

Clean up of Crude Oil-Contaminated Soil

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ABSTRACT

Crude oil contamination of the environment has been an age-long phenomenon and a serious subject of concern. The effect of oil on soil depends on the size, quantity and grade of oil spilled. Crude oil contamination does not damage the soil permanently but has some adverse effects on crops and other vegetation. Over the years attempts have been made to find the cheapest, most efficient and environmentally-friendly method for the clean-up crude oil contaminated soil. This review is aimed at analysing the different methods used in the clean-up of contaminated soil. Crude oil contaminated soil can be cleaned up using physico-chemical, thermal and biological treatments. The first two methods have been found to be grossly inadequate and ineffective, and may result in further contamination. Biological methods (bioremediation and phytoremediation) have received considerable attention during the last few years as the most promising and environmentally benign technique for effective clean-up of crude oil contaminated soil. A wide range of bioremediation strategies is being developed to treat contaminated soil.

Keywords: bioremediation, petroleum hydrocarbon, phytoremediation, pollution

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INTRODUCTION

The discovery of crude oil has changed man's way of life considerably. It has improved our life economically and has also led to the availability of quick and accessible source of energy. Crude oil, or petroleum, is a complex mixture of hydrocarbons of varying molecular weight and structure. It comprises of three main chemical groups, namely, paraffinic (aliphatic), naphthenic (alicyclic) and aromatic. These hydrocarbons range from the simple, highly volatile substances to complex waxes and asphaltic compounds which cannot be distilled (Wadley-Smith 1983; Leahy and Colwell 1990).

Crude oil is obtained below the subsurface of the earth. It generally occurs at depths below 1500 meters and is recovered through boreholes within the earth. The liquid and gaseous phases of crude oil occur naturally underground, within pore spaces of sedimentary rocks. Crude oil occurs naturally in many parts of the world, particularly in the USA, Russia, Persia, Mexico, Romania, Iran, Iraq, Kuwait, Saudi Arabia, Libya, and Nigeria.

The major problem associated with crude oil exploration is the pollution/contamination of the environment. Crude oil contamination is a global phenomenon affecting all aspects of the environment. Cases of crude oil contamination of the soil have been documented (Imevbore 1973; Awobajo 1998).

Contamination of soils by crude oil has remained an emerging issue. Costly damages have been caused on coastal lines in different parts of the world by offshore oil spills (Anderson 1993; Britenbect 1998; Bassam and Battikhi 2005). Crude oil comes into contact with the soil naturally through natural oil seeps or man-made through accidental or deliberate spills and leakages such as intentional or accidental bursting of pipelines (Okpokwasili and Amanchukwu 1988; Leahy and Colwell 1990; Anderson 1996; Okecha 2000).

Over the years, several methods have been devised for the clean up of crude oil contaminated soil using physical, chemical, thermal and biological treatments. This paper presents a critical review of these methods.

EFFECT OF CRUDE OIL CONTAMINATION ON THE SOIL

The effect of crude oil on the soil depends on the size, quantity and grade of oil spilled. Crude oil spillage decreases the porosity of the soil (Lolomari 1979). This is due to the fact that oil tends to force the soil particles to stick together, thereby decreasing the pores. Higgins and Burns (1979) reported that in oil-contaminated soil, oil droplets interfere with the interstices. Additionally, crude oil forms a coat covering the soil surface to retain carbon dioxide from the respiration of soil organisms. Persistence of oil on/in the soil depends on the amount spilled, procedures of clean-up, microbial degradation, climatic conditions, and the type of oil spilled.

Crude oil changes the characteristics of the land, polluting it to the detriment of living organisms. Vegetation, wildlife, crops and farmlands are adversely affected (Okecha 2000). Toxicological studies have identified Polycyclic Aromatic Hydrocarbons (a derivative of crude oil) as being carcinogenic and have been implicated to be the cause of rapid death of living organisms (Onwurah 2000). Oyefolu and Awobajo (1979) reported that a good percentage of oil spills that occurred on dry land between 1978 and 1979 in Nigeria affected farms in which crops such as rice, maize, yams, cassava, and plantain were cultivated.

No permanent damage is however done to the soil except in cases in which the soil is completely submerged by the oil in areas of poor drainage and aeration. With volatilization, scientific principles, microbial degradation, rainfall and aeration, light oil spillage can be cleaned up within 2 to 3 years (Odu 1977).

Klokk (1984) studied the effect of crude oil pollution on the germination and vegetative growth of five species of vascular plants and reported a reduction in overall germination rate. Germination response to oil varied greatly with plant species and members of the same plant species showed differential sensitivity to oil contamination (Adam and Dubcan 2002). Amakiri and Onofeghara (1984) reported that the seeds of *Zea mays* exhibited no germination after exposure to oil for longer than seven days while those of *Capsicum frutescens* exhibited 100% viability after 32 weeks exposure.

Some adverse effects of oil spills and contamination of birds and aquatic animals have been documented. Chronic marine pollution in South Eastern Newfoundland (Canada) waters was reported to have led to the death of 74% of the sea birds that died between 1984 and 1999. The dead birds were found with oil on their feathers (Weise and Ryan 2003). An oil tanker accident involving the oil tanker Prestige in November 2002 where about 63,000 tons of heavy oil reached the Galician Coast (NW Spain) was found to have caused different levels of DNA damage on birds exposed to the spill (Laffon *et al.* 2006). Further studies on the impact of the Prestige oil spill and its clean up activities on the macroinfauna community of the Galician Coast six months after the spill, revealed that the macroinfauna population was drastically reduced, with *Eurydice* and *Scolecopsis squamata* as the most affected taxa (Junoy *et al.* 2005). Khan *et al.* (2007) evaluated the effect of diesel oil on aquatic species (*Oncorhynchus mykiss* and *Daphnia magna*) using acute toxicity testing and found that their mortality rates were significant compared to species not exposed to diesel oil.

TYPES OF SOIL TREATMENT

There are four steps involved in the remediation of any contaminated site. These include:

1. A preliminary assessment – This involves the identification of those conditions at a site that pose an imminent threat to human health and the environment.
2. Selection and implementation of appropriate interim remedial measures – It addresses any imminent hazards that may exist at a site.
3. Site investigation and remediation technology feasibility study – In this stage the nature and extent of contamination are defined, and potential final remedial methods are identified and evaluated.

4. Selection of final remedial methods – Selection processes are taken into account based on results of the site investigation, including effectiveness of different remedial methods, the time necessary for complete clean-up and the overall treatment cost (Atlas and Bartha 1992; Cutright and Lee 1994; Colleran 1997).

There are two major types of soil treatment: *In situ* (where the soil is treated at the site of contamination) and *ex situ* (in which case the soil is excavated and transported to another site for treatment).

PHYSICO-CHEMICAL CLEAN UP OF CONTAMINATED SOIL

Soil excavation

This is the mechanical removal of contaminated soils to off-sites either for burying or burning. Kenil (2006) suggested that the most efficient way to clean up contaminated soil is to remove it. The process is however very expensive as a contractor has to be hired to take away a layer of ground. Another problem with excavation is that the place from which the layer is removed is made prone to erosion and other environment damaging agents (Araruna *et al.* 2004).

Soil washing

Soil washing is an *ex situ* treatment process applicable to a broad range of organic, inorganic and radioactive contaminants in soil (Anderson 1993). It involves the use of liquid/water sometimes combined with chemical additives and a mechanical instrument to scrub soils. This removes hazardous contaminants and concentrates them into smaller volumes (Wood 2002).

Hazardous chemicals easily adhere to silt and clay like sand and gravel particles. During soil washing therefore, the silt and clay are mechanically separated from the uncontaminated coarse soils (Wood 2002). The contaminated fine sand can then be disposed or treated accordingly while the coarse sand is retained as backfill. The effectiveness of this method has been shown to be less than 80% though efficiency increases when hot water is used (Wood 2002). It is therefore mostly used as a pre-treatment method for final cleaning up of soils.

Soil Vapour Extraction (SVE)

This method is a relatively simple physical process of cleaning up crude oil contaminated soils. SVE involves the use of a specially designed system to remove volatile contaminants (e.g. crude oil) from the soil in vapour form (Kosky and Jones 1995).

The process of SVE is carried out by applying a vacuum through a system of underground wells which pull up contaminants to the surface as vapour or gas. Air is some times introduced to enhance the process. Soil vapour extraction is frequently used to remove chlorinated hydrocarbons, especially trichloroethylene (TCE) from the soil (Imamura *et al.* 1997).

THERMAL CLEAN UP METHODS

Thermal desorption

This is a more recent clean up method. It involves heating up crude oil contaminated soils to temperatures of 200-1000°F at which contaminants with low boiling point vapourize and desorb (physically separate) from the soil (Troxler *et al.* 1994; Elgibaly 1999). This method is also termed Low Temperature Thermal Desorption or Low Temperature Thermal Volatilization, due to its use of low temperature. It is also called thermal stripping or soil roasting

(Anderson 1993).

Most times during thermal desorption, contaminating hydrocarbons are vapourized and ignited. The remaining by-products are removed from the system by convection and treated by filters or second stage re-ignition or by an air emission treatment system (Wood 2002). On the other hand, they can generally be treated in a secondary treatment unit (e.g. after burner, catalytic oxidation chamber, condenser or carbon adsorption unit) prior to discharge to the atmosphere. Afterburners and oxidizers destroy the organic constituents while condensers and carbon adsorption units trap organic compounds for subsequent treatment or disposal.

Depending on the organics present and the temperature of the desorper system, thermal desorpers can cause complete or partial decomposition of some of the organic constituents (Anderson 1993; Troxler *et al.* 1994). Afterwards, soil is cooled, remoistened for dust control and stabilized to prepare them for disposal/reuse by depositing them on-site or as landfill covers to be incorporated into asphalt (Anderson 1993). Up to 90% efficiency has been recorded with thermal desorption in removal of crude oil hydrocarbon contaminants from soils. Thermal desorption has three major pitfalls. It is expensive, time consuming and hazardous (Wood 2002). However, thermal desorption seems to be a very promising method for cleaning up crude oil contaminated soil because it is simple and avoids all the difficulties associated with digging up the soil for disposal or cleanup (Elgibaly 1999).

Incineration

This implies burning off the contaminants from the soil surface using fire. According to US EPA, at high temperatures (i.e. between 1,600°F and 2,500°F) incineration takes place, and hazardous wastes including crude oil are destroyed from the soil and toxic elements are reduced to basic elements (mainly hydrogen, carbon, chlorine and nitrogen). The basic elements then combine with oxygen to form stable non-toxic substances such as water, carbon dioxide and nitrogen oxides.

Igniting and burning crude oil is often difficult due to the rapid loss of more flammable and volatile components via evaporation (Kenil 2006). Contaminated soils are normally first excavated and carried to off-site facilities before incineration is effected (Bassam and Battikhi 2005).

Disadvantages of incineration include: high operational cost due to high energy requirement, the large space involved and the dangers of environmental pollution (Araruna *et al.* 2004; Bassam and Battikhi 2005).

BIOLOGICAL CLEAN-UP OF OIL CONTAMINATED SOIL

Biological treatment involves the use of microorganisms, plants and other biological systems to clean-up oil contaminated soil. Biological processes are used to treat excavated soils, saturated and unsaturated soil *in situ*, and recovered ground water (Galaska *et al.* 1990; Eckenfelder and Norris 1993).

Microbial degradation of crude oil (biodegradation)

Biodegradation of organic waste is an increasingly important method of waste treatment (Atlas 1981). Biodegradation has many advantages – it uses inexpensive equipment, environmentally friendly nature of the process and simplicity (Nadean *et al.* 1993).

Microorganisms play an important role in the clean up of crude oil contaminated environment. The use of microorganisms in the clean-up of an oil spill comes in after a large amount of the oil has been removed by various physical and chemical methods (Atlas 1981; Okpokwasili and Amanchukwu 1988; Ijah and Ukpe 1992; Amund and Akangbuo 1993; Ijah and Okang 1993).

Many studies have been carried out on microbial degradation of crude oil (Higgins and Gilbert 1978; Bartha 1986; Leahy and Colwell 1990). Microbial degradation is made possible because microorganisms have enzymatic systems that breakdown the crude oil utilizing it as a source of carbon and energy (Ijah and Antai 1988; Antai and Mgbomo 1989, 1993). These biotransformations can be exploited for treatment of contaminated soils and ground water (Bouwer and Zehnder 1993).

Microorganisms capable of utilizing petroleum hydrocarbons in their metabolism are widely distributed in soils. They are mostly found in the surface soil in the vicinity of an oil field and also in petroleum-contaminated soils (Bossert and Bartha 1984; Antai and Mgbomo 1989). Crude oil-degrading microorganisms have been identified and include bacteria, yeast, filamentous fungi and algae (Atlas 1981; Prince *et al.* 1993; Ezeji *et al.* 2005). The major bacteria genera implicated in crude oil degradation in both soil and aquatic environments comprise mainly *Pseudomonas*, *Achromobacter*, *Athrobacter*, *Actinomyces*, *Flavobacterium*, *Micrococcus* and *Nocardia* (Atlas 1981; Bossert and Bartha 1984; McCarthy and Williams 1992; Odokuma and Okpokwasili 1993; Okpokwasili and Nnubia 1999). Almost any hydrocarbon, even the heaviest paraffin (asphaltic residues) can be attacked by bacteria (Essien *et al.* 1997). Wardly-Smith (1983), however reported that microbial degradation does not always lead to complete disappearance of oil constituents.

The oil-degrading ability of microorganisms in tropical soil has been reported to depend on the adequacy of certain environmental factors such as temperature, nutrients, moisture, pH, oxygen, the viscosity of oil and coarseness of the affected soil (Odu 1977, 1981; Bossert and Bartha 1984; Antai and Mgbomo 1989; Ijah and Okang 1993). Temperature has been described as one of the most important parameters in determining the rate and effectiveness of biodegradation (Atlas 1981). Bossert and Bartha (1984) reported that the range of temperature favourable for microbial degradation of crude oil is 30-40°C. Low temperatures have been found to result in slower degradation rates possibly because it increases the viscosity of oil resulting in low drifting capacity which in turn creates a low surface area for degradation (Atlas 1981).

Nutrient concentration is a key factor affecting biodegradation rates of oil in contaminated soil and beach sediments (Xu *et al.* 2003). A continuous increase in microbial population and sustained stimulation of biodegradation have been reported in response to addition of nitrate and phosphorus containing compound (Mark and Jeffrey 1991; Abu and Ogiji 1996).

Microbes carrying out metabolic transformations require adequate moisture for their growth and activity. In surface soils, inadequate supply of water can severely restrict biodegradation (Alexander 1984). Oxygen availability is also very essential for biodegradation. Low degradation has been reported in depths of seas and sediments where low oxygen is available (Leahy and Colwell 1990).

pH affects solubility and consequently the availability of many constituents of soil which can affect biological ability. Biodegradation is affected negatively by extreme acidity and extreme alkalinity (Leahy and Colwell 1990) and therefore most laboratory-based biodegradation studies are carried out at pH range near neutral.

Bioremediation

Bioremediation is the act of adding fertilizers or other materials to the contaminated environment such as oil spill sites, to accelerate the natural biodegradation process. Bioremediation of petroleum-contaminated soil is adopted principally to improve the biophysico-chemical properties of soil through the augmentation of soil nutrients in order to stimulate growth and multiplication of indigenous microflora (Dragun 1993; Holiday and Deuel 1993).

Bioremediation is considered one of the most promising

methods for dealing with a wide range of organic contaminants, particularly petroleum hydrocarbons (Balba 1993). Two basic methods are available for obtaining microorganisms to initiate the bioremediation: bioaugmentation – in which adapted genetically coded toxicant degrading microorganisms are added (Okpokwasili *et al.* 1986) and biostimulation – which involves the injection of necessary nutrients to stimulate the growth of indigenous microorganisms (Lee and Levy 1989, 1991). Odokuma and Dickson (2003) suggested that a combination of bioaugmentation and biostimulation with indigenous hydrocarbon utilizers would be effective in the remediation of crude oil polluted tropical soils.

Numerous laboratory studies on nutrient enhancement of oil degradation by naturally occurring microorganisms have concluded that this technology is promising for use in stimulating oil degradation (Oliveiri *et al.* 1978; Amanchukwu *et al.* 1989; Ibekwe *et al.* 2006; Ubochi *et al.* 2006). Field deployment of this biotechnology has yielded good results in the treatment of an oil spill following the Exxon Valdez incident in Prince William Sound and the Gulf of Alaska (Chianelli *et al.* 1991; Glaser *et al.* 1991; Ladousse and Tramier 1991; Lee and Levy 1991; Safferman 1991; Tabok *et al.* 1991; Venosa *et al.* 1991).

A wide range of bioremediation strategies is being developed to treat contaminated soil. Blackburn and Hafker (1993) reported that selecting the most appropriate strategy to treat a specific site can be guided by considering three basic principles: the amenability of the pollutant to biological transformation to less toxic product (biochemistry); the accessibility of the contaminant to microorganisms (bioavailability); and the optimization of biological activity (bioactivity).

Some large-scale bioremediation technologies used in treatment of contaminated soils include windrowing, biopiling, bio-venting, land farming and composting. Windrow techniques are constructed by mixing the contaminated soils with the composting material and placed in elongated piles. Bio-pile involves the construction of soil piles above ground with the contaminated soils placed within the bund area. The piles are aerated using air injection or vacuum extraction to either push or pull air through the piles to ensure the transfer of oxygen and therefore aerobic degradation of the organic contaminants.

Bioventing combines the capabilities of soil venting and enhanced bioremediation to cost-effectively remove light and middle distillate hydrocarbons from vadose zone soils and the groundwater table. Soil venting removes the more volatile fuel components from unsaturated soil and promotes aerobic biodegradation by driving large volumes of air into the subsurface (Hoeppel *et al.* 1991; Hinchee and Arthur 1991).

Land farming is a well known biological method used in the treatment of petroleum hydrocarbon contaminated soil. The system involves periodic tiling of the ground to induce aeration, controlled moisture content and addition of nutrients to enhance microbial degradation of the contaminants. The contaminated soil is excavated onto a designed lined bed (to avoid leaching) and mixed with a controlled amount of nutrients and soil additives such as bulking agents (Lodolo *et al.* 2001).

Compost bioremediation refers to the use of a biological system of microorganisms in a mature, cured compost to sequester or break down contaminants in water or soil. Adenuga *et al.* (1992) reported the use of in-vessel composting in the treatment of PAH-contaminated soil.

Bioremediation has the advantage that polluted soil can be treated at the site without having to move them somewhere else. Balba *et al.* (1998) remarked that bioremediation is a site-specific process and therefore feasibility studies are required before full-scale remediation can be successfully applied.

Phytoremediation

Phytoremediation refers to the use of vegetative species for *in situ* treatment of land areas polluted by a variety of hazardous substances (Sykes *et al.* 1999). Different types of phytoremediation have been developed. These include phytoextraction, which relies upon a plant's natural ability to take up certain substances (such as heavy metals) from the environment and sequester them in their cells until the plant can be harvested, phytodegradation, a means by which plants convert organic pollutants into a non-toxic form, phytostabilization, where a plant releases certain chemicals that bind to the contaminant to make it less bioavailable and less mobile in the surrounding environment, and phytovolatilization, a process through which plants extract pollutants from the soil and then convert them into a gas that can be safely released into the atmosphere (Bentjen 2002).

Attempts have been made to improve the efficacy of phytoremediators through genetic modification. Genes from different sources, including mammals and microorganisms are being introduced into plant species, resulting in the creation of novel classes of phytoremediators that have the ability to extract harmful heavy metals from contaminated soil (Gleba *et al.* 1999).

Phytoremediation is environmentally friendly, visually attractive and more cost effective than conventional remediation methods (US EPA 2001). Moreover, the structure of the soil is highly maintained (Khan *et al.* 2000). A number of studies on the use of vegetation in the treatment of oil contaminated soil have been documented (Lee and Banks 1993; Shimp *et al.* 1993). In a field study carried out at the US Naval base, Port Hueneme, California, Banks *et al.* (2003) reported that the total petroleum hydrocarbon concentrations were lower by the end of the study in the vegetated plots when compared to the unvegetated soil.

CONCLUSION

Crude oil contamination is a very serious problem affecting both the terrestrial and aquatic environments. Its impact, especially on the coastal regions has been a source of concern to many people and governments of the world. The need to devise sustainable clean up methods have never been so crucial. This review has tried to highlight the different methods used for the clean up of crude oil contaminated soil. The choice of clean up method should depend on a number of factors which include, how well the soil is cleaned, the ability of the soil to be reused after clean up and how environmentally friendly the clean up method is. Other factors to be considered include the overall cost of the clean up exercise and also whether the clean up method is approved by the Environmental Protection Agency. Lodolo *et al.* (2001) reported some other criteria to be considered in choosing a soil remediation technology to include the technique's ability to clean-up to a desired level (minimum pollutant concentration achievable by the technology), community acceptability, post-treatment costs, soil quality required after the intervention, environmental impacts and risks of remediation activities/processes. Khan *et al.* (2004) observed that no single specific technology may be considered as a panacea for all contaminated site problems.

There are also limitations involved in the use of each clean-up methods. Low permeability, high content and soil heterogeneity limit the performance of the SVE method. There is also the risk of potential release of hazardous compounds during excavation and materials handling (Lodolo *et al.* 2001). The major disadvantages of the thermal methods (incineration and thermal desorption) are the high energy costs involved and the danger of environmental pollution.

The biological methods have been judged as the best remediation methods in terms of efficiency and environmental acceptability. Biological techniques are encouraged because of the advantage of soil sustainability and possibility of the soil to be restored to its original use. EPA has chosen bioremediation as a primary reasonable remedy to treat or-

ganic contaminants in soil, sludge, and sediments at wood-treating sites (USCOTA 1997). The only limitation associated with biological methods is the long time it takes for the complete removal of the contaminants.

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