Assessment of Canadian Regulations and Remediation Methods for Diesel Oil Contaminated Soils

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Abstract: Diesel fuel released into the environment can contaminate ground water, degrade potable water supplies and cause the collapse of fisheries. They are toxic to both animals and humans and can affect the liver, lungs, kidneys, and nervous system leading to cancer as well as immunological and reproductive effects. The objectives of this study were to review current Canadian regulations pertaining to diesel fuel and to evaluate the current remediation methods using five criteria: efficiency, applicability, cost, time and cleanliness. PAHs are deemed toxic under the Canadian Environmental Protection Act but no standards have been set for PAHs in diesel. The Canadian Council of Ministers of the Environment (CCME) has developed Canada-Wide Standards for Petroleum Hydrocarbons in Soil (CWS PHCS) while the Atlantic PIRI has implemented a Risk Based Corrective Action (RBCA) for the Atlantic region. The remediation methods included soil washing, landfilling, incineration, thermal desorption, radio frequency heating, chemical addition, landfarming, biopiling, composting, bioventing, liquid delivery and bioreactors. The bioreactors studied included: static bed, continuous mix, horizontal drum, fungal compost, slurry-phase, DITS, biofilters and packed bed bioreactors. The results showed that the biological methods were more effective than nonbiological ones and the bioreactors scored the highest among the biological methods. Eight criteria were then used for the evaluation of bioreactors: efficiency, time, cost, maintenance, simplicity, release of VOCs to the atmosphere, containment of contaminants and control of operating parameters. The results showed that the continuous mix bioreactor was the most effective system.

Key words: Diesel fuel, remediation, regulation, ecosystem, physical, chemical, biological, evaluation

INTRODUCTION

Diesel Fuel is intended for use in compression engines such as those found in trucks, trains and subtrains. It is composed of a variety of organic compounds as shown in Table 1. As the fuel weathers over time, the concentration of these compounds change due to volatilization and degradation to other compounds.

Accidental diesel spills and the leakage of underground storage tanks have far reaching impacts on the environment. A study on marine iguanas on one of the Galapagos Islands in Ecuador has shown that 62% of the species population has died since the oil tanker spill that occurred 1500 m offshore in 2001. There are over 400,000 petroleum hydrocarbon contaminated sites in the USA alone as a result of spillage and leakage of underground tanks located at airports, refineries and farms. Pockets of oil on these sites can persist in the environment for many years. The study on the 700,000 L diesel spill of 1969 (which is only one sixtieth of that spilled by Exxon Valdez) is still going on by Woods Hole Oceanographic Institute of Massachusetts.

According to Riser-Roberts, hydrocarbons in the soil are considered toxic when they reach concentrations greater than 100 µg/g soil. The soluble compounds of diesel (benzene, toluene, ethyl benzene, and xylenes which are known as BTEX) are toxic to aquatic life as well as animals and humans. Diesel released into the environment can contaminate ground water, degrade potable water supplies and cause the collapse of fisheries. Polycyclic aromatic hydrocarbons (PAHs) in diesel (such as naphthalene) have long term effects on soil, ground water and sediments and can act as endocrine disruptors (i.e. interfere with hormone production and function). The PAHs and BTEX affect the liver, lungs, kidneys and nervous system leading to cancer, immunological, reproductive, fetotoxic and genotoxic effects.
CCMEm summarizes the pathways through which humans and wildlife can be exposed to contaminants (Table 2).

Table 1: Composition of Diesel Fuel #2\[2\]

| Component          | Concentration (% Volume) |
|--------------------|--------------------------|
| C10 paraffins      | 0.9                      |
| C10 aromatics      | 0.4                      |
| C10 cycloparaffins | 0.6                      |
| C11 paraffins      | 2.5                      |
| C11 aromatics      | 1.0                      |
| C11 cycloparaffins | 1.7                      |
| C12 paraffins      | 3.8                      |
| C12 aromatics      | 1.6                      |
| C12 cycloparaffins | 2.8                      |
| C13 paraffins      | 6.4                      |
| C13 aromatics      | 2.8                      |
| C13 cycloparaffins | 4.8                      |
| C14 paraffins      | 8.8                      |
| C14 aromatics      | 3.8                      |
| C14 cycloparaffins | 6.6                      |
| C15 paraffins      | 7.4                      |
| C15 aromatics      | 3.2                      |
| C15 cycloparaffins | 5.5                      |
| C16 paraffins      | 5.8                      |
| C16 aromatics      | 2.5                      |
| C16 cycloparaffins | 4.4                      |
| C17 paraffins      | 5.5                      |
| C17 aromatics      | 2.4                      |
| C17 cycloparaffins | 4.1                      |
| C18 paraffins      | 4.3                      |
| C18 aromatics      | 1.8                      |
| C18 cycloparaffins | 3.2                      |
| C19 paraffins      | 0.7                      |
| C19 aromatics      | 0.3                      |
| C19 cycloparaffins | 0.6                      |

CANADIAN REGULATIONS

Federal Regulations: Many of the regulations that pertain to diesel fuel in Canada relate to its sulphur content\[8\], since the production of \( \text{SO}_2 \) during combustion and exhaust is the leading cause of acid rain. However, diesel-powered vehicles are a significant source of aromatic hydrocarbons in urban areas. Human exposure to diesel containing benzene at any concentration will have adverse health effects. Although PAHs (like benzene) are considered toxic under the Canadian Environmental Protection Act (CEPA), there are no standards for PAHs in diesel\[9\]. CEPA regulations apply to quantities greater than 400 m\(^3\) of fuel produced or imported into Canada that contains any additives. The petroleum industry is required to report sulphur content and any additives in the fuel, other than lead, to the Minister of Environment, where the liquid fuel is from crude oils, coal, or bituminous sands.

Many factors affect the cost of the diesel clean-up in Canada including: (a) the accessibility or remoteness of the spill location, whether the spill is located on land, in a river, or in the ocean, (b) the weather conditions, (c) the quantity spilled, (d) the extent of environmental damage, and (e) the time required for the clean-up. Blondeau\[10\] reported that, based on the data obtained from the Saskatchewan Spill Response Center, the leading causes of spills are equipment failure and accidents during road transport and most of the spills documented were from petroleum, transportation and mining companies.

About 60 % of Canada’s contaminated sites involve petroleum hydrocarbons (PHCs) that can cause fires and/or explosions on these sites and impair the quality and uses of land and water. The Canadian Council of Ministers of the Environment (CCME) developed Canada-Wide Standards for Petroleum Hydrocarbons in Soil (CWS PHCS) in 2001\[7\]. These standards separate soil under four different land uses: agricultural, residential/parkland, commercial, and industrial. Table 3 shows the allowable petroleum hydrocarbon (PHC) fractions in soil, depending on the land use. The CWS PHCS specifies the methods and outcomes for the assessment and management of contaminated sites but timelines are left for individual jurisdictions to decide. When assessing a contaminated site, one must also consider ignition hazards, toxicity, odor, appearance of the contaminants, effects on buried infrastructure, and formation of non-aqueous-phase liquids (NAPL). Table 4 shows the required site characterization. All provinces and territories except Quebec have endorsed CWS PHCS and the legislation for its enforcement.

Regulations in the Atlantic Region: The Atlantic Provinces (Nova Scotia, New Brunswick, Prince Edward Island, and Newfoundland and Labrador) have a harmonizing partnership agreement called the Risk Based Corrective Action (RBCA) agreement with its own set of PHC guidelines. The Atlantic Partnership in RBCA Implementation (PIRI) ensures that RBCA is effective and serves the needs of Atlantic Canadians by returning more sites to safe use at a reduced cost. The RBCA has been in use since 1999 and it differs from the CWS PHCS with respect to criteria for laboratory procedures for the comparison of site data\[11\].

RBCA is a 3-tiered approach to risk assessment and risk management. Tier I uses the risk-based screening levels from Table 5 to determine the need for and the extent of removal of any remedial work required after confirmation of site applicability. It identifies the presence of ecological receptors on or adjacent to the site (within 150 m) and the potential for ecological receptors to be exposed to the release of.
hydrocarbons. Tier II uses the values from Table 6 to determine the need for and the extent of remedial work required. Tier III is triggered by an ecological risk assessment, even if human health risk is managed under RBCA. RBCA is used in residential and commercial land use settings and adult is the default receptor\[^{[11]}\].  

**REMEDICATION METHODS**

Soil remediation can take place either in-situ or ex-situ using one or more of the current remediation technologies. These include: physical, thermal, chemical and biological processes\[^{[2]}\].

**Physical Processes:** Physical remediation technologies include soil washing and landfiling.

**Soil washing:** Soil washing is when a wash solution (water and/or a surfactant) is added to soil to remove contaminants. The contaminant is transferred from the soil to the wash solution, which then must be treated. Residual sludge is often associated with this method. Water alone is not effective in removing PAHs. Haapia and Tuhkanen\[^{[12]}\] reported that the amount of total PAHs in the soil decreased by about 50% after soil washing as the PAHs were transferred into the washing water. Viglianti et al.\[^{[13]}\] found the addition of cyclodextrins significantly improved the soil washing process. Rajput et al.\[^{[14]}\] used soil washing to remove 1,2,4-trichlorobenzene (TCB), aniline, phenol, and 2,4-dichlorophenol (DCP) and found water washing to be suitable for removing all contaminants except TCB which required washing with surfactant first and then rinsing with water. Other solutions such as hydrochloric acid and sodium hydroxide have also been used in soil washing techniques\[^{[15,16]}\].

**Landfilling:** Landfilling is one of the oldest forms of remediation. Contaminated soil is excavated from the site and transported to a landfill where it remains indefinitely. In cases where the soil is brought to a first generation landfill, there is still the potential for the contaminants to enter groundwater or bedrock. PAHs can contaminate landfill leachate and their presence has been reported\[^{[17,18]}\]. However, methods have been devised for the removal of aromatic organics from soil which may help to control these contaminants in landfills\[^{[19]}\].

**Thermal Processes:** There are currently three thermal remediation technologies in use: incineration, thermal desorption and radio frequency heating.

**Incineration:** Incineration is the destruction of contaminants by burning contaminated soil. This method can achieve greater than 99.99% success in destroying carbon tetrachloride, chlorinated benzenes and polychlorinated biphenyls but is very expensive\[^{[2]}\]. Benzene is adequately destructed via thermal incineration at temperatures ranging from 850 to 973 K\[^{[20]}\]. However, catalytic incineration is more efficient than thermal incineration in destroying aromatic hydrocarbons. CuO/CeO\(_2\) and CuO/\(\gamma\)-Al\(_2\)O\(_3\) were found to be effective catalysts in the incineration of toluene and \(p\)-xylene\[^{[21-23]}\]. Arsenijević et al.\[^{[24]}\] used a Pt/Al\(_2\)O\(_3\) catalyst to incinerate ethylene oxide. Tseng and Chu\[^{[25]}\] studied the catalytic incineration of styrene (also known as vinyl benzene) using MnO/Fe\(_2\)O\(_3\) as a catalyst.

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Table 2: Land uses, key receptors and exposure pathways\[^{[7]}\]

| Exposure Pathway       | Agriculture | Residential/ Parkland | Commercial | Industrial |
|------------------------|-------------|-----------------------|------------|------------|
| Soil contact           | Nutrient cycling | Nutrient cycling       | Nutrient cycling | Nutrient cycling |
|                        | Soil invertebrates | Invertebrates         | Invertebrates | Invertebrates |
|                        | Crops (plants)  | Plants                | Plants      | Plants      |
|                        | Human (child)   | Human (child)         | Human (child) | Human (adult) |
| Soil ingestion         | Herbivores     | Wildlife*              | Wildlife*   | Wildlife*   |
|                        | Human (child)   | Human (child)         | Human (child) | Human (adult) |
| Groundwater/ Surface water | Aquatic life | Aquatic life          | Aquatic life Human (child) | Aquatic life Human (adult) |
|                        | Livestock watering | Human (child)         | Human (child) | Human (adult) |
| Vapour inhalation**    | Child (indoor)*** | Child (indoor)       | Child (indoor) | Adult (indoor) |
| Produce (meat and milk produced on site**) | Child | Child (produce only) | -  | -  |
| Off-site (migration of soil/dust)*** | -  | -  | Human/eco |  |

* Wildlife dermal contact and ingestion data may be particularly important for PHCs, but there are unlikely to be sufficient data to develop guidelines that address this exposure pathway.

** Humans only

*** A 30m horizontal offset is assumed between the farm residence and the PHC contamination, consistent with oil and gas development practices. Contamination nearer a farm residence triggers a residential assessment.
Table 3: Tier 1 levels for surface soil

| Land Use           | Soil Texture | PHC (mg/kg) |
|--------------------|--------------|-------------|
|                    | Fraction 1   | Fraction 2  | Fraction 3 | Fraction 4 |
| Agricultural       | Coarse 130   | 450 (150°)  | 400        | 2800       |
|                    | Fine 260 (180°) | 900 (250°)  | 800        | 5600       |
| Residential/ Parkland | Coarse 30°     | 150°        | 400        | 2800       |
|                    | Fine 260 (180°) | 900 (250°)  | 800        | 5600       |
| Commercial        | Coarse 310 (230°) | 760 (150°)  | 1700       | 3300       |
|                    | Fine 660 (180°) | 1500 (250°) | 2500       | 6600       |
| Industrial        | Coarse 310 (230°) | 760 (150°)  | 1700       | 3300       |
|                    | Fine 660 (180°) | 1500 (250°) | 2500       | 6600       |

Where applicable, for protection against contaminated groundwater discharge to an adjacent surface water body.

Where applicable, for the protection of potable groundwater. Assumes contamination near residence with slab-on-grade construction.

Table 4: Site characterization

| Characteristics                  | Description                                                                 |
|---------------------------------|-----------------------------------------------------------------------------|
| Land use                         | Historical, existing, intended, and potential land uses at the site and its surroundings, including the presence or absence of any critical wildlife habitat |
| Proximity                        | Distance between the site and surface water especially drinking water supplies |
| Groundwater depth                |                                                                                      |
| Human receptors                  | Children and adults                                                                |
| Ecological receptors             | Microorganisms responsible for nutrient cycling, soil invertebrates, plants, wildlife, and aquatic life |
| Exposure pathways                | Soil contact, soil ingestion, groundwater/surface water, vapor inhalation, produce, meat and milk produced on site, and off-site migration of soil/dust |
| Stratigraphy                     | Properties of surficial materials, especially soil texture                         |
| Depth to contamination            | Distance to points of exposure or compliance                                         |
| Built environment                | The presence and type of buildings, utility corridors, and conduits                |
| Contaminants                     | Characterization and delineation of contaminants such as toxicity, ignitability, solubility and volatility |

Table 5: Tier I TBSL for soil[11]

| Receptor         | Groundwater Use | Soil Type | Compound of concern in soil (mg/kg) | Modified TPH |
|------------------|-----------------|-----------|-------------------------------------|--------------|
|                  |                 | Benzene   | Toluene    | Ethyl Benzene | Xylenes | Gas | Diesel/#2 | #6 oil |
| Residential      | Potable         | Coarse 0.03 | 0.38      | 0.08        | 11      | 39  | 140       | 690    |
|                  |                 | Fine 0.01  | 0.08      | 0.02        | 2.3     | 140 | 220       | 970    |
|                  | Potable         | Coarse 0.16 | 0.38      | 0.08        | 11      | 39  | 140       | 690    |
|                  |                 | Fine 1.5   | 120       | 430         | 160     | 330 | 4400      | 8300   |
|                  | Non-potable     | Coarse 0.03 | 0.38      | 0.08        | 11      | 450 | 7400      | 10000  |
|                  |                 | Fine 0.01  | 0.08      | 0.02        | 2.3     | 520 | 840       | 4700   |
|                  | Non-potable     | Coarse 1.8  | 160       | 430         | 200     | 450 | 7400      | 10000  |
|                  |                 | Fine 11     | 680       | 430         | 650     | 10000 | 7700     | 10000  |

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|                  |                 | Fine 11     | 680       | 430         | 650     | 10000 | 7700     | 10000  |

Everaert and Baeyens[26] review catalytic oxidation processes for volatile organic contaminants (VOCs).

**Thermal desorption:** In thermal desorption, soil is heated under an inert atmosphere to increase the vapor pressure of organic contaminants causing the contaminants to volatilize and be released from the soil[27]. Merino and Bucala[28] reported that hexadecane can be nearly completely removed at 300°C without risk of pyrolysis. Piña et al.[29] found that low heating rates practically eliminate gas oil from soil matrices while avoiding significant chemical transformations but higher temperatures were required to achieve optimal removal efficiencies.

**Radio frequency heating:** Radio frequency power has been used in the steam reforming of hydrocarbons. These waves are converted to thermal energy in the soil for heating the contaminants and causing their volatilization. This procedure is very expensive but because the heat can be directed, the treatment is more accurate[2]. Al-Mayman and Al-Zahrani[30] cracked Saudi light oil into lower olefins using radio frequency heating. Shih et al.[31] decomposed benzene (C_6H_6) in radio frequency plasma environments and found naphthalene (C_{10}H_{8}) to be a predominant product.

**Chemical Processes:** Peroxide or an alkaline solution with a pH of 10.5 containing cobalt (III) can be added to contaminated soil to oxidize organic contaminants to
Table 6: Tier II Pathway-Specific Screening Level (PSSL) for soil\(^{[11]}\)

| Receptor Groundwater Use | Soil Type | Exposure Pathway | Benzene (mg/kg) | Toluene (mg/kg) | Ethyl Benzene (mg/kg) | Xylenes (mg/kg) | Modified TPH (mg/kg) |
|--------------------------|-----------|------------------|----------------|----------------|----------------------|----------------|----------------------|
| Residential Potable Coarse | Soil ingestion | 390 | 12000 | 7000 | 120000 | 8900 | 5300 | 8300 |
| Soil leaching Indoor air | 0.031 | 0.38 | 0.083 | 11 | 680 | 1100 | 630 | 4400 | >RES |
| Fine | Soil ingestion | 390 | 12000 | 7000 | 120000 | 8900 | 5300 | 8300 |
| Soil leaching Indoor air | 0.0071 | 0.082 | 0.018 | 2.3 | 140 | 220 | 970 |
| Non-potable Coarse | Soil ingestion | 390 | 12000 | 7000 | 120000 | 8900 | 5300 | 8300 |
| Soil leaching Indoor air | 1.5 | 120 | >430 | 160 | 330 | 4400 | >RES |
| Fine | Soil ingestion | 390 | 12000 | 7000 | 120000 | 8900 | 5300 | 8300 |
| Soil leaching Indoor air | Not applicable for non-potable scenarios | 1.8 | 160 | >430 | 200 | 450 | 7400 | >RES |

Commercial Potable Coarse | Soil ingestion | 570 | 18000 | 10000 | 180000 | 13000 | 7700 | 12000 |
| Soil leaching Indoor air | 0.031 | 0.38 | 0.083 | 11 | 2500 | 11000 | >RES |
| Fine | Soil ingestion | 570 | 18000 | 10000 | 180000 | 13000 | 7700 | 12000 |
| Soil leaching Indoor air | 0.0071 | 0.082 | 0.018 | 2.3 | 520 | 840 | 4700 |
| Non-potable Coarse | Soil ingestion | 570 | 18000 | 10000 | 180000 | 13000 | 7700 | 12000 |
| Soil leaching Indoor air | Not applicable for non-potable scenarios | 11 | >680 | >430 | >650 | >RES | >RES | >RES |
| Fine | Soil ingestion | 570 | 18000 | 10000 | 180000 | 13000 | 7700 | 12000 |
| Soil leaching Indoor air | Not applicable for non-potable scenarios | 0.16 | 14 | 58 | 17 | 39 | 140 | 690 |

Table 7: Advantages and disadvantages of nonbiological methods\(^{[67]}\)

| Process | Advantages | Disadvantages |
|---------|-------------|---------------|
| Soil Washing | Relatively efficient Quick Uses water | Transfers contaminants to a different phase Surfactants are often necessary Requires infrastructure |
| Landfilling | Removes contamination from site | Transfers pollution to another site Expensive Land requirement Transportation costs |
| Incineration | Quick Destroys hydrocarbons | Expensive Can cause air pollution Irreversible soil degradation |
| Thermal Desorption | Broad applicability High removal efficiencies No excavation required in situ | At high temperatures contaminants may volatilize and are released from the soil into the air Low temperatures do not achieve optimal removal efficiencies Energy use |
| Radio Frequency Heating | Accurate | Very expensive Products must be disposed of |
| Chemical Addition | Simple Oxidizes organic contaminants Soil left intact | Extracts must be treated Excavation |

CO\(_2\) and CO. Supercritical water can also be used to oxidize hazardous materials\(^{[32-34]}\). Acetone, methanol and ethanol solutions can then be used to extract compounds like benzene and pyrene\(^{[2,35]}\).

**Biological Processes:** There are a number of biological technologies currently in use: landfarming, biopiling, composting, bioventing and liquid delivery. It should be noted that hydrocarbon concentrations of less than 10 µg/l do not usually stimulate microbial growth\(^{[2]}\) and hydrocarbons with rings or many branches are slower to biodegrade\(^{[36]}\). Ghaly and Pyke\(^{[37]}\) reported that hydrocarbons with heavy molecular weight (C\(_{12}\)H\(_{12}\)-C\(_{31}\)-C\(_{64}\)) are slower to biodegrade.

**Landfarming:** Millions of tons of contaminated soil are treated by landfarming annually in the USA and Canada and more than half of which is associated with petrochemical contaminants. The processes involved with this method of treatment include: leaching, adsorption, desorption, photodecomposition, oxidation, hydrolysis, and biological metabolism\(^{[2]}\). Aeration and
Table 8: Advantages and disadvantages of land based biological methods

| Process     | Advantages                                                                 | Disadvantages                                                                                     |
|-------------|-----------------------------------------------------------------------------|---------------------------------------------------------------------------------------------------|
| Landfarming | Least expensive, can be performed ex-situ or in-situ                        | Long residence time, unsuitable in towns (large land area required), potential for contaminating water, air, soil, sensitive to weather, limited capability in degrading complex compounds, possibility of contaminant transport, requires less than 2% grade slope |
| Biopiling   | Effective nutrient supplementation, second least expensive                   | Biodegradation occurs during summer months unless steam is supplied, soil must be accessible, land requirement is relatively large, requires infrastructure |
| Composting  | High microbial diversity, low capital and operating costs, simple operation and design, high treatment efficiency, moisture, nutrient, and pH levels can be controlled, less threat than incineration, no mixing with surface, shorter treatment time than landfarming, can treat high concentrations of organic compounds | Large land requirement, difficulty siting, time required, possible groundwater contamination, difficult to capture off-gasses |
| Bioventing  | Can be low cost, high efficiency                                             | Not suitable for VOCs, water table should be >10ft from surface, not used for surficial soils, site may need to be capped, adversely affected when hydraulic conductivity <10^-4 cm/s, off-gases may need further treatment, should not be used near buildings (explosion hazard), can be difficult to add nutrients, can take years, requires underground infrastructure |

| Process     | Advantages                                                                 |
|-------------|----------------------------------------------------------------------------|
| Liquid Delivery System | Good for fractured rock aquifers, good for shallow water tables |

| Table 9: Advantages and disadvantages of bioreactors |
|----------------------------------------------------|
| Advantages                                         | Disadvantages |
| Shorter timeframe (70 times faster than landfarming), less space required, can be cheap, can capture VOCs, can have aerobic conditions for recalcitrant compounds, simple, work on concentrated residues, can be coupled with other techniques, various sizes (several liters to millions of liters), economic and technical advantage for saturated soils | Some reactors can be expensive, soil sometimes has to be pretreated, require constant mixing, transportation costs, off-gases likely require further treatment |

| Table 10: Evaluation criteria for remediation methods |
|-----------------------------------------------------|
| Criteria | Definition | Score |
|----------|-------------|-------|
| Applicability | Used under various situations with no or little modification | 25 |
| Efficiency | 99% removal warrants 25 but <50% warrants 0 | 25 |
| Time | Removes contaminants within three months | 20 |
| Cost | Inexpensive | 15 |
| Cleanliness | Pollutants are not transferred to other locations | 15 |
| Total score |                                  | 100 |

There are several disadvantages to this form of treatment: (a) it requires a large amount of land area which can be difficult to find in populated areas, (b) it has the potential to contaminate groundwater, (c) it is sensitive to the weather, (d) it has limited capability for degrading heavier components of petroleum oils, (e) there is a chance of contaminant transport (f) it has a slow detention time and (g) it is not suitable when volatile organic compounds (VOCs) are present because they will be released to the atmosphere. However, researchers have tried this technique under wide conditions. Marin et al. used landfarming to reduce the total hydrocarbon content in an oil refinery sludge by 80% in 11 months under semiarid conditions. McCarthy et al. treated soil in Alaska contaminated with petroleum hydrocarbons (such as diesel-range organics, trimethylbenzenes, gasoline-range organics and BTEX compounds) in 55 days using landfarming. Landfarming has also been used in the degradation of oil in the desert.

**Biopiling:** Biopiling is an ex-situ remediation method that is very effective in nutrient supplementation. Biopiles require accessible contaminated soils and sufficient land area. It has been found that contaminants in biopiles show the greatest reduction in concentration over the summer months. While treating diesel contaminated soil, Nano et al. found that sand improved pile porosity (and subsequently oxygen...
diffusion) and surfactants were effective in increasing contaminant bioavailability. Jørgensen et al.\textsuperscript{[45]} mixed nutrients (N, P, K) and microbes in soil biopiles and found mineral oil degradation rates to be the highest in the first months, following a standard first order degradation curve.

**Composting:** Contaminated soil is mixed with a bulking agent such as manure or wood chips, and heaped in a large pile. Manure or sewage sludge are also used for inoculation of the pile to provide high microbial diversity, specifically mesophilic and thermophilic microbes\textsuperscript{[46]}. Jørgensen et al.\textsuperscript{[45]} used bark chips as a bulking agent while composting a hydrocarbon contaminated soil in biopiles. Composting can be carried out in-situ or ex-situ to treat highly contaminated soils. Machinery is used to turn the pile (aerating it). Moisture, nutrient and pH levels are also controlled\textsuperscript{[2]}. Composting has a shorter detention time than landfarming and the final product can be used for landscaping. The contaminated soil is also not worked into the land so there is less potential for contaminants to enter groundwater than with landfarming. There is also less threat to air than that associated with incineration\textsuperscript{[2]}. Namkoong et al.\textsuperscript{[46]} treated diesel oil contaminated soil by composting and only 2\% of the total petroleum hydrocarbons were lost by volatilization.

**Bioventing:** Bioventing is suitable for less volatile contaminants that are biodegradable under aerobic conditions\textsuperscript{[3]}. When volatile compounds are present, off-gases need to be treated, thereby increasing the cost of the operation. This process is most applicable where the water table is greater than 3 m deep from the surface. The site must be capped if the soil and water table are shallow\textsuperscript{[43]}. Bioventing should not be used near building because there is the potential for an explosion. Moisture levels of 40 \%-60 \% of field saturation must be maintained in order for the operation to be successful. Major costs are incurred in installing wells, blowers, controllers, infrastructure and other equipment. Österreicher-Cunha et al.\textsuperscript{[47]} reported that bioventing may be a valuable tool in treating gasoline-ethanol contaminated soil as the process appeared to accelerate soil detoxification. While treating toluene and decane contaminated soil, Malina et al.\textsuperscript{[48]} found bioventing efficiency to be dependent on temperature with respect to remediation time.

**Liquid delivery systems:** Liquid delivery systems require extensive site characterization and are best for sites with fractured rock aquifers, shallow water tables, formations with narrow saturated intervals, or when control of plume migration is mandated. The cost of the operation depends on the type of contaminants present, the amount and extent of contamination, sediment characteristics and source of oxygen. For example, low numbers of microbes are associated with clay soils and the addition of 100 mg/l of hydrogen peroxide as an oxygen source can be toxic to biota\textsuperscript{[43]}. Flores et al.\textsuperscript{[49]} reported that hydrogen peroxide is a major source of OH radicals which are oxidative agents in the decomposition of hydrocarbons in the soil. Ghassemi\textsuperscript{[50]} demonstrated the in situ delivery of liquid and other treatment agents into hydrocarbon contaminated soil for the purpose of its remediation.

**BIOREACTORS**

There are many different types and sizes of bioreactors ranging from a vessel of a few liters to large systems that can hold millions of liters\textsuperscript{[43]}. Bioreactors have a shorter detention time, lower costs than traditional physical, thermal and chemical reactors, take up less space, are simple to use, and offer an economic and technical advantage for contaminated soils having high moisture content\textsuperscript{[2, 39, 51]}. They are divided into three categories based on the state of the medium: solid, liquid and gas bioreactors. Solid reactors can handle contaminated soil and they include static bed reactors, continuous mix reactors, horizontal drum reactors and fungal compost reactors. Liquid reactors are designed for liquid medium or slurry and they include slurry-phase reactors and dual injection turbulent suspension reactors. Gas bioreactors are usually used in a combination with solid or liquid reactors to remove volatile organic contaminants from the exhaust gas of those reactors and they include a variety of biofilters and packed bed reactors.

**Static Bed Reactors:** A static bed reactor consists of a clay or synthetic liner, overhead irrigation system to spray water and nutrients onto the bed of excavated contaminated soil and pipes embedded in sand to collect leachate. It is a closed loop system, preventing contamination from being released to the environment. Soils contaminated with PAH compounds (naphthalene, phenanthrene, and pyrene) and pentachlorophenol were remediated using this system\textsuperscript{[52]}. Diesel fuel was reduced from 683 ppm to 81 ppm in four months using this system\textsuperscript{[2]}.  

**Continuous Mix Reactors:** Continuous mix reactors allow for enhanced diesel fuel turnover in a soil mixture. They are similar to in-vessel composting systems and as such, the moisture levels should be at 50
% of the maximum water capacity. The temperature, pH, moisture content and aeration level can be effectively controlled in these reactors. However, this type of reactors have the potential to form pellets which reduce microbial activity and the degradation of contaminants and are associated with high equipment and operating costs[2]. Antizar-Ladislao et al.[53] used this system to remove 16 U.S. Environmental Protection Agency listed PAHs. Truax et al.[54] used a continuous flow reactor to treat a diesel contaminated sandy soil.

**Horizontal Drum Reactors:** These are horizontal drums that rotate around on their axis like cement mixers thereby keeping soil loosely packed. Temperature, oxygen content and nutrient supply are all controlled. The advantage of these reactors is that they can be used for solid material or slurry[2]. They provide a means for performing chemical processes using high temperatures at near atmospheric pressures. However, their complexity has merely led to practical and expensive designs[55]. A Canadian consulting company (UMATAC Industrial Processes, a division of UMA Engineering Ltd) designed a horizontal drum reactor to pyrolyse oil shale into vapors[56].

**Fungal Compost Reactors:** Fungal compost reactors work on the principle of bound residue formation. Indigenous peroxidase enzymes are stimulated to enhance the rate of bound residue formation. Although this form of treatment is low cost, the reactor can become carbon limited and a supplementary carbon source should be used[2]. McFarland and Qiu[57] removed benzo(α)pyrene from soil using *Phanerochaete chrysosporium* and corn cobs as a supplementary carbon source in a fungal compost reactor. Eggert[58] used white rot fungi (*Pleurotus ostreatus*) for creosote contaminated soil. This process removed 86% of the total 16 PAHs listed by the U.S. Environmental Protection Agency.

**Slurry Phase Reactors:** A slurry-phase bioreactor contains soil that is suspended in water by utilizing a mechanical stirrer. In these reactors, soil and water are mixed with air, nutrients and microbes. These reactors have been used in the bioremediation of soils contaminated with petroleum and its derivative PAHs. The treating time is in the order of days or weeks[44]. Saponaro et al. [59] reported a high removal efficiency for all PAHs from soil after 23 days using a slurry system reactor. Boopathy[60] reported that diesel biodegradation rates in a slurry reactor operating under anaerobic conditions were highest using mixed electron acceptor groups followed by sulfate reducing, nitrate reducing and methanogenic groups. Wang[61] reported that biodegradation of naphthalene in a continuously stirred batch slurry reactor was successful.

**Dual Injection Turbulent Suspension Reactors:** The dual injection turbulent suspension reactor (DITS) is a modification of the slurry reactor. It has a combined air-liquid injector at the bottom. Residence time is approximately 100 hours, which means that the degradation time is 70 times faster than that of landfarming[2]. Geerdink et al.[62] found that after treatment in a DITS reactor, oil was slowly released from the contaminated soil and treatment by another method was required for a further 10 weeks to reach minimal contamination levels.

**Biofilters:** Biofilters are bioreactors used to remove volatile compounds from contaminated air streams. They are made from biologically active material to which the microbes can be attached such as compost or peat[2]. Because this process relies upon an established microbial population within the filter, it can be difficult to operate when mixtures vary over a short period of time[53]. Leson and Smith[63] reported that biofilters remove major petroleum hydrocarbon classes (aromatics, aliphatics) to varying degrees. Maestre et al.[64] reported that fungal biofilters are an excellent choice to treat high loads of toluene.

**Packed Bed Reactors:** These types of reactors are packed with growth supporting medium such as amberlite and are used to treat gas currents. A helical feed reactor optimizes conditions due to its continuous operation and long residence time. As such, treatment is rapid and the reactor is small. The process is sealed so there is no uncontrolled release of VOCs. This type of reactor allows for a quantitative estimation of the oxygen diffusion through the compacted soil[2]. Ogata et al.[65] used a conventional packed bed reactor with ferroelectric materials to decompose benzene. Takaki et al.[66] also used a packed bed reactor with ferroelectric pellets to remove perfluorooethane (C₂F₆).

**Comparative Analyses**

**Remediation Methods:** The advantages and disadvantages of the various nonbiological and biological remediation methods are listed in Tables 7-9 were used as the basis for the comparative analysis performed on these remediation methods. Five criteria were used to evaluate these methods: efficiency, applicability, cost, time and cleanliness. Table 1o shows
Table 11: Assessment of nonbiological remediation methods

| Criteria        | Land Filling | Soil Washing | Incineration | Thermal Desorption | Radio Frequency Heating | Chemical Addition |
|-----------------|--------------|--------------|--------------|--------------------|------------------------|-------------------|
| Applicability   | 25           | 16           | 25           | 20                 | 15                     | 17                |
| Efficiency      | 25           | 20           | 25           | 15                 | 15                     | 15                |
| Time            | 0            | 25           | 25           | 15                 | 15                     | 15                |
| Cost            | 10           | 6            | 0            | 10                 | 8                      | 10                |
| Cleanliness     | 0            | 0            | 10           | 10                 | 5                      | 10                |
| TOTAL SCORE     | 60           | 67           | 85           | 70                 | 63                     | 62                |

Table 12: Assessment of biological remediation methods

| Criteria        | Land Farming | Biopiling | Composting | Bioventing | Liquid Delivery | Bioreactors |
|-----------------|--------------|-----------|------------|------------|----------------|-------------|
| Applicability   | 15           | 15        | 23         | 15         | 10             | 25          |
| Efficiency      | 15           | 23        | 25         | 25         | 23             | 25          |
| Time            | 12           | 12        | 20         | 10         | 8              | 25          |
| Cost            | 10           | 12        | 10         | 10         | 3              | 12          |
| Cleanliness     | 3            | 12        | 5          | 10         | 5              | 10          |
| TOTAL SCORE     | 55           | 74        | 83         | 70         | 49             | 97          |

Table 13: Advantages and disadvantages of specific solid bioreactors

| Bioreactor        | Advantages                                                                 | Disadvantages                                                                 |
|-------------------|-----------------------------------------------------------------------------|------------------------------------------------------------------------------|
| Static Bed        | Environmentally friendly, contaminants contained, temperature and gas flow  | Slow, no pH/moisture control, gases may leak during cleaning or maintenance,  |
|                   | control, simple; easy to operate, efficiency increases with temperature      | other equipment used to capture volatile off gases, relatively costly          |
| Continuous Mix    | Enhanced turnover, consistent aeration, temperature, pH, moisture content   | Expensive, can form pellets that reduce microbial activity                     |
|                   | and aeration effectively controlled, simple hardware; easy to operate, would  |                                                                              |
|                   | not have to be cleaned as often as a batch reactor which could reduce some  |                                                                              |
|                   | costs, VOCs stored                                                           |                                                                              |
| Horizontal Drum   | Can use high temperatures at atmospheric pressures, feed can be dry or in    | Expensive, complex, difficult to control thermal reactions inside drum, may be |
|                   | slurry, soil stays loosely packed, aiding efficiency, contaminants contained,| difficult to operate based on design, biofilms on the inside of the drum      |
|                   | VOCs housed in the drum                                                      | would be difficult to clean                                                   |
| Fungal compost    | Low cost, can treat a wide range of contaminants, fast and efficient,        | Carbon supplementation required, cleaning, fast in terms of bioreactors but  |
|                   | contaminants contained, temperature and other variables can be controlled,  | will take many weeks                                                          |
|                   | VOC traps                                                                    |                                                                              |

Table 14: Advantages and disadvantages of specific liquid and gas bioreactors

| Bioreactor        | Advantages                                                                 | Disadvantages                                                                 |
|-------------------|-----------------------------------------------------------------------------|------------------------------------------------------------------------------|
| Slurry Phase      | Well stirred, efficient, used in situ or ex situ, contaminants contained,   | Will need to separate solids and liquids, volatile off gases must be        |
|                   | conditions controlled, lab microcosms are cheap, easy to clean and operate  | controlled, expenses reasonable; depend on pollutant concentration          |
| DITS              | 70 times faster than landfarming, two zones; separates light material from  | Extract must be further treated for upwards of 10 weeks slow, simply a     |
|                   | heavily polluted material - both contained, off gases captured, bed easily  | modified slurry reactor, cost depends on the outlet concentration of the      |
|                   | removed for maintenance, parameters can be controlled, simple               | pollutant                         |
| Biofilters        | Remove VOCs, simple; easy to operate, low cost, minimal maintenance,        | Rely on microbes, slow                                                       |
|                   | efficient, contaminants contained, temperature, pH, and moisture controlled  |                                                                              |
| Packed Bed        | Rapid treatment, VOCs not released, parameters controlled, easy to operate,  | Small, localized high temperature regions (hot zones) could cause serious   |
|                   | minimal labor, contaminants contained, low cost                             | environmental or safety incidents                                            |
Table 15: Evaluation criteria for bioreactors

| Criteria        | Definition                                      | Score |
|-----------------|-------------------------------------------------|-------|
| Efficiency      | 95-99% removal warrants                          | 20    |
| Time            | Removes contaminants within weeks                | 15    |
| Cost            | Relatively inexpensive                           | 15    |
| Maintenance     | Easy to maintain                                 | 10    |
| Simplicity      | Easy to operate                                  | 10    |
| Release of VOCs | Non degraded VOCs are not released to the atmosphere | 10    |
| Containment     | Pollutants are not leaked out of the reactor     | 10    |
| Control         | Need for pH, temperature or moisture control     | 10    |
| TOTAL SCORE     |                                                 | 100   |

Table 16: Assessment of specific bioreactors

| Criteria     | Static Bed | Continuous Mix | Horizontal Drum | Fungal Compost | Slurry Phase | DITS | Biofilters | Packed Bed |
|--------------|------------|----------------|-----------------|----------------|--------------|------|------------|------------|
| Efficiency   | 10         | 15             | 12              | 10             | 10           | 10   | 10         | 5          |
| Time         | 5          | 15             | 12              | 5              | 10           | 10   | 5          | 12         |
| Cost         | 8          | 5              | 5               | 12             | 10           | 0    | 8          | 5          |
| Maintenance  | 10         | 10             | 5               | 5              | 10           | 10   | 10         | 10         |
| Simplicity   | 10         | 10             | 5               | 10             | 10           | 8    | 10         | 10         |
| Release of VOCs | 5          | 10             | 10              | 10             | 0            | 10   | 10         | 10         |
| Containment  | 8          | 10             | 10              | 10             | 10           | 10   | 10         | 10         |
| Control      | 5          | 10             | 2               | 10             | 5            | 8    | 5          | 0          |
| TOTAL SCORE  | 61         | 85             | 61              | 72             | 65           | 66   | 68         | 62         |

the definition and scores assigned to these criteria. The final results of the comparative analysis are shown in Tables 11 and 12. Among the nonbiological remediation methods, incineration scored the highest (85) followed by thermal desorption (70). The other nonbiological remediation methods had much lower scores (67, 63, 62 and 60 for soil washing, radio frequency, chemical addition and landfilling, respectively). The analysis performed on the bioremediation methods showed that with the exception of liquid delivery and landfarming (which are used under special circumstances), they are more effective than nonbiological remediation methods (except incineration). Bioreactors scored the highest (97) followed by composting (83), biopiling (74) and bioventing (70). Bioreactors have the advantages of: (a) shorter treatment time, (b) minimum space for operation, (c) ability to capture VOCs, (d) operate under aerobic conditions for recalcitrant compounds, (e) work on very concentrated residues, (f) can be operated at various sizes, and (g) can be coupled with other techniques if so needed.

Bioreactors: The advantages and disadvantages of the various bioreactors listed in Tables 13 and 14 were used as a basis for the comparative analysis performed on the bioreactors. Eight criteria were used to evaluate these reactors: efficiency, residence time, cost, maintenance, simplicity of operation, release of VOCs to the atmosphere, containment of contaminants and control of operating parameters such as pH, temperature and moisture control. Table 15 shows the definition and scores assigned to these criteria. The final results of the comparative analysis are shown in Table 16. Among the solid bioreactors evaluated, the continuous mix bioreactor scored the highest (85) followed by the fungal compost bioreactor (72). Both, the static bed bioreactor and the horizontal drum bioreactor scored 61, the first suffered from the release of VOCs and the long residence time while the second had a high cost and difficulties associated with maintenance and control. The Liquid and gas bioreactors, which are usually used for specific cases, scored much lower than the continuous mix bioreactor, 62 for the packed bed reactor, 65 for the slurry-phase bioreactor, 66 for the DITS bioreactor and 68 for the biofilters.

CONCLUSION

Many of the current Canadian regulations relate to sulphur content in diesel fuel because of acid rain. Although PAHs are considered toxic under the Canadian Environmental Protection Act, there are no standards for PAHs in diesel. About 60 of Canada’s contaminated sites involve petroleum hydrocarbons. The factors affecting the cost of clean up include: the accessibility to the site, weather conditions, quantity of spilled fuel, the extent of environmental damage and the time required for the clean up. The Canada-wide standards for petroleum hydrocarbons in soil separate soil into four categories based on land usage: agricultural, residential, commercial and industrial. It
also specifies the methods and outcome for the assessment and management of contaminated sites but timelines are left for individual jurisdictions to decide.

The remediation methods included soil washing, landfilling, incineration, thermal desorption, radio frequency heating, chemical addition, landfarming, biopiling, composting, bioventing, liquid delivery and bioreactors. The advantages and disadvantages of several remediation methods were determined. Five criteria were used for the evaluation of these methods: efficiency, applicability, cost, time and cleanliness. The results showed that the biological methods were more effective than nonbiological ones and the bioreactors scored the highest among the biological methods. Further evaluation was performed on several solid, liquid and gas bioreactors which included static bed, continuous mix, horizontal drum, fungal compost, slurry-phase, DITS, biofilters and packed bed bioreactors. Eight criteria were used for their evaluation: efficiency, time, cost, maintenance, simplicity, release of VOCs to the atmosphere, containment of contaminants and control of operating parameters. The results showed that the continuous mix bioreactor was the most effective system.

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