

Bioremediation of petroleum hydrocarbons through landfarming: Are simplicity and cost-effectiveness the only advantages?

Mphekgo P. Maila & Thomas E. Cloete

1. Introduction

The technologies that involve the biological removal of petroleum products from contaminated soil environments are today well established, and many are applied commercially on a large scale. During the 1970s, when environmental concerns associated with uncontrolled disposal became apparent, and environmental regulations were established and applied in North America and Europe (aimed at minimising the risk of air and groundwater contamination), landfarming gained popularity. This 'low tech' biological treatment method involves the controlled application and spread-out of a more-or-less defined organic bio-available waste on the soil surface, and the incorporation of the waste into the upper soil zone (Genou et al. 1994). In 1983 it was estimated that at least one-third of all United States refineries operated full-scale or pilot scale landfarmers (American Petroleum Institute 1983). The technology has been widely used, as it is simple and cost-effective to implement compared to other treatments (American Petroleum Institute 1983; Harmsen 1991).

Landfarming lost its popularity in 1984 when the United States Environmental Protection Agency (US EPA) issued the land disposal restriction (LDR) as part of the hazardous and solid waste amendments (HSWA) to the resource conservation and recovery act (RCRA). The US EPA went further on 18 August 1992, by publishing a final rule, (57 FR 37194, 37252), establishing treatment standards under the land disposal restrictions program for various hazardous wastes that included petroleum products. Landfarm operators had to either operate their facilities to treat their waste below the EPA specified contaminant levels (referred to as treatment standard), or to submit a petition demonstrating that there was no migration of hazardous constituents from the injection zone (US EPA 1984). As a result, most of the traditional landfarms in North America were closed.

Although there have been some restrictions on the application of the technology, it is still being used to treat petroleum products, with added measures for minimising or treating volatiles and leachates (Genouw et al. 1994; Harmsen et al. 1994; Balba et al. 1998; Picado et al. 2001; Maila 2002).

The petroleum products from the soil during landfarming are largely removed through volatilisation, biodegradation and adsorption (Morgan & Watkinson 1989; Devliegher & Verstraete 1996; Margesin et al. 1999; Hejazi et al. 2003). Lighter (more volatile) petroleum products like gasoline tend to be removed by volatilisation during landfarm aeration process and to a lesser extent, degraded by microbial respiration (EPA 1994). The mid-range petroleum products like diesel fuel and kerosene contain lower percentage of lighter constituents than does gasoline. Bio-degradation of these petroleum products is more significant than volatilisation. The more heavier or non volatile petroleum products like heating oil and lubricating oils do not volatilise during landfarm aeration, the dominant mechanisms that breaks down these petroleum products is biodegradation. Adsorption also plays an important role in the dissipation of petroleum products from the soil. According to Margesine et al. (1999), a third of diesel was removed from the contaminated soil by physicochemical means (adsorption and volatilisation).

The volatile organic compounds (VOCs) from the landfarm area can present air pollution problems if the treatment area is not properly covered to minimise the emissions (Hejazi et al. 2003). Apart from the VOC emissions, other constraints faced by the rehabilitation practitioners considering landfarming as a treatment option include, requirements for large land area for treatment, availability of the pollutant degrading bacteria, effectiveness of the technology at high constituent concentration (more than 50,000 ppm), improved concentration reductions in cases requiring more than 95% of pollution reduction and the flexibility of the technology in integrating the removal of petroleum hydrocarbons with other contaminants that may occur with the petroleum products. Although problems associated with depth of pollution can be solved by ex situ treatment, the polluted soil often requires a large treatment area, which can increase the risk of human exposure to the contaminants. However, such exposure is only temporary, as contaminants will be degraded if environmental conditions are optimal (Ausma et al. 2002).

Although simplicity and cost-effectiveness are the major advantages of the technology, the treatment has physical,

chemical and biological ‘constraints’, which must be addressed. In this paper, we discuss these limitations, benefits, and possible solutions to the constraints.

2. Benefits and constraints of the technology

Bioremediation through landfarming is both simple and cost-effective to implement compared with other treatment technologies (Pearce & Ollerman 1998; Kelly et al. 1998). On average, the costs associated with treating petroleum hydrocarbon-contaminated soil ranges from \$30 to \$70 per ton of contaminated soil compared with a physical treatment like soil venting which is relatively expensive (\$70 to \$200) per ton (Marijke & van Vlerken 1998; Environment Canada 2003). However, as a result of costs associated with soil excavation and transporting the contaminated soil, in situ techniques can be in general about 40 – 50% of ex situ techniques (SCG 2004). The technology is simple in that typical equipments, which are used for landfarming, is used widely in the farming community and is therefore ‘readily’ available. As most of this equipment is designed to till the soil to a depth of 60.5 m, additional costs can be incurred during soil excavation for ex situ treatment (Kelly et al. 1998). Different forms of the technology are shown in Figure 1. For additional landfarm layouts or designs, the reader is referred to Doelman and Breedveld (1999) and to Battelle series (Alleman & Leeson 1999a–c). However, together with these advantages (Table 1), there are physical, chemical and biological aspects of the technology that can hamper the remediation process. The physical aspects include the land area required for treatment, the ability and limitations of aeration equipments, mobility of pollutants in the soil, water requirements; chemical aspects include toxicity, transformation and partitioning of the petroleum products in different environmental media while biological aspects include biostimulation or bioaugmentation for optimal biotransformation of petroleum products in the soil. The constraints of landfarming are listed in Table 1.

3. Physical and chemical aspects of landfarming

Landfarming requires a sizeable area to treat the contaminated soil in cases where the volume of the excavated contaminated soil is large, and this can increase the risk of exposure to pollutants if ex situ treatment is applied. The potential health hazards due to the volatilisation of lighter petroleum products from the soil during the treatment can be avoided by designing the landfarms as shown in Figure 1b. In this way exposure to harmful pollutants and dust will be minimised. However, volatilisation is only important during the loading of the greenhouse, particularly in mild climates.

The treatment of contaminated soil using landfarming can also be limited by the capacity of the aeration equipment. It is important to design landfarms in such a way that the tilling equipments are able to reach the ‘subsurface’ contaminated soil. The depth of the contaminated soil varies, depending on the capacity of the tilling equipments (30–60 cm is commonly used, EPA 1994). Also of importance during the treatment design is the need to incorporate an impermeable membrane with a drainage layer (as shown in Figure 1b). This membrane (high-density polyethylene membrane, ‡250 lm thickness) prevents groundwater contamination.

Soil moisture can also impact the efficiency of removing petroleum compounds from the soil. The level of moisture in most landfarms is kept between 30 and 80% field capacity (Block et al. 1992; Pope & Mathews 1993; Malina et al. 2002). The moisture level ensures the survival of the pollutant-degrading bacteria and enables dust control. However, as the size of the treatment area increases, the amount of water required to maintain the level of moisture ideal for biological activity can be enormous, especially in dry countries, and this can increase the treatment costs.

The interaction between the pollutant and micro-biota can result in the transformation of parent compounds to toxic metabolites which can lead to abortive pathways (Leisinger et al. 1981; Haugland et al. 1990; Lee et al. 1994), while adsorbents like clay and organic matter, which are site-specific can decrease the bioavailability and therefore a lower risk for higher organisms (reduction in toxicity) and lower biodegradation efficiency as contaminants are tightly bound to the soil matrix (Guerin & Boyd 1992; Hatzinger & Alexander 1995; Volkering 1996). The interaction between the pollutant and soil components is shown in Figure 2. While the physical and chemical constraints of landfarming can hamper the efficiency of landfarming, the knowledge that has been generated during the last two decades, which addresses these limitations (Verstraete & Top 1999; Holden & Firestone 1997), has made it possible for the treatment of petroleum products in an environmentally safe manner.

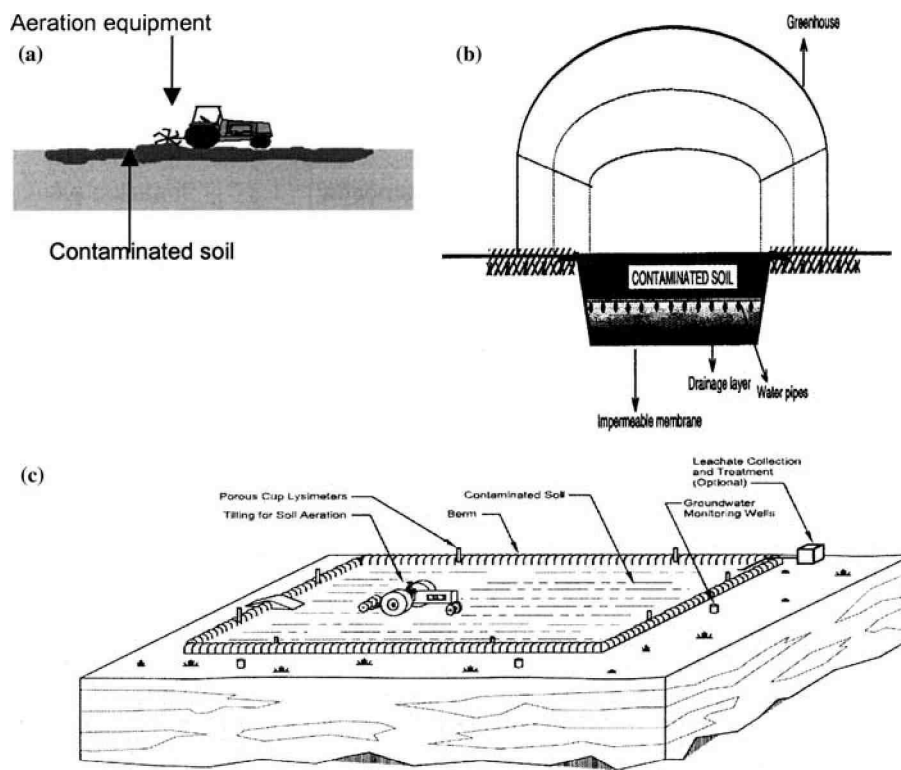


Figure 1. Different landfarm layouts (a) Traditional 'landfarming' system. (b) 'Complex' landfarm system adapted from Picado et al. (2001). (c) Landfarm system without a greenhouse structure adapted from EPA (1994).

Table 1. Benefits and constraints of landfarming

Technology	Benefits	Constraints
Landfarming	<ul style="list-style-type: none"> • Very low capital input required • Technology is simple to design and implement • Large soil volumes can be treated • Can be applied ex-situ • Has small environmental impact • Energy efficient 	<ul style="list-style-type: none"> • Limited to removal of biodegradable pollutants • Large treatment area is needed • Involves risk of pollutant exposure • Substantial cost can be incurred during excavation • Limited knowledge of microbial process or the unravelling limiting factors during bioremediation

4. Bioaugmentation and biostimulation

Bioaugmentation, the process of introducing pollutant-degrading bacteria to contaminated site, has been reported with mixed success (Van Veen et al. 1997). The limitation to successful bioaugmentation in soils have been cited as being due to suppression of added strains by indigenous microbial community (poor survival of the introduced strains) and the use of readily degradable substrates, due to low concentrations and non-biodegradability of targeted pollutants (Alexander 1994). Various efforts have been attempted to improve the success of bioaugmentation in contaminated sites (Del'Arco & de Franca 1999). Strategies employed to improve bioaugmentation process for the effective removal of contaminants from the soil include the use of adapted strains or the Field Application Vector (as tested by Lajoie et al. 1994). However, the most promising approach with regard to bioaugmentation has been attempted by 'seeding' the

biodegradation knowledge to the indigenous microbial populations (Miethling & Karlson 1996; EI Fantroussi et al. 1997; Kastner et al. 1998; Top et al. 1999). This involves the genetic transfer from the augmented strains to the indigenous bacteria.

With biodegradable pollutants like petroleum products (Table 2), biostimulation of microbiological processes at the contaminated site is encouraged. This usually involves the modification of the site by adjusting pH, addition of limiting nutrients to achieve an ideal C:N:P ratio and improving the soil moisture High petroleum








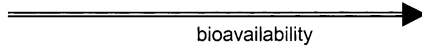
Physical		Chemical	
i  Particulate pollutant	iv  absorbed	Vii  chemically bound	
ii  liquid film	v  in water phase		
iii  surface adsorbed	in pores		
	vi  as separate phase in pores		
Extractability			
Easily extractable	poorly extractable (aged compounds)	non-extractable	
Bound pollutants			
High			low

Figure 2. Different physical and chemical forms of organic pollutants in soil i: solid particles, ii: liquid film, iii: adsorbed onto soil, iv: in the water phase, v: soil pores, vi: as a separate phase in soil pores, vii: chemically bound to soil adapted from Rulkens (1992), Volkering (1996) and Devliegher and Verstraete (1996).

hydrocarbon removal rates have been reported using the ratio of 100:10:1 (Genouw et al. 1994). Table 3 shows some of cases in which biostimulation and bioaugmentation were attempted with relative success. The availability of petroleum hydrocarbon-degrading bacteria should be investigated during the biotreatability studies. The presence of these bacteria at contaminated site indicates that remedial approaches involving biostimulation can be used to 'encourage' the biological removal of petroleum hydrocarbons from the soil.

Biostimulation of indigenous petroleum hydrocarbon-degrading bacteria in landfarms should be

encouraged ahead of bioaugmentation, as the former process relies on the degrading bacteria that have already adapted to the site's conditions.

Bioaugmentation should be implemented in contaminated sites where no indigenous petroleum hydrocarbon degrading bacteria exists, such as sites contaminated by high molecular weight polyaromatic hydrocarbons. The process of bioaugmentation should aim at 'seeding' the knowledge of degrading the pollutants to the indigenous bacteria (Brokamp & Schmidt 1991; Fulthorpe & Wyndham 1992; De Rore et al. 1994; Top et al. 1998, 1999; Verstraete & Top 1999). As the number of microorganisms tends to increase during biostimulation, the increase in the number of degrading bacteria can be used as potential bioindicators during bioremediation (Margesine et al. 1999).

Table 2. The biodegradability of different petroleum products (adapted from EPA 1994)

Biodegradability	Example constituents	Products in which constituent is typically found
<i>Hydrocarbons and biodegradability</i>		
More degradable	<i>n</i> -butane, <i>n</i> -pentane, <i>n</i> -octane	Gasoline
↓	Nonane	Diesel fuel
	Methyl butane, dimethylpentenes, methylloctanes	Gasoline Gasoline
	Benzene, toluene, ethylbenzene, xylenes	Diesel, Kerosene
	Propylbenzenes	Diesel
	Decanes	Kerosene
	Dodecanes	Heating fuels
	Tridecanes	Lubricating oils
	Tetradecanes	Diesel
	Naphthalenes	Kerosene
	Fluoranthenes	Heating oil
	Pyrenes	Lubricating oils
	Acenaphthenes	
	Less degradable	

Table 3. Successes of full-scale landfarming of TPH sites

Technology	Efficiency (%)	Microbial process and pollutants	Duration	References
	82-90	Biostimulation (oil)	12 months	Balba et al. (1998)
	43	Bioaugmentation oil	28 days	Del'Arco and de Franca (1999)
	80-90	Biostimulation PAHs	3 years	Berends and Kloeg (1986), Bossert and Bartha (1986) and Kincannon and Lin (1985)
Landfarming	78	Biostimulation PAH	3 months	Picado et al. (2001)
	15	Biostimulation (heavy molecular weight PAHs)	7 months	Schenk et al. (1992)

5. Lesson learned

The objective of landfarming is to treat petroleum-contaminated sites in an environmentally safe manner by harnessing the removal efficiencies of biological, physical and chemical processes in the soil. This objective is sometimes not realised due to the constraints of the technology. In addition, no standard procedure is available for determining the allowable loading of landfarms and

the time required for biodegradation of the petroleum compounds in the soil. This lack of procedure makes many landfarm designs to become a trial and error procedure with no assurances that the design will be successful in remediating the contaminated soil. While the bio-treatability protocol recommended by Sabate et al. (2004) is relevant, the urgency of the bio-treatability studies makes it difficult to gather the relevant information about optimising the processes involved in the removal of higher molecular weight petroleum compounds or the removal of poorly available part of the contaminants that are removed after the dissipation of the low molecular weight or the easily degradable petroleum compounds. There is a need to incorporate, in the biotreatability studies, investigations aimed at gathering information about the unravelling of the subsequent limiting factors during bioremediation. As this type of study may require a longer time than the 'generic' or well documented bio-treatability studies (EPA 1994; Sabate et al. 2004), the studies can run concurrently with the full scale treatment of the contaminated site. With this approach, the information obtained from the 'urgent' bio-treatability studies, can be used to initiate the full scale treatment, while the information from the 'extended' studies about the subsequent limiting factors, used to optimise the treatment after the removal of the easily degradable petroleum compounds.

Picado et al. (2001) reported a 63% reduction in total polyaromatic hydrocarbons (PAHs) concentration after the first three months of the treatment. The majority of the PAH removed during the treatment period were the 2, 3 and 4 ringed polyaromatic hydrocarbons. High molecular weight PAHs were not removed, probably due to lack of the degrading strains, unfavourable bacterial growth conditions or due to the fact that they required a longer treatment time to dissipate, as they are difficult to degrade. Knowledge about enhancing the removal of the remaining high molecular weight hydrocarbons after the dissipation of low molecular weight hydrocarbons can help in improving the efficiency of landfarming.

Bossert et al. (1986) studied landfarming of 16 PAHs present in oil-contaminated sludge and reported a reduction of about 80–90% after 3 years of treatment. Low removal rates of high molecular weight petroleum compounds and the long treatment periods were experienced in some of the studies (Table 3) due to the lack of process optimisation. According to Harmsen et al. (1994) landfarming include two steps; the first step involves an intensive treatment in which the readily available contaminants are removed. During the second step an extensive (intrinsic) treatment, the poorly available part of the contaminant is removed. In most landfarm operations, these two steps are not properly optimised by either biostimulation, in which an ideal C:N:P ratio is applied or by bioaugmentation in which the bio-degraders are added to degrade petroleum compounds that are difficult to degrade by the site's indigenous biota. In addition, subsequent limiting factors (nutrients, pH, biodegraders, toxic metabolites) during landfarming are not adequately addressed, resulting in long treatment periods.

While landfarming has been able to reduce the concentration of petroleum compounds in contaminated soil (Table 3), concern remains about its effectiveness in reducing the level of recalcitrant hydrocarbons and the potential toxicity of the metabolites generated during the degradation process. Also critical is the amount of time needed to reduce the concentration of petroleum compounds to levels acceptable by the regulators.

Apart from the generic approach of implementing landfarming, to treat petroleum compounds, it is important to take into account the 'added or non-additive effect' of potential limiting factors on bioremediation. This can be achieved by a detailed bio-treatability studies which can run concurrently with the full scale treatment process, or by incorporating an improved monitoring program that include investigation of the unravelling limiting factors.

6. Possible solutions to the constraints

One of the earlier concern about using landfarming to treat petroleum contaminated soil has been the risk of transferring environmental pollutants from one environmental compartment (soil) to another (air or groundwater). This necessitated the need to find solutions to both the physical, chemical and biological constraints associated with landfarming. Treatment standards had to be

met when applying the technology to remove petroleum compounds from the soil. The concern for further environmental contamination due to landfarming led to better treatment designs as shown in Figure 1 (b and c) from the traditional treatment approach (Figure 1a). Landfarming should be designed as shown in Figure 1b. This treatment design is able to prevent or minimise the transfer of contaminants from one environmental media to another. The design encompass a greenhouse structure that avoid or minimise dust and volatilisation of lighter petroleum compounds from the soil and also include an impermeable membrane with an impermeable layer (high density polyethylene membrane, 250 µm thick) which prevents ground water contamination. However, this ‘physical structure’ alone does not guarantee the efficient removal of petroleum compounds from the soil. The condition conducive to the proliferation of petroleum degrading bacteria in the soil has to be created for the efficient removal of petroleum compounds. This has to be established during the feasibility studies. In addition, as treatment standards vary from one country to another, the success of one treatment design in one country is not a guarantee that different treatment standards will be met in another country (Table 4).

Table 4. The landfarming principles

Parameter	Ideal characteristics
Soil	Well drained soil
Nature of pollutants	Pollutants should be biodegradable (by existing microbiota)
Climatic conditions	Greenhouse type structure (required to minimize erosion and precipitation effects)
Microbiological	Indigenous pollutant degrading bacteria and conducive environmental condition (pH, nutrients, moisture content, etc.)

As the technology ‘relies’ on the biological process to remove petroleum compounds, the key to successful remediation is to implement removal approaches that are inline with the petroleum degrading bacteria. It is important to first conduct the feasibility studies which will yield the information about the type and metabolic activity of the indigenous microorganisms at the site, presence of possible inhibitors, biodegradability of contaminants under optimal conditions, influence of nutrients and bioavailability of pollutants in soil. This information will also help the rehabilitation practitioner to decide if biostimulation or bioaugmentation is the relevant approach for cleaning the contaminated soil. However, while this information is useful for intensive treatment of petroleum compounds, it provides very little information about the unravelling of limiting factors during bioremediation and this can have an impact on the efficiency of landfarming. Landfarming design should include a monitoring plan, which addresses the limiting

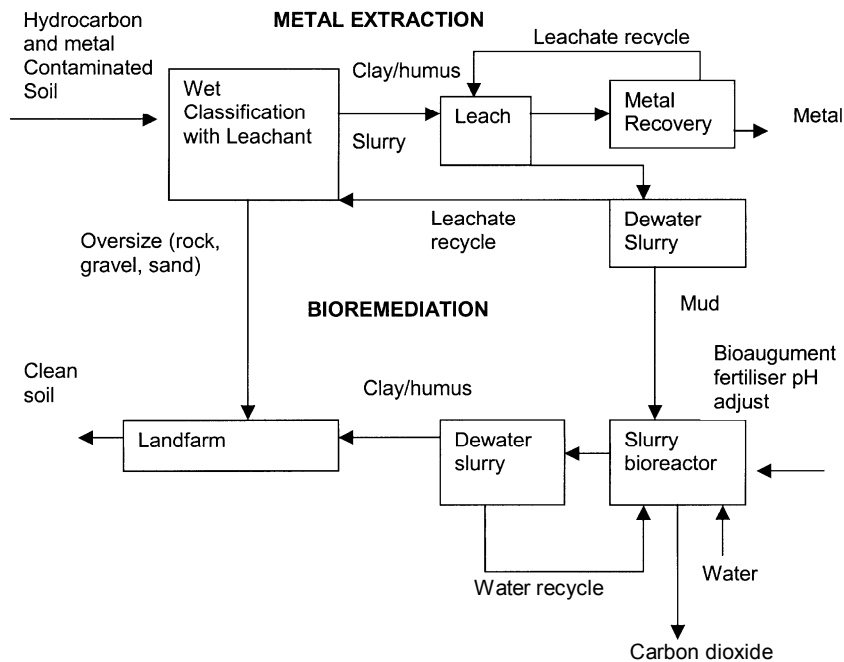


Figure 3. Metal leaching and bioremediation process adapted from US EPA (1992).

factors that may occur during bioremediation, particularly as both the biological, physical and chemical processes in the soil have the potential to alter soil conditions, which may become unfavourable to petroleum degrading bacteria.

Also, the petroleum products are often present in combination with other pollutants (e.g. heavy metals as in motor washbay areas) and this creates problems, as the metals can be toxic to hydrocarbon-degrading bacteria. In this case, a bio-separation process as shown in Figure 3 is recommended. However, soil washing is recommended if the sand fraction of the contaminated soil is large, as clay matrix can be destroyed at low pH (Tichy et al. 1996). With this process, metals can be removed by extraction while the petroleum hydrocarbons can be treated biologically using landfarming (Figure 3).

7. Ecological risk management

The volatilisation of lighter petroleum products and the mobility of petroleum pollutants from landfarms constitute a threat to humans and groundwater resources. The risk to humans and groundwater can be minimised by designing landfarms as shown in Figure 1b or 1c, in which the volatiles and the downward migrating pollutants are minimised or treated.

According to Hejazi et al. (2003), landfarming at the site poses risk of detrimental effects through the air pathway (through the inhalation exposure route) to site workers during the initial period of landfarming. Contaminated soils are excavated and spread on a pad with a built-in system to collect any 'leachate' or contaminated liquids that seep out of contaminant-soaked soil. In some cases, reduction of contaminant concentrations actually may be attributed more to volatilisation than biodegradation (Morgan & Watkinson 1989). When the process is conducted in enclosures controlling escaping volatile contaminants, volatilisation losses are minimized.

Bioremediation through landfarming aims to remove pollutants through conversion to CO₂ and water. However, in many cases, an important fraction of pollutant and its metabolites remain untouched by the cleaning process (Devliegher & Verstraete 1996). This amount of pollutant

remaining in the soil constitutes a major concern and source of debate in relation to risk assessment. The threat posed by the pollutant residues can be minimised by adding adsorbents to form the non-bioavailable residues as suggested by DeVliegheer and Verstraete (1996). Non-bioavailable pollutants can be considered as representing no direct harm to the environment. The different physical and chemical forms of organic pollutants are listed in Figure 2.

8. Future R&D needs

Landfarming is a cost-effective method of treating biodegradable petroleum products in the soil. However, it is important to design the treatment system in such a way that the transfer of pollutants to other environmental media is minimised or prevented. It is also important to modify the contaminated site's conditions to be 'inline' with the normal activities of the indigenous pollutant-degrading bacteria as this can improve the biological removal of petroleum products.

One of the disadvantages of landfarming is the inability of the technology to have concentration reductions of more than 95% (EPA 1994). This pollution reduction may (in some instances) not be adequate to meet regulations or standards from specific petroleum constituencies in some countries. As this can be attributed to the unavailability of the pollutant to biota, agents (like surfactants) that improve the bioavailability of petroleum products in soil must be considered during the design phase of the technology. This should be particularly encouraged where there is a significant risk posed by the remaining residues. However, the effectiveness of this approach must be compared with the addition of adsorbents, which can make the pollutant residues, less available and therefore not harmful to higher organisms.

Landfarming may also not be effective for high constituent concentrations in the soil. As high concentration of the pollutants can be toxic to soil microorganisms, studies should be undertaken during the biotreatability studies to determine the minimum amount of soil or adsorbents (e.g. straws which can also improve soil aeration) that can be added to the soil to reduce toxicity. It is therefore important to corroborate (using other petroleum products) the findings of Del'Arco and de Franca (2001), who reported that the extent of oil bio-degradation is inversely proportional to increasing oil contamination.

Landfarming has been used to treat volatile and biodegradable pollutants with relative success. However, the technology has not been greatly used to treat persistent organic pollutants like the high molecular weight polyaromatic hydrocarbons. There is a need to understand microbial processes and environmental conditions conducive for 'seeding' biodegradation information to the indigenous microbial communities. Remedial approaches involving bioaugmentation with the aim of increasing the removal capacity of the indigenous bacteria should therefore be evaluated at both pilot and large scale to improve the biological removal of persistent petroleum compounds using landfarming. It is also important to understand the unravelling of the subsequent limiting factors during bioremediation of both the low and high molecular weight petroleum compounds.

In conclusion, although simplicity and cost-effectiveness are the major advantages of using landfarming, the technology has 'inherent' physical, chemical and biological constraints. However, these constraints which are generally regarded as disadvantageous to implementing the technology can be addressed by applying the current wealth of knowledge on biodegradation and bioavailability of petroleum hydrocarbons, partitioning of petroleum hydrocarbons between environmental media, genetic transfer of the biodegradation knowledge to indigenous microbial communities, impact of petroleum products on soil microbial diversity and the intensive treatment of contaminated soil where space is a constraint. This wealth of knowledge on biodegradation and bioavailability of pollutants adds on to the advantages that have been well documented about landfarming. Hence, simplicity and costeffectiveness are not the only advantages associated with landfarming. Stimulated biological process and co-metabolism of recalcitrant (heavy molecular weight PAHs) are the other advantages associated with the technology. It is however, important to implement the technology in such a way that 'side effects' are minimised (i.e. there is less risk of transferring the pollution to other environmental media like the air and groundwater).

Acknowledgements

The author would like to thank Dr K Drønen and Dr P Wade for their constructive inputs to the paper. This work was also partially funded by the National Research Foundation-Research Council of Norway grant.

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