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ORIGINAL PAPER

Experimental study of the environmental and geotechnical properties of landfills under long-term leachate effects: macro–microscopic tests on in situ clays

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Abstract

Clay structures are widely used as impermeable barriers to landfill pollution, although waste can degrade for more than 30 years, no studies concerning the long-term service performance of clay layers under leachate have been reported. In this paper, the in situ clay leaching in landfill for more than 20 years was studied by drilling and sampling. The results show that clay particle contents in soils decreased under long-term leachate effects, causing the liquid limit and plasticity index of the soil to reduce substantially, and the plastic limit to decrease to a lesser extent. The internal friction angle of clay increased, its cohesion decreased, and its compressibility increased, which caused the hydraulic conductivity of clay to increase by 2–4 times at constant porosity following leachate interaction. The long-term effects of leachate led to a reduction in the number of clay minerals, such as montmorillonite, in the soil, the coalescence of soil particles, an increase in the volume of large pores by 132%, a decrease in the volume of small pores by 14.4%, and increased pore connectivity. When landfill waste is undergoing middle and later stages of degradation, the leachate pollutant diffusion depth in the bottom clay layer is approximately 1 m. Since the groundwater is not polluted, our results confirm that the long-term safety aspect of landfill meets the bottom clay impermeability requirements specified in GB16889-2008 (hydraulic conductivity $< 10^{-7}$ cm/s, thickness > 2 m).

Keywords Landfill · In situ clays · Leachate · Geotechnical properties · Micromechanics

Introduction

In China, the annual production of domestic solid waste is approximately 235 million tons, around 33% of which is disposed of into landfills. Before 2000, China's domestic waste was mainly disposed of by simple landfills, leading to the formation of about 27,000 obsolete landfills covering more than 534 km^2. A landfill represents a large-scale biochemical reactor and releases a large volume of gas and liquid pollutants and heat due to biochemical degradation reactions. The water level inside landfills in China can be as high as 50–80 m, and the corresponding temperatures can reach 55°C (Chen et al. 2014; Jafari et al. 2018; Whitworth and Ghazifard 2009; Yoobanpot et al. 2017). The leachate contains large amounts of Na$^+$, K$^+$, Ca^{2+}, and other ions (Zhao et al. 2000; Bhatnagar et al. 2017; Somani et al. 2018) with biochemical reactions lasting up to 30 years or more. Old landfills typically use only a compacted clay layer (CCL) as the barrier structure, which exposes to the complex environments, such as waste pile loading, high temperature, and long-term leachate interaction, resulting in deterioration of the engineering properties of the clay layer and thus environmental pollution and geological disasters.

Leachate can have significant effects on the geotechnical properties of clay barrier structures (CCL, GCL, cutoff wall, etc.). The effects of leachate on different types of clay minerals vary widely, and mechanisms of leaching influence include diffuse double layer compression due to cation exchange, chemical erosion, chemical precipitation, and

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microbial siltation (Aldaeef and Rayhani 2014; Francisca and Glatstein 2010; Safari and Valizadeh 2018). Previous investigations into the effects of leachate interaction on the compacted silty clay behaviors showed that compaction, permeability, and the internal friction angle of compacted clay increased with increasing leachate concentration, whereas cohesion decreased with increasing leachate concentration, because of the reduction in montmorillonite due to leachate interaction. Zhao et al. (2016) studied the permeability and strength properties of similar kinds of clays as examined in Li et al. (2013) using synthetic leachate, and found that biomass silting led to the abovementioned differences in engineering properties. Harun et al. (2013) found that the liquid limit and plasticity index of compacted clays decreased with increasing leachate concentration. Previous studies have also used triaxial shear, SEM, XRD, and laser particle size tests to explore the strength and microstructure of compacted clay (Xu et al. 2018, 2019), showing that the stress–strain curve of compacted clay shifts from strain softening to strain hardening and that increasing leachate concentration, decreasing secondary minerals, and increasing pore space were the main reasons for the decrease in strength of compacted clay. Oztoprak and Pisirici (2011) investigated changes in the microstructure of in situ clay under leachate interactions around the landfill prior to using the leachate soaking method. Changes in microstructure, permeability, compressibility, and the stress–strain behavior of clay were investigated, showing that the influence of structural changes in clay on macroscopic mechanical indices was explained by a combination of mineralogical and chemical compositions.

Leachate composition is another major controlling factor influencing the effectiveness of leachate–clay interactions. With increasing landfill age, cation concentrations and COD both increase and then decrease, while pH values decrease and then increase, gradually shifting from acidic values in the early stages to alkaline values in the later stages (Liu et al. 2018). Li-wen et al. (2009) investigated the effect of major cation species in landfill leachate on engineering properties such as permeability, strength, and plasticity of landfill-compacted clay. Zaki et al. (2017) showed that the adsorption behavior of Zn^{2+}, Cd^{2+}, and Pb^{2+} ionic metals on landfill silty clay was mainly influenced by the nature of the clay. The studies noted above analyzed the effects of leachate on the geotechnical properties of clays, mainly by using landfill leachate or chemicals to soak the clay (up to 60 days). The conclusions vary widely depending on leachate composition and concentration, erosion time, and clay properties. In recent years, few works used in situ borehole sampling to estimate the barrier performance of clay layers to leachate contaminants (Zhan et al. 2014). When compared with laboratory tests, in situ samplings and investigation have the following advantages. (1) Test results directly reflect the engineering properties and microstructure of clay under long-term (up to decades) exposure to leachate interactions, whereas leachate immersion is mostly conducted for only a few months, and thus fails to reflect long-term effects of leachate interaction. (2) Laboratory-scale remodeling methods destroy the structural properties of clay, and leachate soaking remodeling methods do not accurately reflect the effects of leachate interactions with the microstructure of clay. Nonetheless, research on the geotechnical properties of in situ clay at the bottom of landfills under long-term leachate interaction has rarely been reported.

Above all, this paper takes an old landfill site in Hubei, China, as this study object. Firstly, contaminated and uncontaminated in situ clay samples at the bottom of the landfill were selected by geotechnical investigation methods. Subsequently, the effects of long-term leachate immersion on clay behaviors and engineering properties were comparatively investigated. Finally, the effects of long-term leachate exposure on the microstructure of clay were analyzed, which helps us to infer and predict the microscopic mechanisms of long-term exposure to leachate on macroscopic behavior, such as the engineering properties of clays. Results in this paper will provide recommendation for the evaluation of engineering safety parameters in old landfill sites.

Materials and methods

Materials

In situ clay samples were obtained from an old landfill site (geographic information is shown in Fig. 1) located in Chibi City, Hubei Province, China, approximately 22 km away from the Yangtze River. This landfill was filled in the 1990s, and field drilling results show that it covers an area of around 20,000 m^2, with a stratigraphy divided into five layers from top to bottom: (1) plain landfill layer (area number I), i.e., the landfill clay sealing cover layer, with a thickness of 0.2–2.0 m (average thickness 1.2 m), reddish brown, and mainly composed of clay soil; (2) garbage soil layer, with a thickness of 5.5–12.2 m, distributed throughout the site, with mixed color, loose state, and mainly composed of organic waste, such as meal scraps, plastics, textile fibers, and a small amount of inorganic waste including glass, ceramic masonry, and clay soil, corresponding to a landfill time of over 20 years; (3) peripheral clay layer (area number II), distributed around the landfill area, with a thickness of 11.0–16.8 m, reddish brown in color, and mainly composed of clay soil; (4) leachate-interacting clay layer (area number III), distributed at the bottom of the garbage layer, with a thickness of 0.6–2.9 m (average thickness 0.9 m), contaminated by garbage leachate soaking, dark brown in color, and mainly composed of clay; (5) bottom clay layer (area number IV), distributed at the bottom of the leachate-interacting soil

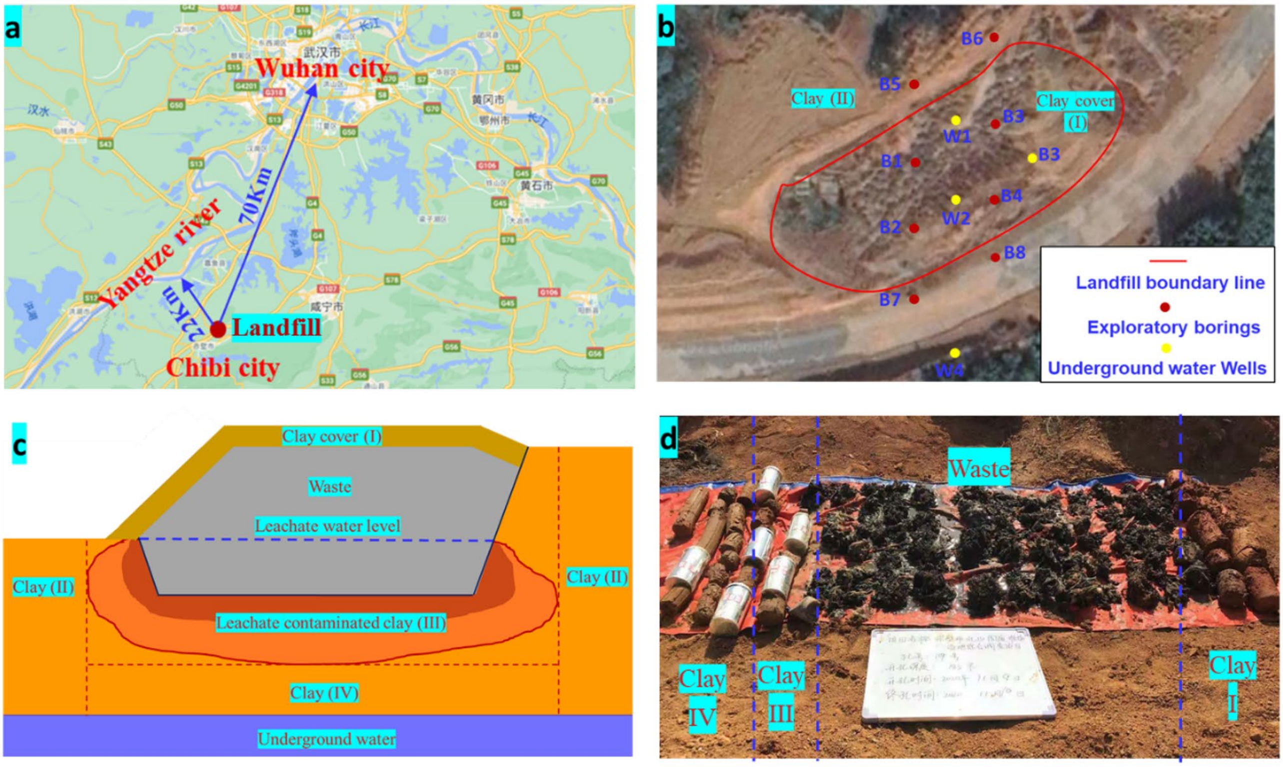


Fig. 1 Site survey map of the old landfill studied herein. **a** Location map. **b** Satellite map. **c** Schematic diagram of the stratigraphic structure of the landfill. **d** Photograph of stratigraphic structure drilling

layer, with a thickness of 10.0–15.0 m, reddish brown in color, and mainly composed of clay.

In situ clay samples were obtained from eight sampling holes using a 110-mm auger; four holes were drilled in the landfill area (borehole numbers B1, B2, B3, and B4) and four in the surrounding area (borehole numbers B5, B6, B7, and B8), with a borehole spacing of 60–100 m. These holes were drilled to the bottom bedrock and, following sampling, materials were sealed in iron sheet boxes and transported back to the laboratory for experimental analysis.

Methods

Test samples were numbered using the area (I–IV)-borehole numbers (1–8)-sampling depth (meters). Four groundwater wells were studied, including three leachate wells (W1–W3) in the landfill site and one off-site groundwater well (W4, in the downstream direction of groundwater within the site).

The analyses conducted herein included four parts: basic physical properties testing, geotechnical testing, microstructural characterization, and environmental chemical analysis. The physical properties test included the determination of natural water content, specific gravity, consistency limits, and particle size distribution. The geotechnical tests included the consolidation and direct shear tests. Microstructural characterization included MIP and SEM analyses. Environmental chemical analyses included groundwater and bottom clay layer contamination measurements. The landfill sealing clay (area number I) and the surrounding clay (area number II) were only tested for their basic physical properties. In contrast, the bottom clay undergoing interaction with leachate (area number III) and the bottom clay layer (area number IV) were tested for the above four aspects.

Physical property testing

According to previous research, the influence of leachate on the basic physical properties of clay soil is predominantly reflected in its consistency limits and particle size distribution. Therefore, the physical property tests conducted in this study mainly include consistency limits and particle size distributions, while the test method refers to the "Standard for soil test method" (GB/T 50123–2019).

Geotechnical testing

The geotechnical tests conducted herein include the shear test, consolidation test, and permeability analysis; these include four groups of leachate-interacted clay in zone III (specimen numbers III-1–11.3, III-2–8.6, III-3–11.0, and III-3–11.4), and four groups of uncontaminated clay in zone IV (specimen numbers IV-1–11.8, IV-1–12.6, IV-2–9.5, and IV-3–15.5). The test method was as follows.

(a) Shear test

Before testing, specimens were saturated with deionized water for 24 h. Prior to shearing, consolidation was conducted at pressures of 50 kPa, 100 kPa, 200 kPa, and 400 kPa. The test method used was based on the Chinese "Standard for soil test method" (GB/T 50123–2019) and employed a quadruple electric direct shear apparatus from the Nanjing Soil Instrument Company.

(b) Consolidation test

Prior to testing, specimens were saturated with deionized water for 24 h. Oedometer tests were conducted according to the Chinese "Standard for soil test method" (GB/T 50123–2019) at consolidation stresses of 25–3200 kPa, with multiplication of pressure at each stage.

(c) Permeability analysis

The hydraulic conductivities of clay under different consolidation pressures were obtained by inversion of Terzaghi's one-dimensional consolidation theory using the results of consolidation tests. Although the direct measurement of *k* is preferred, indirect methods, such as oedometer tests, have been extensively used in the determination of *k* values (Du et al. 2015a, b). The calculation formula is as follows:

$$k = c_v m_v \gamma_w \tag{1}$$

where k is the hydraulic conductivity (m/s), c_v is the coefficient of consolidation (m^2/s) determined using Taylor's (square-root-of-time) method, m_v is the coefficient of volume change (kPa^{-1}), and γ_w is the unit weight of water (kN/m^3).

Microstructure test

The microstructure test mainly includes MIP and SEM analyses, the purpose of which is to compare and analyze changes in pore size distribution and pore morphology of in situ clay following long-term interactions with leachate. The samples consisted of two groups of leachate-interacted clay in zone III (sample numbers III-1–11.3 and III-2–8.6) and two groups of uncontaminated clay in zone IV (sample numbers IV-1–11.8 and IV-1–12.6). Samples were obtained in their original state, freeze-dried, carefully cut into the sizes required, and then tested using MIP and SEM. The test methods are referenced in the literature (Wan et al. 2014).

Environmental chemical tests

These tests included landfill leachate, groundwater quality analysis, and pore solution characteristic contaminant analysis of the clay layer at the bottom of the waste. The test methods were as follows.

(a) Leachate and groundwater water quality analysis

Leachate was sourced from wells W1, W2, and W3 in the landfill site, and groundwater was obtained from well W4 downstream of the landfill site, with a total of 45 test indexes, referring to the China Standard for Pollution Control on the Landfill Site of Municipal Solid Waste (GB16889-2008) and China Standard for Groundwater Quality (GB/T 14848-2017).

(b) Chemical analysis of bottom clay pore solution

Three depths (0.5, 1, and 1.5 m below the waste) were selected at the bottom of the landfill to test the concentration of soil pore solution contaminants. Based on the results of leachate testing in this paper and methods of characteristic leachate contaminant selection in the literature (Zhan et al. 2014), Na^+, Cl^-, and TN were selected as characteristic leachate contaminants to evaluate the migration of leachate contaminants in the bottom clay layer. Pore solution contaminants were tested according to previous work (Du et al. 2006; Zhan et al. 2014). Soil sampling boreholes B1, B2, and B3 were used to obtain average test results at the same depth as the final results.

Results and discussion

Basic physical property test results and analysis

Physical properties

Figure 2 and Table 2 show the linear fitting relationship between soil consistency limits (LL/PL/PI) and clay content based on the test results presented in Table 1. The soils within the survey area are all classified as clay, with properties varying greatly across different areas. According to the physical property test results of the leachate-contaminated clay layer (No. III) and uncontaminated clay layer (No. IV) at the bottom of the landfill, following long-term leachate soaking and interaction, clay properties changed significantly, and the plastic limit, liquid limit, plasticity index, and clay content all decreased significantly, with an average decrease of 22.7%, 12.1%, 30.8%, and 16.2%, respectively.

Basic physical properties

Figure 2 shows the linear fitting relationship between soil consistency limits (LL/PL/PI) and clay content based on the

Table 1 Physical characteristics of clay samples

Specimen code	Liquid limit, W_L (%)	Plastic limit, W_P (%)	Plasticity index, I_P (%)	Clay content (< 2 μm) (%)
I-1–0.2	70.1	27.5	42.6	49.6
I-2–0.2	73.7	27.4	46.3	51.1
I-3–0.2	68.3	31.8	36.5	42.5
I-4–0.2	64.4	26.7	37.7	40.2
Mean, Xm	69.13	28.35	40.78	45.85
Standard deviation, *σ*	3.35	2.02	3.92	4.60
II-5–6.0	57.00	27.30	29.70	35.6
II-6–3.8	63.40	26.50	36.90	45.8
II-7–13.0	66.00	28.10	37.90	51.5
II-8–8.4	56.80	25.10	31.70	41.1
Mean, Xm	60.80	26.75	34.05	43.50
Standard deviation, *σ*	4.01	1.11	3.44	5.86
III-1–11.3	39.80	20.40	19.40	30.1
III-2–8.6	36.90	18.40	18.50	25.0
III-3–11.0	37.20	17.60	19.60	28.1
III-3–11.4	36.40	17.30	19.10	22.3
Mean, Xm	37.58	18.43	19.15	26.38
Standard deviation, *σ*	1.32	1.21	0.42	2.97
IV-1–12.8	44.30	21.20	23.10	25.6
IV-1–13.6	56.00	21.60	34.40	37.4
IV-2–10.1	45.90	22.00	23.90	27.7
IV-3–12.5	48.40	19.10	29.30	35.2
Mean, Xm	48.65	20.98	27.68	31.48
Standard deviation, *σ*	4.49	1.12	4.56	4.94

test results presented in Table 1. The fitting shows a linear increasing trend between the soil consistency limits (LL/PL/PI) and clay particles, indicating that decreasing clay particle content after long-term exposure to leachate is one of the main reasons for decreasing soil consistency limits. However, large local fluctuations indicate that other causes (e.g., changes in the electrochemical properties of soil particle surfaces due to leachate contaminants) also affect soil consistency limits.

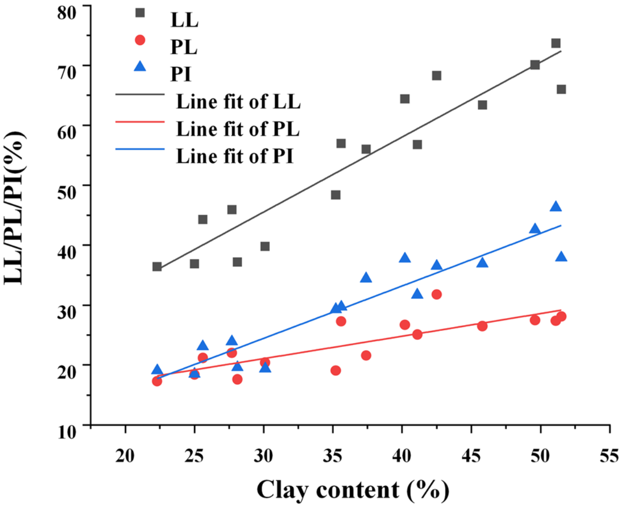


Fig. 2 Relationship between variation of consistency limits and clay content

$$LWC = \alpha CC + \beta \tag{2}$$

where CC is clay content (%); α is the slope; and β is the intercept of the linear relationship.

Geotechnical test results and analysis

Shear test results

Table 3 shows the results of the clay shear test. For the clay layer exposed to leachate (No. III) and the uncontaminated clay layer at the bottom of the landfill (No. IV), it can be seen that long-term leachate immersion and interaction cause the cohesive strength of the in situ clay samples to decrease by

Table 2 Linear fitting parameters of consistency limits and clay content

LWC	*α*	*β*	Adj.R
LL	1.23	7.95	0.87
PL	0.36	10.0	0.69
PI	0.87	−2.01	0.88

Table 3 Results of clay shear tests

Specimen code	*C (kPa)*	*Φ (°)*	Specimen code	*C (kPa)*	*Φ (°)*
III-1–11.3	38.5	23.31	IV-1–12.8	32	24.08
III-2–8.6	17	26.89	IV-1–13.6	54	15.27
III-3–11.0	37.4	26.66	IV-2–10.1	45	19.54
III-3–11.4	15	30.23	IV-3–12.5	48.4	17.91
Mean, Xm	26.98	26.77	Mean, Xm	44.85	19.20
Standard deviation, *σ*	11.00	2.45	Standard deviation, *σ*	8.085	3.2031

39.8%, while the internal friction angle increased by 39.4%. Figure 3 shows the variation of shear strength index in relation to soil clay particle content. Clay cohesion in the same region increases with increasing clay mineral content, while the angle of internal friction decreases with decreasing clay content. Relative to the results of the basic soil property test, the shear test results of the in situ clay are more discrete, which may be related to in situ soil disturbance and large particles in soil samples. Therefore, the decrease in cohesion and increase in internal friction angle of leachate–clay samples in this paper were attributed to decreasing clay particle contents in the soil. Similar findings have been reported in previous literature (Li et al. 2013; Xu et al. 2018).

Consolidation test results

According to the consolidation test results of several groups of in situ clay in the same area and depth, the mean values of porosity and errors of the bottom leachate-exposed soil layer (No. III) and uncontaminated clay layer (No. IV) under the action of consolidation pressure at all levels are shown in Fig. 4. This demonstrates that the porosity of the bottom leachate-exposed clay layer (No. III) is smaller than that of the uncontaminated clay layer (No. IV) under the same consolidation pressure. With increasing consolidation stress, the difference between the two gradually decreases. Differences between the consolidation curves of multiple sets of in situ clays in the uncontaminated clay layer (No. IV) were small (relatively small error bars), such that the consolidation curves of the contaminated soil layer (No. III) were relatively discrete; this may be related to differences in the degree of contamination between different specimens. Oztoprak and Pisirici (2011) and Li et al. (2013) conducted consolidation tests on remolded clay and in situ clay under leachate immersion, respectively, with conclusions similar to the results of this paper. The mechanisms of leachate contamination on clay consolidation curves are understood as follows. Firstly, chemical effects such as high concentrations of cations in leachate can be equated to consolidation pressures (equivalent to an increase in additional consolidation pressure) (Ma et al. 2019; Yao et al. 2020). Secondly, changes in soil pore structure (equivalent to changes in material stiffness) due to leachate interaction are also major factors affecting soil consolidation.

Permeability results

The hydraulic conductivity of in situ clay was obtained through inversion of the consolidation test results. The mean values and errors of hydraulic conductivities in the bottom

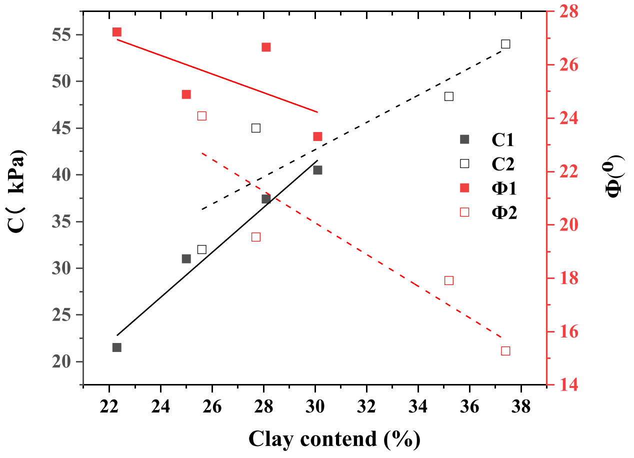


Fig. 3 Relationship between shear strength index and soil clay content

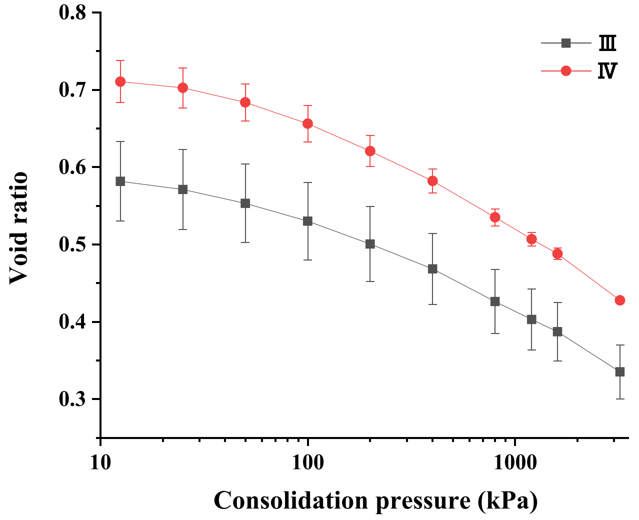


Fig. 4 Results of in situ clay consolidation tests

leachate-contaminated clay layer (No. III) and the uncontaminated clay layer (No. IV) under consolidation stress effects at all levels are shown in Fig. 5. These findings are based on inverse hydraulic conductivities from multiple sets of consolidation test results in the same area. Figure 5 indicates that the mean value of the hydraulic conductivity of contaminated clay under the same consolidation stress is slightly larger than that of uncontaminated clay. However, this difference is not significant. Clay permeability is primarily affected by factors such as soil porosity and specific surface area, such that the relationship between the three can be expressed by Eq. (3), where the specific surface area can be replaced by plasticity index or clay content (Du et al. 2006; Zhan et al. 2014).

$$k = c_s \bullet \left(\frac{\gamma_w}{\mu_w}\right) \bullet \left(\frac{e^3}{\mathrm{SSA}^2(1+e)}\right) \quad (3)$$

where c_s is the pore shape parameters, μ_w is the coefficient of viscosity of water (kPa·s), and SSA is the specific surface area (cm^2/g).

Based on Eq. (3), the mechanism of leachate influence on clay permeability is manifested in two aspects. Firstly, the erosive effect of leachate decreases the plasticity index, clay particle content, and specific surface area of clay, leading to increased clay permeability. Secondly, leachate interactions lead to a decrease in the porosity of contaminated clay under the same consolidation stress (as shown in Fig. 4), causing clay permeability to decrease. Therefore, the effect of leachate on clay permeability is a combination of the above two positive and negative factors. For the same porosity, the permeability of leachate sludge clay is 2–4 times higher than that of uncontaminated clay (Fig. 6).

Based on previous literature, this paper summarizes the hydraulic conductivity of clay under the influence of chemical solution, as shown in Table 4. Geosynthetic clay liner (GCL) is more sensitive to chemical solutions, and the coefficient of hydraulic conductivity tends to increase, whose maximum increase is 284 times (Xue et al. 2012; Wang et al. 2019; Setz et al. 2017; Bradshaw and Benson 2014). The experimental laboratory results on the hydraulic conductivity of compacted clay layers (CCL) under the chemical solutions need further investigation. The permeability properties of compacted clay are affected by various factors, and cations in leachate can compress the electric double layer of clay minerals and increase the permeability of clay. The clogging effect of the biochemical reaction of leachate and the self-weight stress consolidation of the waste dump will both reduce the permeability of the clay. The results of Li-wen et al. (2009) and Aldaeef and Rayhani (2014) are similar to this paper, whose the coefficient of hydraulic conductivity of CCL increased approximately 2 times in the presence of metal ions and simulated leachate. Mortezaei and Karimpour Fard (2017) used methanol as the permeation solution and showed a 10-time increase in hydraulic conductivity coefficient. However, some studies in the literature have shown that the coefficient of hydraulic conductivity decreases (Li et al. 2013; Ozcoban et al. 2013). Therefore, the increase or decrease in the permeability properties of the landfill clay layer under the action of leachate is the result of the combined effect of multiple factors. The results of in situ experiments can more realistically reflect the changes in permeability properties of compacted clay layers under the long-term action of leachate.

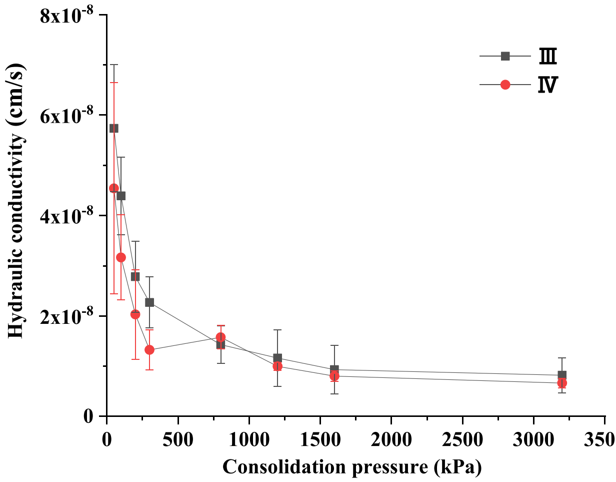


Fig. 5 Variation of the hydraulic conductivity with the consolidation stress of in situ clay

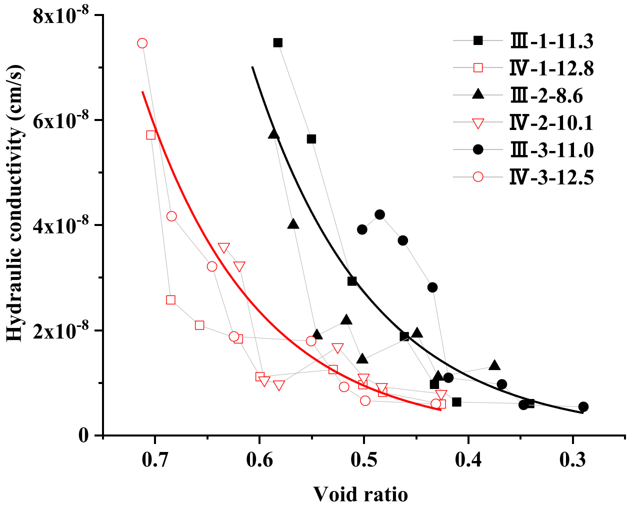


Fig. 6 Variation of hydraulic conductivity against the void ratio of in situ clay

Microstructure test results and analysis

MIP test results and analysis

Figure 7 shows the pore size distribution curves of long-term leachate-contaminated and uncontaminated in situ clay.

Table 4 Comparison of laboratory results of coefficient of hydraulic conductivity

Sample	Solution	Coefficient of hydraulic conductivity	Reference
GCL	$CaCl_2$, 200 mM	Increased 284 times	(Xue et al. 2012)
GCL (natural)	A mixed solution of calcium and sodium chloride	No obvious changes	(Wang et al. 2019)
GCL (sodium)	A mixed solution of calcium and sodium chloride, 1500 mg/L	Increased 86 times	(Wang et al. 2019)
GCL	Ammonium	3.3 times at low concentration (100 mM); 105 times at high concentration (500 mM)	(Setz et al. 2017)
GCL	Landfill leachate	Increased 5.6 times	(Bradshaw and Benson 2014)
CCL	Multiple metal ion solutions, 0.005 mol/L, 0.01 mol/L, 0.02 mol/L	Increase 2 times	(Li-wen et al. 2009)
CCL	Methanol	Increased 10 times	(Mortezaei and Karimpour Fard 2017)
CCL	2 500 mg/L acetic acid solution, 0.5 mol/L $CaCl_2$ solution	No adverse effect	(He et al. 2015)
CCL	Simulated leachate	For different types of clay, the increase is 1.8–2.71 times	(Aldaeef and Rayhani 2014)
CCL	Landfill leachate	Reduced by 1/3	(Li et al. 2013)
CCL	Landfill leachate	Reduced by 1/20	(Ozcoban et al. 2013)

This indicates a bimodal pore distribution curve, featuring a dividing point between the two peaks of around 1 μm, such that large pores (pore sizes greater than 1 μm) occur mainly between agglomerates while small pores (less than 1 μm) occur mainly within agglomerates and between particles (Xue et al. 2014). Based on pore size distribution tests, the volume of large pores in uncontaminated in situ clay (IV-1–12.8 and IV-2–10.1) was calculated to be 0.064 cm^3/g and 0.053 cm^3/g (mean 0.059 cm^3/g), respectively, while the corresponding volumes of small pores were 0.168 cm^3/g and 0.195 cm^3/g (mean 0.181 cm^3/g). In contrast, the large pore volumes of the in situ clays exposed to long-term leaching (III-1–11.3 and III-2–8.6) were 0.132 cm^3/g and 0.102 cm^3/g (mean value 0.117 cm^3/g), while their small pore volumes were 0.155 cm^3/g and 0.154 cm^3/g (mean value 0.155 cm^3/g), respectively. Large pore volume increased substantially (by 132%) and small pore volume decreased slightly (by 14.4%) following long-term leachate interaction. Compression of the clay particles' diffuse double layer due to the high concentration of cations in leachate leads to the agglomeration of clay particles, i.e., the main reason for the increase in macropore volume and decrease in micropore volume in leachate-interacted clay (Ma et al. 2019; Yao et al. 2020). The reduction of expansive clay minerals, such as montmorillonite, during leachate interaction, is also one of the factors leading to the abovementioned changes in pore distribution (Li et al. 2013; Oztoprak and Pisirici 2011).

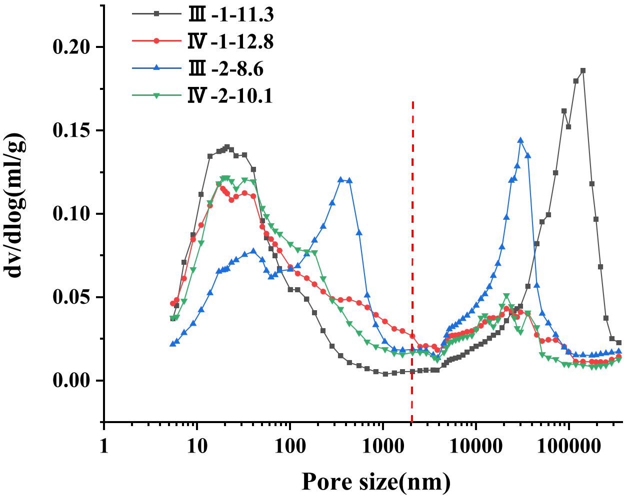
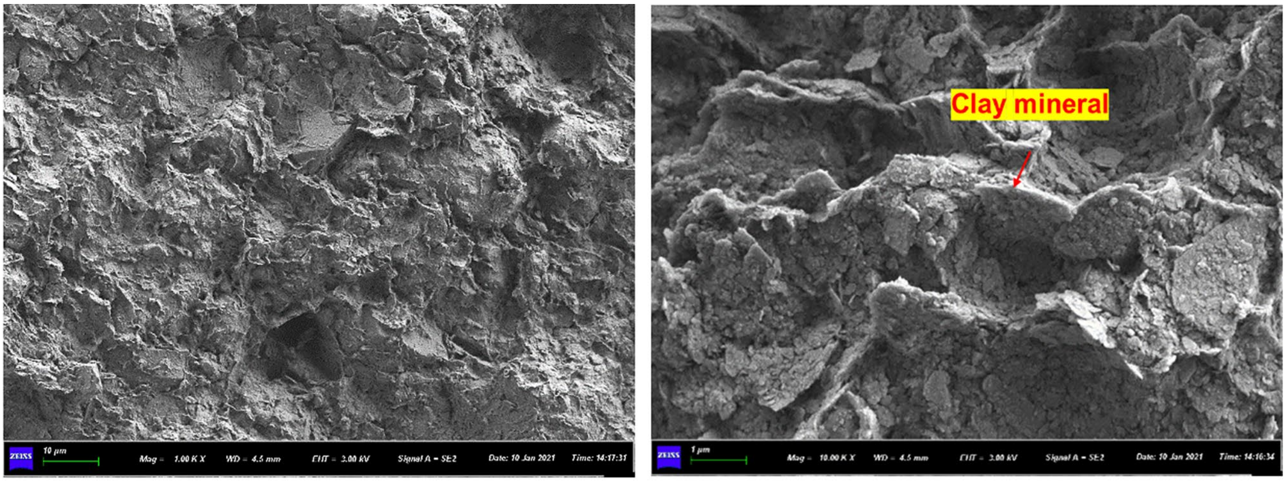


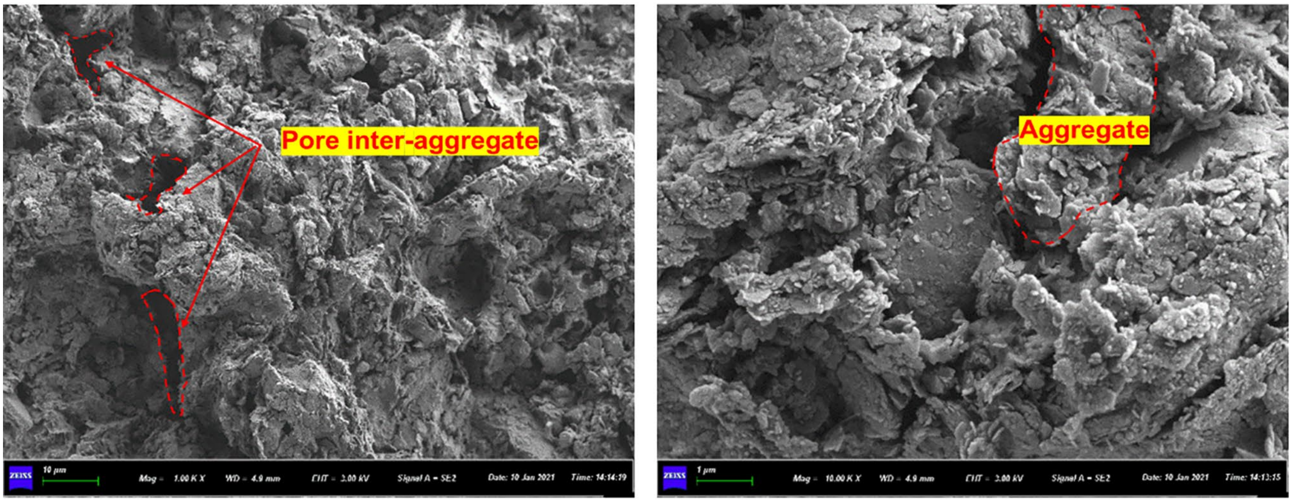
Fig. 7 Pore size distribution curve of in situ clay

SEM test results and analysis

Figure 8 shows results from SEM analyses of long-term leachate-contaminated and uncontaminated clay. This indicates that uncontaminated clay (IV-1–12.8) has a dense structure with lamellar clay minerals mainly in face-to-face contact. Following long-term interaction with the leachate, the microstructure of the clay (III-1–11.3) changed significantly, developing a loose soil structure, macropore development (consistent with MIP results in Fig. 7), and increased pore connectivity. The mechanism of the influence of waste leachate on clay microstructure is mainly reflected in two aspects. Firstly, long-term interaction with leachate leads to the reduction of clay mineral content, such as montmorillonite (see Table 1 and previous studies) (Li et al. 2013;



a. IV **-1-12.8**



b. III **-1-11.3**

Fig. 8 SEM observations of in situ clay. **a** IV-1–12.8. **b** III-1–11.3

Oztoprak and Pisirici 2011). Secondly, the high concentration of cations in leachate compresses the thickness of the diffuse double layer at the surface of clay particles, leading to the coalescence of soil particles and an increase in macropore volume and connectivity.

Environmental chemical test results and analysis

Analysis of groundwater test results and pollution status

According to our survey, the pH of landfill waste ranged from 7.9 to 9.05, i.e., weakly alkaline, while organic matter contents ranged from 4.1 to 17.2%, characteristics of waste degradation between the middle and late stages of maturity. Lignin to cellulose ratios ranged from 0.13 to 0.31, indirectly indicating that the degradation of domestic waste had not yet reached stabilization; i.e., it was not undergoing mineralization and still had biogas production capacity. The test results of leachate showed the following characteristics (Table 5): COD and ammonia nitrogen occurred at low levels, but color, turbidity, and ammonia nitrogen had not yet reached the China Standard for Pollution Control on the Landfill Site of Municipal Solid Waste (GB16889-2008), the pH value is 7.42, i.e., alkaline and similar to the monitoring results of the waste pile, in terms of heavy metal pollutants, mercury, arsenic, lead, and others meet the emission limits specified in GB16889-2008. According to the China Standard for Groundwater Quality (GB/T 14848-2017), fifteen indicators in leachate exceeded the requirements of the standard V limit value, yet many indicators in groundwater did not exceed the requirements of the standard V limit value. Thus, it was judged that the landfill leachate pollutants had not penetrated

Table 5 Leachate and groundwater testing results

Indicator	Leachate	Concentration limits, GB16889	Groundwater	Concentration limits, GB/T14848
Chromaticity	400	40	<5	≤ 15
Smell	Very significant odor	-	No strange smell and odor	No
Turbidity, NTU	444	30	2.6	≤ 3
Visible objects	Large amounts of visible matter	-	No	No
Total hardness (as $CaCO_3$), mg/L	415	-	186	≤ 450
Total dissolved solids, mg/L	2.53×10^3	-	320	≤ 1000
Chloride, mg/L	404	-	3.46	≤ 250
Manganese, mg/L	1.14	-	0.0239	≤ 0.10
Volatile phenols (by phenol), mg/L	0.0058	-	<0.0003	≤ 0.002
Oxygen consumption (COD_{Mn} method, in terms of O_2), mg/L	65.7	100	2.4	≤ 3.0
Ammonia nitrogen (as N), mg/L	152	25	0.32	≤ 0.50
Sodium, mg/L	585	-	6.78	≤ 200
Total coliforms, MPN/100 mL	$>1.6 \times 10^3$	10,000	2	≤ 3.0
Total number of bacteria colonies, CFU/mL	1.8×10^5	-	80	≤ 100
Fluoride, mg/L	1.57	-	0.22	≤ 1.0

the bottom of the landfill and the surrounding clay layer, and thus had not polluted the groundwater around the landfill.

Chemical test results of pore solution and contamination migration analysis

Previous research has shown that a large number of inorganic cations, such as Na^+, K^+, and Ca^{2+}, contained in the leachate can compress the diffuse double layer on the surface of clay particles and trigger their coalescence, leading to an increase in hydraulic conductivity by more than one order of magnitude, as well as a weakening swelling capability and increased compressibility (Benson and Meer 2009; Setz et al. 2017). When compared with the single mineral composition of GCL, the mineral composition of the clay layer is relatively complex. Hence, the laws by which leachate influences the geotechnical properties of the clay layer vary greatly and the mechanism of contaminant-clay interactions is more complicated.

According to the chemical composition of leachate shown in Table 5 and the conclusions of previous literature (Johnson et al. 1989; Quigley and Rowe 1986; Zhan et al. 2014), Na^+, Cl^-, and TN were selected as the characteristic pollutants in leachate at the study site. Variations in Na^+, Cl^-, and TN concentrations in soil pore solutions with depth are shown in Fig. 9; i.e., concentrations in the clay pore solution at the bottom of the landfill decreased rapidly with depth, and the diffusion depth was approximately 1 m. The pattern of variation was similar to that in Zhan et al. (2014). However, there were also notable differences. In previous literature, concentrations of Na^+, Cl^-, and TN in the clay pore solution at the bottom of landfill sites have been found to be significantly higher than those presented herein, with diffusion depths exceeding 3 m (compared with the approximate 1 m diffusion depth in this paper). Two main factors can be invoked to explain this: the first is that the concentrations of leachate contaminants are around one order of magnitude higher than in the leachate reported herein; secondly, the hydraulic conductivity of the clay is around one order of magnitude higher than that of the clay in this study.

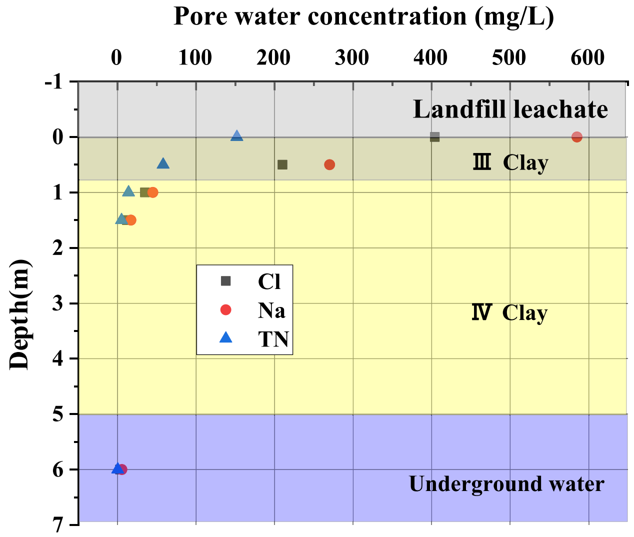


Fig. 9 Results of chemical analysis of the bottom clay pore solution

Conclusion

Due to leachate interaction time and clay structural properties, it is challenging to produce laboratory tests that truly reflect changes in the geotechnical properties of in situ clays after long-term (several decades) leachate interaction. In this paper, macro–microscopic experimental studies of in situ clays interacting with leachate for more than 20 years were carried out by means of landfill drilling and sampling methods. The main conclusions are as follows:

1. Physical property tests show certain differences between the properties of in situ clays in different areas of the landfill; these differences can be explained using statistical patterns. The long-term effect of leachate reduces clay particle content in the soil, such that the liquid limit of the soil undergoes a large reduction while the plastic limit undergoes a smaller reduction. This results in an obvious linear relationship between the soil limit water content and the clay particle content of the soil.
2. Geotechnical test results show that the internal friction angle of clay specimens after long-term leachate exposure increases while the cohesive force decreases. The compressibility of clay increases under the same consolidation stress, and hydraulic conductivity increases slightly under the same consolidation stress; however, under the same porosity conditions, the hydraulic conductivity of leachate-exposed clay is significantly higher than that of uncontaminated clay (2–4 times higher).
3. Microscopic results demonstrated that long-term exposure to leachate led to the reduction of clay minerals such as montmorillonite coupled with an increase of quartz content in the soil; soil particles coalesced, their structure changed from a dense to a loose state, and pore connectivity increased. In particular, the volume of large pores (> 1 μm) increased significantly (by 132%), and the volume of small pores decreased slightly (by 14.4%).
4. The results of environmental chemical tests show that landfill waste degradation at the study site is in the post-intermediate stage, with high sodium salt, chloride, and total nitrogen contents in the leachate, which has not yet reached the discharge limit specified in GB16889. The groundwater COD is slightly higher than the limit value of Class V in GB/T 14848. Concentrations of Na^+, Cl^-, and TN in the clay pore solution at the bottom of the landfill decrease rapidly with the depth, with a diffusion depth of approximately 1 m.

Author contribution Yong Wan: conceptualization, resources, writing—original draft. Dongdong Guo: data curation, methodology, writing—review and editing. Ruiqi Liu: investigation, methodology. Xinminnan Hui: data curation, methodology. Zhiwei Dong: investigation, conceptualization, writing—review and editing. Lei Liu: supervision, writing—review and editing. Qiang Xue: supervision, resources funding acquisition.

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Data availability The authors confirm that the data supporting the findings of this study are included in this article.

Declarations

Conflict of interest The authors declare no competing interests.

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