Heavy metal pollution characteristics and health risk assessment of a high risk site in Jiangsu Province

Quanlong Zhu[a], Yu Song[a,*], Zhifeng Guo [b], Wenjie Xu [a], Shuling Zhang[a]

[a]Jiangsu environmental science and Technology Co., Ltd. Changzhou, China
[b]Shandong Provincial No.6 Exploration Institute of Geology and Mineral Resources, Weihai, China

*Corresponding author: 1321880701@qq.com

Abstract. Taking a high-risk legacy site (the closed electroplating enterprises) in Jiangsu as the research object, the single pollution index method and the comprehensive pollution index method were used to evaluate the soil heavy metal pollution status, and the pollution of heavy metal Ni in the site was clarified. The spatial distribution of Ni in soil was discussed according to Kriging interpolation method, and the health risk assessment of heavy metals in soil was carried out based on HJ 25.3-2019. The results show that the heavy metal pollution in the site is mainly affected by the electroplating process, and the heavy metal Ni pollution decreased gradually with the increase of the distance from the pollution source and the vertical depth. According to the results of ecological risk assessment, the carcinogenic risk of heavy metal Ni in the soil within 3 m depth of the large electroplating workshop in the site is unacceptable, and the pollution degree beyond 3m depth is significantly reduced. The site needs to be controlled or adopt engineering measures such as soil remediation. Studies have shown that the existence of clay layer in the site can effectively reduce the downward migration of pollutants and protect the deep soil. Therefore, we should pay attention to the distribution of clay layer in the later project implementation process to ensure the safety of deep soil and groundwater.

Keywords: Jiangsu; High-risk legacy sites; heavy metal pollution; Ni; risk assessment.

1 INTRODUCTION

In the past 40 years of reform and opening up, China's manufacturing industry has moved from recovery to rise, and has gradually become a major manufacturing country in the world. Electroplating industry is an indispensable link in the basic industrial chain, and the wide application of its products plays an important role in China's economic and social development and the normal operation of the whole industrial system. However, China's electroplating enterprises have the characteristics of small scale and scattered layout, and the production of some enterprises presents the phenomenon of low level of process equipment, inadequate pollution prevention and

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management, so it is very likely to lead to soil and groundwater pollution in their production process [1]. In recent years, various regions have issued corresponding policies to carry out rectification work for electroplating industry, and banned and shut down non-compliant enterprises or production projects according to law or stopped production for rectification. Therefore, the study on the distribution and migration characteristics of contaminants in this type of site is of positive significance for assessing the environmental risks and risk management of contaminated sites inelectroplating industry. In this paper, a closed electroplating enterprise site in Jiangsu Province was taken as the investigation object, soil samples were collected, and soil pH, cadmium, lead, mercury, nickel, copper, arsenic, zinc, hexavalent chromium and other indicators were monitored. The risk assessment of heavy metal pollution provides a basis for the environmental management of heavily polluted enterprise sites.

2 SURVEY METHOD

2.1 Site overview

The electroplating enterprise was established in 2000 and closed at the end of 2018. The original enterprise electroplating types are zinc plating, nickel plating, copper plating, etc. The main raw and auxiliary materials are passivator, sodium hydroxide, cuprous cyanide, copper plate, hydrochloric acid, nickel plating concentrated solution, zinc chloride, zinc plate, anti-discoloration agent, nickel sulfate, nickel chloride, nickel plate and so on. The enterprise covers an area of about 9800 m2, and the layout of various structures in the factory area is shown in Fig.1.

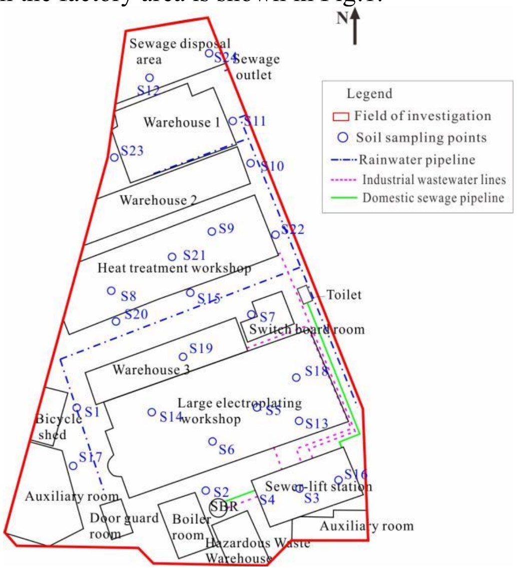

Fig. 1. Investigation site layout and sampling point layout

After site reconnaissance, the structures of the enterprise are in good condition, all the cement on the ground is hardened, no obvious cracks are found on the site, but epoxy floors are not laid in key areas. The workshop wastewater is treated by the sewage treatment station, and finally discharged from the factory area through the main discharge outlet. At present, the north side of the site is a parking lot, while the west and south sides are roads. The weeds on the east side are growing normally. The nearest sensitive area of the site is a residential area about 40 m from the northeast side. According to the local government's explanatory documents, the site is a high-risk legacy plot, which is managed according to the temporarily undeveloped plot.

After site reconnaissance, personnel interviews and data collection and analysis, the soil pollution of the site was identified. It is preliminarily judged that the soil of the site may be polluted by heavy metals such as zinc, nickel and copper. The transportation, storage, feeding, coating and packaging of electroplating solution in the production process, as well as the abnormal electroplating production and the running, running, dripping and leaking in the temporary accident state will cause soil pollution at a single point or a piece of point area in the electroplating production workshop; In the process of sewage collection, there may be soil pollution in the sewage treatment area caused by running, running, dripping and leaking, and the pollution distribution is scattered and the depth is uneven. Because of the influence of groundwater migration, it is not known whether pollutants diffuse outside the plant boundary with groundwater migration.

2.2 Layout of sampling points and sample collection

According to HJ25.1-2019 [2], HJ25.2-2019 [3], and DB32/T 4425-2022 [4], point monitoring was conducted on the soil at the survey site. Based on the preliminary investigation results and professional judgment, the layout of sampling points highlights the key points and takes into account uniformity. Sampling points are evenly distributed across functional areas, with a total of 24 soil sampling points (S1-S24) arranged, as shown in Fig.1.

According to the survey data of the site, the exposed depth of the site is divided into 6 layers from top to bottom according to the characteristics of soil layer: ① miscellaneous fill with an average layer thickness of 1.23 m; ② clay, with an average layer thickness of 4.62 m; ③ Silt mixed with silty clay, with an average layer thickness of 2.66 m; ④ silt, with an average layer thickness of 4.32 m; ⑤ silty clay mixed with silt, with an average layer thickness of 1.41 m; ⑥ Silt mixed with silt, not penetrated.

Therefore, the soil sampling depth is designed to be 3.0 m or 5.5 m, and samples of miscellaneous fill soil layer and clay layer are collected. Three samples are sent for inspection by soil holes (S1-S15) with a depth of 3.0 m, and the collection depths are 0 ~ 0.5 m, 1.5 ~ 2.0 m and 2.5 ~ 3.0 m from top to bottom respectively; Four samples were sent from soil holes with a depth of 5.5 m (S16-S18, S20-S24), and the collection depths were 0 ~ 0.5 m, 1.5 ~ 2.0 m, 3.5 ~ 4.0 m and 5.0 ~ 5.5 m from top to bottom, respectively. Point S19 was designed to be 5.52 m. According to the drilling situation of field sampling, the first 3 meters (0 ~ 3m) of point S19 were miscellaneous fill, and the amount of extracted soil was small. The collected soil was mixed into one sample, and three samples were sent for inspection, and the collection depths were 0 ~ 3.0 m, 3.5 ~ 4.0 m and 5.0 ~ 5.5 m from top to bottom, respectively. To sum up, there are 80 soil samples in this test.

2.3 Sample Analysis Method

According to the characteristics and historical situation of the site, the monitoring indicators are determined as pH, cadmium, lead, mercury, nickel, copper, arsenic, zinc, total chromium, hexavalent chromium, etc. pH in soil was determined by potentiometric method. Arsenic and mercury were determined by atomic fluorescence spectrometry. Cadmium and lead were determined by graphite furnace atomic absorption spectrophotometry; Copper, nickel and zinc were determined by flame atomic absorption spectrophotometry. Determination of hexavalent chromium by flame atomic absorption spectrophotometry

2.4 Evaluation method

2.4.1 Assessment of heavy metal pollution.

According to the planning of the local government, the site is a high-risk legacy plot, which is managed according to the temporarily undeveloped plot. However, there are many sensitive targets around the plot, and the distance is relatively close. Therefore, according to the current situation of its enterprises, the screening values of GB 36600-2018 [5] and DB4403/T 67-2020 [6] are used as reference.

The single pollution index method and Nemero comprehensive pollution index method were used to evaluate the pollution status of heavy metals in soil [7]. Formula for calculating individual pollution index:

$$P_i = C_i / S_i$$

In the formula, Pi is the pollution index of the i-th pollutant; Ci is the measured value of the i-th pollutant; Si is the evaluation standard for the i-th pollutant.

The calculation formula for the comprehensive pollution index:

$$P = \sqrt{\frac{\left(\frac{1}{n}\sum_{i=1}^{n} P_i\right)^2 + P_{i\max}^2}{2}}$$

In the formula, “P” represents the comprehensive pollution index, “n” represents the total number of pollutants, and Pi represents the pollution index of the i-th pollutant, Pimax, which is the maximum value of all pollution element indices.

The grading standards for single factor pollution index and comprehensive pollution index evaluation methods are shown in Table 1.

Table 1. The criterion of pollution grade of soil heavy metals

Grade	Pollution index criterion of indivigual factor		Comprehensive pollution index criterion	
	Pollution index	Pollution grade	Pollution index	Pollution grade
Grade1	P*i*≤1	Clean	P*i*≤0.7	Security
Grade2	1＜P*i*≤2	Light pollution	0.7＜P*i*≤1	Alert
Grade3	2＜P*i*≤3	Medium pollution	1＜P*i*≤2	Light pollution
Grade4	P*i*＞3	Heavy pollution	2＜P*i*≤3	Medium pollution
Grade5	0		P*i*＞3	Heavy pollution

2.4.2 Risk assessment of heavy metals.

According to HJ 25.3-2019 [8], the first type of land exposure assessment model is used to conduct human health risk assessment of heavy metals in soil.

3 RESULTS AND DISCUSSION

3.1 Analysis of soil detection results

According to GB36600-2018 and DB4403/T 67-2020, except for hexavalent chromium, only nickel exceeded the standard and its concentration exceeded the standard seriously, and the exceeding point was S5, which was located in a large electroplating workshop, The depth exceeding the standard is about 3 meters. The coefficient of variation reflects the average variation degree of each sampling point in the overall sample, and the variation size of heavy metal content in a certain area can reflect the distribution of heavy metal elements and the difference of pollution degree in this area [9]. According to Fig.2, except mercury, the variation value of each element shows a downward trend as a whole, and the coefficient of variation of each element below 3m is less than 0.5, which belongs to weak variation and has no significant dispersion, indicating that seven heavy metals are less affected by human beings and more affected by natural factors below 3m; Within 3m, copper, mercury, nickel and zinc all have moderate or strong variation, which indicates that copper, mercury, nickel and zinc are greatly affected by human industrial activities, especially on the surface soil.

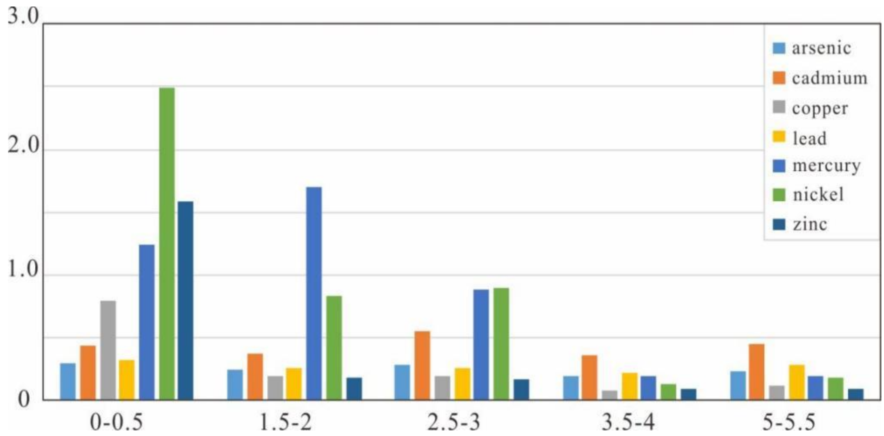

Fig. 2. Coefficient of variation of each element at different depths

3.2 Assessment of heavy metal pollution

Based on the screening values of Class I land in GB36600-2018 and DB4403/T 67-2020, the single factor pollution index of soil heavy metals and Nemero comprehensive pollution index are calculated according to heavy metal concentration. Only one point (S5) has a single factor pollution index of nickel in soil greater than 1 and a depth of about 3m. This point is located in a large electroplating workshop, and the maximum single factor pollution index of nickel is 9.86. The single factor pollution index of cadmium, lead, mercury, nickel, copper, arsenic and zinc in other points is less than 1 in each layer of soil. It shows that only large electroplating workshop soil heavy metal pollution exists in the electroplating plant site and the main pollutant is nickel.

According to the soil test results, the comprehensive pollution index range of the soil layer below 3.0 m in the investigated plot is less than 0.5 m, which indicates that the soil below 3m is clean soil; In 0.5 ~ 3.0 m soil layer, the comprehensive pollution index of some points is between 1 and 2, which indicates that the soil at this point is lightly polluted; In the surface soil, the comprehensive pollution index of some points is greater than 3 m. It shows that the surface soil at such points belongs to heavy pollution level, and the pollution degree gradually decreases with the increase of soil depth.

According to the reconnaissance, the over-standard points are mainly located in the large electroplating workshop, where there are two semi-automatic nickel plating production lines, one zinc plating production line and two copper plating production lines. It is preliminarily judged that the anti-seepage measures on the floor of the workshop are not in place and nickel-containing sewage is infiltrated. Therefore, it is necessary to carry out risk assessment on the soil in this range.

4 RISK ASSESSMENT

4.1 Pay attention to pollutants

The measured value of nickel in soil at some points is obviously higher than the screening value of Class I land in GB36600-2018 and DB4403/T 67-2020. According to HJ25.3-2019, it is necessary to carry out risk assessment on the contaminated site. According to the calculation results, the carcinogenic and non-carcinogenic effects were judged.

4.2 Health Risk Assessment

This site is a high-risk legacy land, which is a sensitive land. The carcinogenic risk and non-carcinogenic effect of nickel are evaluated according to the exposure of people in adulthood. Nickel in the soil is not easy to enter the human body through skin contact, non-volatile, and the groundwater in the site is not used as drinking water. Therefore, this risk assessment mainly considers the risk of nickel in soil entering human body through two exposure routes: 1. Oral intake of soil; 2 ways of inhaling soil particulate matter. Based on the health risk model and Kriging spatial interpolation method [10], Surfer 15 was used to simulate the health risk distribution of nickel in the soil of this plot. The three-dimensional spatial distribution of carcinogenic and non-carcinogenic risk concentration gradient maps of nickel is shown in Fig.3.

The carcinogenic risk of heavy metals in vertical space decreases with the increase of soil blowing depth. According to Fig.3a, when the soil depth is 0 ~ 5.5 m, the carcinogenic risk of nickel in vertical space of soil decreases with the increase of soil depth, and the carcinogenic risk of nickel spreads outward with S5 sampling point as the center. The total carcinogenic risk (CRn) within 3m of soil depth of S5 sampling point exceeds the acceptable carcinogenic risk level of a single pollutant by 10-6, with a maximum of 1.07 × 10-5. According to Fig.3b, the calculated value of non-carcinogenic risk (HIn) of S5 sampling point in the range of 0 ~ 0.5 m is 4.88, which exceeds the acceptable level of non-carcinogenic risk (< 1), and the non-carcinogenic risk is also unacceptable.

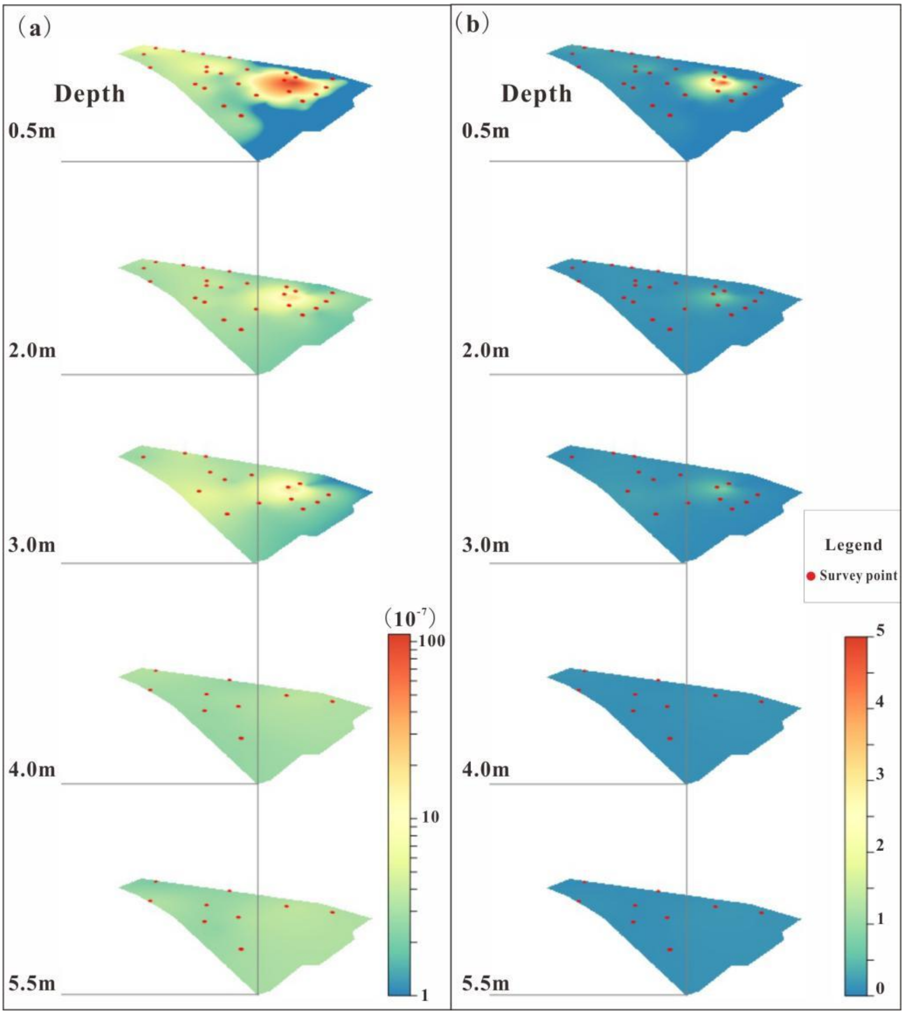

Fig. 3. carcinogenic and non carcinogenic concentration gradient and risk spatial distribution of nickel in soil

To sum up, nickel in the investigation site belongs to the pollution characteristics of point source diffusion, and the most significant pollution area is the large electroplating workshop. On the horizontal level, the farther away from the large electroplating workshop, the lower the soil pollution degree; Restricted by the clay layer in this site, the deeper the distance from the surface layer within 3m, the lower the soil pollution degree.

5 CONCLUSION

(1) There is heavy metal pollution in the soil of the electroplating enterprise site, and the heavy metal nickel accumulates obviously in the surface soil, and the maximum single factor pollution index is 9.86. Copper, mercury, nickel, zinc and other elements are greatly affected by human industrial activities, especially on the surface soil. With the deepening of the vertical depth of soil, the content of heavy metals in soil gradually decreased, and the disturbance of human activities became less and the pollution degree decreased.

(2) The heavy metal pollution in this site is mainly located in the large electroplating workshop, and the pollution is mainly affected by the electroplating process. According to the results of ecological risk assessment, the carcinogenic risk of heavy metal nickel in soil within 3 m depth of electroplating workshop in the site is significant, and the pollution degree beyond 3 m depth is obviously reduced. The total carcinogenic risk (CRn) of S5 sampling site within 3 m of soil depth can reach 1.07 × 10-5, which is unacceptable. At the same time, the non-carcinogenic risk of S5 sampling point in the range of 0 ~ 0.5 m is also unacceptable.

(3) The soil of this plot has been polluted by heavy metals, so it is necessary to control or adopt engineering measures such as soil heterogeneous remediation. The research shows that the existence of clay layer in the site can effectively hinder the downward migration of pollutants and protect the deep soil. Therefore, attention should be paid to the distribution of clay layer in the later project implementation process to ensure the safety of deep soil and groundwater.

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