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Bioremediation of organic contaminated soils*

John R. Ryan
Remediation Technologies, Inc., 1011 S.W. Kickitat Way, Suite 207, Seattle, WA 98134 (USA)
Raymond C. Loehr
University of Texas, Austin, TX 78712 (USA)
and
E. Rucker
American Petroleum Institute, Washington, DC 20005 (USA)

Abstract

The term bioremediation covers a wide variety of engineered systems which utilize microorganisms to degrade, detoxify and immobilize organic contaminants. These systems include:

- solid-phase treatment using unlined land treatment systems or prepared bed bioreactors,
- slurry-phase treatment systems completed either in-place or within tanks or impoundments, and
- *in situ* treatment systems.

Bioremediation is cost effective, available and demonstrated. The technology is highly flexible and can be adapted to a broad variety of situations.

Introduction

The use of biological treatment technologies for the remediation of groundwater and soils contaminated with organic compounds has seen broad application over the past several years. It is not a novel technology. Bioremediation has been used for over 30 years to treat petroleum contaminated soils. Applications of the technology are becoming increasingly important in the field of hazardous waste due to its effectiveness and low cost as compared to other treatment alternatives.

While not a panacea, bioremediation is applicable for the treatment of a broad variety of organic contaminants found in soil at Resources Conservation and Recovery Act (RCRA) and Comprehensive Environmental Response

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Compensation and Liability Act (CERCLA) sites. This paper discusses some of the applicable processes and focuses on the existing applications.

Biological treatment processes

There are a variety of biological treatment processes currently in use which are often collectively referred to as bioremediation systems. These processes typically rely on the use of aerobic indigenous microorganisms at a site or enrichment with naturally occurring microorganisms to degrade the contaminants of interest. Generally these processes are grouped into three broad categories:

- solid-phase bioremediation
- slurry-phase bioremediation
- in situ bioremediation

Solid-phase bioremediation

Organic contaminants in soils are susceptible to aerobic digestion, and this process occurs naturally to a limited degree. The rates may be greatly enhanced by fertilizing, irrigating, and tilling the soil to increase the availability of nutrients, moisture and oxygen to the soil microorganisms, a process also described as landfarming, or treatment. In this process, the organisms used are most often the indigenous population, unless the environment is too severe. In these cases, parameters of the soil matrix can often be adjusted, and organisms known to degrade the contaminant can then be mixed into the soil matrix. Through scheduled tilling and maintenance of the moisture content, pH, and nutrients, decomposition and immobilization of the contaminants occur within both the upper soil layer (zone of incorporation), and the underlying layers. This type of treatment is one of the older and most widely used treatment technologies for hazardous waste treatment. In particular, the technology has been used successfully throughout the United States, especially at petroleum refinery sites treated under RCRA, and also with creosote-contaminated sludges and soils.

A variant of this technique is the use of prepared bed reactors. A general schematic of a prepared bed reactor is shown in Fig. 1. Although operations are similar to land treatment, the prepared bed reactors involve a greater degree of engineering controls including the use of liners, leachate collection systems, irrigation systems and nutrient delivery and inoculation systems. A number of prepared bed reactors have been used at CERCLA sites to degrade organic contaminants in soil. In some cases, the prepared bed reactors have been enclosed within greenhouse structures and equipped with air emission control systems.

A variant of the prepared bed reactor is composting. This type of treatment has been shown effective for the treatment of highly contaminated material,

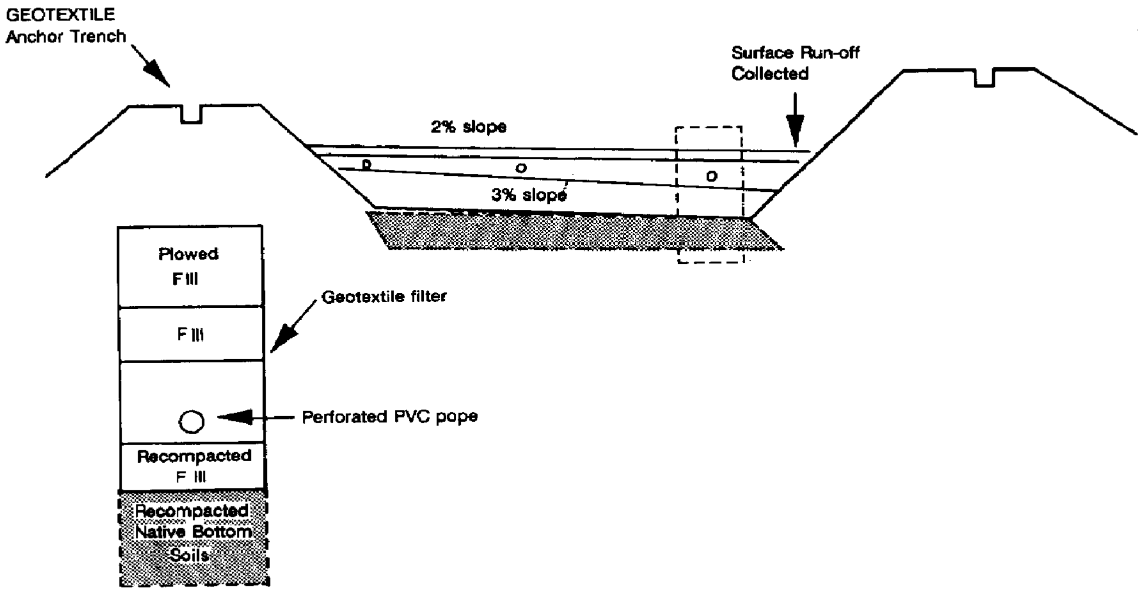

Fig. 1. Typical prepared bed bioreactor.

and consists of piling the contaminated soil, mixed with a bulking agent, at heights of three to six feet (0.9–1.8 m).Aeration is provided by either forcing air through a contained system, or by mechanically turning the pile. The bulking agents increase the total volume of the treated material, and facilitate mixing requirements and oxygen transfer. The system is amenable to moisture, pH, and nutrient control by simple irrigation techniques, and to volatile emission control, when the system is enclosed. Where temperature is critical to increasing removal rates, the compost pile can be amended with other sources of organic matter to increase the biological activity, and the temperature of the system.

Slurry-phase treatment

Treatment of soils in an aqueous media or slurry offers a significant control over important operating parameters resulting in rapid and effective treatment. The system relies on efficient mass transfer controlled by adequate mixing and aeration conditions. This type of treatment is similar in concept to conventional activated sludge treatment, but is usually conducted in batch mode, resulting in the discharge of active organisms at the same time as the cleaned soil is discharged from the treatment tank. The time required to re-establish an acclimated microbial population in subsequent batches can be minimized by recycling portions of the previous batch of solids and/or the decant.

Slurry-phase treatment requires entirely different equipment for materials handling than solid-phase treatment, and the issue of how best to promote

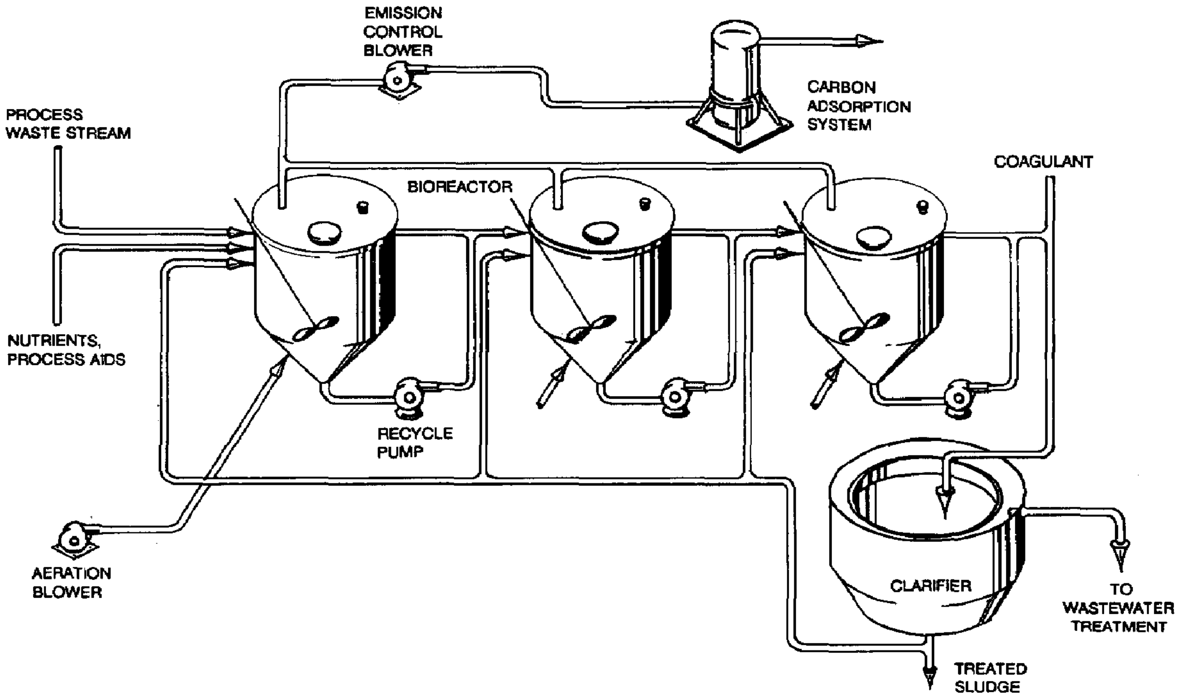

Fig. 2. Schematic of slurry-phase treatment system.

mass transfer of oxygen and soluble contaminants is a major concern for engineering design. The solids content of the resulting slurry depends on several factors including the type of soil, the type of mixing and aeration equipment available, the rates of removal to be achieved. The two rate limiting steps in an ideal system are the degree of solubilization of the organic contaminant, and the level at which active microbial populations can be maintained during the treatment.

The sophistication of the treatment system can range from lined lagoons constructed with earth materials at the site, to carefully designed and engineered reactors, depending on the objectives of treatment. Figure 2 illustrates a schematic of an engineered tank based system.

While the cost of treatment with liquid slurry treatments (LSTs) is highly influenced by the conditions of the site, the associated capital costs are over 50% less than alternative technologies such as incineration. The use of LSTs potentially could provide control of volatile emissions when they are of concern, and may also eliminate some statutory concerns related to contaminant migration.

In situ *treatment*

There are many cases where *in situ* biological treatment should be considered, and especially where soil excavation would be difficult or extremely expensive. The most common type of *in situ* treatment involves the biodegradation of contaminants which are adsorbed onto soils within the saturated zone of a site. The process involves the addition of small amounts of ammonia and phosphate, and large quantites of an oxygen source, typically (but not limited to) hydrogen peroxide. This is accomplished by injecting nutrient-enriched solutions into the contaminated zone through a series of wells or trenches, and recovering groundwater down gradient. Figure 3 provides a schematic of such a system.

In order for the process to be effective, the injection/recovery system must provide for the transport of nutrients throughout the entire contaminated region, following the pathway taken by the contaminant, if possible. This is particularly difficult at sites where the geology is highly irregular, or has been disturbed because of past construction, and at sites with multiple or unknown sources of contamination. The *in situ* remediation process is usually accompanied by surface treatment of the recovered groundwater.

The engineering parameters associated with this process are highly dependent on soil permeability, which becomes the rate-limiting step for mass transfer of oxygen to the aerobic organisms. In a few instances, *in situ* bioreclamation has been used for treatment in unsaturated soils. However, these cases are limited to fairly shallow depths over groundwater which is already contaminated. These treatment situations are difficult to control, and rely on introducing nutrient rich water through percolation, or through pressure injection with multiple injection points. Air is then drawn through the soil, using vacuum pumps, to enhance the air exchange in the soil matrix. *In situ* treatment is most cost-effective with contaminants which are easily degraded, but which have low solubilities in water.

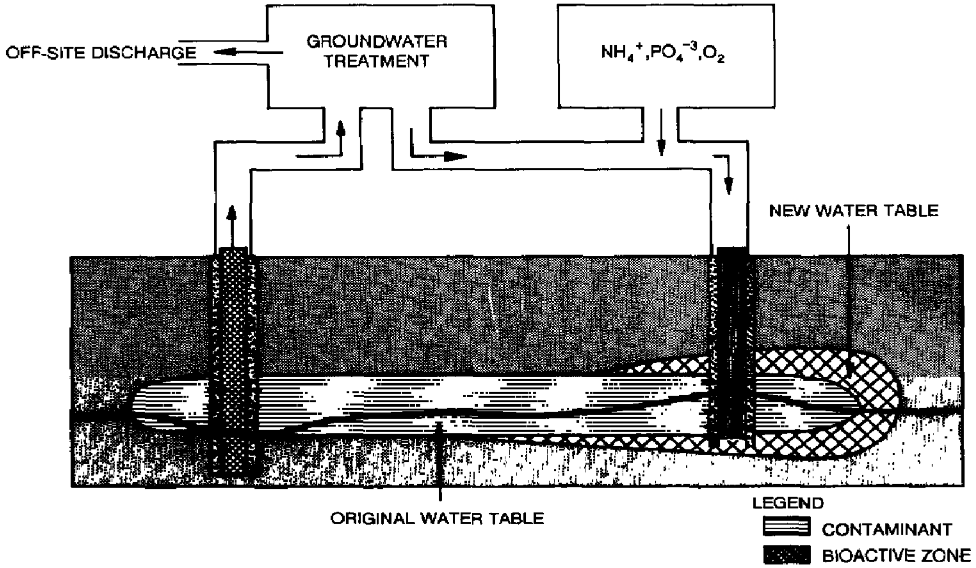

Fig. 3. *In situ* bioremediation of contaminated aquifer.

Applications of bioremediation

Solid-phase bioremediation has been used for over 30 years for the remediation of petroleum contaminated soils in unlined land treatment systems [1]. The use of prepared bed reactors was introduced in the last decade with one of the first applications involving the bioremediation of creosote contaminated soils at a Superfund site in Minnesota [2]. Prepared bed systems have also been used to treat petroleum contaminated soils using forced aeration composting techniques [3].

In situ applications were pioneered in 1972 by Sun Refining to remediate a gasoline spill [4]. Since that time, there have been a number of engineering advancements in nutrient and oxygen delivery systems. It is estimated that over 100 *in situ* projects have been implemented. Most of the applications to date have been related to light petroleum derivatives associated with gasoline and diesel contamination. Several field demonstrations have been initiated in the past several years directed towards heavier coal tar derivatives as well as halogenated aromatics [5].

Slurry-phase systems are a fairly recent innovation. Most applications to date have involved treatment of sludges and contaminated soils resulting from the closure of impoundments containing petroleum refining wastes, petroleum production wastes or petrochemical wastes [6].

There are approximately 20 CERCLA sites for which "Records of Decision" (RODs) have been issued for soil bioremediation. In addition, there are ap-

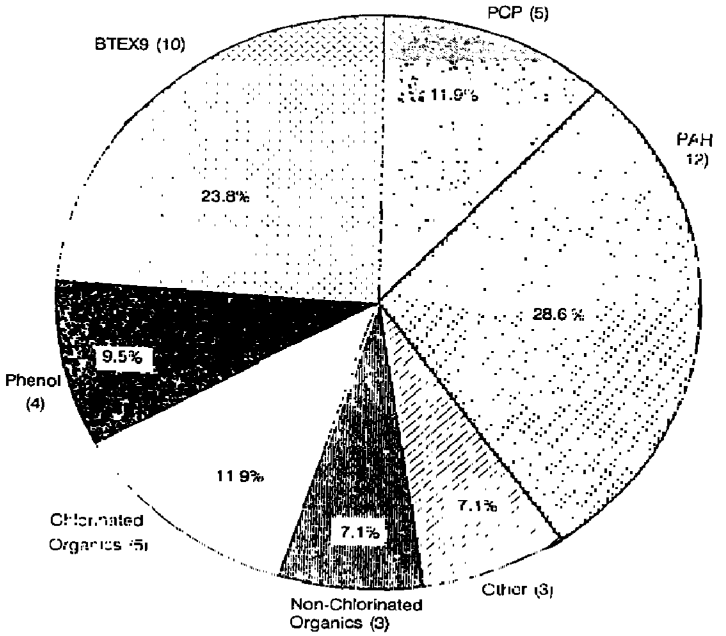

Fig. 4. Source control-bioremediation sites (major chemicals found at these sites).

TABLE 1

Superfund Record of Decisions (RODs) - Summary of soil bioremediation remedies (update January 1991)

Site	Waste type/contaminants	Bioremediation method	Status	Est. soil volume (yards^3)
Fiscal Year 1989				
American Creosote Works, FL	PAH, PCP	Slurry-phase	Design	23,000
Ordinance works Disposal areas, WV	Chemical formulation, PAHs	Prepared bed, solid-phase	Remedial design	13,000
Galesburg, IL	Wood preserving, PAH/PCP	prepared bed solid-phase	Ongoing, field demo	15,000
Cliff/Dow, MI	Charcoal manufactoring, BTX/PCE/phenols	Forced aeration composting	Remedial design	9,000
Sheridan Disposal, TX	Commercial waste disposal, VOC/PCB	Slurry-phase treatment	Ongoing field demo	44,000
Vogel Paint and Wax, IA	Paint manufactoring, solvents	To be determined	Remedial design	3,000
Somers, MT	Wood preserving, PAH	Prepared bed solid-phase	Remedial design	12,000
Libby, MT	Wood preserving, PAH/PCP	Prepared bed solid-phase	Full-scale	30,000
Oroville, CA	Wood preserving, PAH/PCP	*In situ*	Completed field demo	110,000
Whitmoyer Labs, PA	Organic arsenic formulation	To be determined	Remedial design	Unknown
Fiscal Year 1988				
Iron Horse Park, MA	Indust. complex and Railyard, VOC/PAH	To be determined	Remedial design	23,000
LA Clarke and Sons, VA	Wood preserving, PAH	*In situ*/land treatment	Remedial design	119,000
Live Oak, FL	Wood preserving, PAH	Prepared bed solid-phase	Full-scale	8,000
Clovis, NM	Railyard, phenols	*In situ*/land treatment	Full-scale	28,000
French LTD, TX	Petrochemicals, PAH/PCP/VOC	Slurry-phase	Completed field demo	10,000
North Cavalcade, TX	Wood preserving, PAH	Slurry-phase/land treatment	Design	22,000
Fiscal Year 1987				
Renora, NJ	Waste oil disposal, PAH/VOC	To be determined	Unknown	Unknown
Fiscal Year 1986				
Leetown Pesticide, WV	Pesticide storage and disposal	Unknown	Treatability completed	Unknown
Brainerd, MN	Wood preserving, PAH	Prepared bed solid-phase	Full-scale	12,000
Fiscal year 1984				
Old Inger, LA	Oil refining and waste oil reclamation, PAH	prepared bed solid-phase	Full-scale	150,000

proximately 17 sites which call for ground water bioremediation using above ground biological reactors and five ROD's which specify in situ treatment of groundwater. Some of the groundwater sites are included in the number of soil bioremediation sites. Table 1 identifies the soil bioremediation sites and Fig. 4 identifies the major types of contaminants which are being treated.

A large majority of the Superfund sites involve the use of solid-phase bioremediation using prepared bed bioreactors. The liner systems have been constructed using either clay or synthetic materials. Over half the sites are contaminated with polynuclear aromatic hydrocarbons (PAH) and/or light aromatics (BTEX). The majority of the RODs have been issued since 1988 and many of the sites are currently in the "Remedial Design" (RD) phase. Several are currently in full-scale treatment or have completed treatment.

In addition to these 20 sites, ReTeC (Remediation Technologies, Inc., Seattle, WA) is currently involved in a fund financed bioremediation program as part of an emergency response at a former wood preserving site in Alton, Missouri. The program involves treating over 14,000 cubic yards (10^4 m^3) of creosote contaminated soil in a solid-phase prepared bed reactor.

Within the RCRA program, there are numerous examples of bioremediation as part of treating on-going process wastes as well as for closure of impoundments. The broadest use has been in the petroleum refining sector which has used unlined land treatment systems for treating oily wastewater sludges and contaminated soils for over 30 years. There are approximately 100 RCRA permitted land treatment facilities in the U.S. Many of these sites are currently contemplating closure in response to the "Land Disposal Restrictions" (LDRs).

Over 40 facilities have filed "No-Migration Petitions" in order to obtain a variance from the LDR's. Bioremediation is also being used as part of closing RCRA surface impoundments. Most of these programs involve *in situ* slurry-phase treatment sometimes in conjunction with solid-phase prepared bed bioremediation. ReTeC has completed over 15 of these projects involving wood preserving, petroleum refining and petrochemical residuals. The treated residuals have subsequently been stabilized and capped in-place.

Solid-phase bioremediation is a common treatment technology for non-hazardous petroleum contaminated soils. Many of these projects are driven by state regulations and are difficult to quantify in terms of numbers. This subset, however represents the broadest experience base. Solid-phase techniques which have been applied to these materials include unlined land treatment systems, prepared bed bioreactors and forced aeration systems.

The above list (Table 2) is by no means exhaustive. However, it provides a perspective on the variety of bioremediation approaches which are currently in use as well as the broad applicability of these treatment systems. It should be noted that in addition to the existing experience base continual advances are being made in the field with respect to process development as well as microbial techniques. Notable amongst these are the use of anaerobic processes, applications of selective strains of bacteria or fungi and the use of co-metabolites to induce degradation of recalcitrant compounds [3].

Performance

Evaluating the performance of bioremediation systems has largely been based on defining the rate of biodegradation of specific compounds. Numberous factors affect biodegradation rate which are both compound and matrix specific as well as process related [7,8]. A number of well documented laboratory studies are available which define degradation rates for specific compounds under a broad variety of environmental and process conditions [9,10]. Most of these studies have focused on solid-phase bioremediation and most of the data relates to PAH, light aromatics and phenolic compounds present in petroleum refining or wood preserving wastes.

Comparative studies of solid-phase and slurry-phase treatment have generally found that degradation rates are faster within slurry-phase systems although economic factors often favor solid-phase systems [11].

There are limited studies available which compile the large amount of performance data collected on field systems. Much of this data has been collected as part of field demonstrations completed at CERCLA sites or as part of land treatment demonstrations completed as part of RCRA permitting efforts. Field studies completed by ReTeC on the performance of solid and slurry-phase bioremediation system have consistently reported over 90% removal of aromatic and polynuclear aromatic compounds [12,13].

EPA evaluated data from 67 different studies to characterize the effectiveness of soil treatment technologies to treat different chemical groups [14]. Included in this evaluation was bioremediation, although there was no effort made to differentiate between different bioremediation technologies. The results of the survey indicate that bioremediation can successfully treat many halogenated aliphatic compounds, nonhalogenated aromatic, polynuclear aromatic, heterocyclics and other polar compounds in excess of 99%.

Removal efficiencies are a function of treatment time. A broad variety of chemicals can be successfully degraded to environmentally acceptable levels under appropriate treatment conditions and adequate treatment times. Typically, waste specific treatability and/or field studies are necessary to define the optimal treatment conditions and attainable end point concentrations for a specific situation.

In addition to degradation rate, several studies have focused on the relative mobility and toxicity of the contaminants before and after treatment. These studies have shown that bioremediation is capable of detoxifying and immobilizing the treated residuals [9,10].

Summary and conclusions

The term bioremediation covers a wide variety of engineered systems which utilize microorganisms to degrade, detoxify and immobilize organic contaminants. These systems include:

- solid-phase treatment using unlined land treatment systems or prepared bed bioreactors,
- slurry-phase treatment systems completed either in-place or within tanks or impoundments, and
- *in situ* treatment systems.

A broad experience base exists with the application of bioremediation techniques to the treatment of contaminated soils. There are at least 40 RODs which specify bioremediation for the treatment of soils and/or groundwater at CERCLA sites. In addition, there is a wealth of experience in the RCRA sector as well as related to the treatment of non-hazardous petroleum contaminated soils.

There is a large data base concerning the performance of these systems although the information is highly disagregated and often lacks quality assurance data. The data has shown that a broad variety of chemicals can be successfully degraded to environmentally acceptable levels under appropriate treatment conditions and adequate treatment times.

Bioremediation is cost effective, available and demonstrated. The technology is highly flexible and can be adapted to a broad variety of situations.

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