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

*Mohammed Alsakit[a,b], Abdelsalam Abugharara[c,d], Amer Aborig[c,e] , Stephen Butt[c]

[a]Faculty of Science, Memorial University of Newfoundland, St. John's, NL A1B 3X7, Canada
[b]Faculty of Agriculture, Sebha University, Sebha Libya
[c]Faculty of Engineering & Applied Science, Memorial University of Newfoundland St. John's, NL, A1B 3X5, Canada
[d]Faculty of Energy and Mining Engineering, Sebha University, Sebha Libya
[e]Faculty of Engineering, University of Tripoli, Tripoli Libya

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**ABSTRACT**

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 major problem for the petroleum industry. Nowadays, the focus has been on the awareness of the treatment methods of 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-I focuses on the physical and chemical remediation technologies of oil-contaminated water.

# الابتكارات التكنولوجية للمعالجة الفيزيائية والكيميائية للمياه والتربة الملوثة بالنفط: دراسة مرجعيه (الجزء الأول)

*محمد الساكت[1,2] و عبد السلام أبوغرارة[3,4]و ,عامر أبوالريق[3,5] و ستيفن بت[3]

[1]كلية العلوم، جامعة ميموريال بنيوفاوندلاند، سانت جونز، كندا
[2] كلية الزراعة، جامعة سبها، ليبيا
[3]كلية الهندسة والعلوم التطبيقية، جامعة ميموريال بنيوفاوندلاند سانت جونز، كندا
[4]كلية هندسة الطاقة والتعدين، جامعة سبها، ليبيا
[5]كلية الهندسة،جامعة طرابلس،ليبيا

**الكلمات المفتاحية**
المياه الملوثة بالنفط
الملوثات البترولية
تقنيات العلاج
صناعة النفط
فصل الزيت عن الماء

**الملخص**

تنتج صناعة النفط كميات كبيرة من المياه الزيتية، إما من خلال عملية إنتاج النفط المسماة بالمياه المرتبطة، أو من خلال عملية تكرير النفط، أو بسبب تسرب خط الأنابيب من خلال نقل النفط. تعد المعالجة المسموح بها بيئيًا للمياه الزيتية مشكلة كبيرة لصناعة النفط. في الوقت الحاضر، تم التركيز على الوعي بطرق معالجة المياه الزيتية. ولذلك أصبح فصل الزيت عن الماء عقبة كبيرة حديثة، ويجب على الصناعات النفطية استكشافها

*Corresponding author:
E-mail addresses: ma7326@mun.ca, (A. Abugharara) a_nasar@mun.ca, (A. Aborig) ) amaborig@mun.ca , (S. Butt) sdbutt@mun.ca
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وحلها للوفاء بالمعايير المحددة وحماية البيئة. ومع ذلك، خلال العقود الأخيرة من تزايد التحضر والتصنيع في جميع أنحاء العالم، زاد استهلاك الزيوت ومشتقاتها بشكل كبير. بسبب بعض العوامل مثل سوء الإدارة، والاحتراق غير الكامل، وتسرب خطوط الأنابيب، وغيرها من الظروف العرضية، يتسرب النفط إلى الماء والتربة ولا يمكن تجنبه 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 الحلول الفعالة لتقنيات معالجة المياه والتربة للمواقع الملوثة بالنفط ضرورية للسلامة البيئية والنمو المستدام. تركز الدراسه المرجعيه الحاليه في جزئها الاول على تقنيات المعالجة الفيزيائية والكيميائية للمياه الملوثة بالنفط.

## 1. Introduction

Crude oil is a mixture of organic molecules called hydrocarbons, mainly carbon and hydrogen [1]. Such organic molecules are formed from the remains of the living organisms that lived and were buried millions of years ago and, over time, have been subjected to high pressures and temperatures that vary from depth to depth to transform into other substances, such as oil, gas and solids.

Oil spills due to offshore drilling rigs, the shipping sector (tankers, vessels, etc.), and industrial wastewaters (petroleum refineries, etc.) are the major sources of oily waters (Fig. 1).

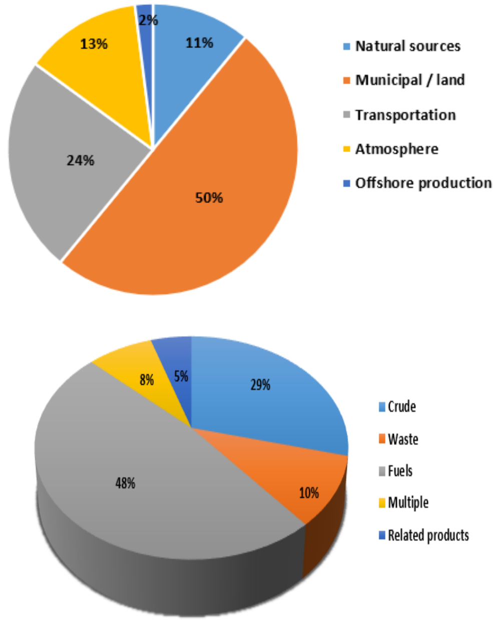


**Fig. 1:** Top: Sources of oil leaked into the seas worldwide, bottom: Types of oils spilled into sea [2,3]

Oily wastewater treatment produced from any source is necessary to avoid endangering aquatic and wildlife, as well as to respond with clear ecological regulations, which require a total oil and grease concentration in discharge waters of 10-15 mg/l [4,5,6,7]. In general, oil concentrations in wastewater produced in oil fields can be from 100 to 1,000 mg/l [4]. The physio-chemical characteristics, concentration, and drop size of the oil will decide the traditional method for handling oily wastewater.

Recent statistics reveal that worldwide, the primary causes of oil spills into the sea are attributed to tanker Accidents, pipeline leaks, natural disasters, etc. Table 1 provides recent statistical data on worldwide causes of oil spills into the sea with percentage contribution and regions.

**Table 1: Recent sstatistical worldwide causes of oil spills into the sea**

| Cause of Spill | Percentage Contribution | Region | References |
|---|---|---|---|
| Tanker Accidents | 10-15% | Global, notably in North America and Asia | ITOPF, 2024; NOAA, 2024 (ITOPF) (NOAA Response Restoration) |
| Pipeline Leaks | 15-20% | North America, Russia, Middle East | |
| Equipment Failures | 20-25% | Global | |
| Natural Disasters | 5% | Coastal regions (hurricanes, tsunamis) | ITOPF, 2024; NOAA, 2024 (ITOPF) (NOAA Response Restoration) |
| Human Error/Negligence | 20-30% | Global, high maritime traffic areas | |
| Offshore Drilling Operations | 5-10% | Gulf of Mexico, North Sea, Persian Gulf | |
| Deliberate Acts (War, Sabotage) | 5% | Conflict zones, Middle East | ACME Environmental, 2024; NOAA, 2024 (ACME Environmental) |
| Illegal Dumping | 5% | Less regulated regions | |

Methods for treating oily wastewater can be categorized as primary, secondary, or tertiary treatment. The primary treatment stage is removing the free oil (>40µm) using a centrifuge to separate the emulsion. The secondary treatment stage is removing the dispersed oil (>20µm), which includes the coagulation process and/or flocculation of the oil drops to separate the emulsion and an alteration in pH to neutralize the emulsion stabilizers. The target of the tertiary treatment is to decrease the concentration of inorganic salts and dissolved soluble oil fractions. The most popular methods are thermal processes (evaporation), reverse osmosis, and carbon adsorption [7]. Physical, chemical, and biological methods are commonly used to treat oily wastewater. Most traditional methods, such as centrifugation, sedimentation, coagulation, and filtration, are ineffective for treating stable oil-water mixture (O/W) emulsions, mainly when the oil droplets dispersed the concentration very low and distributed on a large area. In general, these techniques can only reduce oil concentrations by 1% by the volume of the total oil-water mixture quantity and cannot effectively remove oil droplets below 10µm [7] (Fig. 2).

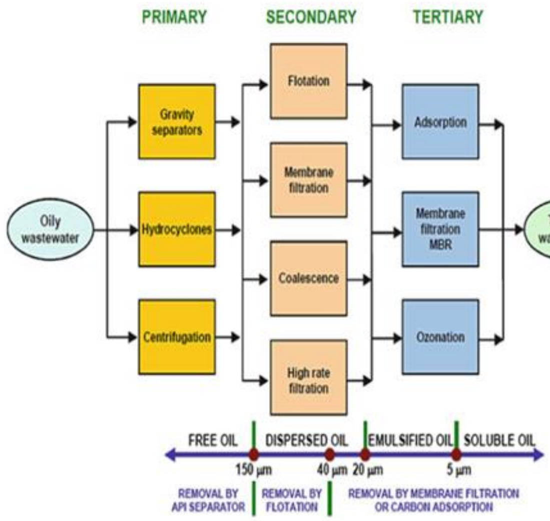


**Fig. 2:** Processes in the treatment of oily wastewater [7]

## 2. Water Remediation

In general, Water pollution is a major international problem caused by industrial, domestic, and environmental impacts. The United Nations estimates that 300-500 million tons of heavy metals, solvents, and other waste enter the world's water supply each year as hazardous by-products of industrial activity [8,9]. Water pollution can also be of natural origin. For example, countries such as West Bengal (India), Nepal, and Bangladesh have serious problems with arsenic contamination due to weathering of naturally arsenic-bearing rocks. Moreover, as the world's population continues to grow, it is predictable that human pressure on water supplies will increase, thereby increasing the potential for contamination. [8,10,11].

## 3. Water remediation technologies

### 3.1 Membrane technology

Membrane processes are becoming increasingly common in the remediation of oil-water mixtures. Recently, this technology has become a popular and important separation technology for the oil-water mixture. There are many advantages to using this technology, such as it is easy to handle, no chemicals needed lower energy requirements, etc. [12]. Membrane separation technology shows better performance than traditional techniques such as flotation, coagulation and biological treatment, and the efficiency of the technology is dependent on the membrane itself. In general, the pressure-driven membrane separation technology can be categorized into four types depending on the separation and pore size type as follows:

- (MF): Pore size for this group ranges from 0.1 - 5µm.
- (UF): The pore size ranges from 0.01 - 0.1µm.
- (NF): The pore size of NF technology ranges from 0.001 - 0.01µm.
- (RO): The pore size of RO ranges from 0.0001 - 0.001µm.

MF and UF membranes are both capable of removing particulate matter from a liquid phase, and UF has a high ability to remove viruses, bacteria, and emulsions. NF membrane is used to separate materials of nanometer size and to remove salts and organic compounds as well [13]. A technology called reverse osmosis (RO) is used to remove salt from seawater. This technology blocks the passage of sodium chloride (NaCl) through the membrane with an efficiency of more than 95% [14]. However, this membrane will reject most of the ionic species. Membranes can be bipolar or carry a positive or negative charge. The membrane technology, as an efficient method, is one of the most widely used technologies in the fuel cell industries for the separation of oil-water wastewater or emulsions. According to Yu L. et al. [15], the oily wastewater was purified from an oil field using a tubular UF module with polyvinylidene fluoride membranes. The results show that the oil content was below 1 mg/l the solid particle was less than 2 µm, and the oil removal efficiency was up to 99%. [16] was generated by a tubular carbon matrix obtained from carbonization microfiltration carbon. The results show that an oil content of less than 10 mg/l and an oil removal efficiency of up to 97% can be achieved using this technology.

### 3.2 Flotation technology

In general, oil spills increased significantly in the last decades of the twentieth century significantly, because of the very frequent of oil in the water bodies, there is a need arose to develop a cost-effective and easy-to-use oil/water separation technology to minimize the impact of these spills, help continue the economic activity without damage, and restore the natural life of the marine environment [17]. The method of floating arms is one of the methods used to treat the spill at the oil spill site (sea or ocean). However, the importance of this technique lies in limiting the spread of the oil spill above the (sea or ocean) surface and preventing discharge into the basins and adjacent water channels. The flotation method is one of the traditional methods of treating oily wastewater [18,19,20]. Flotation is pouring into the water in the form of fine bubbles, the tiny air bubbles in the adhesion of oil particles suspended in the water; because the floating density of oil is less than that of water, the formation of a scum layer is separated from the water [21]. The flotation method can be divided into air flotation, dissolved air flotation, electroflotation, and nozzle air flotation. Thus, due to different densities (oil and water), the floating oil layer separates from the water so that it can be separated, and floating oil is removed easily. However, there are many advantages of the flotation method, such as less sludge, separation efficiency, and high potential to treat oily wastewater. In addition, there are many disadvantages of flotation technology, such as high energy consumption, generation of a huge amount of air, and technicians.

### 3.3 Coagulation technology

According to Varjani S. et al. [22], the oil industry has developed a composite coagulant, and then when applying this composite, the oil removal efficiency reaches 98%. The coagulation technique is carried out by reducing the surface charge of the droplets and separating the oil droplets by coagulants, which promote the dispersion of the emulsion.

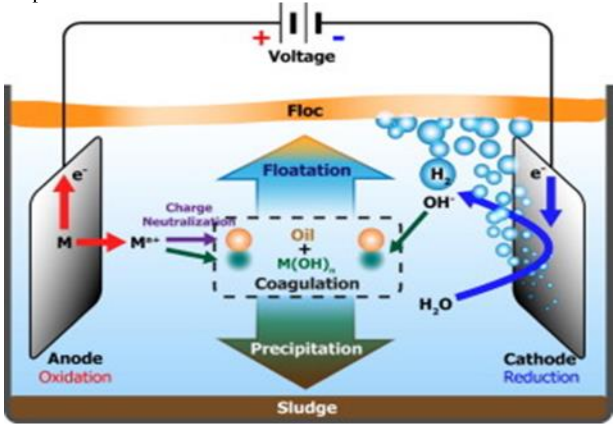


**Fig. 3:** Diagram of the mechanisms of electrocoagulation process[25]

Thus, this method is usually followed by separating the oily and aqueous stages by dissolved air flotation or precipitation [23]. However, the coagulation process (Fig. 3) is one of the traditional methods of oily wastewater treatment, which was heavily used previously, but because of some disadvantages, including high cost, high amount of coagulant materials, corrosion issues because of low pH, increased concentration of heavy metals in effluents and very expensive sludge production [24,25,26].

### 3.4 Biological technology

Biological methods typically remove oils and fats using biodegradation and are less costly than chemical alternatives. In this process, water dissolution occurs through microbial metabolism and colloidal organic contaminants, which are converted into healthy, harmless substances [15]. In general, there are types of bacteria that multiply and grow faster in an oil spill area. These types of bacteria contribute significantly to reducing the impact of oil spills through biotherapy. These bacteria can convert oil spills into non-harmful secondary products such as fats, proteins, and organic solvents. Besides, these bacteria also contain genes to produce proteins, through which they can cut the long hydrocarbons that originated from petroleum materials into small pieces and convert them into non-harmful materials. One of the most important methods to treat the oil spill is biological treatment [27]. However, it is ineffective in the short term, as it can take a long time to eliminate the effects of oil.

On the other hand, these bacteria may suffer from a deficiency in the primary nutrients that contribute to their growth, such as nitrogen, phosphorous, and oxygen. Therefore, the competent authorities add additional nutrients to the water environment to increase microbial community activity; thus, biological treatment can improve, and it is called (the biostimulation) process [24].

**3.5 Electrochemical technology**

In general, electrochemical is one of the most effective methods of oily wastewater treatment (Fig. 4). Recently, several electrochemical methods have been applied to treat oily wastewater from different sources [28,29].

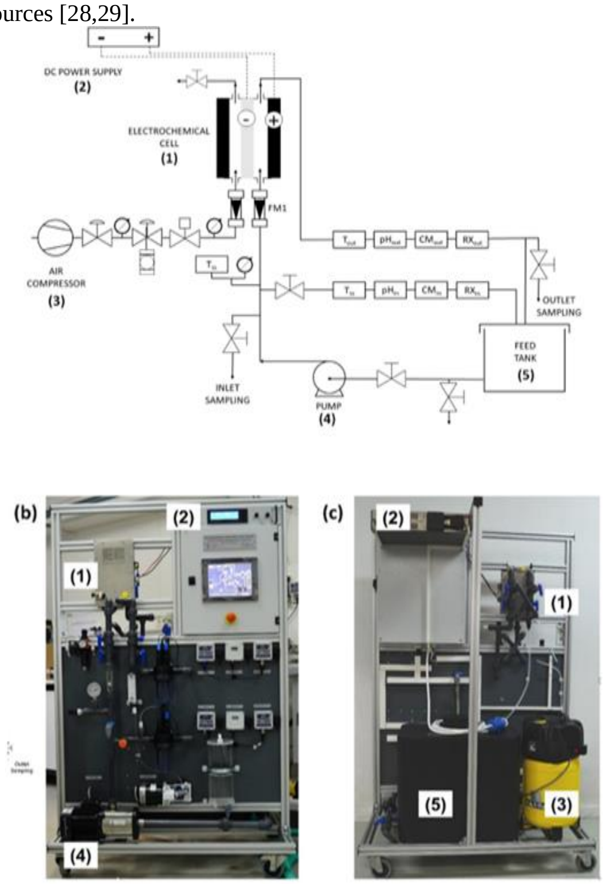


**Fig. 4:** Diagram of the electrochemical pilot-scale plant [28]

One of the most important approaches for removing chemical oxygen demand (COD) and petroleum hydrocarbons from oily wastewater is chemical oxidation. Electrochemical oxidation utilizes redox reactions to degrade organic matter into easily degradable materials directly or indirectly. In addition, the electrochemical oxidation method has many advantages, such as requiring no additional chemical activity, low space requirements, being simple to automate, and providing efficient treatment in a short period of time [30,31].

**4. Adsorbent materials for water remediation**

The Adsorption process (Fig. 5) is a method for separating the oil-water mixture by using solid adsorption materials with a large surface area as well as high porosity as a sorbent to absorb small and medium-sized oil molecules on the surface [32,33,34].

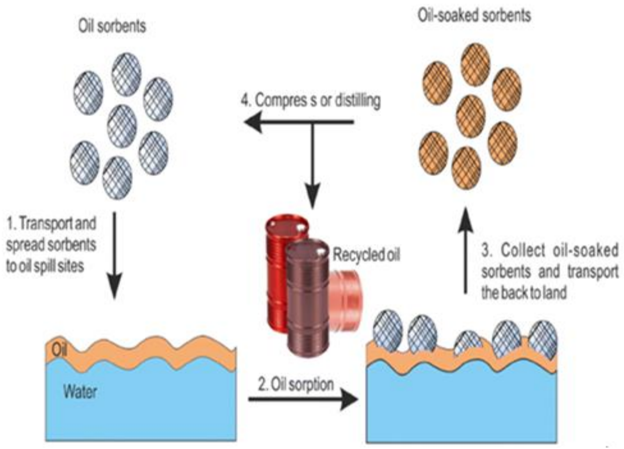


**Fig. 5:** The oil-spill cleanup and recycling of oil sorbents [34]

Therefore, when using adsorbent materials to treat oily wastewater, the adsorbent material can reduce the Chemical Oxygen Demand (COD) and adsorb indecomposable hydrocarbon organics as well as decolorize and deodorize the oily wastewater [35].

For this reason, the adsorption method has been commonly used in shipping oily wastewater treatment because it can handle sewage to a reusable level. Nowadays, researchers are working to develop high-efficiency sorbents from different materials for treating oily water, such as metal-organic framework, biomass, foam, chitosan, magnetite nanoparticles, sponge and cotton [36].

**4.1 Biochar**

Generally, the sorbent method is one of the important methods used in treating oily wastewater [34]. Biochar (BC) is an adsorbent material, and it has many advantages and great efficacy for oil cleanup, including low cost, availability on a commercial scale, easily recoverable, and safe disposition. Moreover, biochar is derived from available agricultural by-products or waste and often has a large surface area, and due to its characteristics, it can contribute to increased oil sorption and oily wastewater treatments. Generally, biochar (BC) describes a carbonaceous substance produced from biomass through pyrolysis under nearly anoxic conditions. BC is obtained by pyrolysis of sustainably produced biomass using pyrolysis technology under controlled conditions. The carbonaceous materials are produced from any carbon source. Biomass is waste materials suitable for biochar, including crop residues (both field residues and manufacturing residues such as nutshells, fruit pits, bagasse, etc.) as well as yard, food, and forestry waste and animal manure [37,38,39]. Biochar is manufactured to treat oily wastewater as a sorbent to eliminate contaminants from the gases and fluids [40,41].

Standard techniques for oily wastewater treatment include the utilization of sorbent materials; adsorption is widely utilized for remediation purposes to eliminate an enormous of contaminants, such as heavy metals and organic combinations. However, carbonaceous materials have an excellent capacity for adsorption due to their high surface area and colossal porosity as well. Generally, porosity and surface area increase when the activation temperature increases. Under a circumstance, carbon condensation is attributed to the development of dense, highly disordered graphene layers named turbostratic crystallites attributable, therefore forming greater micropores and nanopores within the biochar structure. However, Pyrolysis involves the thermal decomposition of biomass without oxygen, resulting in solid (char), gas, and liquid products (the latter can be extracted from the condensable gas fraction). Biochar, an enduring and resistant organic carbon material, is formed through biomass pyrolysis at temperatures ranging from 300 to 1000 °C under minimal oxygen conditions.

According to Manya, J. J. [42], the conditions of the pyrolysis process of biochar are significantly influenced by the temperature, which is the most important parameter influencing the characteristics of biochar, especially the surface area. In general, the surface area of biochar can be extraordinarily increased through a physical or chemical activation process. For example, the physical activation processes progress to a significant increase in the biochar surface area through the factor of temperature (high or low), and chemical reaction progress happens after that through adding catalysts such as $ZnCl_2$, etc. [43,44,45]. However, high pyrolysis temperature leads to greater specific surface area and aromaticity of biochar. Biochars are efficient adsorbents for the removal of various pollutants due to their special properties such as surface area and porosity. Biochar's have become increasingly relevant as a solution for the remediation of contaminants in the industrial and agricultural sectors to improve environmental conditions. Hence, biochar (BC), is used as an adsorption material for the treatment and removed contaminants from oily wastewater and sediments.

### 4.2 Metal-organic frameworks (MOFs)

Metal-organic frameworks (MOFs) have appeared as a large class of crystalline materials with high porosity (more than 90%) and massive internal surface areas extending more than 6000 $m^2/g$. Thus, these properties, degree of contrast, and homogeneity in structures make MOFs of interest for potential applications in oil-water separation [46,47]. MOFs are also of great importance through their work as storage media for gases such as hydrogen and methane and high-absorbent capabilities to meet different separation needs [48]. Metal-organic frameworks are crystalline structures containing mineral ions and organic linkers; these components will form a tridimensional structure [49]. The content is known for its largest pore and high surface area. According to Jeffrey, R et al. [50], MOFs are porous structures constructed from the coordinative bonding between metal ions and organic linkers or bridging ligands (Fig. 6). The singular features of MOFs, such as their great surface areas and variety of structures, will make them suitable for a broad domain of industrial processes [51,52]. The metal-organic framework's structure can be prepared to capture specific gases and chemicals while other substances pass through. The MOFs are, therefore, well-suited for efficient separation processes [53,54].

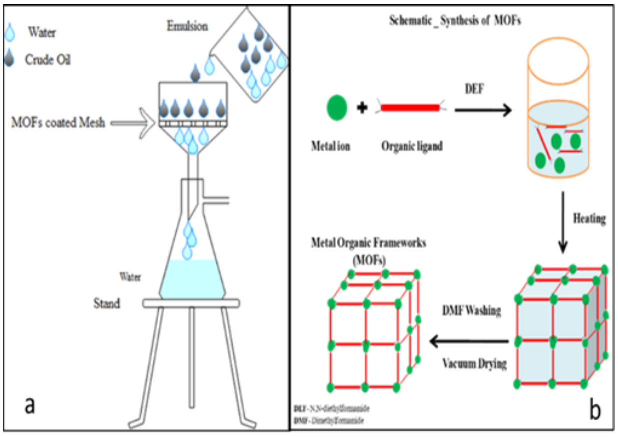


**Fig. 6: (a)** Schematic diagram of the separation process using the MOFs coated mesh (two-stage process mesh and polish). **(b)** Schematic diagram of the synthesis of the (MOFs).

Many researchers have concentrated on the pore size and pore surface of MOF-5 in order to enhance its efficiency for different applications. Intense work has been carried out over the last two decades on MOFs, which have developed and increased the variety of porous materials [55,56]. According to Shervani S [57], the separation efficiency of the mesh was 89%, and after the polishing phase, it was more than 99%. Recently, the nanomaterials of MOFs have become important due to their great properties, such as high surface area and porousness, when compared with other materials [58].

### 5. Groundwater Contamination

Groundwater is the main source of fresh water for the entire world's population, and it is often used for domestic, agricultural and industrial purposes. Groundwater is the primary source of drinking water for nearly a third of the world's population [59,60]. Recent decades have seen a significant increase in the focus of groundwater investigations on chemical contamination (Fig. 7). Although groundwater contamination poses a serious concern to human populations, it also allows scholars to understand better how our subsurface aquifers have changed over time and help decision-makers understand how we can protect the quality and quantity of these resources [62].

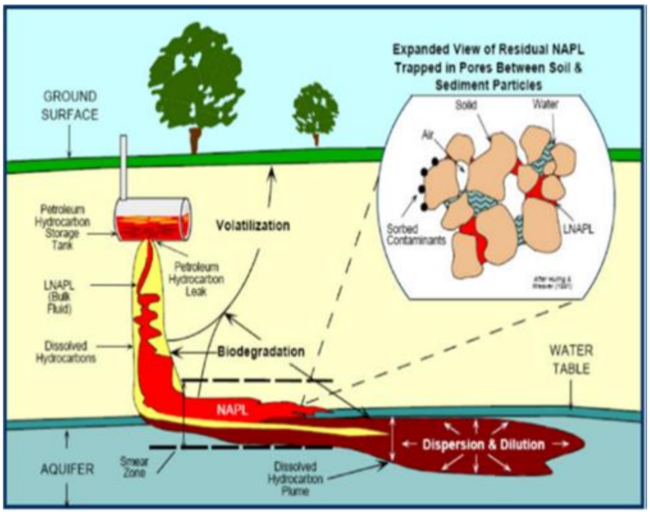


**Fig. 7:** Groundwater pollution due to leaching of petroleum hydrocarbons [61]

The Canadian government defines Groundwater contamination as adding organic or inorganic pollutants to groundwater due to human activity. Groundwater contamination can be caused by petroleum, microorganisms, brines, fertilizers, and chemicals [63].

### 6. Groundwater remediation technologies

Scientists have recently created very effective methods for purifying water from numerous pollutants. The main subject of these procedures was the remediation of surface water resources, such as rivers, lakes, and water reservoirs, because these are the primary sources of groundwater. Groundwater is roughly 30% of the world's freshwater reserve and has become a crucial water source in most regions. Because of that, scientists and environmental researchers are increasing awareness of the risks of the pollutants and removing them from underground water using safe technologies [60].

### 6.1 Pump and Treat Technology

Pump and treat are one of the traditional technologies used for groundwater remediation. Removing the dissolving chemicals, heavy metals, solvents and fuel oil that contaminated groundwater is the most critical point of groundwater remediation. In this process, contaminated groundwater is piped to treatment units or ground lagoons, where it is treated using various techniques, including activated carbon or air stripping. Generally, through well-established groundwater remediation technology, pollutants in surface-pumped groundwater can be reduced to very low levels. The removal of all toxins from the subsurface is not assured by pumping contaminated water from an aquifer. However, the behaviour of contaminants in the subsurface, site geology and hydrogeology, and extraction system design all have an impact on how much contamination can be removed [64]. Thus, pump and treat technology can handle high amounts of contaminated groundwater and simple equipment; there are several disadvantages, including the high cost, the spread of toxins into the ecosystem, the lengthy operation period, and the possibility of reversing the hydraulic gradient [60,65].

### 6.2 Air Sparging Technology

The air-sparging process is one of the most commonly used techniques to remediate groundwater contaminated with volatile organic pollutants (VOCs).

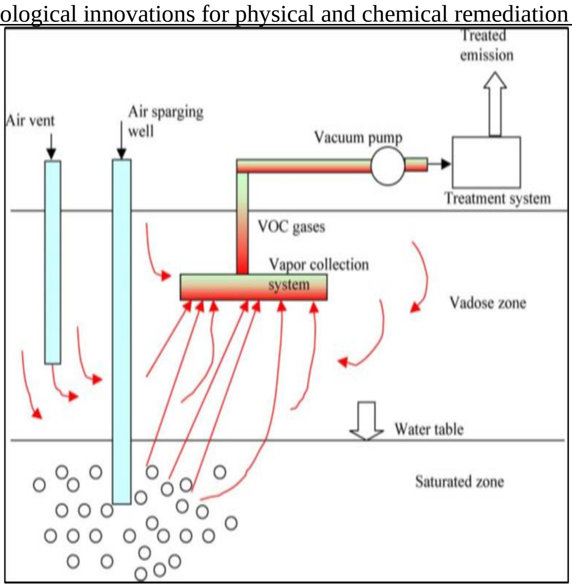


**Fig. 8:** Air sparging with soil vapor extraction process [67]

It is considered efficient, fast, and relatively economical [66]. In this process, compressed air is injected into the nadir of contaminated groundwater (Fig. 8).

It purifies subsurface water by changing the volatile hydrocarbon state to a vapor state. Pumping air below the saturation zone removes contaminants from the aquifer and provides oxygen for biodegradation [68]. To remove toxic impurities, the extracted air must be treated with a vacuum extraction system [69]. A limitation of this technology is that it is an expensive method when working with hard surface areas, or many deep wells are required for remediation. Furthermore, soil heterogeneity can lead to the uneven treatment of contaminated groundwater [60].

A detailed table summarizing various technological innovations for physical and chemical remediation of oil-contaminated water including information on the type of process, type of separation, entry level, size of oily wastewater droplets, efficiency percentage, materials used, risk involve is contained in table (2).

**Table 2: A detailed table summarizing various technological innovations for physical and chemical remediation of oil-contaminated water.**

| Type of Process | Tech-nology | Entry Level | Size of OilyWater Droplets | Efficienc % | Materials Used | Risk Involved | Authors |
|---|---|---|---|---|---|---|---|
| Physical | Membrane Filtration | Advanced | Micron to sub-micron | 85-95% | Polymeric membranes, ceramic membranes | Membrane fouling | Zhao et al. (2024) |
| Chemical | Electroc-oagulation | Inter-mediate | Micron | 70-90% | Aluminum or iron electrodes | Low electrical risk | Wang et al. (2024) |
| Chemical | Advanced Oxidation Processes (AOPs) | Advanced | Sub-micron to nano | 80-98% | Ozone, hydrogen peroxide, UV light | Chemical handling | Li et al. (2024) |
| Biological | Bioremed-iation | Basic | Macro to micron | 60-85% | Microorganisms, nutrients | Low | Chen et al. (2024) |
| Physical | Centrifugal Separation | Inter-mediate | Macro to micron | 70-85% | Centrifuges | Mechanical risk | Kumar et al. (2024) |
| Physical/Chemical | Nanotech-nology-based Adsorption | Advanced | Nano to sub-micron | 75-90% | Nanoparticles, functionalized materials | Low | Zhang et al. (2024) |

## 7. Conclusion

The review outlines the challenges and considerations in remediating water environments contaminated with petroleum hydrocarbons. It highlights several important key points:

**(i)** Variety of Remediation Technologies: Although many technologies are available, no single technology suits all pollutants and site conditions universally. **(ii)** Site-Specific Considerations: Choosing a remediation strategy depends on factors like site conditions, types of pollutants, sources of contamination, and potential remediation measures. (**iii)** Combination of Technologies: Often, multiple technologies must be used together or sequentially to address contamination effectively. Process trains combining different methods can enhance pollutant removal. **(iv)** Importance of Research and Development: Significant research focuses on developing technologies for treating petroleum hydrocarbons and improving oil-water separation processes. Ongoing research is crucial for understanding mechanisms and enhancing remediation effectiveness. **(v)** Importance of Research and Development: Joint research efforts are advocated to prevent, monitor, and eliminate environmental pollution. Collaboration helps resolve conflicts in decision-making. **(vi)** Practical Application: Identified technologies should be investigated for practical application in specific situations of interest, considering efficiency and cost-effectiveness.

Overall, the review stresses the need for a holistic approach to remediation, integrating multiple factors and techniques to achieve efficient and sustainable restoration of contaminated environments

## Nomenclatures

| | |
|---|---|
| **µm** | Micrometres |
| **BC** | Biochar |
| **COD** | Chemical Oxygen Demand |
| **MF** | Microfiltration |
| **MOF-5** | $Zn_4O(BDC)_3$ |
| **MOFs** | Metal-organic frameworks |
| **NF** | Nanofiltration |
| **O/W** | Oil-water mixture. |
| **RO** | Reverse Osmosis |
| **UF** | Ultrafiltration |
| **VOCs** | volatile organic pollutants |

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