test for the flotation recovery of chalcopyrite and galena with 0.97% and 0.3% of the proportion. In this study area, heavy metals were gradually depleted in the oxidation zone and enriched in the deeper cementation zone. Therefore, it is necessary to adjust the process appropriately according to the different ores and deposition times when the cyanide tailings are utilized as secondary resources.

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