Numerical Evaluation and Management Suggestions for Heavy Metal Pollution Risks in a Sludge Landfill: A Case Study from Fuyong Landfill, Shenzhen, China

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Despite the extensive attention paid to the transport of heavy metals in sludge landfills, the processes of transporting these pollutants from a landfill to the underground environment are quite complicated and subject to significant uncertainty. In this study, the transport of typical heavy metal pollutants in a sludge landfill through saturated and unsaturated soil zones during rainfall was investigated via numerical modeling. The objectives of the study are to evaluate the heavy metal pollution risk from a sludge landfill under rainfall infiltration conditions and to propose several management suggestions. The results indicate that, during rainfall, heavy metal concentrations at the top of the unsaturated sludge layer decrease rapidly, but they decrease more gradually at the bottom of the layer. The maximum concentration appears in vertical distribution and decreases gradually through the saturated zone. Nickel is the first heavy metal pollutant to break through the low-permeability natural silt barrier. The transport parameters not only influence the simulated time for heavy metal pollutants to break through the silt layer and cause underground environmental pollution but also affect the extent to which the heavy metal pollutants in pore water exceed the guidelines. On the basis of these results, for dredged sludge with heavy metal concentrations significantly exceeding the standard, the concentration of heavy metals in pore water should be reduced before the sludge is landfilled, and a covering layer should be established on the sludge surface to control rainfall infiltration.

1. Introduction

Solidification/stabilization technology is widely used to treat sludge dredged from rivers and lakes before they are transported to landfills [1]. However, pollutants in the dredged sludge, such as heavy metals, cannot be removed through solidification/stabilization [2, 3]. These heavy metal pollutants may migrate to underground environments via mechanisms such as convection, diffusion, dispersion, and adsorption, especially in simple landfills common in China [4, 5]. These landfills generally do not have either a bottom liner system or a closure cover system, and their antiseepage properties are dependent on the low permeability of natural clay layers [6, 7]. Under the effect of rainfall infiltration, the dredged sludge in landfills is associated with potential environmental risks. Therefore, the transport of heavy metal pollutants in dredged sludge has already become a research focus in the field of sludge processing and disposal.

Previous studies have proposed a number of methods for modeling pollutant transport in underground environments [7–9]. In contrast with pollutant transport in groundwater, pollutant transport in landfills generally occurs in unsaturated porous media. Under rainfall, what the laws of heavy metal concentration in unsaturated porous media are and how the pollutants in landfill migrate into the surrounding area, the simulation study on this phenomenon is still a novel issue. In addition, the pollution level of landfill for the underground environment is closely related to the site
address. Therefore, investigations that use representative site conditions will possess more practical significance.

In this study, a sludge landfill in the south of China was chosen as the research location, and the transport of typical heavy metal pollutants in the landfill during rainfall was investigated using a saturated-unsaturated pollutant migration numerical model. The objectives of the study are to evaluate the heavy metal pollution risk from landfilled sludge under rainfall infiltration conditions and to propose several management suggestions. This study provides a reference for the prediction and evaluation of the long-term safety of landfill environments as well as landfill design and management.

2. Overview about Fuyong Sludge Landfill

Fuyong sludge landfill is located on the east side of Lingdingyang Bay in the Pearl River estuary, west of Shenzhen, China. The landfill has a land area of 230,000 m² (Figure 1) and the quantity of landfilled sludge after solidification/stabilization is up to 500,000 m³. A 4 m high dam has been built around the landfill. The natural sill layer is used as the impervious barrier, and no closure cover is built. The antiseepage barrier is taken at the wing dam only, as shown in Figure 2.

Sludge samples were collected from the landfill and analyzed (Figure 3). The typical heavy metal pollutants and their mean concentrations in the solidified sludge pore water samples from the Fuyong sludge landfill are summarized in Table 1. According to the threshold concentrations for heavy metal pollutants in the Chinese national standards for groundwater quality [10], Pb and Zn concentrations in pore water samples reached the Class IV standard for underground water, and Cd, Cu, and Ni concentrations exceeded the Class V standard. Therefore, the overall pore water of solidified sludge in this landfill was Class V. If the Class III standards for underground water were applied as the pollution threshold for heavy metals, concentrations of the five heavy metals would exceed the standards by between 2.6 and 167 times (Table 1).

3. Materials and Methods

3.1. Numerical Model. Heavy metal transport was simulated over long periods and under rainfall conditions using a numerical model of solute transport in flowing saturated-unsaturated fluid using the finite element method (FEM). The FEM program DTRANSU 2D, designed for calculating fluid flow with mass transport via coupled solute advection-dispersion, was employed [9]. This program has been verified in numerous applications pertaining to seepage flow and contaminant migration [4, 5, 11, 12]. The equation governing solute transport and dispersion, taking saturated-unsaturated fluid flow into account, is as follows [9]:

\[
R_d \frac{\partial c}{\partial t} = \frac{\partial}{\partial x_i} \left( \theta \rho D_{ij} \frac{\partial c}{\partial x_j} \right) - \frac{\partial}{\partial x_j} (\theta \rho V c) - \theta \rho \lambda R_d c - Q_c,
\]

where \( R_d \) (unitless) is the retardation factor, \( \theta \) (unitless) is the moisture content, \( \rho \) (kg/L) is the fluid density, \( c \) (kg/L) is the solute concentration, \( D_{ij} \) (m²/s) is the hydrodynamic dispersion coefficient, \( V \) (m/s) is the seepage velocity, \( \lambda \) (unitless) is the attenuation coefficient, and \( Q_c \) is the source term. The soil-water characteristic curves for soil were described using the Van Genuchten model [13].

3.2. Model Generation. In accordance with the stratigraphic distribution in the sludge landfill (Figure 2), a saturated-unsaturated calculation model with a height of 17 m was developed, as shown in Figure 4. Some parameters for the model layers were obtained according to the geological investigation report, as shown in Table 2. The longitudinal dispersivities, diffusion coefficients, and retardation factors used to model the transport of the different heavy metal pollutants through the different materials were taken from previous publications and are shown in Table 2 [14–17].

3.3. Initial and Boundary Conditions. The mean heavy metal concentrations of the solidified sludge pore water samples from the sample survey were set as the initial concentrations (Table 1). The initial concentration \( c_0 \) of each pollutant at the nodes of the solidified sludge was as follows:

\[
\tau(x_i, 0) = c_0(x_i, 0), \quad (0 \leq x_i \leq 4 \text{ m}).
\]

The background concentrations of the heavy metal pollutants were assumed to be negligible, so the initial pollutant concentrations at the nodes of the other layers were 0 mg/L, as shown in the following equation:

\[
\tau(x_i, 0) = 0, \quad (4 < x_i \leq 17 \text{ m}).
\]

The initial groundwater level was set at 1 m below the base of the solidified sludge. The solidified sludge layer and the top meter of the natural silt layer were modelled as unsaturated zones. The model top was set as the rainfall infiltration boundary. Because the annual average rainfall of the landfill area is about 1665 mm, 900 mm was deducted as evaporation. The infiltration rate was set at 1065 mm/y, modelled as a continuous infiltration of 3 mm/d.

\[
\bar{q}_b(x_i, t) = 3 \text{ mm/d}, \quad (x_i = 0 \text{ m}),
\]

where \( q_b \) (m²/s) is the volumetric flow rate at the top boundary. The heads at the bottom boundary were held constant at 12 m, as shown in the following equation:

\[
\bar{h}(x_i, t) = 12 \text{ m}, \quad (x_i = 17 \text{ m}).
\]

4. Calculation Results

4.1. Transport Process of a Representative Pollutant in Saturated and Unsaturated Zones. The long-term unsteady flow was calculated assuming a continuous rainfall intensity of 3 mm/d, and the transport behavior of a representative heavy metal was determined first. Figure 5 shows the variation of the Cd concentration distribution in the saturated-unsaturated model over time. Owing to the effect of rainfall infiltration, the Cd concentration on the surface of the unsaturated solidified sludge decreases rapidly, while it
decreases more gradually at the bottom of the solidified sludge. The peak concentration appears in vertical distribution, and the pollution scope expands gradually. The Cd concentration declines gradually in the solidified sludge layer, and the decreasing range decreases with depth and time. The Cd concentration in the saturated layers rises gradually, and the increasing range also decreases gradually with depth and time. In places closer to the top of the unsaturated zone, the peak Cd concentration appears earlier and the peak value is higher. The concentrations at various vertical points decrease gradually after reaching peak values.

Therefore, the peak concentration decreases from 0.03 mg/L to 0.01 mg/L during the simulation period of 50 years. Additionally, the location of the peak concentration moves downward, from 3 m from the top of the unsaturated zone in the 5th year of the simulation to 8.5 m from the top of the unsaturated zone in the 50th year.

As for the reasons for the above results, when rainwater recharges the top of the model domain, the moisture content at the top of unsaturated solidified sludge increases rapidly while the influence in the saturated zone is relatively weak and the impact time also lags. Then, the flow velocity in the
unsaturated solidified sludge pore water increases, and the advection rate rises accordingly, which promotes faster transport of the pollutants. With the gradual infiltration of rain, the pollutant transport depth also increases. The results also demonstrate the rationality of the calculations.

4.2. Breakthrough Behaviors of Various Heavy Metal Pollutants. In the simulated landfill, the natural silt layer was modelled as a low-permeability barrier and the sand layer as a permeable layer. Therefore point P, at the junction of the natural silt layer and the sand layer (see Figure 4), was chosen as the location to report the concentration changes of the various heavy metal pollutants. Figure 6 shows the breakthrough curves of the heavy metals Cd, Cu, Ni, Pb, and Zn in the sludge landfill. The concentrations of these five heavy metals at point P all show an upward trend. However, the rates of increase vary. The Cd concentration at point P increases slowly with time in the beginning, and an inflection point appears on the breakthrough curve after approximately 10 years of simulated infiltration. Then, the concentration increases linearly and exceeds the pollution threshold concentration after 29.58 years. This indicates that the breakthrough time of Cd is 29.58 years. The Cd concentration then rises more slowly after 45 years and reaches 0.0097 mg/L after 50 years, which is 32.43% of its initial concentration (Figure 6(a)). The Cu concentration reaches 0.68 mg/L (19.37% of the initial concentration) after 50 years but does not exceed the pollution threshold concentration (Figure 6(b)). The breakthrough curve of Ni is similar to that of Cd. However, Ni exceeds the pollution threshold concentration after 16.20 years and reaches 1.14 mg/L (33.99% of the initial concentration) after 50 years (Figure 6(c)). Owing to the large retardation factor, the Pb concentration rises slowly, and it is only $2.55 \times 10^{-8}$ mg/L in the 50th year (2.84 × 10^{-5}%)

Table 1: Measured concentrations of heavy metal pollutants in the landfill.

<table>
<thead>
<tr>
<th>Heavy metal pollutants</th>
<th>Cd</th>
<th>Cu</th>
<th>Ni</th>
<th>Pb</th>
<th>Zn</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mean concentration (mg/L)</td>
<td>0.03</td>
<td>3.51</td>
<td>3.34</td>
<td>0.09</td>
<td>2.60</td>
</tr>
<tr>
<td>Class III standard of underground water (mg/L) [10]</td>
<td>0.005</td>
<td>1.00</td>
<td>0.02</td>
<td>0.01</td>
<td>1.00</td>
</tr>
<tr>
<td>Excess multiple</td>
<td>6.00</td>
<td>3.51</td>
<td>167.00</td>
<td>9.00</td>
<td>2.60</td>
</tr>
</tbody>
</table>

Figure 3: Sample collection in Fuyong sludge landfill.

Figure 4: Schematic diagram of the calculation model.
of the initial concentration). The pollution threshold concentration is not reached by Pb within 100 years (Figure 6(d)). The Zn concentration reaches 0.59 mg/L (22.68% of the initial concentration) after 50 years but also does not exceed its pollution threshold concentration (Figure 6(e)). Zn does not break through the natural silt

<table>
<thead>
<tr>
<th>Parameters</th>
<th>Solidified sludge</th>
<th>Natural silt</th>
<th>Sand</th>
<th>Silty clay</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hydraulic conductivity (m/s)</td>
<td>$1.0 \times 10^{-7}$</td>
<td>$1.0 \times 10^{-8}$</td>
<td>$1.85 \times 10^{-5}$</td>
<td>$1.1 \times 10^{-8}$</td>
</tr>
<tr>
<td>Porosity (%)</td>
<td>50.0</td>
<td>60.0</td>
<td>33.0</td>
<td>53.0</td>
</tr>
<tr>
<td>VG-Parameters</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$\alpha$ (m$^{-1}$)</td>
<td>1.0</td>
<td>1.0</td>
<td>5.75</td>
<td>1.0</td>
</tr>
<tr>
<td>$n$ (-)</td>
<td>1.2</td>
<td>1.2</td>
<td>1.6</td>
<td>1.2</td>
</tr>
<tr>
<td>$\theta_f$ (-)</td>
<td>0.541</td>
<td>0.541</td>
<td>0.541</td>
<td>0.541</td>
</tr>
<tr>
<td>$\theta_r$ (-)</td>
<td>0.2</td>
<td>0.2</td>
<td>0.0</td>
<td>0.2</td>
</tr>
<tr>
<td>Longitudinal dispersivity (m)</td>
<td>0.1</td>
<td>0.1</td>
<td>1.0</td>
<td>0.1</td>
</tr>
<tr>
<td>Diffusion coefficient ($\times 10^{-10}$ m$^2$/s)</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Cd</td>
<td>8.90</td>
<td>8.90</td>
<td>8.90</td>
<td>8.90</td>
</tr>
<tr>
<td>Cu</td>
<td>7.90</td>
<td>7.90</td>
<td>7.90</td>
<td>7.90</td>
</tr>
<tr>
<td>Ni</td>
<td>0.59</td>
<td>0.59</td>
<td>0.59</td>
<td>0.59</td>
</tr>
<tr>
<td>Pb</td>
<td>3.60</td>
<td>3.60</td>
<td>3.60</td>
<td>3.60</td>
</tr>
<tr>
<td>Zn</td>
<td>3.30</td>
<td>3.30</td>
<td>3.30</td>
<td>3.30</td>
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<tr>
<td>Retardation factor (-)</td>
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<td></td>
<td></td>
</tr>
<tr>
<td>Cd</td>
<td>2.0</td>
<td>2.0</td>
<td>1.0</td>
<td>2.0</td>
</tr>
<tr>
<td>Cu</td>
<td>3.3</td>
<td>3.3</td>
<td>1.0</td>
<td>3.3</td>
</tr>
<tr>
<td>Ni</td>
<td>2.0</td>
<td>2.0</td>
<td>1.0</td>
<td>2.0</td>
</tr>
<tr>
<td>Pb</td>
<td>30.0</td>
<td>30.0</td>
<td>1.0</td>
<td>30.0</td>
</tr>
<tr>
<td>Zn</td>
<td>3.0</td>
<td>3.0</td>
<td>1.0</td>
<td>3.0</td>
</tr>
</tbody>
</table>

Figure 5: Variation of Cd concentration distribution with time.
layer. Therefore, Ni is the first heavy metal to break through the natural silt layer after 16.2 years.

5. Discussion

The five heavy metal pollutants modelled in this study have different initial concentrations, pollution threshold concentrations, and transport parameters. As a result, the different heavy metal pollutants caused underground environmental pollution at different times under the same site conditions and rainfall conditions. In other words, the breakthrough times for the different heavy metals varied. Among these five heavy metal pollutants, Ni transport was the fastest and its breakthrough time was the shortest. Although the initial concentration of Ni is not the highest, its retardation factor is the lowest, so its absolute (1.14 mg/L)
and relative concentrations (33.99%) after 50 years are the greatest reported at point P, and it exceeds the pollutant threshold concentration most significantly (66.8 times). Cu has the highest initial concentration, but it exceeds the pollutant threshold at point P by only 3.51 times. Pb transport is the slowest because its retardation factor is more than 10 times greater than the other heavy metals. The silt layer adsorbs Pb in pore water on a large scale, greatly slowing down the transport of Pb through the layer. Therefore, the transport parameters not only influence the time for heavy metal pollutants to break through the low-permeability natural silt barrier and cause underground environmental pollution. They also affect the extent to which the simulated heavy metal pollutants in pore water exceed the guidelines.

For dredged sludge with heavy metal concentrations that greatly exceed the applicable standards, the concentration of heavy metals in the pore water should be reduced before the sludge is landfilled. Where rainfall in the region of a landfill is abundant, a cover layer should be arranged on the sludge surface following the sludge disposal process [18]. Landfill covering should be conducted after the sludge is processed to control rainwater infiltration with an impervious barrier [19]. Finally, closure management should be conducted for landfills and leachate should be collected until the landfill becomes stable [20].

The management of landfill involves a combination of environmental, economic, and social factors [21]. The indexing method used in this study is a commonly used method for landfill risk assessment in the world [22]. The time for heavy metal pollutants breakthrough the natural clay liner was calculated in this study, but the complexity of the landfill determines that there are still many uncertainties [23]. Therefore, in the future, factors such as regional characteristics, urban planning, and the conditions of the landfill itself can be further considered to manage the landfill throughout its life cycle [24].

6. Conclusions

In this study, the vertical transport of typical heavy metal pollutants through an unsaturated soil zone in a sludge landfill to the saturated soils underneath was investigated using a numerical modeling approach. The results can provide a reference for the prediction and evaluation of the long-term safety of landfill environments as well as landfill design and management. Several conclusions and suggestions could be drawn and are shown below.

Under the effect of rainfall, the concentrations of heavy metal pollutants on the surface of an unsaturated sludge layer reduce rapidly while concentrations at the bottom of the unsaturated sludge layer decrease more gradually. The peak concentration appeared in vertical distribution and dropped gradually in the saturated zone. The location of the peak concentrations also declined.

Under the effects of rainfall infiltration, Ni was the first pollutant to break through the impervious barrier of natural silt. Therefore, the transport parameters not only influence the time for heavy metal pollutants to break through the low-permeability natural silt barrier and cause underground environmental pollution, but they also affect the extent to which the simulated heavy metal pollutants in pore water exceed the guidelines.

Data Availability

The data used to support the findings of this study were supplied by Hohai University under license and cannot be made freely available. Requests for access to these data should be made to Shi Shu, shushi@hhu.edu.cn.

Conflicts of Interest

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

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