

# Non-Destructive Detection and Evaluation of Heavy Metal Pollution in Tailings Reservoir

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## Abstract

Heavy metal pollution is a negative effect generated in the process of utilizing non-ferrous mineral. Studies about heavy metal migration detection are very important. A new method for rapid detection of heavy metal migration based on ground penetrating radar (GPR) was provided. Comparative tests were studied from field to lab with GPR and X-ray fluorescence analysis (XRF). A tailings reservoir in the Xiangjiang River basin at Hunan Province was taken as experimental site. The downward transfer rule of heavy metal migration was confirmed through tests on systematically arranged survey lines and sampling points in tailings site. Results showed: 1) Through GPR image recognition, tailings reservoir had 3 layers. Reclaimed soil layer (the first layer) and tailings layer (the second layer) had a clear interface. However, tailings layer (the second layer) and subsoil layer (the third layer) had an obscure interface on radar images. It was concluded that heavy metal component had migrated downwards. 2) Chemical component analysis verified image recognition conclusions. Concentrations of As, Cd and Pb were significantly out of limit, while concentration of Cr was under limit according to analysis results on samples from different depths. 3) Pollution degree was evaluated. Downward migration was the main form of heavy metal migration in tailings site, upward migration occurred through adsorption at the same time.

## Keywords

Tailings Site, Heavy Metal Migration, Comparative Tests, Ground Penetrating Rada

## 1. Introduction

Metal element which has specific gravity over 5 is generally called as heavy metal. Heavy metal pollution is defined as environmental problems caused by heavy

metals and heavy metal compounds. It is mainly generated from man-made factors, such as extraction of minerals, irrigation with industrial waste water, discharge of waste gas and so on. In the field of heavy metal pollution research, it is mainly focused on heavy metals of strong toxicity, such as cadmium, chromium, plumbum, arsenic, mercury. With rapid economic growth, large amounts of resources had been consumed and serious problems of environmental pollution had been caused at the same time in China. In May 2013, the Guangzhou Food and Drug Administration sampling results showed that, for rice and rice products samplings, 8 out of 18 batches (as high as 44.4%) were found excessive levels of cadmium. In July 2013, it was reported that nearly 30% soils were contaminated with heavy metals at the Pearl River in southern China by media. Heavy metal pollution made a great contribution to endemic disease in contaminated areas. For example, from 1930s to 1970s, the residents suffered from a cadmium intoxication disease named itai-itai disease in the Toyama Prefecture, Japan. Cadmium intoxication is derived from eating local rice irrigated by wastewater of smelting plants. In 1956, Residents suffered from Minamata disease (methyl mercury intoxication) through eating contaminated fishes and shellfishes in Kumamoto Prefecture of Japan.

In mining engineering, the tailings were stacked in the tailings reservoir. They were important sources of heavy metal pollution. Hunan Province, which is called "Land of China's non-ferrous metals", is seriously contaminated area by heavy metals. A plenty number of tailings reservoirs have been built in Hunan. In order to prevent the pollution, the research on the mechanic and detection of heavy metals migration is very important [1] [2]. It is not only the foundation of evaluation on surrounding soil, water and other environmental factors in tailing site, but also an important basis for taking pollution control measures. Currently, research have been mainly focused on the heavy metal migration in soil environment [3]-[5], garbage [6], plants and animals [7]-[9].

Non-destructive detection technology (NDT) is one of detection technologies by using sound (light, magnetism, electricity or other features) to detect whether the target body exists in the test object. On the other hand, the target body properties including the size, location, nature and quantity information are given without destruction of the test object. Ground penetrating radar (GPR) is non-destructive detection device to detect subsurface information using electromagnetic wave. When electromagnetic wave travels through subsurface medium, its travel path, field strength and waveform will differ with medium changes and geometry varies. It has been widely applied in engineering geological investigation and monitoring [10]-[12], engineering quality testing [13]-[16] and soil contamination detection [17]-[23] and other fields.

It is a novel focus of research to achieve rapid evaluation and analysis on heavy metal migration. Based on general principles of non-destructive detection, a case test was studied. A new method for rapid non-destructive detection of heavy metals migration was provided, and its reliability was proved by lab analysis of samples.

## 2. Project Overview

### 2.1. Introduction of Experimental Site

Xiangjiang River undertook 60% population and 70% GDP of Hunan Province. It suffered from over 60% pollutants of Hunan. It was the most seriously polluted river by heavy metals in China. Dongjiang Lake is one of the water sources of Xiangjiang River. Therefore, pollution status of Dongjiang Lake area is very important to the heavy metal treatment of Xiangjiang River.

An abandoned tailings reservoir was taken as the research object. It is located in Qingjiang town, Zixing County, Hunan Province. It was built in 1980s and closed in 2008. Due to the lack of design documents and standard regulation, parameters of this tailings site were inadequate. Based on site survey, it was found that tailings had been discharged directly on hillside. Tailing dam was constructed with waste rocks. The thickness of accumulated tailings was approximately 3 m. On the east of this tailings site, a path led to X023 county road. A river beneath the feet of tailing dam flowed into Dongjiang Lake. Minimum distance between the tailings dam and Dongjiang Lake was less than 2 kilometers, as shown in **Figure 1**.



**Figure 1.** Surrounding environment of the tailings site.

### 2.2. Principle of Non-Destructive Detection

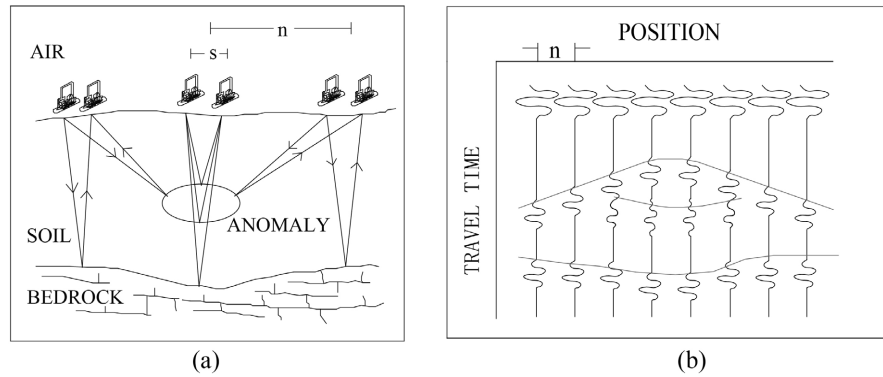
Ground penetrating radar (GPR) is a high-frequency electromagnetic technique which is used in geophysical explorations. It involves the emission of an electromagnetic wave that travels through the medium. The signal is reflected by sharp changes in the electromagnetic properties of the materials, which is recorded by the radar system. The travel time, frequencies and amplitude characters of the GPR wave are recorded.

Relationship of travel time, depth of reflection and wave velocity is shown in Equation (1).

$$t = 2H/v \quad (1)$$

In the formula,  $h$  represents distance,  $m$ ;  $v$  represents velocity.

The most common mode of GPR operation is single-fold, fix-offset reflection profiling as illustrated in **Figure 2(a)**. This mode of operation gives rise to data such as shown conceptually in **Figure 2(b)**.



**Figure 2.** (a) Schematic illustration of common-offset single-fold profiling. (b) Format of GPR reflection section with radar events shown for features as depicted in (a).

### 2.3. Arrangement of Survey Lines

Six survey lines were arranged on reclaimed tailings site considering site conditions (**Figure 3**). Five of those lines (from line No. 1 to No.5) were set as X direction, the other one (line No.6) was set as Y direction. Respectively, the length of survey lines from No.1 to No.6 was 85 m, 94 m, 88 m, 66 m, 48 m, and 130 m.

Reclaimed tailings site could be divided into three layers (reclaimed soil layer, tailings layer, and subsoil layer) according to ingredient. It's known that the deepest depth of tailings layer was only about 3 m. Antennas frequency of 100 MHz were selected for the testing in this situation.



**Figure 3.** Design of GPR detection lines.

## 3. Non-Destructive Testing

### 3.1. Parameters of GPR

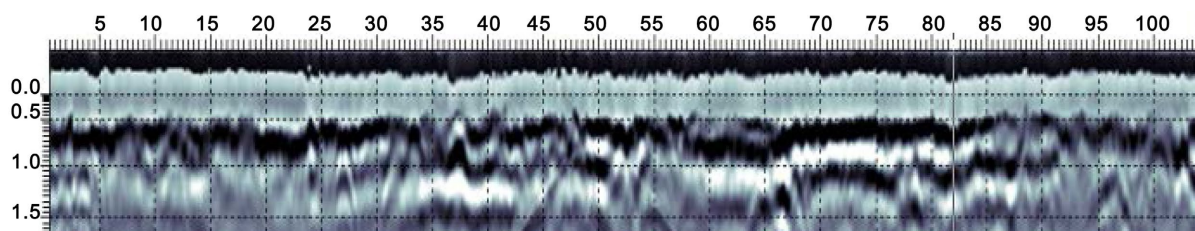
Considering the estimated depth (3 m) of tailings layer, GPR antennas of 100 MHz

were selected, and time window was set as 335 ns. During the data processing, electromagnetic wave velocity in wet sand was set as reference velocity. It presented subsurface information in approximately 10 meters.

## 3.2. GPR Images Recognition

### 3.2.1. Interfaces

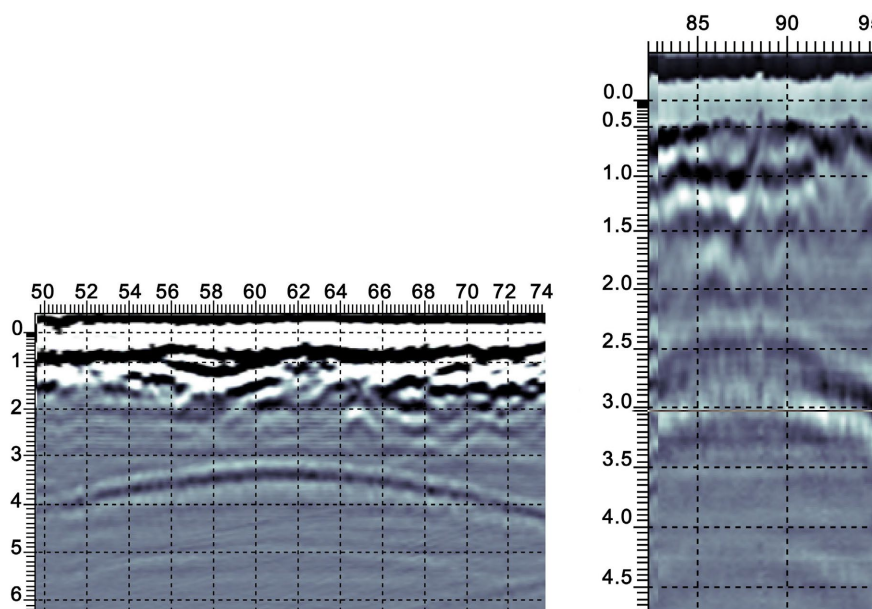
There was quite clear interface between reclaimed soil and tailings layer showed in each survey line's result. There were little changes of thickness in reclaimed soil. Thickness ranged from 0.6 m to 1 m (seen in **Figure 4**).



**Figure 4.** Results from surface to 1.7 m depth of survey line 6.

### 3.2.2. Anomalies

There was an anomaly detected (shown in **Figure 5**) by radar near the intersection of line 1 (60 - 65 m) and line 6 (85 - 90 m). The anomaly was about 3 meters deep. It was speculated to be a geometrical shape of tubiform based on the form of reflection. It was estimated to be pipe lines installed before.



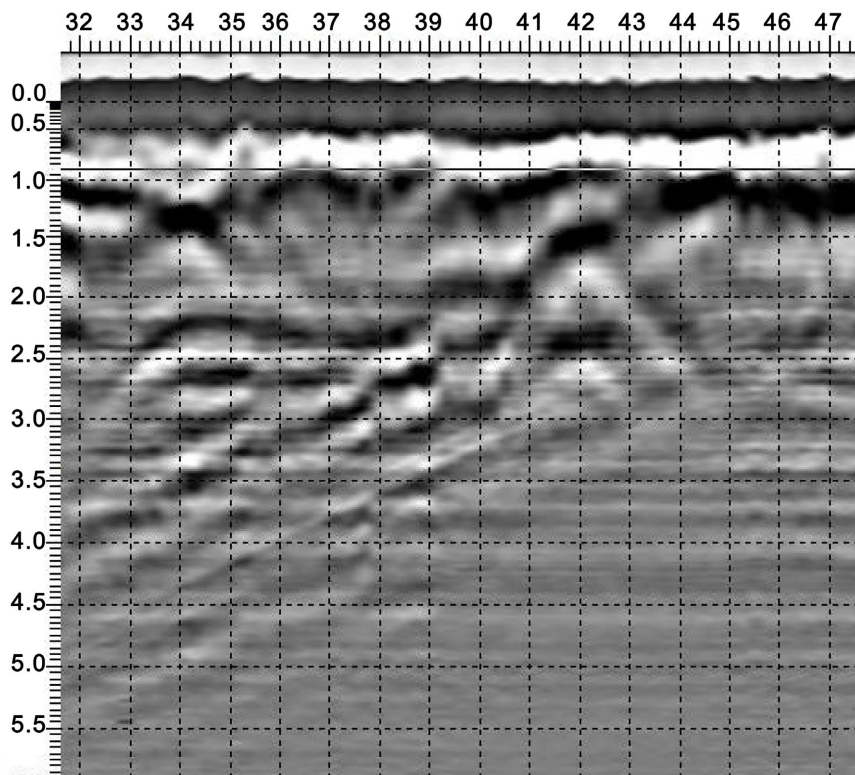
**Figure 5.** Anomaly detected in the intersection of line 1 (above) and line 6 (below) at depth about 3 m.

It was found that uneven distribution of a large number of scattered small reflector appeared at depths range from 0.5 m to 1.5 m in each survey line. Because

tailings dam was constructed with backfill materials, they were speculated to be reflection of that structure. such as gravels, bricks and other wastes. Survey line 5 was closest to the filling dam, so it was found of the largest number of that scattered small reflections. As shown in **Figure 5**, there was an inclined reflection presented in survey line 5. It could be caused by the structure of tailings dam as well.

### 3.2.3. Interference

When GPR was working near the wire, it was interfered. Significant signal disturbance appeared in the results of three survey lines. After excluding the interference of wires, there were rarely abnormal reflections received under the depth of 3 m, as shown in **Figure 6**. Therefore, it was concluded that there was rarely interface of two different medium in terms of electromagnetic properties. In other words, it could be speculated that subsoil had been mixed up with tailings. Downward migration of heavy metals occurred in this tailings site.



**Figure 6.** Scattered small reflectors and inclined reflection presented in survey line 5.

## 4. Chemical Composition Analysis

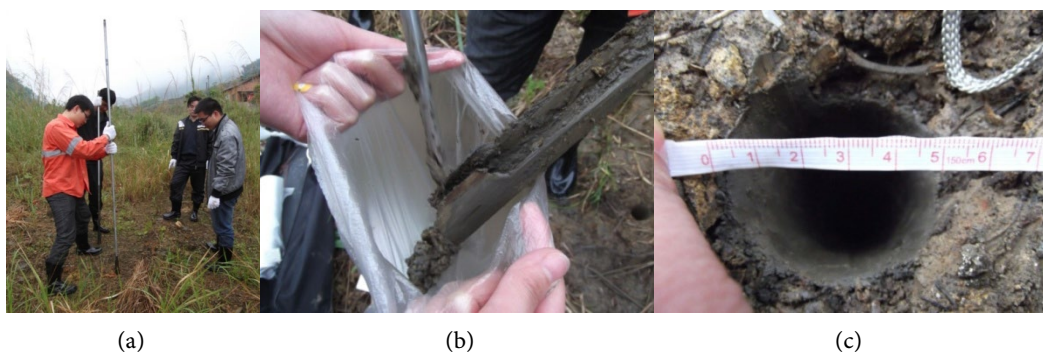
### 4.1. Analyze Method

In order to verify the results of GPR detection, two points along survey lines were selected as the sampling holes (shown in **Figure 7**). Samples were analyzed by X-ray fluorescence method to determine chemical composition. The depths and numbers of each samples were shown in **Table 1**. National standard GB15618-1995 of China was set as reference to determine contamination degree in all

samples.

**Table 1.** Statistical of samples.

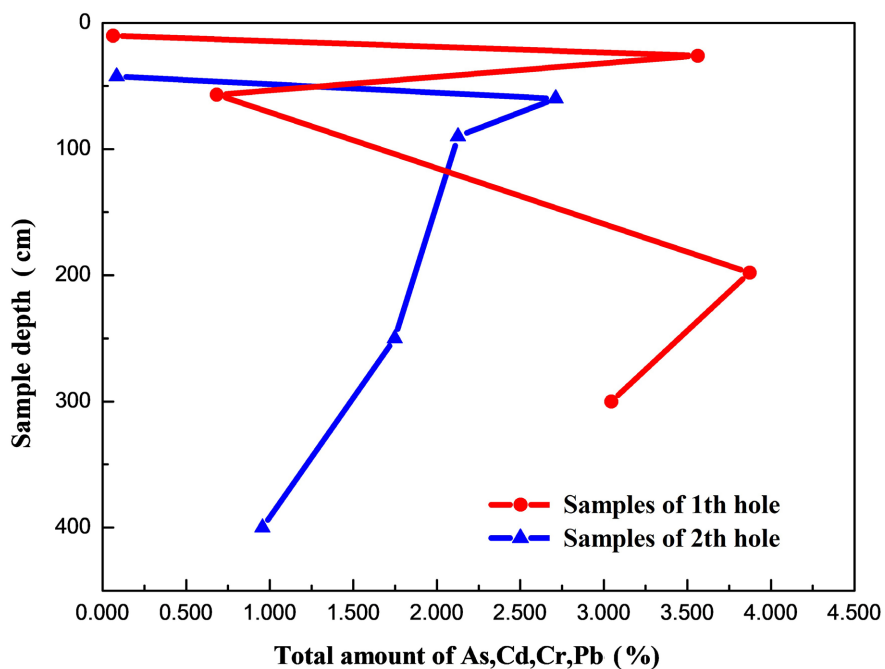
Position of sample	Near surface	Subsurface reclaimed soil	Interface	Tailings layer	Deepest sample
Number	1-1	1-2	1-3	1-4	1-5
Depth (cm)	10	26	57	198	300
Number	2-1	2-2	2-3	2-4	2-5
Depth (cm)	42.5	60	90	250	400



**Figure 7.** Sampling with luoyang shovel (a); sample collection (b); dimension of sampling holes (c).

## 4.2. Chemical Analysis

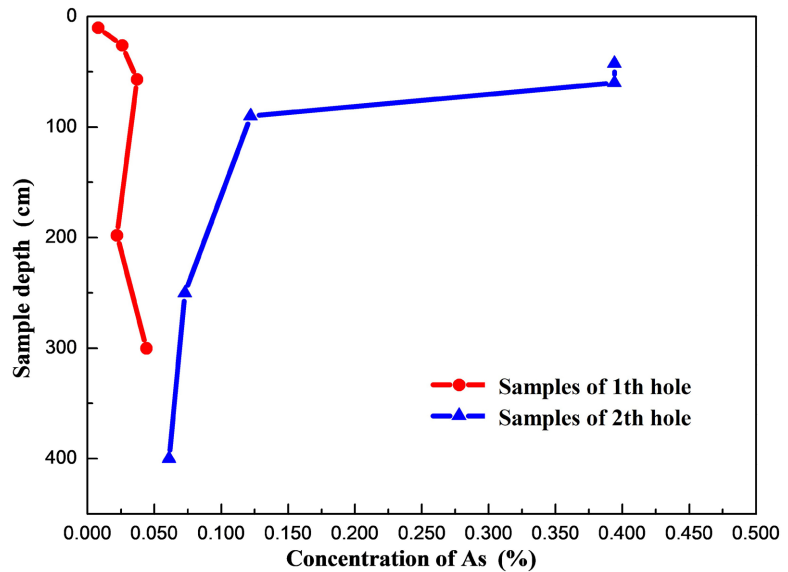
Concentrations of four strong toxic heavy metals (As, Cd, Cr, Pb) were statistically analyzed. Analysis results indicated:



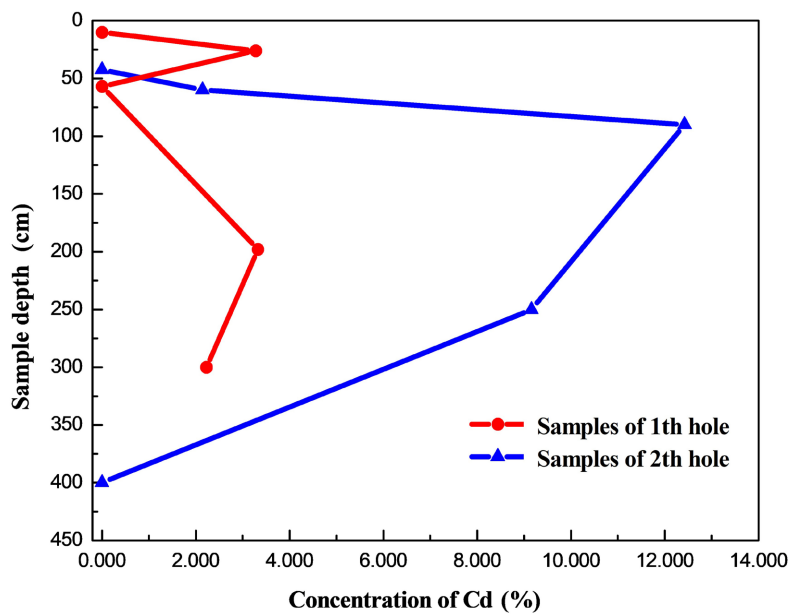
**Figure 8.** Total amount of As, Cd, Cr, Pb vs Sample depth.

1) Total amount: The total amount of the four elements was illustrated in **Figure 8**. It could be found that there were also heavy metals in surface soil, and the concentration were the lowest, but peak value of heavy metals concentration was not found in deepest samples.

2) As: Arsenic was found in both sampling holes (trivalent arsenic). Concentration of arsenic in all the samples was found exceeded class III soil standard specified in GB15618-1995. The highest concentration of As in samples was 98.5 times limit value of dry land (**Figure 9**).

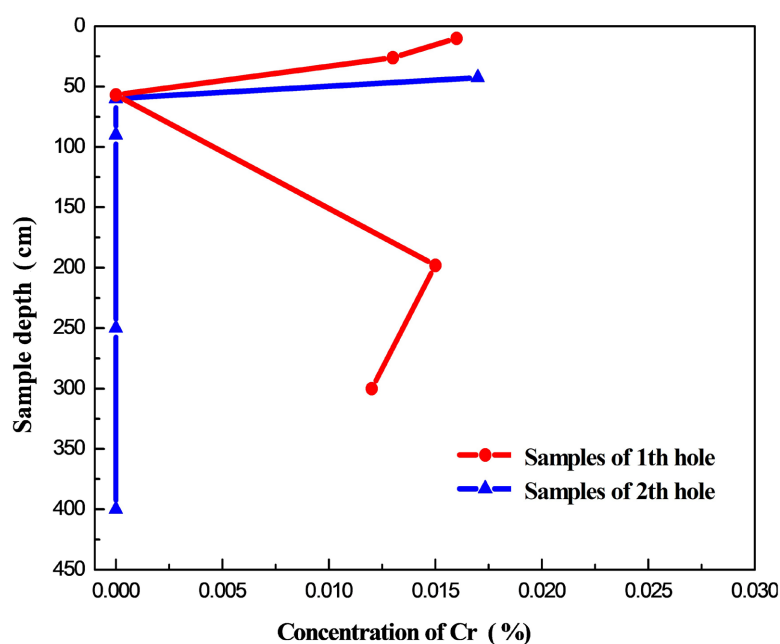


**Figure 9.** Concentration of As vs Sample depth. Critical concentration of As specified in class III national soil quality standard is 0.003% for paddyfield, 0.004% for dryland.

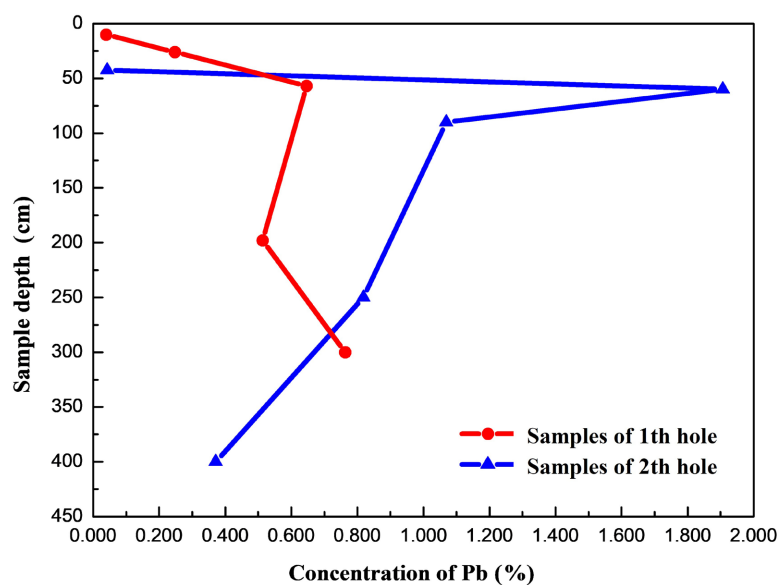


**Figure 10.** Concentration of Cd vs Sample depth. Critical concentration of Cd specified in class III national soil quality standard is 0.0001%.

3) Cd: There was no cadmium found in four samples. The number was 1-1, 1-3, 2-1, 2-5, respectively. Cadmium concentrations of remaining samples exceeded the standard value. Highest value of cadmium concentrations was 21,000 times standard value. Cadmium in soil usually existed in the form of cadmium carbonate, it was difficult to be dissolved and migrated with water flow. Therefore, it was not evenly distributed in the soil. It was indicated that distribution of Cd was random in the contaminated soil area (Figure 10).



**Figure 11.** Concentration of Cr vs Sample depth. Critical concentration of As specified in class III national soil quality standard is 0.04% for paddyfield, 0.03% for dryland.



**Figure 12.** Concentration of Pb vs Sample depth. Critical concentration of Pb specified in class III national soil quality standard is 0.05%.

4) Cr: Concentration of Cr in all samples was lower than level specified in national standard, Cr component was distributed in the soil randomly, Owing to the insolubility of chromium oxide. There was no Cr found in some samples (Number 1-3, 2-2, 2-3, 2-4, 2-5 showed in **Figure 11**).

5) Pb: It was found in all samples. Concentration of Pb was not substandard in surface soil samples. However, Concentration of Pb in other samples was all exceeded to standard (**Figure 12**).

### 4.3. Pollution Degree

The concept of pollution degree (W) was introduced to reflect soil pollution effectively caused by heavy metals. Critical concentration of national soil quality standard is shown in **Table 2**.

**Table 2.** Critical concentration of national soil quality standard.

Heavy metal elements	Critical Concentration
As	0.003%
Cd	0.0001%
Cr	0.04%
Pb	0.05%

$$W = \left( \frac{\text{Concentration of test data}}{\text{Critical concentration}} \right) - 1 \quad (2)$$

On the basis of calculation results, pollution degree of soil was divided into different ranks.  $W < 0$  (degree I): no pollution,  $0 < W < 1$  (degree II): light pollution,  $1 < W < 2$  (degree III): moderate pollution,  $W > 2$  (degree IV): heavy pollution. After the calculation, **Table 3** below was got:

**Table 3.** Pollution degree of two sampling holes.

Number	Depth (cm)	As	Cd	Cr	Pb
1-1	10	II	—	I	I
1-2	26	IV	IV	I	IV
1-3	57	IV	—	—	IV
1-4	198	IV	IV	I	IV
1-5	300	IV	IV	I	IV
2-1	42.5	IV	—	I	I
2-2	60	IV	IV	—	IV
2-3	90	IV	IV	—	IV
2-4	250	IV	IV	—	IV
2-5	400	IV	—	—	IV

Based on the analysis and calculations above, in general, this land was heavily

polluted. The concentration of As, Cd and Pb in this area was much higher than normal.

## 5. Comparatively Analysis

It was showed that there were a lot of small anomalies in reclaimed soil. They were presumed to be waste rock backfills, bricks and so on. During the sampling process, it was failed to sample in some drill holes due to the presence of waste rock. Some sampling points had to be relocated. However, reflectors and abnormalities decreased gradually more than 1.5 m depth. It was indicated that medium in deeper layer was more uniform than medium of shallow subsurface. During sampling process, in fact, the gravels hadn't been found excess the depth of 1.5 m. GPR imaging results were verified by samplings as well.

The interface was clearly evident between reclaimed soil and tailings layer. But, there was no reflection signal made by the interface of primary soil and tailings discharged in the radar images. It's speculated that tailings layer and primary subsoil layer had been mixed up with each other, and downward migration of heavy metals had emerged at the same time. It could be found through chemical analysis that: Concentrations of Cd and As were found substandard in surface soil only, the concentration of every heavy metal was found exceeded to national soil standard of type III with the increasing depth. the concentration of heavy metal was much higher than the standard value under the tailings layer as well. Downward migration of heavy metals was found in this tailing site, and it was the dominating form of heavy metals migration. Upward migration occurred through adsorption at the same time.

## 6. Conclusions

1) GPR was not only a non-destructive detection device for geological information and structures, but also a non-destructive testing method for vertical migration of heavy metal pollution in tailings site. Through image recognition, it was found that tailings reservoir had 3 layers. Reclaimed soil layer (the first layer) and tailings layer (the second layer) had a clear interface. However, tailings layer (the second layer) and subsoil layer (the third layer) had an obscure interface on radar images. The downward migration was the main form of heavy metal migration in this area. Chemical component analysis of soil samples verified these conclusions.

2) In the tailings site, Cd, Cr, As and Pb were the main pollutants. Cd and Cr's concentration had a large range of variation. While As and Pb's concentration changed little in different samples.

3) Generally, soil in this tailings site was heavily polluted based on pollution degree analysis. The concentration of As, Cd and Pb in this area was seriously out of limit.

4) Heavy metal pollution had the ability to migrate vertically upwards and downwards. Downward migration was the main transfer form, and upward migration occurred meanwhile by adsorption.

## Foundation Item

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## Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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