Research Article

Survey and Risk Assessment of Contaminants in Soil from a Nitrogenous Fertilizer Plant Located in North China

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China is the world’s largest consumer of fertilizer, with fertilizer plants widely distributed throughout the country. With the removal and closing of fertilizer factories in recent years, pollutant surveys and risk assessments (human health risks) for these sites have become increasingly necessary. However, there has been little research on contaminated fertilizer factory sites. This study aimed to characterize the distribution of pollutants, assess the health risk of the site, and calculate the remediation area and volume in a typical fertilizer plant site in North China. A total of 443 samples were collected in 2019; they indicated that the study site had high concentrations of copper (Cu), ammonia-nitrogen (NH₃-N), total petroleum hydrocarbons (TPH), and fluoride at maximum ratios (the ratio of the highest value of all test data for a particular pollutant to the standard value of the pollutant) of 3.30, 2.55, 19.69, and 1.10, respectively. The health risk assessment results suggested that some hazard quotients exceeded the threshold safe level (>1 established by environmental regulations). The risk control values of soil were 2000mg/g (Cu), 826mg/g (TPH), and 1549mg/g (NH₃-N), and the total remediation soil volume was 72860.71m³. The results provided basic information on soil pollution control and environmental management in a contaminated fertilizer plant site.

1. Introduction

Nitrogen is the first element in soil for plant growth. The nitrogenous fertilizer has been intensively used in agricultural systems to achieve higher yields in China. China is the largest consumer of fertilizer and has the biggest nitrogen production in the world [1–3]. Fertilizer plants are widely distributed throughout China. A large amount of chemicals have been released from these plants into soil and groundwater during their production. In recent years, with the change of fertilizer demand from chemical fertilizer to organic fertilizer, lots of nitrogen fertilizer plants have been closed. The land of these plants had been planned to be used for residential or recreational purposes [4]. Investigation and assessment of contaminants left in these sites is necessary for their safe redevelopment. A large number of studies have focused on the investigation and assessment of contaminants on petroleum [5], coking [6], and chemical-contaminated sites [7]. However, there has been little concern about the fertilizer-contaminated sites, but in fact, this kind of site could have serious environmental impacts and human health risks.

Ammonia-nitrogen (NH₃-N) is the most typical pollutant found at fertilizer-contaminated sites [8]. As a major element determining plant growth and productivity, inorganic and organic forms of NH₃-N in the soil can be absorbed by plants through their roots [9, 10]. However, high levels of NH₃-N have become an increasingly significant environmental problem [11], along with the acidification and eutrophication of ecosystems and climate change [12]. Although NH₃-N is not considered a carcinogenic pollutant when assessing human health risks, the exposure
to NH$_3$-N at high concentrations could be harmful to the respiratory tract, eyes, and skin [13, 14]. Compared with NH$_3$-N, other typical pollutants, such as copper (Cu), total petroleum hydrocarbons (TPH), and fluoride, are at relatively low concentrations in fertilizer-contaminated sites. Even so, they cannot be neglected because of potential bioaccumulation and carcinogenicity.

Understanding the fate and transport of the contaminants present on fertilizer plant sites and identifying their environmental exposure risks are not only the preconditions for soil pollution prevention and control but also to provide important information for making decisions on polluted site remediation. The specific objectives of this study were to (1) characterize the distribution of pollutants in the fertilizer-contaminated site, (2) assess the health risk of pollutants in the study site and identify health risk exposure pathways, and (3) calculate the remediation soil area and volume.

2. Materials and Methods

2.1. Contaminated Site Characterization. The fertilizer plant in this study is located in Linzhang County in the southern part of Hebei Province, belonging to Taihang Mountain’s foreland and flood plain. Construction waste and miscellaneous fill (0.1–2.8 m) are distributed on the surface of the study area, and below them are silt (8.8–11.4 m), silty clay (0.6–2.3 m), and silt (0.4–7.0 m) formed in alluvial and di-fluvial sediments of quaternary. The site began operating in 1975 and shut down completely in 1999; it covers an area of $53 \times 10^3 \text{m}^2$ (Figure 1). The plant received anthracite, water, electrolytic copper, and catalyst as the raw material. The main product of the plant was ammonia ($30 \times 10^3 \text{t/a}$), and the byproduct was ammonium bicarbonate ($100 \times 10^3 \text{t/a}$). However, poor pollution controls and production technology during the operational period caused serious environmental pollution. Many hazardous substances were released into the soil, resulting in potential damage to the surrounding environment and the plant site. A detailed survey of the contaminants present along with a risk assessment is essential if the site is to be redeveloped, in particular as residential land.

2.2. Soil Sampling. A total of 55 soil sampling sites were planned in the plant (Figure 2). The depths of soil sampling from a soil-drilling core were decided by soil lithology and the transport character of contaminants. The soil cores were collected by a drilling rig (SH-30, drilling rig manufacturer). The sampling standard is that one sample is taken from 0.5 m on the surface, and the vertical sampling interval is less than 2 m between 0.5 m and 6 m; if the sampling distance is less than 6 m, one sample is taken every 3 m until no obvious pollution odor is found on the spot. The specific locations of the sampling points this time are 0–0.5 m, 1–1.5 m, 3–3.5 m, 5–5.5 m, 8–8.5 m, 11–11.5 m, 14–14.5 m, 17–17.5 m, and 20–20.5 m. The vertical sampling interval was less than 2 m in 0.5–6 m, and the sampling interval was 3 m at most when the depth exceeded 6 m. To avoid cross-contamination, the sampling personnel replaced their disposable gloves after retrieving each sample. The samples to be analyzed for TPH and NH$_3$-N were transferred to 250 mL brown glass bottles as soon as possible, and the samples to be analyzed for copper and fluoride were transferred to valve bags. All the samples were stored at 4°C and transported to the laboratory for analysis immediately. In total, 443 samples were collected in April and August of 2019.

2.3. Sample Analyses. All analyses were completed by the Pony Testing International Group. Table 1 provides the selected analytical methods employed to measure copper, TPH, fluoride, and NH$_3$-N. To corroborate the results and determine the accuracy of each method, 82 duplicate samples were analyzed by Hebei Shipu Testing Technology Service Co., Ltd. The relative deviation and relative standard deviation can meet the national standard (technical code for soil environmental monitoring HJ/T 166-2004).

2.4. Assessment Standard. TPH and copper concentrations were converted using the risk screening and risk intervention values in accordance with the soil environmental quality risk control standard for soil contamination of development land (GB36600-2018) [18]. However, fluoride and NH$_3$-N, which could not be found in GB36600-2018, were calculated using the technical guidelines for the assessment of contaminated sites (HJ25.3-2014) [19].

The risk assessment of soil contaminants was carried out according to HJ 25.3-2014 [19]. To simplify calculations, the soil was divided into 6 layers as given in Table 2. It is worth mentioning that because the pollutants did not exceed the standard in the 20–25 m depth layer, the layer was ignored.

According to the plan for land utilization of Linzhang County, the future land use pattern of the fertilizer site was residential. Therefore, both adults and children were considered the sensitive human receptors under the residential scenario. The soil exposure of the local population was estimated by considering six different routes: oral ingestion, dermal contact, inhalation of soil particles by mouth, inhalation of gaseous pollutants from the surface soil in the outdoor air, and inhalation of gaseous pollutants from the underlying soil in outdoor and indoor air. The hazard quotient (HQ) of exposure pathways was estimated using the equations given in Table 3.

2.5. Contaminated Soil Remediation. According to the health risk assessment guidelines, human health might be harmed when the hazard quotient exceeds 1, and soil remediation is necessary. The risk control values of soil (RCVS) were calculated using the equations given in Tables 4 and 5. Furthermore, to avoid excessive remediation, RCVS was compared with the screening value of GB36600-2018 [18].

3. Results and Discussion

3.1. Contaminant Characteristics and Sources. The soil sample results are given in Table 6. Cu concentrations
ranged from 5.94 mg/kg to 6590 mg/kg, with an average of 107.82 mg/kg and a maximum ratio of 3.3. As shown in Figures 3(a) and 4, samples with higher concentrations were mainly distributed among the copper washing workshop and production area entrance, where copper ammonia acetate solution was received as a raw material for the
Table 1: Target pollutants and analytical methods.

| Target pollutants | Test method                     | Reference |
|-------------------|---------------------------------|-----------|
| Cu                | Atomic adsorption spectrometry  | [14]      |
| TPH               | Gas chromatography              | [15]      |
| Fluoride          | Ion selective electrode method   | [16]      |
| NH₃-N             | UV-visible scenery photometry    | [17]      |

NH₃-N, ammonia-nitrogen; TPH, total petroleum hydrocarbons.
### Table 2: Calculating models of hazard quotient.

| Exposure pathway                             | Explanation                                                                 | Calculation model                                           | Equation     |
|----------------------------------------------|----------------------------------------------------------------------------|-------------------------------------------------------------|--------------|
| Oral ingestion                              | 6 ways receptors are exposed to pollutants                                | \( \text{HQ}_{\text{oi}} = \text{OISER}_{\text{nc}} \times \frac{C_{\text{sur}}}{\text{RfD}_{o}} \times \text{SAF} \) | (1)          |
| Dermal contact                              |                                                                           | \( \text{HQ}_{\text{dcS}} = \text{DCSER}_{\text{nc}} \times \frac{C_{\text{sur}}}{\text{RfD}_{d}} \times \text{SAF} \) | (2)          |
| Inhalation of soil particles by mouth        |                                                                           | \( \text{HQ}_{\text{pis}} = \text{PISER}_{\text{nc}} \times \frac{C_{\text{sur}}}{\text{RfD}_{i}} \times \text{SAF} \) | (3)          |
| Inhalation of gaseous pollutants from the surface soil in outdoor air |                                                                           | \( \text{HQ}_{\text{iov1}} = \frac{C_{\text{sur}}}{\text{IOVER}_{\text{nc1}}} \times \frac{1}{\text{RfD}_{i}} \times \text{SAF} \) | (4)          |
| Inhalation of gaseous pollutants from the underlying soil in outdoor air |                                                                           | \( \text{HQ}_{\text{iov2}} = \frac{C_{\text{sub}}}{\text{IOVER}_{\text{nc2}}} \times \frac{1}{\text{RfD}_{i}} \times \text{SAF} \) | (5)          |
| Inhalation of gaseous pollutants from the underlying soil in indoor air |                                                                           | \( \text{HQ}_{\text{iiv1}} = \frac{C_{\text{sub}}}{\text{IIVER}_{\text{nc1}}} \times \frac{1}{\text{RfD}_{i}} \times \text{SAF} \) | (6)          |

### Table 4: Major parameters in the calculation models of hazard quotient.

| Parameter | Explanation                                                                 | Value       | Unit  |
|-----------|------------------------------------------------------------------------------|-------------|-------|
| SAF       | Reference dose distribution ratio for exposure to soil                       | 0.5         | /     |
| \( C_{\text{sur}} \) | Concentration of contaminants in soil                                      | 434         | mg/kg |
| \( C_{\text{sub}} \) | Concentration of contaminants in underlying soil                            | 434         | mg/kg |
| \( \text{RfD}_{i} \) | Reference dose for inhalation                                               | \( 7 \times 10^{-2} \)  | /     |
| \( \text{IOVER}_{\text{nc1}} \) | Exposure dose by inhaling gaseous pollutants from the surface soil in outdoor air | \( 9.88 \times 10^{-6} \) | /     |
| \( \text{IOVER}_{\text{nc2}} \) | Exposure dose by inhaling gaseous pollutants from the underlying soil in outdoor air | \( 1.14 \times 10^{-6} \) | /     |
| \( \text{IIVER}_{\text{nc1}} \) | Exposure dose by inhaling gaseous pollutants from the underlying soil in indoor air | \( 1.60 \times 10^{-4} \)  | /     |

### Table 5: Calculation models of risk control values of soil (RCVS).

| Exposure pathway | Explanation                                                                 | Calculation model                                           | Equation     |
|------------------|------------------------------------------------------------------------------|-------------------------------------------------------------|--------------|
| Oral ingestion   | Formulas for calculating the amount of contaminants ingested orally          | \( \text{RCVS}_{\text{ais}} = \frac{\text{ACR}}{\text{OISER}_{\text{nc}}} \times \text{SF}_{o} \) | (7)          |
| Dermal contact   | Formula for calculating the amount of contaminants ingested by skin contact  | \( \text{RCVS}_{\text{dcS}} = \frac{\text{ACR}}{\text{DCSER}_{\text{nc}}} \times \text{SF}_{d} \) | (8)          |
| Inhalation of soil particles | Inhaled soil particle pollutant quantity calculation formula                  | \( \text{RCVS}_{\text{pis}} = \frac{\text{ACR}}{\text{PISER}_{\text{nc}}} \times \text{SF}_{i} \) | (9)          |

### Table 6: Sample analysis results.

| Variable | Pollutant concentration (mg/kg) | Screening value (mg/kg) | Control value (mg/kg) | Detection rate (%) | Overstandard rate (%) | Maximum ratio |
|----------|---------------------------------|------------------------|----------------------|--------------------|-----------------------|--------------|
| Cu       | 6590.00                         | 2000                   | 8000                 | 100                | 1.81                  | 3.30         |
| NH₃-N    | 3951.00                         | 1549                   | —                    | 100                | 10.16                 | 2.55         |
| TPH      | 16263.00                        | 826                    | 5000                 | 30.70              | 1.81                  | 19.69        |
| Fluoride | 1150                            | 1050                   | —                    | 100                | 0.23                  | 1.10         |

\( \text{NH}_3\text{-N} \), ammonia-nitrogen; TPH, total petroleum hydrocarbons.
adsorption of CO, CO₂, O₂, and H₂S. Thus, it can be concluded that the main reason for high Cu concentrations was the corrosion and aging of the machine that accelerated the release of wastewater [20, 21].

TPH ranged from <3 mg/kg to 16263 mg/kg with an average concentration of 129.19 mg/g. The maximum ratio was 19.69, and the distribution of TPH was relatively concentrated (Figure 4). However, TPH was not a raw material used in the fertilizer plant. It could be concluded that TPH was from the lubricating oil leakage of machines in the process of operation or migration (Figure 3(b)). The pollution level decreased with increasing depth due to

Figure 3: Overstandard points of the study area. (a) Cu, (b) TPH, (c) NH₃-N, (d) fluoride. NH₃-N, ammonia-nitrogen; TPH, total petroleum hydrocarbons.
Figure 4: Polluted areas from different soil layers. (a) The first layer, (b) the second layer, (c) the third layer, (d) the fourth layer, (e) the fifth layer, (f) the sixth layer.
interception by the soil (Table 7). The low volatility, low solubility, and high hydrophobicity and sorption capacity of TPH made remediation difficult [22, 23].

As the main product of the fertilizer plant, NH$_3$-N concentrations ranged from 0.14mg/kg to 3951mg/kg, with a mean concentration of 445.1mg/kg and a maximum ratio of 2.55. The main layers of the plant site were silt and silty clay with low water permeability. With the migration of rainfall and wastewater, NH$_3$-N could permeate down to a high depth. The deepest depth that the sample drill could reach was 27m, and NH$_3$-N was present in those samples. As shown in Figures 5(c) and 3, NH$_3$-N was distributed near the copper washing and carbonization transformation workshops. The pollution level was not typically consistent with the depth, which suggests that the area had been polluted with NH$_3$-N for many years (Table 7). This could be attributed to the leakage of wastewater and the leaching of solid waste. Furthermore, poor pollution control and environmental protection awareness during the operational period could have been key factors affecting the contamination of the site.

Fluoride ranged from 127mg/kg to 1150mg/kg with a mean concentration of 424.94mg/kg and a maximum ratio of 2.14. The overstandard point was at the ammonia pool of the carbonation transformation workshop. The fluoride-polluted area was relatively small compared with the other contaminants, and fluoride was only present in the surface soil (first layer) (Table 7). The presence of fluoride may be attributed to the atmospheric deposition of coal burn or the high background value [24, 25].

### Table 7: Pollutant levels from different layers and locations.

| Pollutant | Layer | Polluted area (m$^2$) | Maximum pollutant concentration (mg/kg) | Maximum ratio | Distribution region |
|-----------|-------|-----------------------|----------------------------------------|---------------|---------------------|
| Cu        | 1     | 908.09                | 6590                                   | 3.30          | Copper washing compression workshop (T6) |
|           | 2     | 1038.06               | 6162                                   | 3.08          | Production area entrance (T45) |
|           | 3     | 253.45                | 5050                                   | 2.53          | Copper washing compression workshop (T6) |
| TPH       | 1     | 2730.22               | 16263                                  | 19.69         | Compression workshop (T40) |
|           | 2     | 318.48                | 2130                                   | 2.58          | Compression workshop (T8) |
| NH$_3$-N  | 1     | 1829.33               | 3951                                   | 3.30          | Carbonization transformation workshop (T24) |
|           | 2     | 5739.5                | 3316                                   | 2.14          | Carbonization transformation workshop (T50) |
|           | 3     | 1516.05               | 2799                                   | 2.40          | Copper washing compression workshop (T40) |
|           | 4     | 2844.85               | 3713                                   | 1.81          | Copper washing compression workshop (T35) |
|           | 5     | 2484.11               | 3785                                   | 2.44          | Carbonization transformation workshop (T28) |
|           | 6     | 150.97                | 1747                                   | 1.13          | Copper washing compression workshop (T33) |
| Fluoride  | 1     | 7                     | 1150                                   | 1.10          | Carbonization transformation workshop (T12) |

NH$_3$-N, ammonia-nitrogen; TPH, total petroleum hydrocarbons.

3.2. Health Risk Assessment. The hazard quotients in soils of the study area were calculated from Table 3 and are given in Table 8. Based on the values obtained, the main potential exposure pathway of Cu, fluoride, and TPH could be from oral ingestion. Inhalation of soil particles and dermal contact were the second most likely pathways for fluoride and TPH, respectively. However, the values with different exposure pathways of NH$_3$-N varied significantly in the following order: inhalation of gaseous pollutants from the surface soil in outdoor air > inhalation of soil particles. The contribution of inhalation of gaseous pollutants from the underlying soil in the outdoor and indoor air for NH$_3$-N was insignificant and may be ignored.

In general, the contribution rates of oral ingestion, dermal contact, and inhalation of gaseous pollutants from the surface soil in the outdoor air were 43.3%, 30.6%, and 25.9%, respectively. These results suggested that wearing safety masks would be an effective measure to reduce the negative health effects for workers and residents in the study site.

As given in Table 8, as the soil depth increased, the concentration of most of the pollutants tested decreased gradually. In the first two layers (0–6.5 m), the hazard quotients of Cu, NH$_3$-N, and TPH were greater than 1. In the third layer (6.5 m–10 m), Cu and NH$_3$-N were higher than the standard HQ. Until the fourth (10 m–15 m) and fifth
layer (15 m–20 m), the HQs of NH$_3$-N were 2.32 and 2.34, respectively. Li et al. also found that the surface soil was more vulnerable to contamination by pollutants compared with deeper soil. Those results indicated that the comprehensive HQs of the study site were relatively high and the soil should be remediated [27, 28].

3.3. The Target Value of Contaminated Soil Remediation and the Amount of Pollution. The RSVs of Cu, TPH, and NH$_3$-N were calculated and are presented in Table 9, except for fluoride, which had an HQ $< 1$. The calculated RSV of Cu and TPH were lower than GB36600-2018 [22]; thus, the final RSVS was based on GB36600-2018.
The remediation area and volume for the targeted pollutants for each different layer were determined by interpolation and manual correction; results are given in Table 10. The spatial distribution of the remediation area for Cu, TPH, and NH₃-N is shown in Figure 6. Cu and TPH need to be remediated only in the first three layers. The Cu and TPH remediation areas overlapped almost completely in the second and third layers. In contrast, the area that requires remediation of NH₃-N comprises almost five layers. The total remediation volume was 72860.71 m³ after superposition. These results show that a risk assessment is necessary where fertilization plants have been operational, and they can be used to compare, develop, and select remediation options.
Figure 6: Remediation areas from different soil layers: (a) the first layer; (b) the second layer; (c) the third layer; (d) the fourth layer; (e) the fifth layer.
4. Conclusion

In this study, a pollutant survey and risk assessment of soils were carried out in a fertilizer plant in Linzhang County, North China. The results indicated that the site was polluted by four main contaminants: fluoride, copper, TPH, and NH₃-N. These contaminants were mainly attributed to coal burning, the leakage of wastewater, and lubricating oil. Their presence also reflected the poor pollution controls and environmental protection awareness during the operation period. The total areas polluted by fluoride, Cu, TPH, and NH₃-N were 7 m², 2199.6 m², 3048.7 m², and 14564.81 m², respectively. The hazard quotients found were relatively high and indicate that oral ingestion, inhalation of soil particles, and inhalation of gaseous pollutants from the surface soil in the outdoor air could be the main exposure pathways of this site without any remediation steps. RSVS was calculated and compared with GB36600-2018, and the final total remediation soil volume was 72860.71 m³ after superposition. To solve the problem of soil pollution and protect public health in the study area, the enterprise and local government should make more efforts to remediate the soil pollution. This study provides useful information and reference for environmental management in fertilizer plant sites.

Data Availability

The data used to support the findings of this study are included within the article, and any data in the full-text can be quoted and does not involve the situation of leakage.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

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