

IN SITU RESTORATION TECHNIQUES FOR AQUIFERS CONTAMINATED WITH HAZARDOUS WASTES

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(Received November 8, 1985; accepted April 3, 1986)

Summary

Improper disposal of hazardous wastes is a threat to the nation's ground water supply. Methods which prevent contamination are probably the most effective techniques to protect ground water. Once contamination problems occur, there are a number of in situ techniques that can be used to cleanse the ground water and at least partially restore the aquifer. Before any treatment program can be implemented, a thorough investigation of the hydrogeology and contamination problems of the site must be made. Plume management techniques such as barriers to ground water flow or hydrodynamic control can be effective when properly installed. One of the options used frequently is to remove the contaminated material to a secure site; while this cleans up the contaminated site, the material is not treated and the potential for contamination of the second site exists. Chemical and physical treatment techniques include processes such as neutralization, chemical reaction, extraction and immobilization. Biological techniques for in situ treatment generally involve enhancing the degradative capacity of the indigenous microflora or the addition of organisms acclimated to degrade the contaminants. Combinations of chemical and biological processes are often effective. Aquifer restoration is likely to be costly, time-consuming, and often only partially effective.

Introduction

In 1978, the Environmental Protection Agency estimated that there were 25,000 active industrial impoundments and landfills in the USA and that many of these could impact usable aquifers [1]. Many of these sites may require remedial action to restore the ground water to usable condition. As Josephson [2] pointed out, protection of the aquifers is probably the best solution to the problem since it is likely to be difficult and expensive to clean up ground water contaminated by improper disposal of hazardous materials. However, once an incident of ground water pollution has occurred, there are a number of techniques that can be used to clean up the polluted aquifer. Available techniques range from physical containment to in situ treatment with chemicals or microbes to various forms of chemical, bio-

logical or physical treatment following removal or withdrawal of the contaminated soils and ground waters. A thorough investigation of the hydrologic conditions of the site and the extent and type of contaminants that are present must be completed before any treatment can be started [3]. Installation of a number of monitoring wells followed by an extensive monitoring program is usually required to determine the aquifer characteristics and the extent of contamination. Geophysical techniques such as ground penetrating radar, electric resistivity soundings, borehole geophysics, seismic refraction profiling, remote sensing, and magnetometry may fill in the gaps in the hydrogeologic information without the costs of additional well installation [4]. If the soil is too impermeable, then it will be difficult to circulate treatment agents or to even withdraw the polluted water [3]. Soils with high organic matter content will tend to strongly adsorb organics but may release them later. The type of contamination also is important; contaminants that are highly water soluble must be handled differently than those that float on the water table like gasoline. Free product or the source of the contamination should be removed before a treatment program is initiated.

Plume management techniques

Physical containment

Removal

Containment and/or removal were the most frequently employed techniques in a survey of 169 remedial actions by Neely et al. [5]. Excavation involves removal and transport of the contaminated soil to a secure site such as a landfill; ground water is usually pumped out and treated by any of several techniques. Excavation is usually accomplished by a dragline which can reach a maximum depth of 60 feet (18.3 m) or a backhoe that can go a little deeper, 70 feet (21.3 m) [6]. Excavation costs between \$1.75 to \$4.50 per cubic yard (\$2.30 to \$5.90 per cubic meter) plus additional costs for transportation of the contaminated material and disposal in an approved facility. However, an estimated 50–60% of the landfills with interim status under the Resource Conservation and Recovery Act (RCRA) may be contaminating ground water, including several RCRA facilities that have received materials from Superfund cleanups [7]. It also may be difficult to excavate all of the contaminated soil in instances where contamination is widespread or when the contaminated aquifer is deep below the land surface.

Barriers to ground water flow

Other plume containment measures include slurry trench walls, grout curtains, sheet piling, block displacement, infiltration controls, and passive interceptor systems [8]. Slurry trench cutoff walls are typically constructed by excavating a narrow trench and backfilling the trench with a soil–bentonite

or cement-bentonite mixture [3]. The resulting barrier has a permeability that ranges from 10^{-6} to 10^{-8} cm/s depending upon the materials used. Slurry trench cutoff walls are reported to have long service life, are easy to install, and are highly cost-effective under certain conditions. However, the durability of the slurry wall, when exposed to organic contaminants in ground water, is not known [4]. Grout curtains are constructed by injecting chemical or suspension grouts under pressure into staggered, closely spaced holes so that the soil pores are filled. Chemical grouts, usually silicates, can penetrate anywhere water moves and prove most useful in fine sediments. They typically consist of two or more liquids that will gel when they come in contact [8]. Suspension grouts are made of Portland cement or bentonite depending upon the size of the particles in the formation; bentonite grouts can be used in finer materials than cement grouts [3]. Suspension or particulate grouts solidify in place as the grout reacts with water [8]. Grout curtains offer many of the same advantages of the slurry wall in that they have a long service life, can be constructed quickly, and can be used in both unconsolidated and consolidated geologic material, but they may be limited in their use since it is difficult to ensure that the grouts from each hole will overlap [3]. In addition, certain chemical grouts are toxicants [8]. Costs of grout cutoff systems have been estimated between \$150 and \$350 per cubic foot (\$5,300 and \$12,360 per cubic meter) depending upon the choice of the grout. Sheet pilings are formed by driving interlocking steel sheet piles through the soil. In coarse, dense material it may be difficult to form an effective barrier and it may not be possible to drive pile in soils with boulders. The life of the steel may be affected by the corrosivity of the contaminated water or soil. Costs for sheet pilings are estimated to be between \$6.30 and \$9.50 per square foot (\$67.80 and \$102.30 per square meter). Few applications of this technology have been made. Block displacement is a method, currently under development, for placing an underground barrier around and underneath a block of earth [6]. It may be useful in situations where unweathered bedrock or other impermeable stratum is not near the surface. The bottom barrier is formed when a pressurized slurry is pumped into fractures in the soil or into notches formed at the bases of the injection holes. A perimeter barrier is installed using traditional techniques.

Unless there is an impermeable bottom to secure a ground water barrier, it may be impossible to prevent flow under or around the barrier without establishing supplemental hydrodynamic control via installation of pumping and injection wells [9].

Hydrodynamic control

Passive ground water controls

Interceptor drains and trenches collect ground water without the use of mechanical devices such as pumps [8]. In these systems, trenches are dug, perforated pipes are placed in the trenches, and the trenches are backfilled

with gravel. The water flows into the interceptor system where it is collected for treatment or is diverted away from the site. Advantages of subsurface drains include relatively simple and inexpensive construction, inexpensive operation since few mechanical devices are utilized, and collection of more concentrated wastes than well-point systems. Limitations on their use include the need for frequent monitoring to assure adequate leachate collection, especially of dissolved components. Subsurface drains are poorly suited for use in soils with low permeability and installation will probably not be feasible in existing sites, although they can be installed upgradient and used to divert water away from the waste site.

Well systems

Hydrodynamic control is accomplished by proper placement of withdrawal and injection wells. Contaminated ground water is withdrawn to prevent it from moving further and may be reinjected, after treatment, downgradient of the plume. These wells can serve to create a ground water divide that separates polluted ground water from clean ground water [3]. A thorough understanding of the hydrogeology of the site must precede the installation of an interceptor system; this is usually accomplished by installation of a network of monitoring wells and characterization of the aquifer through pump tests, analysis of soil properties, and other methods. Free product can be recovered in the event of a spill by several techniques [8]. A single pump system utilizing one or more wells requires minimal equipment and drilling costs, but produces a mixture of product and water that must be separated. A two pump, two well system utilizes one well to produce a water table gradient that allows the second well to recover the floating product. Another type of system utilizes a single well with two pumps, a lower pump to produce a gradient and an upper pump to collect the free product. Well systems, probably the most extensively utilized method of ground water pollution control, represent a proven method for control of many hazardous waste contamination problems. But they are not without disadvantages: high maintenance and operation costs; limited application to fine soils; removal of clean water along with contaminated water which increases the costs of treatment; and long operation times, especially to remove trapped or sorbed contaminants.

Chemical and physical treatment techniques

In situ treatment technologies are largely still under development and are highly dependent on the nature of the contaminants and the soil environment [1]. In situ chemical detoxification techniques include injection of neutralizing agents for acid or caustic ground waters, addition of oxidizing agents to destroy organics or precipitate inorganic compounds, addition of agents that promote photodegradation or other natural degradation processes, extraction of contaminants, immobilization, or reaction in treatment beds.

One example of in situ chemical treatment would be the precipitation of free cyanide with sodium hypochlorite; 2,500 parts per million (ppm) hypochlorite as available chloride has been used to treat cyanide contamination from industrial wastes, although metal complexed cyanides were not treatable by this technique [10]. Additional examples include the reduction of hexavalent chromium with ferrous sulfate and precipitation of heavy metals with alkali agents [6]. Injection of chemical treatment agents may displace pollutants due to the added volume or other hazardous compounds may be produced by undesired reactions. Permeable treatment beds can intercept the plume and provide a reactor for either chemical treatment or precipitation. Materials that can be used in this type of treatment scheme include limestone or crushed shell to neutralize acids and remove heavy metals; activated carbon to remove non-polar organic compounds; glauconitic green sand, a clay from the Mid Atlantic states, that can adsorb from 60 to greater than 90 percent of the heavy metals in solution; zeolites and synthetic ion exchange resins to remove heavy metals, although they are likely to have a short lifetime, high costs, and are difficult to regenerate. Permeable treatment beds may plug or exhibit channeling which will reduce their effectiveness. Extraction of contaminants with solvents, such as organic solvents, acids, chelating agents, ammonia, or water, can be used to remove some compounds; however, the potential exists for further contamination of ground water with these agents. Mobilization of the contaminants by surfactants during soil washing may be a viable option [11]. A four percent solution of two non-ionic surfactants was able to remove greater than 90 percent of polychlorinated biphenyls and a high boiling distillation fraction of crude oil from test soil columns with washes of 10 pore volumes.

The contaminants also can be immobilized by precipitation or encapsulation in an insoluble matrix [1]. A spill of acrylate monomer was treated with catalyst and activator and converted to a solidified polymer; an estimated 85 to 90 percent of the liquid monomer was solidified [8]. Other in situ treatment techniques have been suggested including use of radio frequency for in situ heating [12] and in situ vitrification using an electric current to melt the soils and waste in place [6].

Biological treatment

In situ biological treatment relies on the action of microbes to degrade contaminants with the microorganisms deriving energy and increased cell mass from the process. Microorganisms can degrade many organic compounds, but environmental constraints such as dissolved oxygen, pH, temperature, toxicants, oxidation-reduction potential, availability of inorganic nutrients including nitrogen, phosphorus and others, salinity, and the concentration and nature of the organics may control biodegradation. The number and types of organisms present in the subsurface also may play an important role. Another technique for in situ biological treatment involves

the addition of acclimated microorganisms to enhance the degradation of the hazardous materials.

In situ treatment

Treatment in place utilizes the indigenous microbial population to degrade the contaminants [13]. Microbial populations in excess of one million organisms per gram (dry weight) soil have been found in several shallow water table aquifers [14]. Organisms from pristine aquifers have been shown to be capable of degrading a number of organic compounds ranging from styrene, toluene, and chlorobenzene, to bromodichloromethane [15]. Organisms from contaminated sites may be capable of degrading a wider range of compounds once they become acclimated to the organic compounds, a process that may require several months [16]. At many sites contaminated with hazardous organic wastes such as aromatic compounds, the levels of dissolved oxygen may control biodegradation [17]. In situ treatment processes usually involve the circulation of both oxygen and inorganic nutrients through the aquifer so that indigenous organisms can degrade the contaminants [13,18].

Supplying dissolved oxygen to the ground water is likely to be the limiting factor in the biostimulation process, especially in low-permeability aquifers [13]. Diffusers that sparge compressed air into the ground water can not exceed the solubility of oxygen in water, 8 to 10 ppm. Use of pure oxygen can increase the dissolved oxygen content to 40 to 50 ppm, but pure oxygen is expensive and the supersaturated oxygen is likely to bubble out of solution (degas) before the microbes can utilize it [19]. Hydrogen peroxide, which decomposes to form water and oxygen, can supply much greater oxygen levels. However, concentrations of hydrogen peroxide as low as 200 ppm may be toxic to microbes and levels above 100 ppm may degas to form air bubbles that block the formation. It may be possible to overcome these limitations by acclimating organisms to increasing levels of hydrogen peroxide and by stabilizing the hydrogen peroxide solution. Column tests with four levels of oxygen supplied by air, a nitrogen and pure oxygen mixture, pure oxygen, and a stabilized hydrogen peroxide solution showed an increase in the numbers of bacteria and the degradation of gasoline with increasing levels of oxygen. Field evidence for this effect has also been generated [20]. Dissolved oxygen concentrations in monitoring wells at a site contaminated with gasoline increased from a high of 4 ppm with air sparging to 10 ppm after addition of 100 ppm hydrogen peroxide; a concurrent increase in the numbers of gasoline utilizing organisms was observed. A reduction in the size of the gasoline plume and a decrease from a high of 4 ppm to 2.5 ppm hydrocarbon occurred, although it was difficult to assess the relative contribution of the hydrogen peroxide addition among the other restoration measures employed.

Ozone also can be used as a source of oxygen, but suffers from the same limitations as hydrogen peroxide in that it is toxic to bacteria and may

generate gas bubbles that block the pores in the formation [21]. Ozone was used to treat a hydrocarbon contaminated aquifer in West Germany. The water was pumped out, treated with ozone, and recirculated to the aquifer via injection wells. Dissolved oxygen levels and microbial counts increased in the wells and a decrease in the amounts of dissolved organic carbon and mineral oil hydrocarbons in the water was noted. Reaction of the hydrocarbon with ozone may have accounted for some of the destruction of the organics.

Bioremediation has been chiefly used, with reasonable success, to treat aquifers contaminated with petroleum hydrocarbons [13]. Most of the free product is removed and the levels of dissolved hydrocarbons are reduced. An example of the success of the bioremediation process was at a site in LaGrange, Oregon, contaminated with gasoline. All of the free gasoline was removed during the first seven months of the nutrient supplementation program, but gasoline odors and a cloudy sheen were detected in the ground water with dissolved organic carbon levels (DOC) averaging 20 ppm. After operation for an additional three months, DOC levels were reduced to below 5 ppm for most samples. The bioremediation process has also been used with good results at a site in Waldwick, New Jersey, contaminated with methylene chloride, acetone, n-butyl alcohol and dimethylaniline. Bioremediation is useful for hydrocarbons and certain other compounds, especially water soluble compounds and low levels of other compounds that would be difficult to remove by other means. It is environmentally sound since it destroys organic contaminants and in most cases does not generate waste products. The treatment moves with the plume and thus can reach organics trapped or sorbed by the soil matrix. However, bioremediation does not work with heavy metals and some organics which are toxic or recalcitrant. Bacterial growth can plug the subsurface and reduce circulation. The introduction of nutrients and the residues generated by the organisms can adversely affect water quality. Bioremediation may not work well in aquifers of low permeabilities which do not permit adequate circulation of the nutrients and dissolved oxygen.

A laboratory study to address the use of bioremediation techniques in low permeability formations showed that gasoline-utilizing bacteria could penetrate sand columns prepared with sand packs of coarse 20-mesh sand to very fine 80-mesh sand, with effective permeabilities ranging from 200 darcies (1.93 cm/s) to 3.5 darcies (0.04 cm/s), and consolidated sandstone cores, with effective permeabilities of 19 and 75 millidarcies (1.8×10^{-4} and 7.2×10^{-4} cm/s) [13]. Bioremediation has been used in aquifers composed of dolomite, a highly permeable sand and alluvial fan deposits composed of sand, gravel and cobbles, with some clay and silt. A field demonstration in a very gravelly clay loam was largely unsuccessful due partly to low permeability (3.9×10^{-5} to 3.3×10^{-3} cm/s) which made it difficult to inject nutrients and produce water [22]. The extreme complexities of the site and contamination problems also contributed to the poor success of this field

demonstration. A laboratory study had shown that many of the contaminants including n-alkanes and chlorobenzenes were biodegradable under aerobic conditions whereas chlorinated aliphatics were probably degradable only under anaerobic conditions [23]. Problems encountered in the field demonstration portion of the project included possible mobilization of lead and antimony by the hydrogen peroxide treatment, although the levels of these metals in the ground water did not increase, and reductions in the permeability of the soil due to precipitation of the nutrients. No significant evidence of biodegradation was observed in the first two months of hydrogen peroxide addition but the nutrients had not reached most wells by this time [24]. After 23 weeks, elevated carbon dioxide levels, a metabolic byproduct, were noted in production wells where the nutrients had broken through [22]. Reductions in the concentrations of total hydrocarbon and individual organic pollutants such as chlorobenzene were noted.

Another approach for in situ treatment of aquifers contaminated by hazardous disposal sites is to enrich the growth of a particular population of microbes with specific metabolic capabilities. Wilson and Wilson [25] proposed a method for the removal of trichloroethylene from contaminated ground water. Trichloroethylene is a common ground water contaminant having been found in more than a quarter of the wells sampled in a survey in New Jersey. Similar compounds can be transformed into products which are as bad as the original compound; under anaerobic conditions, tetrachloroethylene can be transformed by reductive dehalogenation to trichloroethylene, dichloroethylene, and vinyl chloride, but is not completely degraded [26]. The method for removal of trichloroethylene proposed by Wilson and Wilson [25] involves enriching the microbial population for methanotrophs and propane oxidizers by the addition of natural gas. The methanotrophs possess an enzyme, monooxygenase, that can oxidize and dechlorinate halogenated methanes. The propane oxidizers then epoxidate the ethylene which can be degraded further to carbon dioxide. This aerobic process results in the complete degradation of trichloroethylene. Other examples of this type of treatment are possible, but have not yet been developed.

Many compounds resistant to aerobic degradation can be transformed under anaerobic conditions; examples are chloroform, bromodichloromethane, dibromochloromethane, bromoform and 1,1,1-trichloroethane [27]. Different redox conditions may also affect the transformation of a compound; Bouwer and McCarty [28, 29] showed that chloroform and 1,1,1-trichloroethylene were degraded under methanogenic conditions, but not under denitrification conditions. Many contaminated aquifers will be anaerobic if the microbial population is capable of degrading the contaminants and it may be possible to use anaerobic in situ techniques to treat some compounds. If the redox conditions can be controlled to achieve conditions under which specific compounds can be degraded, dehalogenation can be promoted, and particular organisms or enzyme systems can be selected.

Addition of acclimated microorganisms

Microbes can be added to either in situ or conventional biological wastewater treatment processes to attempt to enhance biodegradation by increasing biomass and/or by reducing the time necessary for acclimation to occur. Organisms can be selected by enrichment culturing where organisms are exposed to contaminants in increasing levels over long periods under conditions favorable to acclimation. Inoculating acclimated organisms into a new environment has met with variable success. The organism must be able to locate and degrade the compounds of interest at what are often very low concentrations, must be able to survive in the environment in which it is placed, and must retain its selectivity for the compounds to which it was adapted [30]. Genetic engineering techniques can also be used in which organisms with unique metabolic capabilities are fashioned. However, no conclusive evidence has been found that genetically engineered microorganisms have been established in aeration basins to in natural environments already having an active microbial population [31]. Recently, a great deal of concern has been expressed over the release of genetically engineered organisms in the environment. It may be some time before this issue is resolved and the use of genetically engineered organisms to clean up hazardous waste sites is allowed [32]. In general, acclimated and/or genetically engineered organisms will not survive or offer significant advantage in treatment of hazardous wastes unless environmental parameters (dissolved oxygen, temperature, nutrient conditions, etc.) can be controlled to promote survival of the added organisms.

Acclimated organisms have been used in remedial actions to clean up contaminated ground water [21]. At an unidentified site where ethylene glycol and propyl acetate had been spilled, initial treatment by air stripping and clarification was followed by addition of acclimated bacteria, nutrients, and air to the aquifer. The total organic carbon in the water was reduced from 40,000 to 1 ppm. The importance of the acclimated organisms in this reduction could not be assessed. At another site, a spill of 130,000 gallons (492,700 l) of organic chemicals entered a shallow, unconfined aquifer separated from a major drinking aquifer by thick silty clay. Initial treatment was by clarification, adsorption onto granular activated carbon and air stripping. When levels of the contaminants had fallen below 1,000 ppm, the indigenous microflora and a specific facultative hydrocarbon degrader added to the soil were able to rapidly degrade the contaminants. An in situ treatment program using these acclimated organisms along with the addition of nutrients and oxygen accelerated the removal of the pollutants in the soil and succeeded in reducing the levels in the ground water to below 1 ppm. In laboratory studies conducted previously, the acclimated organisms were unable to reduce the levels of the organics beyond that achieved by the indigenous population, so the observed decrease in the organic concentrations may have been due to the native organisms and not to the added hydrocarbon degraders. This

treatment program demonstrates the effectiveness of a combination of traditional physical—chemical and innovative biological treatment technologies.

Conclusions

Many in situ techniques exist to treat aquifers contaminated by hazardous wastes. These include controlling the flow of the polluted ground water, removal to a secure site, and treatment by chemical or biological techniques. Before any process can be implemented, a thorough understanding of the hydrogeology and contamination problems of the site must be obtained and used to design the treatment system. It is likely that the treatment will be time consuming and expensive, with costs ranging from tens of thousands of dollars for simple treatment programs up to the tens or millions of dollars for complex, large sites. The past history of remedial actions at hazardous waste sites suggests that containment and conventional treatment technologies may often be unsuccessful. With more experience and development of innovative treatment techniques, it should be possible to more successfully restore hazardous waste disposal sites [33]. Most treatment schemes currently in use are not completely effective and do not offer permanent solutions for containment or remediation. Some methods may create additional uncontrolled hazardous waste sites. The magnitude of the hazardous waste disposal problem in the U.S. demands a concerted exploratory research effort to develop more effective ground water restoration technologies and to better understand the problems of ground water contamination [34].

Disclaimer

Although the research described in this article has been supported by the United States Environmental Protection Agency through assistance agreement No. CR-812808 to Rice University, it has not been subjected to Agency review and therefore does not necessarily reflect the views of the Agency and no official endorsement should be inferred.

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