Distribution of lead in soils within the plant area and adjacent farmland of a typical lead-related enterprise

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Abstract. In this study, an environmental site investigation was conducted of a typical lead (Pb)-related production plant. Based on a combination of manual and Geoprobe sampling, Pb concentrations in soils were measured using standard analytical methods to clarify the distribution of Pb concentrations in soils of the plant’s production workshops, open spaces within the plant, and in the adjacent farmland. The results show that the maximum depth of Pb pollution in the soil of the production workshop was 1.5 m. Pb pollution was found in the soil of the adjacent farmland within a range of 50 m from the plant and occurred at a maximum depth of 0.15 m. Probably as a result of the characteristics of Pb species, there was little horizontal movement of Pb observed in the sampled soils. Further, the solid ground surfaces in the plant (e.g., hardened concrete) effectively prevented the pollution of subsurface soil by Pb.

1. Introduction

With the constantly accelerating industrialization in China, there are a growing number of industries that discharge heavy metals, including mining and metal smelting as well as the chemical, printing and dyeing, leather, pesticides, and feed production industries [1-3]. Moreover, there are severe problems related to some polluting enterprises, such as illegal use of polluting materials and excessive sewage discharge, which result in a high incidence of heavy metal pollution incidents [4-6].

The hazards caused by lead (Pb) pollution have received much attention [7-9]. In China, the industries associated with the most serious Pb pollution risks include battery manufacturing, Pb smelting, and the demolition and breaking down of old ships [10-11]. Pb-related enterprises, as the main industrial source of Pb pollutants, produce smoke and dust containing Pb pollutants, which can reach the soil surrounding the plant and adjacent areas through atmospheric deposition. As a result of solid waste stockpiling and disposal, the leaching solution containing Pb pollutants reaches the soil through percolation. Thus, the soil becomes a sink for Pb pollutants [7,12-16].

In this study, we selected the area within the Pb-related production plant (hereafter the ‘plant area’) and the adjacent farmland in Jiangsu Province, China as our target sites. There were no similar pollution sources in the vicinity of the farmland. We measured the concentrations of Pb and other heavy metals in the soil, and clarified the levels of pollution and distribution patterns of heavy metals in the soil. The results will provide a reference for investigations of similar sites with heavy metal pollution. Additionally, this study provides baseline information for soil environmental quality assessments and comprehensive control of soil heavy metal pollution in the region, which is important for protecting the health of residents.
2. Materials and methods

2.1. Site information
The study area is located in Taizhou, Jiangsu Province, China. The factory of concern, which had produced lead oxides, including PbO, PbSO₄ and Pb₃O₄. In 2011, the facility was shut down by the local government. The site area is about 5500 m². Besides, this research was also conducted on the farmland to the north and east of the vicinity of this closed plant.

2.2. Sample collection and pretreatment
In the plant area, soil samples were obtained at a depth of 3 m (longitudinal sampling interval, 0.3 m). In the adjacent farmland, soil samples were collected to a maximum depth of 0.5 m (sampling depths: surface, 0.15 m, 0.3 m, and 0.5 m). The collected soil samples were air-dried. Impurities, including stones and plant roots and stems, were removed. The samples were ground, passed through 10-mesh and 100-mesh nylon sieves, and then stored until use. Figure 1 shows the soil sampling locations in the adjacent farmland area.

![Figure 1. Soil sampling locations of this site.](image)

2.3. Analytical methods
Soil pH was measured following the methods described in “Determination of Soil pH in Forests” (LY/T 1239-1999). Eight heavy metals, including copper (Cu), zinc (Zn), Pb, cadmium (Cd), chromium (Cr), nickel (Ni), arsenic (As), and silver (Ag), were analyzed and quality controlled by national standard methods, including GB/T 17138-1997, GB/T 17141-1997, HJ/T 491-2009, GB/T 17139-1997, GB/T 22105.2-2008 and HJ 350-2007, Appendix A.

3. Results and discussion

3.1. Screening of heavy metal pollution in the soil
Soil samples were collected at different locations and depths in the plant area and adjacent farmland to accurately reflect the effects of the pollution source on the soil at these sites. Standard analytical methods were used to measure soil pH and the concentrations of eight typical heavy metals (Pb, Cd, Cu, Zn, Ni, As, Cr, and Ag). The results are detailed in Figure 2. The soil pH ranged from 7.4 to 9.8 in the plant area and from 7.6 to 8.8 in the adjacent farmland. Ag was detected at very low levels (<0.5 mg/kg) in only a few soil samples within the plant area and adjacent farmland. Thus, this metal was omitted from further analysis. Cu, Zn, Cd, Cr, Ni, and As were present in the following ranges in the plant area and adjacent farmland, respectively: 4–46 and 31–57 mg/kg, 54–166 and 92–169 mg/kg, 0.01–0.46 and 0.05–0.84 mg/kg, 48–96 and 70–102 mg/kg, 17–66 and 35–60 mg/kg, and 3.5–21 and 6.9–15 mg/kg. The distribution of heavy metal concentrations was not significantly correlated with soil depth.
Figure 2. Distribution of soil pH and heavy metal concentrations in the plant area (□ Soil heavy metal concentrations in the adjacent farmland, ○ Soil heavy metal concentrations in the plant area, -■- Distribution frequency of soil heavy metal concentrations in the adjacent farmland, and -●- Distribution pattern of soil heavy metals in the plant area).

These results indicate that soil pH, Cu, Zn, Cd, Cr, Ni, As, and Ag in the plant area and adjacent farmland were not affected by the plant’s presence. Based on the distribution heavy metal concentrations that we found, which were mostly within previously reported normal distributions, we regard these values as conforming to normal values in this region [17-19].
Soil Pb concentrations in the plant area and adjacent farmland were found to be in the ranges of 16–160,000 mg/kg and 26–4230 mg/kg, which represents a very wide span of concentrations. The Pb concentrations detected in some soil samples exceeded the corresponding assessment standards. For the plant area, we used the Class II land use screening value in “Soil environmental quality Risk control standard for soil concentration of development land” (GB36600-2018), which is 800 mg/kg, as the standard of reference. For the adjacent farmland, we used the risk screening value that provides the maximum allowable values to protect agricultural production and safeguard human health found in the “Soil environmental quality Risk control standard for soil concentration of agricultural land” (GB 15618-2018); and this value is 240 mg/kg.

Based on the above results, we suggest that the soil in the plant area and adjacent farmland of this Pb-related enterprise has been polluted above acceptable standard levels. Thus, we chose Pb as the target pollutant and the other heavy metals were excluded from further analyses.

3.2. Distribution of soil Pb in the plant area

Ten soil-sampling points were designated in the plant area. Point #1 was in the coal pile area and the remaining points were in production workshops. The levels of Pb concentrations at different depths for each sampling point were obtained based on the measurement results (Figure 3). For point #1 in the coal pile area, there were no significant differences in the concentration of soil Pb at different depths; further, the values were in the range of the selected standard for soil Pb assessment. For the nine points in different production workshops, soil sampling was conducted at locations with broken ground surfaces. The results show that at each of the nine points, the maximum concentration of Pb occurred at the soil surface and exceeded the relevant standard (given above). At these nine points, the Pb concentrations markedly decreased with soil depth. The maximum depth of pollution was 1.5 m. These results indicate that there is a high likelihood of Pb pollution being present in the soil under broken ground surfaces in the production workshops. In future investigations of other heavy metal-polluted sites, soil-sampling points should be designated at locations with a broken ground surface. This will help investigators to accurately measure the full extent of pollution, and guide subsequent management and pollution control at the site.

We also collected soil samples from different depths under the concrete floor and nearby bare ground in the plant area (sites SD2 and SD1, 0.5 m apart, Figure 3). The sampling depth was 0.75 m and sampling interval was 0.05 m. The differences in Pb concentrations at these two points at different depths are illustrated in Figure 4. The soil Pb concentration under the hardened cement floor in the plant area showed no significant correlation with depth. Furthermore, the values were similar to the Pb concentrations detected at point #1 in the coal pile area. We concluded that the soil at under the hardened cement floor did not contain pollution from the plant.

In contrast, the maximum Pb concentration in the surface soil of the nearby bare ground (SD1) was higher than 15,000 mg/kg, which is much greater than previously established baseline values. The soil Pb concentration at this site markedly decreased with depth and fell within the range of baseline values at depths below 0.3 m. Together Figure 3 and Figure 4 show that there was little vertical movement soil Pb in this site. Thus, Pb-related enterprises should carefully maintain unbroken floor surfaces to prevent soil pollution by Pb dust [20-21].

Comparing the Pb concentrations at the same depths at the SD1 and SD2 sampling sites (0.5 m apart), we note that there was also no significant movement of Pb in the horizontal direction [22-24]. Soil Pb at 0.2 m deep at the SD1 (bare ground) had not polluted the soil at the same level at the SD2 (under the concrete floor). This lack of horizontal movement is likely related to the species of Pb present in the soil. Pb often exists in its elemental or oxide form, which has stable physicochemical properties and is insoluble in water [25, 26], with a slow rate of horizontal movement. Based on the characteristics of Pb species, we recommend classifying similarly polluted sites into different types (e.g. “workshop” or “building”) to specifically accurately determine the pollution range for the site type.
Figure 3. Vertical distribution of soil Pb concentrations at different sampling points in the plant area.

Figure 4. Vertical distribution of soil Pb concentrations at SD1 and SD2.

3.3. Distribution of soil Pb in adjacent farmland

According to the site conditions, we selected a group of sampling belts radiating to the east (E), west (W), north (N), and northeast (NE) of the pollution source. The distances of the sampling points from the plant boundaries were 10 m, 50 m, and 100 m (a simplified map of sampling points is shown in Figure 5). For the points 10 m from the plant boundaries, the Pb concentrations in the surface soil were significantly higher than those at the 50-m and 100-m points; these levels also exceeded the selected standard for farmland soil Pb levels. At these points, Pb pollution was found at a maximum...
depth of 0.3 m, and the Pb concentrations tended to decrease with depth (consistent with the trend in the plant area). This confirms that there was limited Pb movement in the soil. This limited movement may relate to the depth of soil tillage during farmland cultivation [27-30]. Only one soil sample at 50 m from the plant boundary contained an amount of Pb exceeding the assessment standard; the soil Pb values for the other samples at 50 m and 100 m from the plant were below the relevant standard. This suggests that the emission of Pb-containing exhaust polluted farmland soil within a range of 50 m from the plant.

Based on the Pb concentrations, we fitted the distribution of soil Pb concentrations at different depths in the farmland adjacent to the plant using. Figure 5 clearly show the distribution of soil Pb concentrations at the surface and 0.15 m deep. According to the fitting curves, Pb pollution occurred at the surface and at 0.15 m deep within a range of ~50 m from the plant boundaries. The degree of pollution increased with decreasing distance from the plant area, as demonstrated in prior studies [27-29]. Besides, the 3D-distribution of soil Pb concentrations was fitted in Figure 6. The most contaminated areas are around E1, N1 and W1.

Figure 5. 2D-distribution of soil Pb concentrations at the surface and at 0.15 m deep in the adjacent farmland (the red curve indicates the selected standard for farmland soil assessment).

Figure 6. 3D-distribution of soil Pb in the adjacent farmland.

4. Conclusions
This study analyzed the distribution pattern of Pb in soils within the plant of a typical Pb-related enterprise and in the adjacent farmland at different locations and depths. The main conclusions are:

(1) Pb was the only soil pollutant in the plant area and adjacent farmland to exceed standard levels. This seems to be a result of pollution by the plant.

(2) Soil Pb pollution was commonly found under surfaces of broken ground in the production workshops, and the maximum depth of pollution identified was 1.5 m.

(3) A solid ground surface (e.g., hardened cement) effectively blocked Pb pollution of the subsurface soil.
(4) The emission of Pb from the plant likely resulted in pollution of soil in the adjacent farmland within a 50-m range, and the maximum depth of this pollution was 1.5 m.

(5) Probably as a result of the characteristics of Pb species, there was little horizontal transport of Pb among the soil samples measured.

These conclusions are important for guiding future environmental investigations of similar sites that may suffer from pollution by Pb and other heavy metals. This study provides a useful case study and baseline data for developing future environmental site investigations in the area.

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