

## **IMPACTS OF LEAD NITRATE CONTAMINATION ON THE GEOTECHNICAL PROPERTIES OF CLAYEY SOIL**

MAHDI O. KARKUSH\*, SHAHAD D. ALI

College of Engineering, University of Baghdad, Aljadriah, Baghdad, Iraq

\*Corresponding Author: mahdi\_karkush@coeng.uobaghdad.edu.iq

### **Abstract**

Increasing the demand for using brownfields and to achieve sustainable development of reusing brownfields, it's important to study the impacts of contamination on the geotechnical properties of soil. The main aim of this work is studying the impacts of lead nitrate contamination on the geotechnical properties of clayey soil. The soil samples were obtained from the site of the Al-Ahdab oil field in the east of Iraq. The soil samples were contaminated artificially with three concentrations of lead nitrate (6666.67, 13333.33, and 20000) ppm. The contaminant solution consists of lead nitrate and 10 litres of distilled water was added to the intact soil. The soil sample remains soaked with the contaminant solution for one month. The chemical, physical, and mechanical properties of intact and contaminated soil samples were measured to investigate the effects of lead nitrate on these properties. The lead nitrate causes decreasing the percentage of particles having the size of clay (5.4 to 24.3%), Atterberg's limits, permeability (19.3 to 42.2%), and optimum water content of the soil (14.4 to 21.3%). In addition, the coefficient of consolidation and shear strength parameters of soil were decreased after contamination. Also, the specific gravity, maximum dry unit weight, initial void ratio, compression index, and recompression index were increased with increasing the concentration of lead nitrate in the soil by 1.5-3.3%, 2.5-4.9%, 2.9-13.5%, 89-195%, and 18.2-39.4% respectively.

Keywords: Cohesive soil, Geotechnical properties, Lead nitrate, Soil contamination.

## 1. Introduction

Recently, soil contamination with heavy metals (HMs) is one of the main problems facing environment. Several sources lead to contaminant the environment and soil with heavy metals such as agricultural, industrial and military activities. There are two types of HMs, the first type is pivotal to the human metabolic system, but when it is higher than the permissible limits lead to the adverse effects and the second type of heavy metals are not useful to human health and may cause negative effects even at low concentrations.

Also, the different properties of clayey soil can be changed when contaminated with heavy metals and this change relying on several main factors such as the mobility and chemical activity of contaminants in the soil, especially when the concentrations of contaminants existed in THE soil are above the accepted limits [1-3]. In Iraq, there are several sites contaminated with heavy metals like Al Suwaira, Khan Dhari, Al Mishraq and Ouireej, therefore, it is important to study the effects of these contaminants on the soil properties [4]. Because of the urban and industrial development and use soil in various engineering projects. The availability of soil that is empty from any type of contaminants reduced, so it would be a benefit to utilize the contaminated soil in the foundations and embankments of buildings, and roads, but it requires the special and thorough knowledge of their geotechnical properties [5].

The cohesive soils are electrochemically active and influenced whenever the environment is contaminated by wastes. The major factors that affect the existence of contaminant in the soil or in groundwater are the adsorption characteristics and permeability of the soil. Properties and chemical composition of soil are the major factors to specify the amount of contaminant in the soil [6]. There are several researches studied the effects of different types of contaminants on the geotechnical properties of soil. Ali [7] and Khamehchiyan et al. [8] studied the effects of oil contamination on the geotechnical characteristics of cohesive and cohesionless soils. The results showed that the shear strength parameters, hydraulic conductivity, maximum dry unit weight, optimum moisture content, and Atterberg's limits were decreased with increasing oil content in the soil.

Karkush et al. [1] described the influence of kerosene,  $\text{NH}_4\text{OH}$ ,  $\text{pb}(\text{NO}_3)_2$  and  $\text{CuSO}_4 \cdot 5\text{H}_2\text{O}$  on the different properties of soil. The results demonstrated different impacts of contaminants on the geotechnical and chemical properties of clayey soil depending on the type and concentration of contaminant in the soil. Karkush and Al-Taher [9] studied the impacts of total petroleum hydrocarbons (TPH) on the geotechnical behaviour of clayey soil. The results demonstrated that the TPH have slight to significant effects on the different geotechnical properties of clayey soil samples.

Karkush and Kareem [6] studied the effects of (10 and 20) % of medium fuel oil (MFO) on the geotechnical behaviour of cohesive soil. The experimental results detected that the mechanical properties of oil-contaminated soil samples were significantly affected by MFO, but other properties of soil (chemical and physical) were slightly affected. In the present study, the impacts of three percentages of lead nitrate contaminant on the geotechnical properties of cohesive soil have been investigated in details.

## 2. Sources of Heavy Metals in Soil

Weathering and human sources are the main reasons that led to soil contamination with heavy metals (HMs). Wuana and Okieimen [10] reported that anthropogenic sources of metal contamination can be isolated into five major groups:

- **Fertilizers:** Agriculture was the first major human effect on the soil. Some soils do not contain heavy metals or content, but in a very few percent, which is not enough for the growth of plants, so large amounts of fertilizers are added to the soil for the healthy growth of the plants. Continue adding chemical fertilizers to the soil lead to higher rates of HMs in the soil.
- **Pesticides:** In the past, many of pesticides utilized broadly in farming and horticulture contained substantial concentrations of metals, so lead to accumulation of HMs in soil.
- **Biosolids and Manures:** The application of various wastes solids such as (livestock manures, composts, and municipal sewage sludge) to the land unknowingly leads to the collection of heavy metals in soil. Some animal wastes are added to agricultural land as fertilizer but lead to the accumulation of heavy metals in the soil such as copper, arsenic and zinc.
- **Wastewater:** There is a common practice all over the world that is application sewage and industrial water flow on the ground. The farmers do not attend to the collection of contaminants in the environment and soil as much as they care to plants growth and increasing production, so Irrigation by using the wastewater, in the long run, led to the accumulation of heavy metals in the soil.
- **Metal Mining and Milling Processes and Industrial Wastes:** Mining and smelting industries in many countries, causing contamination of the soil and the environment significantly. Lead and zinc are the contaminants that are concentrated in the soil as a result of mining operations, which have a significant impact on human health and the environment. Other contaminants are generated from industries such as textile and tanning industries and the petrochemical industry by throwing wastes on the ground a little bit of this material can be useful in the process of agriculture, but many of them contain hazardous materials such as lead, zinc and chromium.

## 3. Soil Sampling and Material Used

The intact soil samples were brought from the site of Al-Ahadab oil field that located in Al-Ahrar city located in the north-west of Wasit province ( $E = 569974$ ,  $N = 359254$ ) at the east of Iraq. The saturated soil sample was obtained by drilling to a depth of 3 m below the existing ground level (EGL). The groundwater table was 2.65 m from EGL.

The undisturbed soil samples were obtained using Shelby tubes of sharp ends and undisturbed soil samples were obtained directly from the excavation. Plastic sacks were used to put the disturbed soil samples in them while using tightening wax to cover the undisturbed soil samples and then all soil samples were labelled and transported to the laboratory.

Figure 1 shows the shapes of some soil samples. The natural moisture content and field unit weight of intact soil were 28.6% and 19.85 kN/m<sup>3</sup>. The intact soil was classified as fat clay (CH) according to the unified soil classification system

(USCS). The fine-grained textured soil has a high response to environmental changes. Also, the chemical reactions between this type of soil and contaminants are higher than other soil types. The clayey soil has a large specific surface area, a high number of available active sites and a dynamic crystalline structure [5, 11].

Lead nitrate was added to soil samples as a chemical solution with different concentrations (6666.67, 13333.33 and 20000) ppm to investigate the effects of various concentrations of lead nitrate on the geotechnical properties of soil. The reason of choosing lead nitrate as an example of heavy metals contaminants is the availability of lead in large amount in the environment and this contaminant becomes the target of many studies to investigate its effects on the environment and soil [12]. Lead nitrate ( $\text{Pb}(\text{NO}_3)_2$ ) used in this study has a density of  $4.53 \text{ g/cm}^3$ , solubility in water of  $565 \text{ g/L}$ , and molar weight of  $331.2 \text{ g/mol}$ .



**Fig. 1. Soil samples.**

#### **4. Experimental Work**

The experimental work conducted in the present study includes the preparation of artificially contaminated soil samples and the measurement of the chemical and geotechnical properties of soil samples before and after contamination. The chemical, physical and mechanical properties of soil samples were measured according to the ASTM [13] specifications.

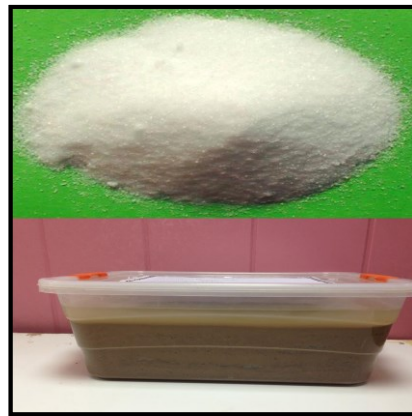
##### **4.1. Artificial contamination of soil samples**

Three amounts of disturbed soil samples (each part weighs about 15 kg) were put in three separate plastic containers and soaked with the contamination solution and covered with tighten covers and left for one month to permit the adsorption of pollutants by soil. The chemical solution consists of the contaminant and distilled water, where the concentration of lead nitrate in soil samples were (6666.67, 13333.33 and 20000) ppm. In the present study used different names of soil samples as given in Table 1.

The distilled water used to mix with lead nitrate is 10 L. This quantity of distilled water used because of the distilled water must be sufficient to submerge the soil sample and provide the height of water above the soil surface about equal to 3 cm in order to allow contaminant to permeate deeper in the soil and to assist in mixing of the contaminant into the soil simply [1, 14]. Figure 2 shows the lead nitrate and soaked soil sample preparation of clayey contaminated soil samples.

**Table 1. Designation of soil samples.**

Symbol	Definition	Concentration of $Pb(NO_3)_2$ (ppm)
<b>S0</b>	Undisturbed intact soil sample	0.0
<b>S0R</b>	Remoulded intact soil sample	0.0
<b>S1</b>	Soil sample contaminated with 100 gm of $Pb(NO_3)_2$	6666.67
<b>S2</b>	Soil sample contaminated with 200 gm of $Pb(NO_3)_2$	13333.33
<b>S3</b>	Soil sample contaminated with 300 gm of $Pb(NO_3)_2$	20000



**Fig. 2. Lead nitrate and soaked soil sample.**

#### 4.2. Chemical and geotechnical properties of soil samples

The chemical properties of soil sample are important in the constitution of soil structure and affected the physical and mechanical properties of the soil. The measured chemical properties are three sulphate ions ( $SO_3$ ), chloride content ( $Cl^{-1}$ ), silicon dioxide ( $SiO_2$ ), calcium oxide ( $CaO$ ), organic matter content (OMC), Gypsum, total suspended solids (TSS), and  $pH$  value. The physical properties of soil play an important role in the adequacy of the site for construction and using the soil as a construction material. The contamination of soil alters its properties.

The studied properties are particle-size distribution, specific gravity ( $G_s$ ), Atterberg's limits ( $LL$  and  $PL$ ), compaction curve, and permeability ( $k$ ). The construction of foundation requires a careful study for the mechanical properties of soil especially with increasing the demand for using the brownfields. The mechanical properties are important to calculate the soil bearing capacity and the expected settlement under the footing of the structure. Therefore, the investigation of soil

mechanical properties like the soil compressibility, which obtained by 1-D consolidation test; and shear strength of soil, which measured by direct shear test (DST), unconfined compression test (UCT), and undrained unconsolidated triaxial test (UUT).

## 5. Results and Discussion

This section includes the results of different tests, chemical, physical and mechanical tests of soil samples.

### 5.1. Chemical properties of soil samples

The chemical tests results are given in Table 2. X-ray diffraction tests were carried on two soil samples *S0* and *S2* because *S0* is the intact soil sample and *S2* is the soil sample that contains the average concentration of lead nitrate to study the effects of contamination on clay and non-clay minerals as given in Table 3. The concentration of trioxide sulphate ( $\text{SO}_3$ ), chloride ( $\text{Cl}^-$ ), total soluble salts (TSS) and gypsum increased in contaminated soil samples, while the *pH* value of the soil is decreased by the contamination.

The values of OMC and CaO are decreased in *S1* and *S2* soil sample but increased in soil sample *S3*. Also, the lead nitrate causes a decrease in  $\text{CaCO}_3$  and increasing the percent of Quartz and clay minerals. The geochemical factors affect the absorption of contaminants, so it is important to know the chemical composition of the intact soil. The main geochemical factors are organic matter content, cation exchange capacity (CEC), *pH* value and clay minerals [15].

**Table 2. Chemical properties of tested soil samples.**

Soil sample	$\text{SO}_3$ %	$\text{Cl}^-$ %	$\text{SiO}_2$ %	CaO %	OMC %	Gypsum %	TSS %	<i>pH</i> value
<i>S0</i>	0.036	0.5442	32.37	18.31	0.620	0.04	3.6	7.6
<i>S1</i>	0.761	0.569	33.21	17.91	0.516	1.203	3.36	7.4
<i>S2</i>	0.277	1.382	33.34	17.18	0.419	2.91	4.09	7.3
<i>S3</i>	0.374	0.693	31.65	18.18	1.214	3.81	3.74	7

**Table 3. Results of X-ray diffraction tests.**

Soil sample	Non-clay minerals, %				Clay minerals, %
	$\text{CaCO}_3$	Quartz	Dolomite	Dialuminium	
<i>S0</i>	46	25.9	11.7	12	4.4
<i>S2</i>	44.4	47.8	5.4	-	7.8

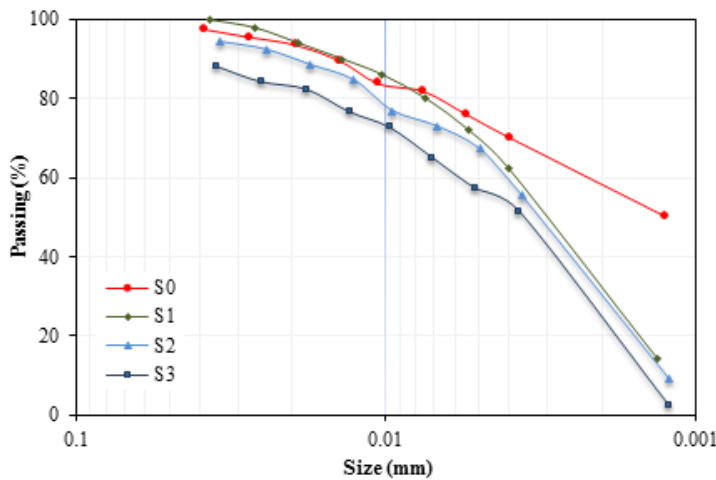
### 5.2. Physical properties of soil samples

The results of this test are given in Table 4 and shown in Fig. 3. The results showed that on the surface mineral of any particle, the contaminant with the oxygen atoms created covalent bonds. Also, the results demonstrate that the presence of lead in soil samples increases the percentages of particles have the size of silt and sand in the soil, which is compatible with results by Hussien [16].  $\text{CaCO}_3$  and OMC are the main agents to cement soil particles and are responsible for both sticking and stability of soil particles [17].

The electronegative is the main factor in determining any adsorption of metals by the soil in better form. The metals have a high value of electronegative, which form strong covalent bonds with oxygen atoms found on the mineral's surfaces. Depended on the basis of electronic activity, the expected order of bonding preference will be  $Cu > Ni > Co > Pb > Cd > Zn > Mg > Sr$  [18]. Lead has the ability to make covalent bonding better than several metals, so it effects on the sizes of soil particles appeared clearly.

**Table 4. Physical properties of tested soil samples.**

Property	Soil sample			
	S0	S1	S2	S3
Sand, %	0.020	0.102	0.187	0.273
Silt, %	25.98	29.898	31.813	43.727
Clay, %	74.00	70.00	68.00	56.00
G <sub>s</sub>	2.74	2.78	2.81	2.83
LL, %	55	45	45	43
PL, %	27	26	24	21
$\rho_{dmax}$ , g/cm <sup>3</sup>	1.678	1.720	1.754	1.760
$\omega_{opt}$ , %	21.6	18.5	17.5	17.0
$k \times 10^{-8}$ , cm/s	3.22	2.60	1.98	1.86



**Fig. 3. Particles-size distribution curves of tested soil samples.**

The specific gravity test results of intact and contaminated soil samples are given in Table 4. For contaminated soil samples, the specific gravity increases with increasing the concentration of contaminant in soil. The percentage of increase in specific gravity of soil sample S1, S2 and S3 are 1.5, 2.6, and 3.3% respectively. This increase is beyond to the high density of the contaminant existed in the soil, which leads to an increase in the density of soil [1]. The results of Atterberg's limits are given in Table 4.

The results show that the lead nitrate causes reduction of Atterberg limits with increasing the concentration of lead nitrate in the soil. There are several factors

affect the liquid and plastic limits such as the clay content, which is a major factor and when reduced will lead to a reduction in Atterberg limits and the HMs coated the clay particle. Also, the dissolution of salts may reduce the thickness of the diffused double layer and flocculation of the clay particles [19]. The results of falling head tests are given in Table 4.

According to the results, the permeability of contaminated soil samples is less than that of intact soil. The reduction in permeability of soil sample *S1*, *S2* and *S3* are 19.3, 38.5, and 42.3% respectively. The contamination of soil with HMs causes filling the voids between particles with salts [20]. The compaction curves are shown in Fig. 4 and the results are shown in Table 4. Increasing the contamination causes a clear change in the compaction curve shape, so the contamination increases the maximum dry density and decreases the optimum moisture content ( $\omega_{opt}$ ). The decrease in optimum moisture content results from the reduction of the voids among the particles due to sedimentation of salts in pores. The reduction in  $\omega_{opt}$  causes an increase in the maximum dry density. The contact points on the clay molecules are covered by heavy metal, so the engineering performance of the soil and the ability of clay to dissociating water molecules is decreased. The contamination of clayey soil with HMs will take more time or need more compaction effort to get the desired maximum dry density [21, 22].

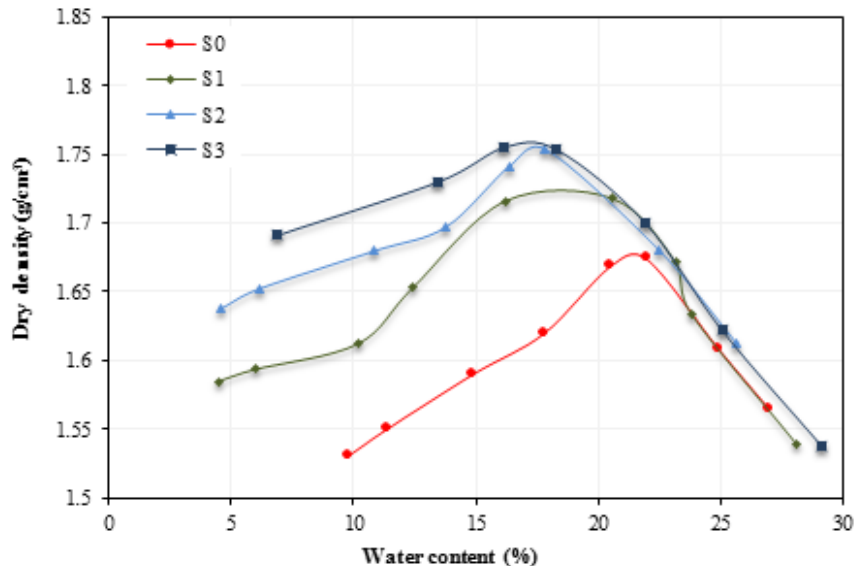


Fig. 4. Compaction curves of intact and lead-contaminated soil samples.

### 5.3. Mechanical properties of soil samples

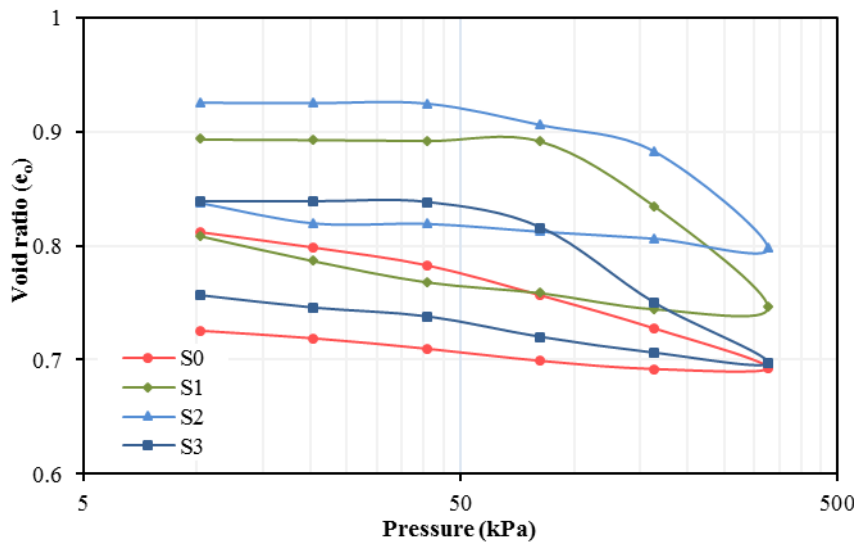
Consolidation test was conducted on intact and contaminated soil samples because it is considered as a major of mechanical tests and gives several indices, which show the behaviour of soil samples [23]. The initial void ratio ( $e_0$ ), compression index ( $c_c$ ), recompression index ( $c_r$ ), pre-consolidation pressure ( $P_c$ ) and coefficient of consolidation ( $c_v$ ) are given in Table 5 and shown in Fig. 5. A significant decrease in the coefficient of consolidation is noticed with increasing



the concentration of lead nitrate in the soil. This decrease may be attributed to the decrease in the permeability of contaminated soil [20]. Also, the results show increasing the initial void ratio, compression index, and recompression index. The initial void ratio was increased due to decreasing the percentage of particles have the size of clay particles, which create a large space among the soil particles of contaminated samples. Also, the increasing in  $c_c$  and  $c_r$  because of the presence of lead nitrate in the soil pores help to slide of soil particles and perhaps due to the charge of the molecules and the nature of the fluid, which impacts the adsorbed cations [9, 24].

**Table 5. Results of 1-D consolidation tests.**

Soil sample	$e_o$	$c_c$	$c_r$	$P_c$ (kPa)	$m_v$ (m <sup>2</sup> /kN)	$c_v$ (cm <sup>2</sup> /s)
S0	0.816	0.100	0.033	69	0.00020	0.00054
S1	0.894	0.252	0.042	118	0.00032	0.00043
S2	0.926	0.295	0.039	150	0.00027	0.00038
S3	0.840	0.189	0.046	70	0.00026	0.00036



**Fig. 5. Variation of void ratio versus pressure of tested soil samples.**

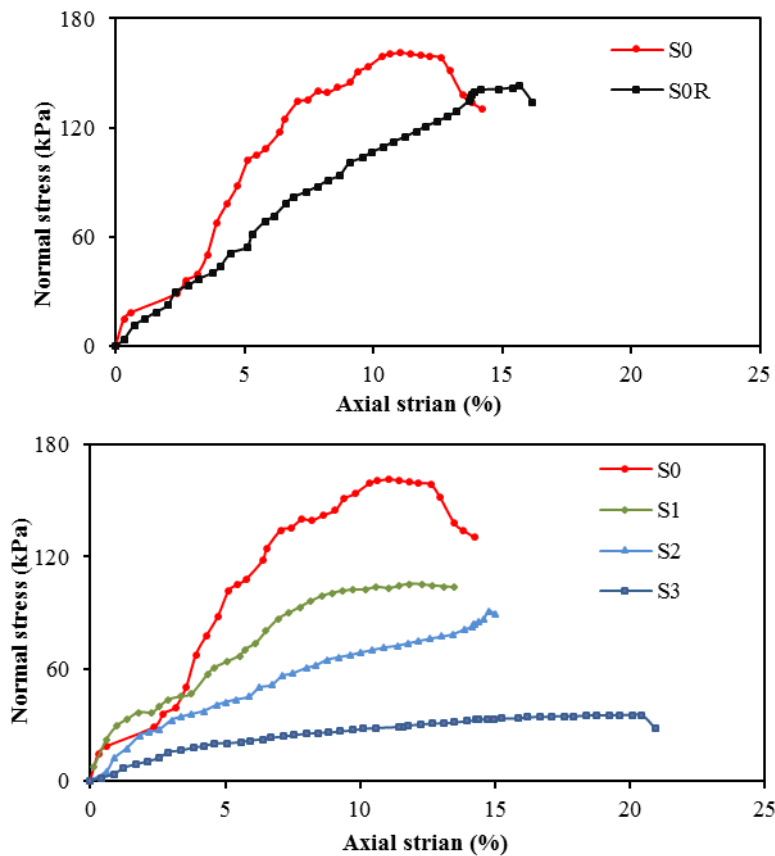
Direct shear was conducted to obtain the shear strength parameters, cohesion ( $c$ ) and angle of internal friction ( $\phi$ ). The results indicated a reduction in  $c$  and  $\phi$ , which resulted from the lessening in friction' and bond' that grips the soil' particles' together in the soil mass. The heavy metal contaminants are infiltrated between the particles of soil in the voids when it spills on soil and forms a thin layer of coats around the particles, in this way prevent the expansion of the cohesive forces between molecules that responsible about the bond of the particles of cohesive soil [25]. The UCT was carried on two types of intact soil (undisturbed and remoulded) to measure the influence of sampling disturbance on the shear strength of the soil. The variation of the stress-strain relation of soil samples are shown in Fig. 6. The sensitivity of soil ( $S_t$ ) can be calculated using Eq. (1). According to Eq. (1), the

sensitivity of soil is about 1.125 and the soil sample can be classified as slightly or low sensitive clays [26, 27]. The heavy metals contamination has negative effects on the cohesion of soil. This behaviour may be reflecting the coating of soil solids with heavy metals that causes of solid particles. The shear strength parameters of soil obtained from DST, UCT, and UUT are given in Table 6.

$$S_t = \frac{\text{Umdrained shear strength of undisturbed soil sample}}{\text{Umdrained shear strength of remolded soil sample}} \quad (1)$$

**Table 6. Results of shear strength tests.**

Soil sample	DST		UCT		UUT	
	<i>c</i> (kPa)	$\phi$ (Degree)	$\epsilon$ (%)	<i>q<sub>u</sub></i> (kPa)	<i>c<sub>u</sub></i> (kPa)	<i>c</i> (kPa)
S0	108	13.4	11.050	161	81	125
S0R	-	-	15.700	143	72	105
S1	59	6.0	11.842	105	53	65
S2	26	5.7	14.750	91	46	56
S3	18	6.0	20.434	35	18	30



**Fig. 6. Stress-strain relationship of unconfined compression tests.**

The triaxial test consist of several stages, in the first stage the saturation of the soil sample must be done, the second stage includes applying cell pressure ( $\sigma_3$ ) and then in the third stage is applied the deviator stress until failure occurs. The cohesion of soil particles is highly influenced when the contaminant entered to the soil matrix. The bounded between soil particles are broken when the salts of heavy metal introduce into the soil, so lead to decreases the value of cohesion after contamination soil. The variation of deviatoric stress versus axial strain are shown in Fig. 7.

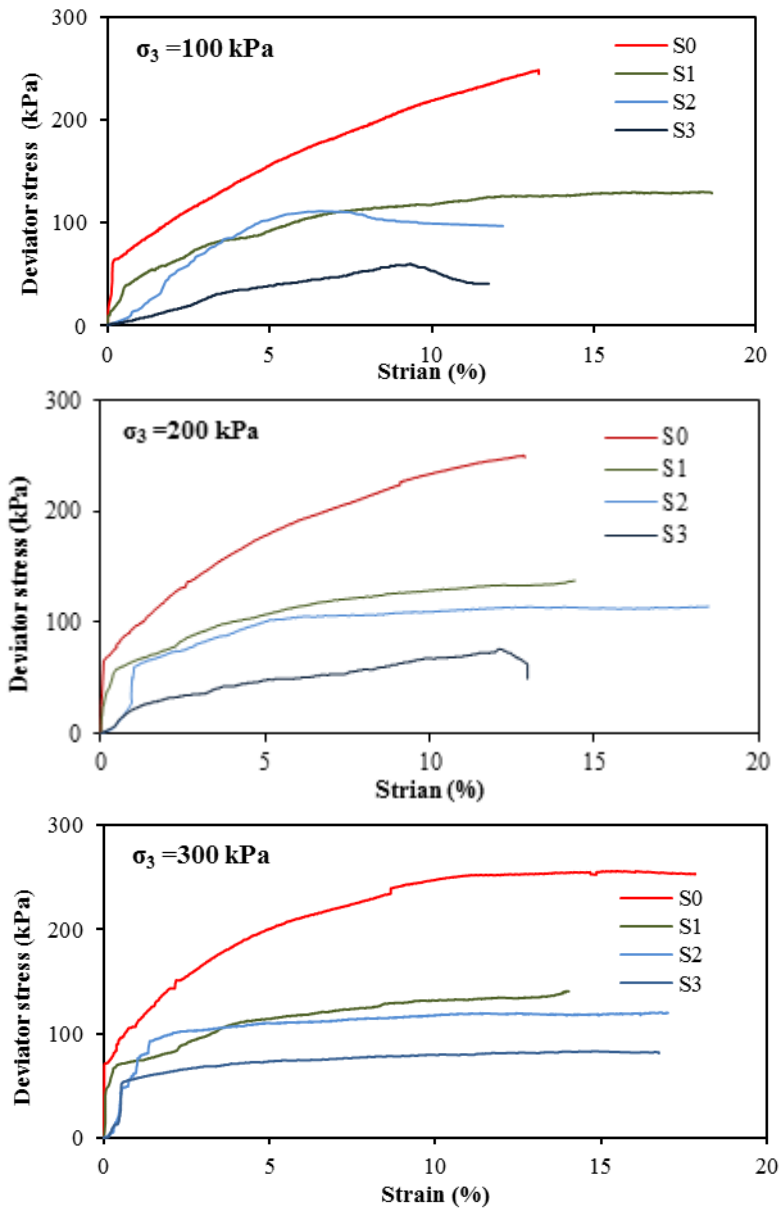


Fig. 7. Results of unconsolidated undrained triaxial tests.

## 6. Conclusions

Depended on the results of experimental tests can be extraction the next conclusions:

- The lead nitrate contaminant has negative effects on the various properties of soil (physical, chemical, and mechanical) and the effects mainly rely on the concentration of contaminant in the soil samples.
- The contamination causes decreasing the percentage of fine particles (size less than 0.005 mm) by 5.4 to 24.3% with increasing the concentration of lead nitrate in soil from 6666.67 to 20000 ppm.
- Also, contamination with lead nitrate causes a reduction in the hydraulic conductivity of soil by 19.3 to 42.2%, optimum water content by 14.4 to 21.3%, but the contaminant lead to increasing the specific gravity by 1.5 to 3.3% and the maximum dry density of soil by 2.5 to 4.9%.
- The coefficient of consolidation of contaminated soil samples decreased by 20.4 to 33.3%, while, the initial of void ratio, compression and swelling indexes and the coefficient of volume change are increased. Generally, the shear strength parameters (cohesion and angle of the internal friction) are decreased in contaminated soil samples.

### Nomenclatures

$c$	Cohesion of soil, kPa
$c_c$	Compression index
$c_r$	Recompression index
$G_s$	Specific gravity
$HC$	High plasticity clay
$k$	Coefficient of permeability, m/s
$LL$	Liquid limit, %
$PL$	Plastic limit, %
$pH$	Potential of hydrogen

### Greek Symbols

$\rho_{d,max}$	Maximum dry density, g/cm <sup>3</sup>
$\varphi$	Angle of internal friction, degree
$\omega_{opt}$	Optimum moisture content, %

### Abbreviations

ASTM	American Society for Testing and Materials
CEC	Cation Exchange Capacity
DST	Direct Shear Test
EGL	Existing Ground Level
HMs	Heavy Metals
MFO	Medium Density Fuel Oil
OMC	Organic Matter Content
TDS	Total Dissolved Solids
TPH	Total Petroleum Hydrocarbons
UCT	Unconfined Compressive Strength Test
UUT	Unconsolidated Undrained Triaxial Test

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