IMPACT OF SELECTED REMEDIATION METHODS ON SOILS CONTAMINATED WITH PETROLEUM SUBSTANCES

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Abstract

Human activities play a significant role in affecting soil alterations. Alterations in land structure and utilization methods accurately reflect the socio-economic progress in each region. The drive to meet the livelihood needs of human populations is a key determinant of the degree of land degradation. This pertains not only to improper crop selection but primarily to heavy metal pollution, sewage, waste disposal, and unregulated leakage of petroleum substances. The objective of this study was to identify a suitable method for reclaiming soil contaminated with petroleum substances based on its type. Types of soils used for testing include brown soil, humus, loamy sand, clay, Vistula sand and loess. To conduct the experiments, various petroleum derivatives were prepared, including gasoline, diesel oil, heating oil, hydraulic oil and used engine oil. It was found that soils like clay and loess are highly resistant to the penetration of petroleum substances into their structures. Also we noticed that soil contaminated with used engine oil exhibited the highest level of contamination, necessitating several reclamation operations such as liming.

Keywords: oil spills, methods of cultivation

Introduction

The chemical degradation of the environment is not solely dependent on the quantity and quality of pollutants introduced into it but also largely depends on the resistance and susceptibility of soil and vegetation to specific types of pollution (Alagu Abirami et al., 2019; Kasztelewicz and Sypniowski, 2010; Steliga et al., 2018). Reclamation is conceptualized as the restoration of the utility of an environment degraded due to human economic and residential activities, as well as natural occurrences such as floods, water and wind erosion, mass soil movements, fires,

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and ecological disasters (Das et al., 2011; Medina-Beliver et al., 2005; Sah et al., 2022; Truslewycz et al., 2019). The primary objective of reclamation treatment is to adjust the quality of soil, topography, water and oxygen conditions, chemistry, and environmental reactions to fulfill specific ecological and economic functions (Chunyan et al., 2022; Cabral et al., 2022).

However, reclamation seldom restores the soil to its original state and allows earlier use of the soil. The effectiveness of soil remediation and the type of remediation method employed depend on the extent of pollution, field conditions, methods of spilled product removal and the duration since the incident occurred (Wu et al., 2017; Zainab et al., 2023; Dickson et al., 2019; Matus et al., 2023). Lands contaminated with lighter fuels pose fewer challenges for reclamation, whereas all other petroleum products saturate the soil with heavy hydrocarbons that are difficult to degrade (Yang et al., 2023; Lin et al., 2010). Therefore, the objective of this study was to identify the suitable method for reclaiming soil contaminated with petroleum substances based on its type.

Remediation methods

It is common practice to burn off the remaining ground contaminated with petroleum residues (Li at al., 2020). However, this results in soil scorching, which significantly hampers and prolongs the biodegradation of hydrocarbons over time. Regardless of the extent of soil degradation, it is relatively straightforward to create conditions conducive to plant growth and the intensive biodegradation of petroleum hydrocarbons. Such soils harbor microorganisms capable of mineralizing hydrocarbons (Varjani, 2017). Agrotechnical methods should aim to provide the soil with nutrients, ensure adequate aeration, and adjust pH levels by liming (Ron and Rosenberg, 2014).

Remediation efforts may be started at any time of the year, but the most suitable periods, considering soil moisture and atmospheric conditions, are autumn and spring. Liming the soil typically initiates the reclamation process (Cao et al., 2022). The dosage of lime should be specified in the remediation plan and cannot be determined based on standard agricultural criteria. It must neutralize the existing and potential acidity throughout the entire oily layer, to a depth of at least 30 centimeters. However, there is no need to be concerned about over-liming the soil, as the slow action of lime means that an excess is more favorable than a deficiency. The applied lime should be thoroughly mixed with the topsoil using appropriate agricultural equipment, such as a tiller. After mixing of lime with the soil, nitrogen and phosphorus fertilization should be applied, followed by deep plowing after approximately 4 weeks.

For soils of medium to heavy texture, a second application of lime may prove to be necessary, similarly mixed into the soil as in the initial treatment. Fertilizers utilized in the reclamation process should be incorporated into the soil using a cultivator or heavy disc harrow. In spring or summer, approximately 2 weeks after the second reclamation treatment, the area should be seeded with:

– a grass mixture if the land is intended for turfing rather than being used as a meadow or pasture,

– field crops intended for on-site plowing or as green manure for adjacent areas. The plant material collected from these areas cannot be used for animal feed. In the second year of reclaiming oily soils, cereal and oilseed crops can be grown for seed production. Full soil productivity and crop suitability can be expected to be restored by the third year. Reclaiming freshly oiled soil is much easier than reclaiming soils with aged oiling. Liquid oil residues on the surface should be removed without burning. Depending on the soil and terrain conditions, various methods can be employed, such as flushing oils into natural or engineered depressions, including ditches, followed by their collection and disposal. Greasy hydrocarbons remaining on the ground must be collected along with a thin layer of soil and subjected to biodegradation. After the initial removal of liquid and greasy hydrocarbons from the soil, agrotechnical reclamation can be undertaken. Soil and subsoil oiling are very challenging to eliminate, as they contain a high concentration of hydrocarbons. The hydrocarbon-saturated subsurface layer of the soil suffers from a severe oxygen deficit, especially in areas with shallow groundwater retention. Reclaiming bottom-up oily soils, based on hydrocarbon biodegradation, can yield the desired results only if supported by procedures such as: removal of petroleum substances from deeper soil layers using technical methods, soil aeration, pumping or drainage of oily groundwater, or anaerobic biodegradation of petroleum components in deeper soil layers. Drainage serves as a means of soil aeration. Reclaiming bottom-up oily soils is a highly complex issue, and establishing the most effective reclamation method is impossible without assessing the extent of oiling in deeper soil layers and the groundwater status. One approach to reclaiming oily soils from below involves removing contaminated topsoil and replacing it with clean soil. The stripped soil should be cleaned through biodegradation or thermal decomposition of petroleum components, or safely disposed of in a landfill. Zoning land for agricultural or construction purposes necessitates complete removal of oily deposits, particularly in cases where groundwater pollution is a concern. The extensive method involves substantial fertilization, liming, aeration, plowing or tilling of the top layer of the deposit during the initial reclamation stage, followed by cultivation of plants in subsequent stages. Intensive fertilization with sewage sludge, compost, and well-distributed manure is particularly beneficial. The intensive biodegradation method of oil derivatives involves aerating the entire layer of the deposit, applying lime to a layer at least 50 cm thick, high nitrogen and phosphorus fertilization, and an appropriate bacterial inoculation. Sprinkler irrigation during warm, dry weather and water drainage during excessive rainfall are also essential components. With biological ex-situ reclamation methods, contaminated soil is excavated and returned to its original location after undergoing the reclamation process. In-situ methods involve reclaiming contaminated soil at its natural location. Unlike ex-situ methods, thorough geological and soil science research is necessary before proper reclamation may be undertaken. Physical and chemical methods of oily soil reclamation involve washing the soil using expensive installations. Contaminated soil washing employs extraction or leaching methods to transfer impurities from the soil to the liquid phase of the washing solution. The first step in reclaiming excavated soil is mechanical preparation, including segregation, classification and comminution processes. An essential component of physical and chemical installations in land reclamation, as with most purification installations, is the need to clean the exhaust air. Thermal methods are particularly effective for reclaiming soils contaminated with organic pollutants, and unlike soil flushing methods, they do not produce waste concentrates. Two methods can be employed: combustion and pyrolysis.

The term "hydrogeological methods" refers to the creation of one or several depression funnels in groundwater horizons using dredge wells. Through this process, contaminated water is extracted from the aquifer, effectively preventing the spread of contaminants in the direction of groundwater flow. Hydrological methods may be active or passive. Soil air extraction techniques are suitable for removing easily volatile compounds from the soil, particularly effective for reclaiming unsaturated soil layers contaminated with halogenated and aliphatic hydrocarbons. This method involves suctioning air from the soil through specially installed squeegees, lances, or piezometric holes, followed by purification.

The shielding method aims to halt the spread of emissions without altering the chemical and physical properties of the contaminated area. From a construction standpoint, shielding methods can be categorized as follows: covering system or surface sealing, vertical sealing system or sheet piling, and sealing system from below or foot-tight.

Immobilization or solidification of contaminants in the soil involves converting harmful impurities into sparingly soluble compounds or binding them in a solidifying material. These methods can be implemented either in in-situ or ex-situ conditions.

Material and methods

Types of soils used for testing include brown soil, humus, loamy sand, clay, Vistula sand and loess. To conduct the experiments, various petroleum derivatives were prepared, including gasoline, diesel oil, heating oil, hydraulic oil and used engine oil.

Brown soils are typically found in temperate climates and often develop under deciduous and mixed forests. They form due to diverse geological origins and soil structures rich in bases or acid rocks, as well as dust formations such as loess and loess-like formations. Brown soils are known for their fertility and high humus content, typically around 3–4%. They are distributed across all continents.

Black earth, or black mold, is highly fertile soil characterized by deep, black humus layers. These soils usually form under steppe vegetation on rocks rich in calcium carbonate. Black earth is commonly found on loess or loess-like dusts.

Loose and slightly loamy sands consist of over 90% quartz, with some variations. They typically contain small percentages of aluminum oxide (Al_2O3), iron oxide (Fe_2O_3), magnesium oxide (MgO), calcium oxide (CaO) and potassium oxide (K_2O). Slightly loamy sand is slightly richer in these ingredients compared to loose sand. Light loam and strong loam sands usually contain 85-90% SiO_2, 4–6% Al_2O_3, 1–1.5% K_2O, and approximately 0.5% CaO and MgO. The presence of metallic components depends largely on the colloidal clay content, except for iron, which can be accumulated or removed through oxidoreductive processes. This applies to all types of sands, not just clay sands.

Clay is a geological formation comprising clay, silt, sand and skeletal particles. It exists in various types, including sandy, light, medium, heavy, and very heavy clay, distinguished mainly by the proportion of clay particles. In sandy loam, clay particles make up 21-25% of the composition, while in very heavy loam, they can constitute even 76–90%. In Poland, clays are predominantly of glacial origin, found in alluvial river valleys and weathering origin mountains. Light and medium clays with high dust particle content, known as silty clays, are considered among the best soil-forming rocks in Poland. Light clay typically contains around 85% SiO_2, 5.8% Al_2O_3, 1.5–2.5% Fe_2O_3, 1–2% K_2O, 0.5–1% CaO and 0.5–1% MgO. Medium clay contains higher amounts of aluminum, iron, potassium, magnesium, and calcium compared to light clay, with SiO2 content usually close to 75%. Heavy clay consists of 70–75% SiO_2, 10–12% Al_2O_3, 3.5–5% Fe_2O_3, 2.5–3.5% K_2O, 2–3% MgO and 1–2% $CaCO_3$.

Loess is a geological formation of aeolian origin, typically composed of ordinary dust and clay dust, occasionally mixed with clay dust. It exhibits high porosity, low plasticity, and high susceptibility to water and wind erosion when dry. Loess formations constitute the largest complex among dust soil formations, having a mechanical composition of clay and loamy dusts, with rare occurrences of ordinary dust. Loess soils are generally fertile and contain an average of 75% SiO_2, although the SiO_2 content can vary widely (66–83%). They are not rich in iron, with an average Fe_2O_3 content of 2.2–2.7%, and their potassium content is similar to that in light clays.

Figure 1 shows soil samples used for the following tests: (A) – black soil, (B) – clay, (C) – sand.

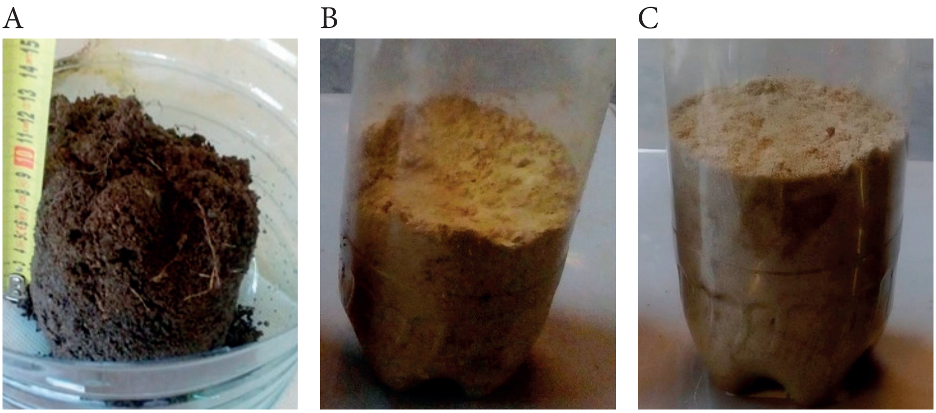

Figure 1. Soil samples used for testing (A) – black soil, (B) – clay, (C) – sand

In test 1, use was made of 1 dm^3 of permeable soil, namely sand. A quantity of 0.25 dm^3 of used engine oil was added to the prepared sample deliberately for better contrast in the experiment. Similarly, 0.25 dm^3 of hydraulic oil was poured into the prepared sample of 1 dm^3 of black mold. During the initial 20 minutes, a slow seepage of oil into the soil sample was observed. The sample was left to stand for an hour. After this time, the hydraulic oil had permeated the soil to a depth of 4 to 5 cm. These experiments show that the density and type of liquid, as well as the structure and density of the substrate, have a significant impact on the penetration of oil derivatives into the soil. The used engine oil notably contaminated the structure of the sand, which will have a considerable impact during the subsequent reclamation process. It can be inferred that the extent of contaminated soil will vary depending on the type of oil derivative, thus influencing the choice of the appropriate remediation method.

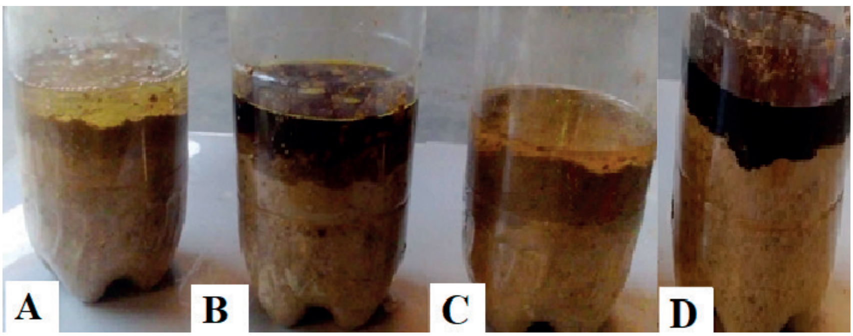

Figure 2. Oiling of clay: (A) – gasoline, (B) – hydraulic oil, (C) – diesel oil, (D) – engine oil

Next, clay was tested, with gasoline, hydraulic oil, diesel oil, and engine oil being used. Petroleum liquids in quantities of 0.25 dm^3 were poured into the prepared, measured soil samples, each consisting of 1 dm^3 of soil. Subsequently, an analysis of the rate of penetration of petroleum liquids into the structure of each soil sample was conducted (Figure 2).

Results

Figure 3 presents the time of absorption into loamy soil of individual liquids: diesel oil, gasoline, hydraulic oil, used engine oil.

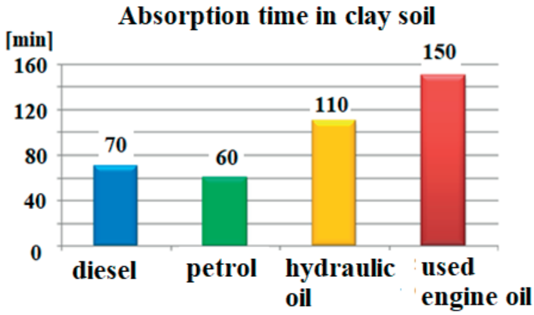

Figure 3. Absorption time of petroleum liquids into loamy soil

The subsequent tests involved sand, with heating oil, gasoline, and hydraulic oil being utilized. Subsequently, the rate of penetration of petroleum liquids into the soil structure was analyzed. Each test involved 1 dm^3 of soil, with 0.25 dm^3 of liquid used in each instance. Figure 4 illustrates the absorption time of individual liquids – namely, heating oil, gasoline, and hydraulic oil – through the sand.

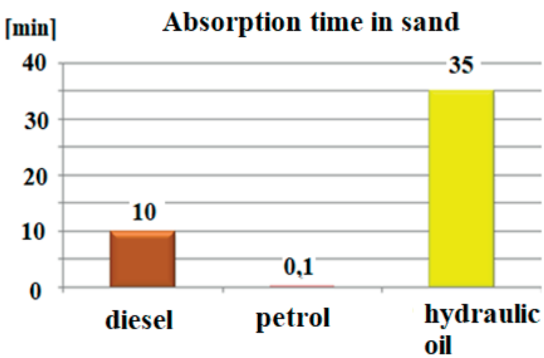

Figure 4. Absorption of petroleum substances in sand

Another investigation focused on clay, with gasoline and diesel being employed. The petroleum liquids were poured into the prepared, measured soil samples. Subsequently, an analysis of the rate of penetration of petroleum liquid into the structure of the respective soil was conducted. Based on the aforementioned research, a graph was constructed to compare the permeability of soils following leaks of petroleum substances.

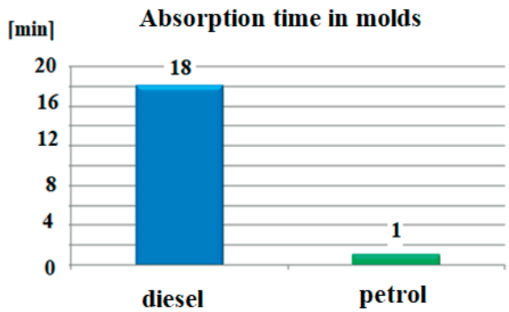

Figure 5. Absorption time of hydrocarbon substances in mold

In the successive test, the scenario of tank truck failure carrying hydraulic oil was simulated. Following the accident and the unsealing of the tank, a leak occurred. To protect the soil from contamination due to oil spillage, a protective layer of sorbent was applied in the direction of the oil spill. In this test, a prepared 5 dm^3 sample of loess was spread on a flat surface measuring approximately 40 cm × 40 cm. The edges were protected with sorbent, and 0.5 dm^3hydraulic oil was poured. The sorbent layer, which shielded the soil from further contamination by petroleum substances, was approximately 4 cm thick. The spilled hydraulic oil formed a layer about 4 cm high on the surface of the loess. Initially, as the liquid spilled onto the loess surface, its flow direction was observed, leaving behind traces of oil that contaminated the soil. It was found that hydraulic oil spilled on the surface did not penetrate through the sorbent immediately. The experiment demonstrated that the soil was effectively protected against petroleum contamination by halting the spread of oil. Additionally, potential leaks can be diverted by creating protected channels or ditches leading to a large trench, secured by a layer of sorbent, where the leaking petroleum liquid can be contained. The liquid accumulated in the trench after a failure can be pumped out and collected along with the used sorbent for proper disposal. The trench and channels should be lined with a thick layer of material, such as petroleum-resistant foil, to prevent further contamination.

In terms of liming, the entire dose should neutralize the acidity in the entire oily layer, minimum 30 centimeters thick. Excess lime in the soil is more beneficial than a deficiency due to its slow action. For soils in the first class, the entire lime dose should be applied at once. In other cases, liming can be divided into two, three, or four parts. The lime should be mixed thoroughly using equipment like a tiller. Mixing three times is typically sufficient for sandy, slightly loamy and light clay soils.

After an hour, all the oil-soaked black earth from sample #1 was transferred into a container. The soil was then mixed and subjected to the liming process, followed by another round of mixing. The total lime amount used was 0.2 dm^3. Additionally, the entire oil-contaminated sand was collected in a separate container after retrieving the oil layer remaining on the soil surface. Significant contamination of the sand was noted, with approximately 80% of the soil being saturated and blackened by the oil. The heavily oil-soaked soil underwent liming, mixing, and prisms, followed by a third round of liming for soil remediation.

The resulting experimental heap underwent a fourth round of liming. In all four liming sessions of this experiment, a total of 0.5 dm^3 of lime was used. The limed pile formed from the contaminated sand was then covered with a one-centimeter layer of clean brown soil. An analysis of the experiment indicates that heavily oil-contaminated soil, prone to petroleum liquid leakage, would necessitate the removal of a substantial soil layer from the affected area. The use of four rounds of liming in the experiment was meant to reduce the pH of the heavily contaminated soil. The remediation process should be conducted on prepared and secured land in designated heaps.

In the subsequent samples, consisting of layers of sand and mold, gasoline was poured. Two soil samples were utilized in the experiment, namely sand/mold and mold/sand, each with a volume of 1.5 dm^3. A volume of 0.25 dm^3 of gasoline was poured into each sample, and the contents were left for an hour. Upon pouring the measured gasoline into the samples, it was observed that the petroleum substance poured into the sand/mold sample was absorbed by the first layer. Due to the high permeability of the sand layer, gasoline penetrated into the subsequent layer within a few seconds. The samples were left to stand for two hours. After this time, the presence of gasoline at the bottom of the container was observed, indicating that the gasoline poured into the sample had penetrated further into the second layer of soil.

In the sample (B-humus sand), it was observed that the same volume of gasoline poured seeped through the first layer of soil much more slowly. The sample was left to stand for two hours, after which no gasoline had accumulated at the bottom of the vessel. Thus, all the poured-out oil derivative remained in the first layer and partly in the sand layer. Similar tests to those conducted for sand and mold were performed on a sample of forest soil and sand. Slower gasoline penetration through the loess soil structure was observed as compared to black mold. These samples were then poured into experimental trays and formed into heaps, followed by liming. They were left to stand for 12 hours at ambient temperature (approximately 15°C). After this time, the experimental heaps were cut to examine the cross-sectional structure. It was found that visible traces of liming were not retained in the structure of the heap. No gas was observed in the trays. It can be assumed that some of the gasoline had evaporated.

Based on the conducted research, it can be concluded that the decisive factors affecting the extent of soil contamination after a petroleum substance leakage are: the types of soil, the surface on which the substance leaked, the density of the soil, and consequently its permeability, as well as the type of the petroleum substance involved. In the aforementioned tests, sand exhibited the highest permeability compared to black mold, forest soil, or clay. This is attributed to the granulation of the sand fraction, as well as its low moisture content. Clay and loess, on the other hand, proved to be the least absorbent. Clay in its natural state displayed minimal permeability, retaining most of the tested liquid on the surface.

Based on the above research, it should be assumed that the greater the permeability of a given soil, the greater the likelihood of contamination spreading across the area and penetrating into its soil profile. In the event of an oil spill spreading on highly permeable soil, responders dispatched within the first few dozen minutes may be unable to collect the liquid from the surface soil, for example using a sorbent.

Examination of the permeability of selected soils to petroleum substances and analysis of the selection of reclamation methods in laboratory conditions

The experimental procedure involved four types of soil (sand, Vistula sand, black loam, and brown soil). Six samples of 150 cm^3 each were prepared for the study. These measured samples were placed on a tripod within a specialized container equipped with a strainer at its bottom. Subsequently, 25 cm^3 of a petroleum derivative was added to the dish containing the soil, while simultaneously measuring the time it took for the liquid surface to disappear and the time it took for the leakage to stop.

In the next stage, the impact of selected soil remediation methods on contaminated soils was investigated, and the effectiveness of each method was analyzed based on the quantity of oil derivative that permeated through the soil and was collected in a measuring container. The following treatments were utilized for this purpose: washing oily soil with cold tap water, washing oily soil with warm tap water, washing contaminated soil with warm water containing dishwashing liquid, and washing contaminated soil with cold water containing dishwashing liquid. Figure 6 illustrates the test setup.

In the initial test, 150 cm^3 of sand was utilized, to which heating oil was added three times in increments of 25 cm^3 to saturate the soil with oil. Similar attempts to oil soils such as Vistula sand, humus, and brown soil were subsequently carried out.

The subsequent phase involved the washing of contaminated soils. Cold water, warm water, and water with the addition of dishwashing liquid containing a surfactant were introduced into the container with oily soil for this purpose. An aqueous solution of the dishwashing liquid was prepared in a 4:1 ratio (80 ml of water mixed with 20 ml of surfactant). This allowed an assessment of the effectiveness of oil removal from the soil, thereby enabling conclusions to be drawn regarding the appropriate selection of remediation methods. Based on the laboratory tests conducted, it can be concluded that the disappearance time of the liquid surface, the permeation time, and the amount of condensed liquid vary depending on the type of soil used, as well as on the petroleum substance and the remediation method employed.

Figure 6. Test stand

Calculations of the permeability of selected soils were carried out during the laboratory experiment of the measurements. The formula for the calculation was:

$$P = \frac{V}{S}t \tag{1}$$

where:
P – transmittance [cm/s],
V – volume of liquid absorbed by the soaking surface [cm^3],
S – soaking area [cm^3],
t – bleeding time (total) [s].

Figure 7 shows the permeability results of black mold and Vistula sand using diesel oil.

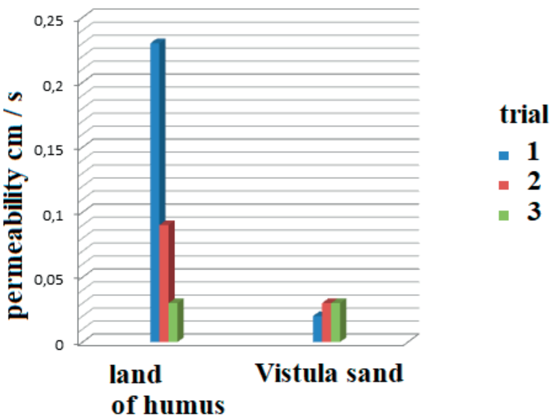

Figure 7. Permeability results of soil samples when using heating oil

Restricting only to simple tests simulating the remediation of oily soil, it was only possible to perform leaching of petroleum substance from soils under laboratory conditions. Figure 8 shows tests for samples of diesel oil contaminated soils and Figure 9 for fuel oil.

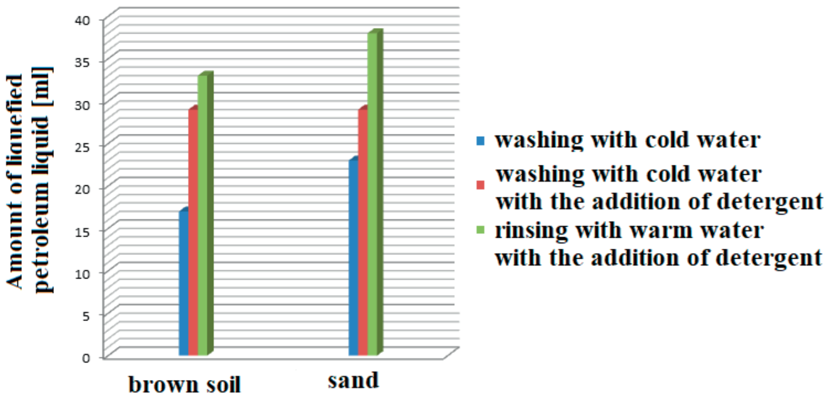

Figure 9. Productivity results of selected methods for remediation of oily soil with fuel oil

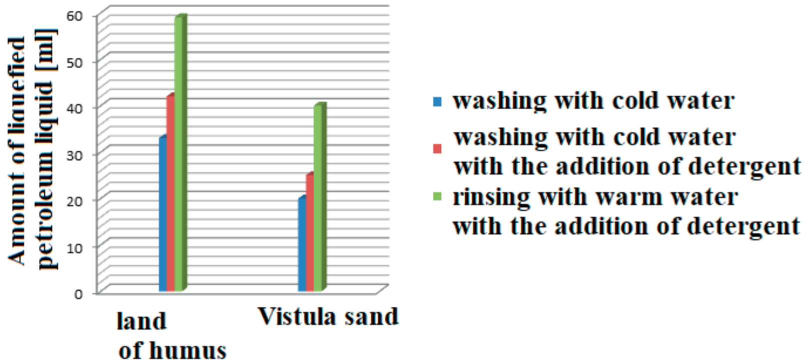

Figure 8. The results of the effectiveness of selected methods of soil contamination with diesel oil

After observations and calculations, the research allows a conclusion that black mold is a soil that offers the highest permeability to oil derivatives, followed by sand, and the least permeable is brown soil. The most effective remediation method in this experiment was washing out soil samples with warm water containing a surface-active substance, as most of the petroleum liquid was removed. The results obtained from testing soil permeability illustrate a noticeable decrease in permeability values with an increase in the amount of absorbed petroleum liquid.

Conclusions

Reclamation is a comprehensive process aimed at restoring the original utility of the environment, which has been degraded due to human economic and living activities. The main goal of reclamation is to adapt the quality of land, water, oxygen conditions, and environmental pH to perform specific ecological and economic functions. Unfortunately, reclamation processes only rarely restore the original condition of the soil, and therefore do not always enable its original reuse. Most land and water bodies have undergone reclamation processes and have lost their utility values to varying extents. This also applies to land contaminated with oil derivatives as a result of breakdowns, accidents or disasters. Soil properties largely determine reclamation methods and post-reclamation use. The utilization of reclaimed land is also crucial and depends on the needs of the local community or further investment plans in those areas.

After analyzing reclamation processes and research outcomes, the following conclusions have been drawn:

- Soils like clay and loess are highly resistant to the penetration of petroleum substances into their structures. There is a strong likelihood that the contaminated area will be relatively small, and contamination deep into the soil profile would be superficial.
- Gasoline, due to its properties, penetrates soil structures the fastest among all the tested substances, especially through sandy structures, within seconds. Gasoline also evaporates relatively quickly from the soil surface, posing a significant fire risk. As an effect, reclaiming highly permeable soils immediately after an event might be challenging and may require off-site soil collection and remediation.
- Soils contaminated with used engine oil exhibited the highest level of soil structure contamination, necessitating several reclamation operations such as liming. The contaminated soil had a heavily oily consistency. Liming should be the first step in the reclamation process and excess lime would not be problematic owing to its properties.
- Washing contaminated soils with warm water and a surface-active substance proved to be the most effective method, resulting in the highest amount of condensed petroleum liquid removal in each case.
- Leaching hydrocarbon substances with cold water yielded the least effective results. It resulted in the smallest amount of oil derivative removal from the tested soil samples.
- Soil oiling is most commonly associated with petroleum transport incidents such as accidents involving road tankers, railroad catastrophes, or tanker ships. Therefore, the reclamation of oily soils near transport routes (e.g., expressways, railroads) should be considered, as it may hinder soil reclamation processes.

– Economic conditions often play a crucial role in selecting the appropriate reclamation method. Agrotechnical methods may be effective for reclaiming areas contaminated with petroleum substances up to a depth of 0.5 metres, especially when contamination is not extensive. However, for deeper contamination, agrotechnical methods may not be as effective.
– When choosing a remediation method, human safety and environmental impact should be prioritized over economic considerations.

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