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Technological Innovations for Physical and Chemical Remediation of Oil-contaminated Water and Soil: A Review (Part-II)

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Keywords: Ecosystem Oil-contaminated soil Petroleum industry Petroleum pollutants Soil remediation technology **A B S T R A C T** The petroleum industry produces significant amounts of oily wastewater, either through the process of oil production named associated water, through the process of oil refinery, or because of pipeline leakage through oil transportation. Environmentally permissible oily wastewater treatment is a significant problem for the petroleum industry. Nowadays, the focus has been on awareness of the treatment methods for oily wastewater. Therefore, oil-water separation has become a recent obstacle, and it must be explored and resolved by petroleum industries to meet set standards and protect the environment. However, during the last decades of growing urbanization and industrialization worldwide, the consumption of oils and their derivatives has significantly expanded. Due to some factors such as poor management, incomplete combustion, pipeline leakage, and other incidental circumstances, petroleum leaks into water and soil and are nearly unavoidable during the process of petroleum extraction, transportation, processing, and utilization. Since the relative toxicity of petroleum and its products, petroleum pollutants in water and soil can seriously harm the local ecosystem and human health. Therefore, efficient water and soil remediation technologies solutions for oil-contaminated sites are essential for environmental safety and sustainable growth. The current review of part II focuses on the physical and chemical remediation technologies of oil-contaminated soil.

ال بتكارات التكنولوجية للمعالجة الفيزيائية و الكيميائية للتربة امللوثة بالنفط: دراسهمرجعيه(الجزء الثاني)

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الكلماتاملفتاحية النظام البيئي التربة امللوثة بالنفط صناعة النفط امللوثات البترولية تقنيات معالجة التربة **امللخص** تنتج صناعة النفط كميات كبيرة من املياه الزيتية، إما من خالل عملية إنتاج النفط املسماة باملياه املرتبطة، أو من خلال عملية تكربر النفط، أو بسبب تسرب خط الأنابيب من خلال نقل النفط. تعد المعالجة المسموح بها بيئيًا للمياه الزبِتية مشكلة كبيرة لصناعة النفط. في الوقت الحاضر، تم التركيز على الوعي بطرق معالجة المياه ً الزيتية. ولذلك، أصبح فصل الزيت عن املاء عقبة كبيرة حديثة، ويجب على الصناعات النفطية استكشافها وحلها للوفاء بالمعايير المحددة وحماية البيئة. ومع ذلك، خلال العقود الأخيرة من تزايد التحضر والتصنيع في جميع أنحاء العالم، زاد استهالك الزيوت ومشتقاتها بشكل كبير. بسبب بعض العوامل مثل سوء اإلدارة، واالحتراق غير

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تقريبًا أثناء عملية استخراج النفط ونقله ومعالجته واستخدامه. نظرًا للسمية النسبية للبترول ومنتجاته، فإن الملوثات البترولية في الماء والتربة يمكن أن تلحق ضررًا خطيرًا بالنظام البيئي المحلي وصحة الإنسان. ولذلك، فإن ً ا
أ الحلول الفعالة لتقنيات معالجة المياه والتربة للمواقع الملوثة بالنفط ضرورية للسلامة البيئية والنمو المستدام. تركز الدراسه المرجعيه الحاليه في جزئها الثاني على تقنيات المعالجة الفيزيائية والكيميائية للتربة الملوثة بالنفط.

1. Introduction

The crude oil components are determined via many factors, such as the type of organic residues, sediments, and environmental factors surrounding the area. An oil spill is the occurrence of discharges of oil quantities, often in water environments such as seas and oceans, and this phenomenon represents one of the most significant issues facing the environment of the seas and oceans. Because of the increase in oil extraction, production and use of giant tankers, thousands of cases occur annually from oil spills resulting from transportation and manufacturing, in addition to the natural spill at the bottom of the ocean [1]. Oily wastewater (O/W) is produced from several sources, and when it enters the natural ecosystem, it poses a significant environmental risk, often with severe implications for human activities. Recently, with the rapid development, especially in the advanced industries such as the petrochemical industries, oil refining, heavy metal, mechanical, food processing, textile and leather industries, vehicle industries, and maritime transport are primary sources of oil-contaminated wastewater [2]. Furthermore, urban wastewater, specifically effluent coming from domestic operations, increases the amount of oily wastewater. In general, gasoline, grease, fats, heavy and light hydrocarbons, tars, lubricating oil, cutting oils, wax oils, and other substances are frequently found in oily wastewater. The oil concentration in the wastewater stream, on the other hand, varies greatly depending on the source of the effluent. For instance, domestic wastewater's oil and grease content ranges between 50 and 100 mg/l [3]. The majority of oil-contaminated water is produced during oil exploration and extraction processes, where the wastewater contains oil and grease with extremely high concentrations (around 4000-6000 mg/l) [4]. Furthermore, the second-largest source of oilcontaminated wastewater is the metal processing industry, which includes grinding oils, cutting oils, lubrication fluids, and coolant oils in both insoluble and emulsified oil form with concentrations ranging from 100 to 5000 mg/l. In addition, food-processing industries such as poultry, slaughterhouses, and dairy are significant contributors to oily wastewater. Furthermore, the fish and meat packaging industries generate a significant amount of oil and fatty materials during the slaughtering, washing, and subsequent by-product processing processes. Thus, the effluent's oil and grease content in food packaging houses may be as high as 1000 mg/l [5].

Contamination of soil by petroleum-based substances alters its properties, both in terms of its physical and chemical composition, as well as its biological structure. These changes can lead to shortages in water and oxygen, along with reduced availability of nitrogen and phosphorus resources [6]. Consequently, the productivity of contaminated soil decreases, with losses becoming particularly significant in areas where the soil's nutrient content is already low. The presence of petroleum-based compounds in the soil exacerbates nitrogen and phosphorus deficiencies, disrupts the water balance, and disturbs the biological equilibrium. Once these substances infiltrate the soil, salinity rises, compacting the soil and affecting its permeability. Additionally, studies have shown that contaminated soils exhibit imbalanced ratios of carbon and nitrogen, further impacting their chemical properties and enzyme activity [7]. According to Alsakit M. et al. [8,9], The influence of factors of total petroleum hydrocarbons (TPHs) and the separation and removal efficiencies were studied on the oily water. Moreover, the experimental method achieved a separation efficiency of 98%, effectively reducing total petroleum hydrocarbons (TPHs) to 5 ppm, demonstrating its strong potential in oil-water treatment. In addition, the challenges and considerations in remediating oil-contaminated water and soil highlight several vital essential points, such as various remediation technologies, site-specific considerations, the combination of technologies, the importance of research and development, and practical application [10,11].

This review paper provides a comprehensive overview of the challenges and considerations involved in remediating soil. Moreover, the review highlights a comprehensive remediation strategy to achieve efficiency and incorporate various sustainable technologies.

2. Soil Remediation

Soil contamination is defined as a change brought about by human activity in the materialistic and chemical properties of the soil that lowers its quality and productivity [12-14]. Soil pollution has become a problem due to the recent rise in industrial pollutants such as petroleum hydrocarbons and heavy metal usage [15,16].

It is widely recognized that petroleum hydrocarbon contamination of soils (Fig. 1), sediments, surface waters, and groundwater poses a significant threat to human health [17].

Fig. 1: Source of soil contamination by PHs [17]

Remediation methods and techniques play an essential role in the cleaning, containment, removal, regeneration, and restoration of contaminated environments. The remediation approach chosen for each contaminated environment is related to the site-specific, and then it depends on the characteristics and composition of the contaminant, all conditions (physical, chemical, and biological) of the affected environment, and the presence or necessary enhancement of the microbial community. Additionally, when selecting an appropriate remediation technique, mechanisms, regulatory requirements, cost, and time constraints are considered. However, in response to the growing demand for responses to environmental pollution, the primary focus of environmental scientists is now on adopting risk-based management approaches to remediate polluted environments. In most cases, authorities specifically focus their remediation priority on the most urgent cases that are less difficult to remediate. Affected environments are mainly soils, surface waters, sediments, and groundwater, and usually contain high and low molecular weight petroleum hydrocarbon compounds, volatile organic compounds (VOC), semi-volatile organic compounds (SVOC), Contaminated with polycyclic aromatic hydrocarbons (PAHs), etc. These pollutants can spread far from their original location, threatening plant and animal communities in the affected areas. Remediation options adopted by pollution professionals can serve as a means of dealing with contaminated environments [18]. The management system is based on multiple ex-site and in-site remediation treatment techniques for surface water, groundwater and soil, including biological technology, chemical, physico-chemical, thermal, electrical, retention, segregation, electromagnetic and ultrasound therapy. In addition,

knowing the characteristics of the pollutants that are obtained from the preliminary experiments helps environmental researchers to choose the appropriate remedial methods from the different treatment methods. In addition, water and soil remediation technology can be done together or separately, depending on the amount and type of contamination. The success of any treatment approach in a particular contaminated site or environment depends on the design and tuning of system operation based on contaminant properties and system performance. Simultaneous or sequential integration of one therapeutic approach with another may result in synergistic or combined effects between the therapeutic approaches employed [19]. According to Xu, X. et al [20], spills and discharges of petroleum hydrocarbons frequently happen during the production, storage, and transportation of petroleum, as well as during the refining and processing stages. These incidents can be attributed to blowouts during oilfield development, leaks from oil pipelines and storage tanks, oil tanker leaks, oil well waxing, and refinery and petrochemical production equipment overhauls. Petrol stations and refineries are the primary sources of soil contamination in many nations, as they expose the soil to minute but continuous petroleum leaks [21]. However, in Russia, accidental spills of oil and oil products mostly happen during pipeline transportation. In the case of oil spills, a total petroleum concentration (TPH) in soil often ranges between 2 and 5%, but in some cases, it can reach up to 50%. In oil and gas condensate production, accidental spills are accompanied by the release of highly mineralized formation waters and drilling fluids, which salinize surface oil, subsoil, and groundwaters [22].

3. Soil remediation technologies

As mentioned earlier, petroleum hydrocarbons have a tremendous impact on the environment and human health. Polluted environments should be cleaned and freed from contaminants using clean technology and sustainable remediation approaches. Therefore, we must choose the best remediation approach to remove petroleum hydrocarbons from the contaminated environment. Variables such as the composition and type of contaminant, the contaminated environment, the biological and chemical conditions, and the microbial species required for growth, as well as time, cost, regulatory requirements and procedures, all need to be considered. In addition, special attention should be taken to adapting new sustainable approaches to mitigate the significant risks of soils contaminated with toxic petroleum hydrocarbons [19].

3.1 Soil washing technology

Soil washing technology is an ex-situ method based on physicochemical separation and leaching processes. In this technology, the dispersion of toxic hydrocarbon contaminants between the soil particles and the washing solution [23]. Petroleum hydrocarbon molecules bind to soil silt and clay particles, increasing soil and water pollution, especially groundwater. In addition, chemical additives such as surfactants are also used for soil and water remediation [24,25]. Soil washing technology uses liquids (usually water only or water mixed with solvents) for soil remediation (Fig. 2).

Fig. 2: Schematic diagram of soil washing system [26]

Solvents are selected based on their ability to dissolve specific

contaminants and their environmental and human health impact [27,28]. Furthermore, when washing the soil, separate fine soil (clay or silt) from coarse soil (sand or gravel). Since hydrocarbon contaminants tend to bind and adsorb to small soil particles (mainly clay and silt), separating small soil particles from larger ones reduces the amount of contaminated soil [27,29]. Thus, a small amount of soil, which contains the majority of clay and silt particles, can be further treated by other methods (such as bioremediation or incineration) or disposed of according to federal regulations [30]. Pollution groups targeted for soil washing include semi-volatile organic compounds (SVOCs), petroleum and fuel residues, heavy metals, PCBs, and PAHs [31-35]. As a result, soil-washing technology can remove a wide range of organic and inorganic contaminants from coarse-grained soils [28]. Soil washing remediation technology is more cost-effective as it reduces the amount of material that needs further processing with another technology.

3.2 Soil vapour extraction technology

Soil vapour extraction (SVE) is one of the most commonly used technologies to remediate soil and groundwater contaminated by volatile organic pollutants (VOCs). Soil vapour extraction (SVE), also known as soil vent or vacuum extraction, is a recognized and costeffective technology for remediating unsaturated soils contaminated with volatile organic compounds (VOCs) and semi-volatile organic compounds (SVOCs) [27,36]. Moreover, soil vapour extraction (SVE) involves installing vertical and horizontal wells in areas of soil contamination. Furthermore, the blowers are often used to assist the evaporation process (Fig. 3). Thus, a vacuum is applied through a well near the source of pollution to evaporate the volatile components of the contaminated mass and extract them through an extraction well.

Fig. 3: Typical components of in-situ soil vapour extraction system [37]

Finally, the extracted vapours are treated (usually by adsorption materials such as carbon) before being released into the atmosphere [38]. This technology is also used for groundwater pumping and air stripping to treat groundwater contamination. SVE is generally more suitable when the contaminated unsaturated zone is homogeneous and relatively permeable. However, due to infiltration and airflow, the contamination site needs to be covered with an impermeable surface layer [27,39,40]. SVE is generally most effective when applied to lighter, more volatile petroleum products such as gasoline fuel. Heavy fuels such as heating oil, diesel fuel, and kerosene are not easily removed by soil vapour extraction technology. Heated air injection increases the volatility of these heavier petroleum products but requires high energy, which makes them economically unfeasible [27,33,38]. Benzene, toluene, xylene, naphthalene, biphenyl, perchloroethylene, trichloroethylene, trichloroethane, and gasoline are all effectively removed from contaminated soils by soil vapour extraction technology [27,30,41].

3.3 Soil flushing technology

In general, soil flushing is one of the traditional in-situ technologies for soil remediation. The soil flushing method is the extraction of soil

contaminants using water or other solvent solutions. Soil flushing technology is an innovative remediation technique that 'floods' contaminated soil with a contaminant-carrying solution where it can be removed [27,30]. Moreover, soil flushing involves an injection/recirculation process, unlike soil washing technology, where contaminated soil is excavated and treated on the surface. The soil flushing technology process starts by drilling, injection, and extraction wells in the ground where soil contamination is found. The number of injection and extraction wells, as well as the site and depth, should be determined based on several engineering considerations and geological factors. In addition to arranging the wells, other equipment, such as the wastewater treatment unit, must be transported to the site or built on-site. The soil flushing process equipment pumps the treatment solution into the injection wells (Fig. 4). The treatment solution moves into the soil and picks up contaminants as it moves to the extraction wells.

Fig 4: In situ soil flushing process [27]

Extraction wells collect rinses containing contaminants. A mixture of solutions and contaminants is pumped out of the ground through extraction wells. The mixture is typically treated in a wastewater treatment unit to remove and recover contaminants [27,30,42]. The soil flushing method is usually the most effective where the soil has pores through which treatment fluids can pass. If the soil has a high amount of silt or clay, the treatment solution cannot pass through the contaminated soil and quickly come into contact with contaminants. This reduces the effectiveness of the soil flushing technology [27,30,38].

3.4 Excavation technique

Excavation is one of the remediation methods for crude oilcontaminated soil, as it allows for quick removal and isolation of the pollutants. However, this approach comes with its own set of challenges and drawbacks [43]. In general, excavation can be expensive and time-consuming due to transporting the contaminated soil to disposal sites. Moreover, creating landfill sites adds to the overall cost and environmental impact [44]. Landfill mines, in particular, can pose environmental risks, as they contribute to air pollution by releasing contaminants into the atmosphere. Additionally, excavation may not always be the most advanced or sustainable method for soil remediation. While it effectively removes contaminated soil from the immediate environment, it does not address the underlying pollution issues or provide long-term solutions for soil restoration. Alternative approaches, such as on-site treatment or offsite treatment at specialized facilities, offer potential advantages in terms of cost-effectiveness and environmental sustainability. These methods may involve technologies like chemical oxidation, bioremediation, or thermal treatment, which can treat the contaminated soil in place or at designated facilities without the need

for excavation and transportation. Ultimately, the choice of remediation method depends on various factors, including the extent of contamination, site-specific conditions, regulatory requirements, and cost considerations. While excavation remains a viable option for specific scenarios, efforts should be made to explore and implement more advanced and sustainable remediation techniques where feasible [45,46].

3.5 Thermal desorption technology

Thermal desorption is an innovative remediation method that involves drilling, screening, and heating contaminated soil to release oil [38]. The soil is heated to temperatures between 100 and 600 degrees, so contaminants with boiling points in this range evaporate and are separated from the soil. Evaporated contaminants are collected and otherwise treated [27,30,47]. The actual process of thermal desorption technology consists of heating soil in a chamber that can vaporize organic contaminants and certain metals. Then, the vacuum system carries the vaporized contaminants to an external treatment system. Thermal desorption technology can be divided into high and low thermal desorption based on the temperature [27,48]. Thermal desorption works on most soils contaminated with hydrocarbons. However, the thermal desorption method at low temperatures is typically used to treat VOCs and various fuels. In contrast, thermal desorption technology at high temperatures is mainly used for PAHs, PCBs, and pesticides [38,48].

3.6 Chemical oxidation method

In general, chemical oxidation effectively removes hazardous waste from soil, particularly at oil spill sites. The effectiveness of this method highly depends on the soil's structure and characteristics. (Fig. 5) [49]. Fenton's hydrogen peroxide and ferric ion reagent are commonly employed for chemical oxidation. Hydrogen peroxide acts as a potent oxidizing agent, generating hydroxyl ions during the Fenton reaction, while ferric ion serves as a catalyst. Hydroxyl ions are highly reactive and efficiently degrade contaminants in the soil. Studies have shown that using Fenton's reagent at lower pH levels can significantly enhance oil removal from sand compared to natural pH conditions or peat [50,51]. Ozone is another effective oxidant used for removing crude oil from soil. It is advantageous due to its ease of generation, storage, and handling, especially for in-situ treatment. Ozone exhibits higher reactivity towards polycyclic aromatic hydrocarbons (PAHs) than alkanes. It can support soil microbial communities by generating oxygen during degradation, enhancing bioremediation. Thus, chemical methods provide a rapid solution for treating contaminated soil but come with potential risks.

Fig. 5: Examination of In Situ Chemical Oxidation [49]

Moreover, chemicals in oxidation processes can threaten nearby soil and organisms due to leaching or side reactions. [52]. Therefore, assessing environmental impacts and taking appropriate measures to mitigate risks during soil remediation is crucial.

Table 1 provides a detailed comparison of technological innovations for remediating oil-contaminated soil, including the type of separation, implemented remediation technologies, their effectiveness and efficiency, materials used, and worldwide use.

4. Conclusion

This study provided an overview of the remediation of soil and water environments contaminated with petroleum hydrocarbon compounds. Various remediation methods are available, but no single method is optimal for all types of contaminants in the affected environment. As discussed earlier, you can use various remediation technologies to remediate the contaminated site. However, no single technique is suitable for all contaminant types and the various site-specific conditions found at various contaminated sites. The potential impact of site conditions, types of contaminants, sources of contamination, source control measures, and potential remediation measures will determine the choice of remediation strategies and techniques. Effectively addressing contaminated site problems often requires multiple remediation techniques. Treatment processes can be combined, and usually combined, into process trains to remove pollutants and toxic substances that are more effectively removed at contaminated locations.

Therefore, several techniques that can provide efficient and costeffective restorations should be considered as possible candidates in the restoration selection process. Choosing one or more specific remediation techniques for a contaminated site is an important decision. In addition, most research has focused on developing remediation technologies, especially for petroleum hydrocarbon compounds treatment, and on oil-water separation processes. Thus, a better understanding of the mechanisms will lead to a more effective and rational design to remediate oil-contaminated (water and soil) and provide new opportunities for remediation technology deployment and management. Joint research and development should be promoted to prevent, monitor or eliminate Environmental pollution. Several essential parameters, which are inherently conflicting, play an important role in this decision. Shortlisted technologies can be further investigated for practical application in situations of interest. **Nomenclatures**

- PHs Petroleum Hydrocarbons SDS Sodium Dodecyl Sulphate SVE Soil Vapor Extraction SVOC Semi-Volatile Organic Compounds TPHs Total Petroleum Hydrocarbons
- VOCs Volatile Organic Pollutants

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