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Lime treatment of a diesel-contaminated coarse-grained soil for reuse in geotechnical applications

Natalia de Souza Correia^{1*†}[®], Fernando Henrique Martins Portelinha^{1†}[®], Igor Santos Mendes^{1†} and Jose Wilson Batista da Silva^{2†}

*Correspondence: ncorreia@ufscar.br [†]Natalia de Souza Correia, Fernando Henrique Martins Portelinha, Igor dos Santos Mendes and Jose Wilson Batista da Silva contributed equally to this work ¹ Department of Civil Engineering, Federal University of Sao Carlos, Sao Carlos, Sao Paulo, Brazil Full list of author information is available at the end of the article

Abstract

Potential reuse of oil-contaminated soils guidance policies are demonstrating ways to reuse these materials in engineering applications. However, the engineering properties of these materials are highly altered after contamination. Alternatively, chemical stabilization treatments can improve oil-contaminated soil properties for its reuse in geotechnical applications. This study investigates the effect of a diesel contamination on a coarse-grained soil and assesses the viability of a lime treatment. Laboratory tests included pH, Atterberg limits, compaction, unconfined compression strength (UCS), California Bearing Ratio (CBR) and tensile strength, RX-diffraction and scanning electron micrographs before and after lime treatment. Tests were performed on natural soil, diesel-contaminated soils (2, 4, 8, 12, and 16% of diesel) and lime treated dieselcontaminated soils (2, 4, 6 and 8% of lime). Diesel drastically changed soil plasticity and strength properties. Lime stabilization of diesel-contaminated soils was efficient to recapture soil natural properties with low lime contents. The influence of oil and lime in soil mechanical properties and mineralogical characteristics was evidenced in this study. The presence of oil drastically altered soil mechanical properties. Crystallization of calcite was present in both natural and oil-contaminated soils treated with lime, increasing particles flocculation. The presence of oil favored dolomite formation. Lime was significate in enhancing oil-contaminated soil mechanical properties, such as UCS, indirect tensile strength and CBR, mainly due to carbonation reactions. Oil-contaminated soil mixtures showed increase in mechanical properties after lime treatment and curing period.

Keywords: Soil mechanics, Oil-contaminated soil, Lime stabilization, Diesel, Soil pollution, Geotechnical properties

Introduction

Oil-contaminated soils result from leaking underground storage tanks and petroleum wells, or oil spills during transportation, tanker accidents, damaged pipelines, oil drilling processes and surrounding petroleum refineries. Soil contamination is a serious problem worldwide and poses a major environmental hazard [1-3]. Its exploitation, stevedoring,



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Fuel underground storage tanks represent a potential source of contamination worldwide. The most common causes for tanks leaking are related to structural deficiencies, mainly a result of inappropriate installation and corrosion of their internal and/or external lining. Petroleum hydrocarbon contamination of soil causes alterations in its physical, chemical and geotechnical properties and the degree of alteration depends on the soil type and the type and concentration of the contaminant [5–7]. However, potential reuse of petroleum-contaminated soils guidance policies are demonstrating ways to reuse non-hazardous petroleum-contaminated soils in civil engineering applications, such as asphalt concrete, cold-mix asphalt, construction material, roadway sub-bases and alternative daily cover materials for landfills [8, 9].

According to Pandey and Bind [5], for a potential reuse, it is necessary to determine the effects of oil contamination on soil engineering properties since they may drastically change and made the soils unsuitable for supporting engineering structures. Therefore, engineering properties of oil-contaminated soils have been the subject of many researches over the last years, due to increasing occurrence, and its importance in the context of reuse and environmental impacts minimization [1, 4, 7, 10–12].

Kermani and Ebadi [13] suggests, based on the study of engineering properties of oilcontaminated soil, the use of improvement methods to convert these soils into a reusable material, for road subbases or parking lots surface materials, after mixing it with stabilization agents, making an economic and time-efficient technique. The stabilization can be done by the addition of fly ash, lime or Portland cement, or even a combination of these, which often results in a pozzolanic reaction. In addition, although the addition of cement is the most successfully used technique, the high cost of cement motivated the search for alternative stabilizers.

Tuncan et al. [14] studied a petroleum-contaminated soil stabilized with 5% cement, 10% fly ash and 20% lime and showed superior strength properties among other contents tested. Shah et al. [15] evaluated a fuel oil contaminated clayey soil treated with different stabilization agents in terms of improvements in the geotechnical properties and observed superior results when the soil was treated with a combination of 10% lime, 5% cement and 5% fly ash, which was attributed to dispersion of oil, cation exchange, agglomeration, and pozzolanic reactions.

A mixture of oil-contaminated sand and cement kiln dust was evaluated by Nars [3] in terms of compaction and strength behavior for the construction of rural roads. 2% oil content and comparing with not stabilized contaminated sand, CBR values increased by about 37% and 47.6%, respectively, when the cement kiln dust content increased from 10 to 15%. In the research of Ochepo and Joseph [2], the influence of lime curing period on the strength properties of an industrial oil-contaminated soil was evaluated using unconfined compression strength tests. Results showed that curing period of 28 days allowed oil-contaminated soil stabilizes with 1% lime to achieve the strength properties of natural soil. George et al. [16] carried out studies to evaluate the efficacy of fly ash as stabilizing agent on the geotechnical properties of a diesel-contaminated sandy soil. Results showed compaction characteristics were observed to be not uniform with increase in fly ash, and CBR values decreased

with addition of contaminant, regaining increase with fly ash addition. Yu et al. [17] presents an experimental investigation on the stabilization of contaminated soils with Portland cement, motivated by reclamations of industrial lands in coastal cities of China. Experimental results showed that the geotechnical properties of plant oil-contaminated soils were very poor, but with application of cement, the oil-contaminated soil plasticity index decrease dramatically and the strength property was improved. Nasehi et al. [18] evaluated the use of nanoscale hydrated lime on 9% gascontaminated clayey soil. Results showed that adding 5% of lime to the contaminated soil increased UCS, shear strength parameters, LL, PL, optimum moisture content and decreased plasticity index and maximum dry density of the soil after 24 days. Chen et al. [4] evaluated a diesel-contaminated soil treated with cement, showing that different combinations of curing time, diesel and cement content led to different strength increase processes. According to Chen et al. [4], the lower viscosity of the diesel, in comparison to crude oil, makes it a powerful contamination potential in cases of oil spillage and soil infiltration.

The present study aims to evaluate a diesel-contaminated lateritic soil treated with different contents of lime. Comparative results of diesel-contaminated soil and natural soil–lime mixtures were used to evaluate the influence of oil contamination and lime as stabilizer. Tests include pH analysis, Atterberg limits, compaction properties, unconfined compression and tensile strength properties. Additionally, X-ray diffraction and scanning electron micrographs were used to evaluate soil structure and mineral neoformation.

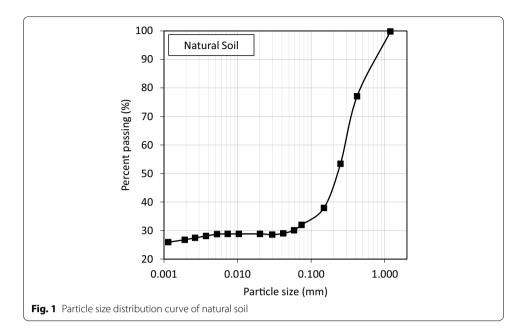
Materials and methods

Soil

Soil samples used in this study were collected in Sao Carlos, in the state of Sao Paulo, Brazil, and was not contaminated in its natural condition. Based on the Unified Soil Classification System [6], the coarse grain soil is classified as SC soil and mineralogical characteristics include quartz, feldspars, iron oxides and Kaolinite Hematite and Chlorite as secondary minerals. Characterization tests were conducted to obtain soil physical and compaction properties, including: specific gravity analysis [19], Atterberg limits analysis [20], Proctor tests [21], grain size distribution tests [22], CBR [23] and pH analysis [24]. Figure 1 presents particle size distribution curve of natural soil. Table 1 shows physical and compaction properties of the natural soil and particle size distribution.

Diesel oil and lime

The diesel oil used in the present study as the organic contaminant is a commercially available diesel oil named S500. Diesel properties include relative density of 0.834 at 20 °C [25], kinematic viscosity of $2.0-5.0 \text{ cm}^2 \text{ s}^{-1}$ at 40 °C [26], pH of 6.0 and biodiesel up to 7.0% v/v. Diesel oil contain compounds from BTEX group, represented by benzene, toluene, ethylbenzene, and xylene, as well as polycyclic aromatic hydrocarbons. For the chemical stabilizer, a commercially available hydrated lime type CH-III category was used in the experiment.



Physical properties	Quantity
Particle-size distribution	
Coarse sand (4.75–0.42 mm)	22.3
Fine sand (0.42–0.075 mm)	45.7
Fines (0.075 mm)	32.0
Specific gravity, Gs	2.84
Liquid limit (%)	33.4
Plastic limit (%)	16.0
Plastic index (%)	17.4
Maximum dry unit weight (kN/m³)—Standard effort	19.6
Optimum moisture content (%)	12.2
pH (in water)	5.4
CBR (%)	29.6

Table 1	Properties of	f the natural so	oil sample used in the tests
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Preparation of oil-contaminated and lime-treated soil samples

In this research, diesel oil was directly mixed with dry soil to prepare oil-contaminated soil samples. The different ratios of diesel to dry soil were set to 4, 8, 12 and 16% to simulate different levels of contamination in the field [2–4]. During samples preparation, soil was oven-dried in a temperature of 105 °C for 24 h, sieved in #4.76 mm, and mixed to form homogeneous mixtures. Changes in final oil content due to evaporation were not considered in this research.

Shah et al. [15] found that the weight percentage of a real fuel oil contamination from leakage of storage tanks in India varied between 7% and 10% of dry soil weight. Then, in order to analyze the influence of lime treatment on a diesel-contaminated soil, the proportion of diesel to dry soil of 8% was chosen. Then, lime treatment was added to 8% diesel-contaminated soils dry weight, in contents of 2, 4, 6 and 8%.

Additionally, in order to analyze the influence of lime in natural soil, contents of 2, 4, 6, and 8% of lime to dry soil were evaluated.

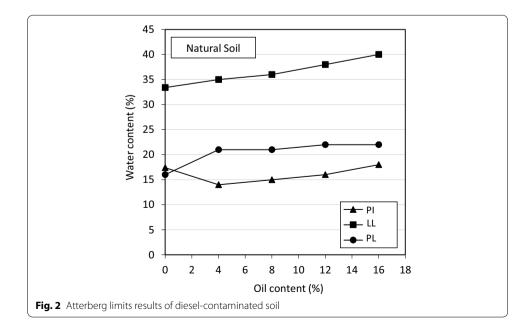
Results

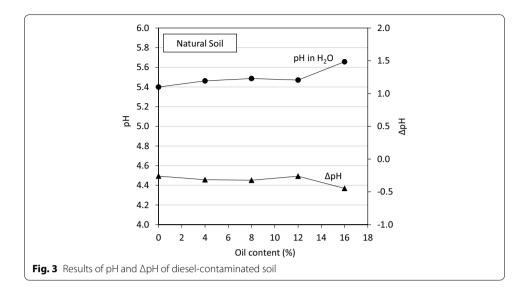
Effect of diesel-contamination on soil properties

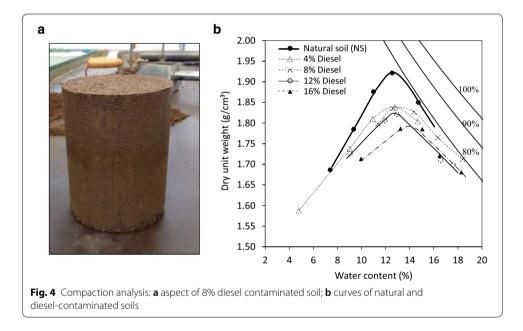
This topic describes the effect of diesel contents on the characteristic properties of the natural coarse-grained soil. Atterberg limits and pH are assumed as representative of characteristic properties that involves soil physicochemical changes. Figure 2 presents the effect of diesel contents on Liquid Limit (LL), Plasticity Limit (PL) and Plasticity Index (PI) of the natural coarse-grained soil. The influence of diesel content indicated increase in soil LL due to the presence of diesel. PL values also increased with oil diesel addition, maintaining constant values with higher diesel contents. Consequently, PI of diesel-contaminated soil increased with diesel content increases. In the research of Kermani and Ebadi [13] where a silt soil contaminated with crude oil was used, plasticity and liquid limits increased as oil content increased. This behavior is expected in the soil particles and water interaction due to oil presence.

The pH exerts a great influence on tropical soils due to the presence of minerals of variable charges and the ΔpH (pH_{KCl} – pH_{H20}) can be an indicator of mineralogical instability [27], which indicates the magnitude of chemical changes of soil. Figure 3 shows the results of soil pH (in water) with different diesel contents, as well as results of ΔpH . Results of natural and diesel-contaminated soils converge to the same values of pH up to 12% of diesel. Values of pH increased in natural soil only for 16% of diesel content, mainly due to Diesel pH value (6.0) which is higher than soil pH. No significate changes were observed in ΔpH values, remaining negative for all diesel contents, thus not altering soil electric charges with diesel content increase.

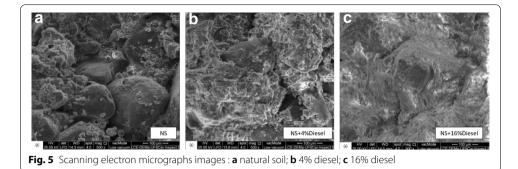
Figure 4 presents the aspect of a compacted soil sample with 8% of diesel content and the influence of oil in the compaction properties of natural soil. Results in Fig. 4b show

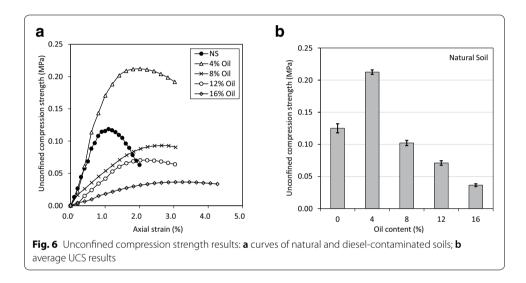






the shape of compaction curves obtained from Standard Proctor [21] for the natural soil and diesel-contaminated samples. It is observed that the behavior of soil compaction curves with diesel addition was significantly altered when compared to natural soil. Significant decrease in maximum dry unit weight (ρ_{dmax}) and an increase in optimum moisture contents (OMC) with diesel addition were observed. Safehian et al. [28] also indicated a decline in maximum dry density and an increase in optimum fluid content in presence of diesel in an illite soil. Accordingly, Safehian et al. [28] suggest that water has a greater influence on the compaction characteristics of illite in comparison to diesel, which is nonpolar and the clay particles do not have any tendency to adsorb them. Similarly, Khamehchiya et al. [29] found significant reduction in soil maximum dry unit weight using the same contents but for crude oil and different types of soils.





Scanning electronic microscope (SEM) analysis was performed on compacted samples of natural and diesel-contaminated soils (4%, and 16% of diesel) and are presented in Fig. 5. Samples were compacted at OMC and 95% compaction degree. According to Fig. 5a, in the natural compacted soil, aggregates of clay minerals and quartz particles are observed. In Figs. 5b and c, mixtures of soil sample with diesel oil caused a flocculated fabric in the soil particles, which increased with increasing diesel contamination. As observed by Nasehi et al. [18] in a study using CL soil and gas oil, this behavior is due to a combination effect of water and oil, which extended the flocculation in soil specimens.

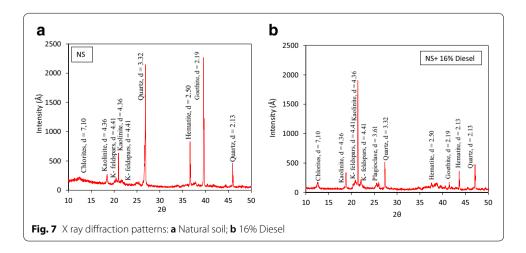
Unconfined compression strength (UCS) tests were conducted according to ASTM D2166M [30] in order to estimate soil mechanical strength alterations due to diesel contamination. Tests were duplicated with samples compacted at OMC and 95% of compaction degree. Figure 6 presents typical unconfined compression strength curves obtained in this study and average UCS values. Results in Fig. 6b demonstrate up to 70% reduction in soil UCS with diesel content increase. Coefficient of variation of maximum 5.8% were observed in the replicate results, validating samples used in this analysis. This behavior was found in several researches and was expected since oil was added to the soil structure and oil interaction. However, for 4% diesel content, a significant increase of 80% in soil UCS was observed, as well as comparative soil stiffness as in natural soil (Fig. 6a). The same behavior was observed for 4% crude oil in a clay sand used by Khamehchiyan et al. [29]. Ijimdiya [10] used a mixture of sand and crude oil and observed that for 2 and 4% of contamination, UCS increments were observed in comparison to natural soil. The author attributed these results to a bond-ing effect occurring between particles and oil, which results in aggregation of finer particles. Eissa et al. [1], Karkush et al. [6] and Nasehi et al. [7] likewise showed similar behavior for a specific content of oil contamination. Other studies found similar trend in higher UCS values for initial oil contents in the soil [1, 31].

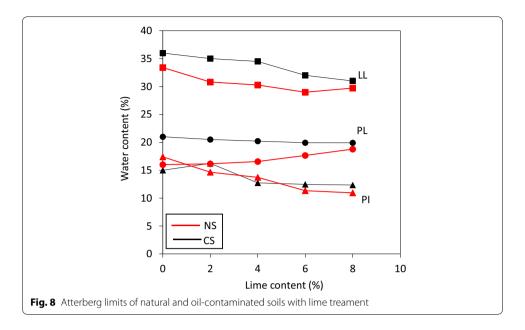
Figure 7 shows X-ray diffractometer analysis of both natural and diesel-contaminated soils that were carried out to evaluate the neo-formation of mineral/chemical compounds due to oil contamination. According to results in Fig. 7a, the natural soil mineralogy comprised mainly quartz, feldspar and clay minerals such as Kaolinite, Hematite, Goethite and Chlorite. In general, X-ray difractograms showed that the soil mineralogy was not altered with the addition of diesel. Echeverri-Ramírezet al. [32] found similar results for a soil contaminated with industrial soap, and suggests that the oil exposure time may influence soil mineralogy alterations.

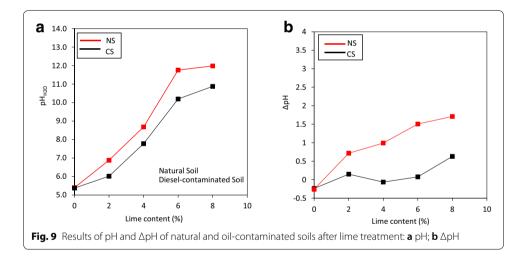
Effect of lime stabilization on diesel-contaminated soils

In this study, lime contents of 2, 4, 6 and 8% were added to an 8% diesel-contaminated soil (CS) in order to improve its geotechnical properties. For comparison purposes, natural soil (NS) was also stabilized with 2, 4, 6 and 8% of lime. Figure 8 presents the lime contents on Liquid Limit (LL), Plasticity Limit (PL) and Plasticity Index (PI) of natural coarse-grained soil and 8% diesel contaminated soil. Figure 8a shows that the addition of lime in both NS and CS soil provided reductions in LL. Values of PL remained constant for CS soil with lime addition and slightly increased with lime contents increase in natural soil. As regards PI results of NS and CS soils, this parameter was found to decreases with lime content increasing.

Figure 9 shows the influence of lime addition on pH and Δ pH of natural and oil-contaminated soils. Differently from the results of the influence of diesel on soil pH. The presence of lime increased pH and Δ pH values in both soils condition due to its alkalinity, even for small lime contents. Nasehi et al. [18] showed that adding 5% of lime to a

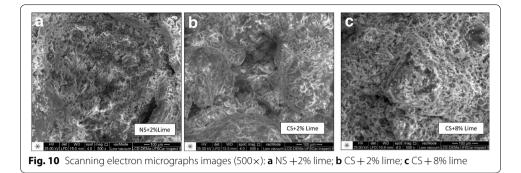


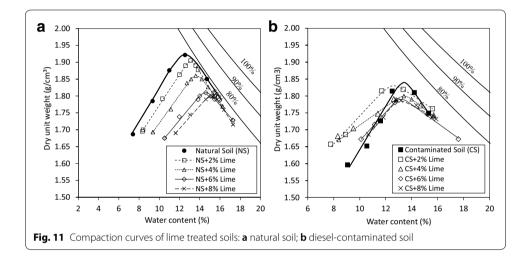




clean and gas-contaminated soil samples caused a considerable increase in pH. According to Nasehi et al. [18], the general consensus about reaction mechanisms of lime treated soils is that when lime is added to the soil, it dissociates in the presence of water into Ca^{2+} and OH^- ions thereby increasing the soil pH.

Figure 10 illustrates the SEM analysis of natural soil after 2% lime treatment and diesel-contaminated soil treated with 2 and 8% of lime. Samples were compacted at OMC and 95% compaction degree and images are shown in 500×. After the addition of 2% of lime to the natural soil (Fig. 10b), lime penetrated into spaces between the soils particles and a significant reduction in soil porosity was evidenced. Calcite minerals are predominate and an increase in particles flocculation was observed, compared to natural soil structure (Fig. 5a). The addition of 2% and 8% lime in the diesel-contaminated soil (Fig. 10b and c) resulted in a flocculated structure soil similar to that of natural soil after 2% lime treatment. According to Nasehi et al. [7], secondary cementitious products due

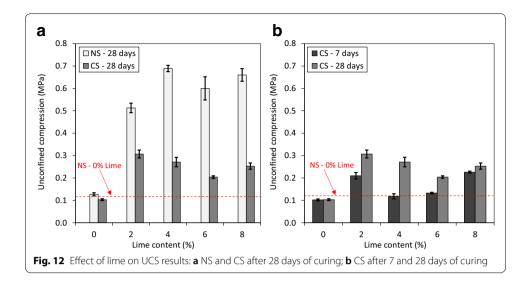




to the pozzolanic reactions between clay minerals and lime take place in presence of water, increasing the soil cohesion and its resistance.

Figure 11 shows changes in compaction curves of natural soil and diesel-contaminated soils treated with different lime contents. Samples were compacted using Standard Proctor Energy as described in ASTM D698 [21]. As expected, the compaction curve of the natural soil stabilized with lime (Fig. 11a) showed significant reductions in soil dry unit weight and increase in OMC, both due to lime fine particles specific gravity, which is lower than the soil tested. In general, the compaction parameters diesel-contaminated soils were less altered by the presence of lime than natural soil. Both natural and oil-contaminated soils maximum dry unit weight reduced with lime increase. According to Nasehi et al. [18], the drop in maximum dry unit weight is because of the flocculated and agglomerated clay particles occupying larger spaces leading to a corresponding decrease in dry density. The reason for increasing optimum moisture content suggested by Nasehi et al. [18] is that lime requires more water for the pozzolanic reaction and more water is required for the dissociation of lime into Ca^{2+} and OH- ions to supply more Ca^+ ions for the cation exchange reaction. However, the presence of the diesel oil corroborated to the lime not absorb water and thus not altering soil optimum moisture contents.

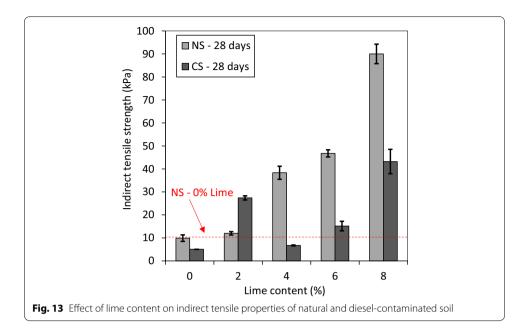
Figure 12 presents results of average UCS values obtained in this study for natural soil and diesel-contaminated soils treated with different lime contents (7 and 28 days). Tests were duplicated with samples compacted at OMC and 95% of compaction degree and

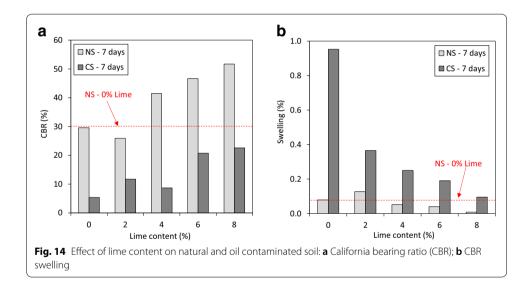


data dispersion in presented in the figures. Maximum coefficient of variation between samples of 8.6 and 7.8% were found respectively for NS and CS analysis. In Fig. 12a, as expected, lime effect was more pronounced in NS than CS soil. In addition, 2% of lime was enough sufficiently increase in 300% the UCS values of CS (8% diesel-contaminated soil) in comparison to NS, evidencing possible carbonation reactions in the mixture. Chen et al. [4] and Yu et al. [17] also showed similar results of UCS increase in soils contaminated with lower contents of oil, but stabilized with cement. Figure 12b presents the influence of curing period (7 and 28 days) on UCS values of CS with lime addition. The period of lime curing was significate in enhancing soil mechanical properties due to carbonation reactions that occurred during time.

Soil tensile properties are important in some oil-contaminated reuse application options, such as pavement or landfill cover materials. Figure 13 presents tensile strength results of lime treated NS and CS mixtures cured during 28 days. Diametric compression test were conducted according to ASTM D3967 [33], usingduplicated samples compacted at OMC and 95% of compaction degree and replicate tests. Data dispersion is presented and maximum coefficient of variation between samples of 14% were found. Natural soil was found to have a pronounced increase in indirect tensile strength property with lime addition due to carbonation reaction and calcite formation, especially with 8% lime addition where pH values favored this behavior (Fig. 9a). Lime allowed diesel-contaminated soil to recover the mechanical properties of natural soil. Diesel-contaminated soil treated with 2% lime showed a significant increase in comparison with natural soil and other increase after 6% of lime addition. This behavior may be attributed to a combination of carbonation reaction and oil interactions with soil particles.

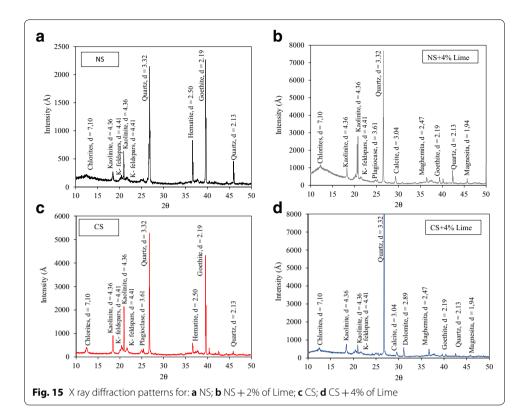
Figure 14 shows the California Bearing Ratio (CBR) samples cured in 7 days and swelling of natural and oil-contaminated soil treated with lime after 4 days of water immersion. Lime increased natural soil CBR, as expected. For diesel-contaminated soil samples, CBR values increased with lime increase, although not achieving CBR of





natural soil (29.6%). In addition, lime reduced the high swelling of diesel-contaminated soil and recovered natural swelling of soil.

Figure 15 shows diffractogram patterns of natural and oil-contaminated soils with 4% lime treatment in order to observe minerals neoformation. Crystallization of calcite is present in both natural and oil-contaminated soils treated with lime (Fig. 15b and d) affecting the soil mineralogy. Dolomite was not found in the treated natural samples (Fig. 15b), indicating that the presence of oil favored dolomite formation (Fig. 15d). This findings corroborates with treated oil-contaminated soils mechanical properties improvement. In general, the magnitudes of the peaks of calcite in the X-ray analyses are greater in the lime-natural soil mixtures than in the



oil-contaminated mixtures, corroborating to the strength responses observed in this study.

Conclusions

- An extensive experimental program was conducted in order to assess the effect of lime treatment on a diesel contamination soil on geotechnical properties of a coarse-grained soil. The experimental program involved the evaluation of soil characteristics properties such as plasticity index, compaction parameters and pH, as well as soil mechanical properties. The following conclusions can be drawn: The presence of diesel affected consistency limits of natural soil, while pH values were not affected. Soil compaction curves with diesel addition was significantly altered when compared to natural soil, decreasing maximum dry unit weight and increasing optimum moisture contents. UCS results demonstrated up to 70% reduction in soil due to diesel, while at 4% of oil-contamination a significant increase of 80% in soil UCS was observed due bonding effect occurring between particles and oil. Mineralogical analysis showed that mixtures of soil samples with diesel oil caused a flocculated fabric in the soil particles. In general, X-ray difractograms showed that the soil mineralogy was not altered with the addition of diesel.
- Lime treatment in diesel-contaminated soil increased pH and ΔpH values, while reduced consistency limits. After lime addition, calcite minerals were predominate in natural and oil-contaminated soils. The addition of lime in the diesel-contaminated soil resulted in a flocculated structure soil similar to that of lime-treated natural soil.

- Compaction curves of natural soil stabilized with lime showed significant reductions in soil dry unit weight and increase in OMC due to lime fine particles specific gravity. Compaction parameters of diesel-contaminated soils were less altered by the presence of lime. Lime allowed 8% diesel-contaminated to sufficiently increase in 300% the UCS soil and recover some mechanical properties of natural soil, evidencing possible carbonation reactions in the mixture. The period of lime curing was significate in enhancing oil-contaminated soil mechanical properties, such as indirect tensile strength and CBR, mainly due to carbonation reactions that occurred during time.
- Crystallization of calcite was present in both natural and oil-contaminated soils treated with lime affecting the soil mineralogy. Dolomite was not found in the treated natural samples, indicating that the presence of diesel oil favored dolomite formation. This findings corroborates with treated oil-contaminated soils mechanical properties improvement.

Geotechnical properties assessed in this research indicate a potential reuse of oil-contaminated soils in geotechnical applications after lime treatment. Some concerns involving oil leachate and chemical compatibility must also be evaluated. Results obtained in this study cannot be directly extrapolated to others soils and oil types and contamination contents.

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Authors' contributions

NSC coordinated the study, analysis, drafted and reviewed the manuscript. FHMP performed the analysis, drafted and reviewed the manuscript. ISM performed experimental data and analysis. JWBS performed experimental data and analysis. All authors read and approved the final manuscript.

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Competing interests

No conflict of interest.

Author details

¹ Department of Civil Engineering, Federal University of Sao Carlos, Sao Carlos, Sao Paulo, Brazil. ² Federal University of Sao Carlos, Sao Carlos, Sao Carlos, Sao Paulo, Brazil.

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