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A Field-Scale Demonstration of a Novel Bioremediation Process for MGP Sites

V. J. SRIVASTAVA,[1] R. L. KELLEY,*[,1] J. R. PATEREK,[1]
T. D. HAYES,[2] G. L. NELSON,[3] AND J. GOLCHIN[4]

[1]*Institute of Gas Technology, Chicago, IL;* [2]*Gas Research Institute, Chicago, IL;* [3]*Midwest Gas Company, Sioux City, IA;*
and [4]*State of Iowa, DNR, Ames, IA*

ABSTRACT

The Institute of Gas Technology (IGT) has developed and demonstrated an integrated chemical/biological treatment (CBT) process to efficiently remediate soils and sludges contaminated with hazardous compounds such as polynuclear aromatic hydrocarbons (PAHs), volatile hydrocarbons (e.g., BTEX), and polychlorinated biphenyls (PCBs). Bench-scale studies as well as the field-scale tests show that the CBT process is effective in significantly enhancing the rate as well as the extent of degradation of these contaminants.

In this paper, the results of bench-scale tests conducted with a variety of PAHs and a set of field-scale tests conducted with soils from a former manufactured gas plant (MGP) site (now a Superfund site) that contained coal-tar associated PAHs. The field tests results show that the chemically enhanced bioremediation using the CBT process results in up to 90% improvement over conventional bioremediation for total PAHs (2–6 ring compounds) degradation and over 100% improvement over conventional bioremediation for carcinogenic PAHs (4–6 ring compounds) degradation. In this paper, the preliminary results of the second field test being conducted using the contaminated soils from another MGP site are discussed.

Index Entries: Bioremediation; field-scale tests; soils.

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*Author to whom all correspondence and reprint requests should be addressed.

INTRODUCTION

History of Manufactured Gas Plants

It has been estimated that more than 1500 manufactured gas plants (MGP) were in existence in the United States during the nineteenth century and the first half of the twentieth century. The first manufactured gas (town gas) plant was built in England in 1812 by London and Westminster Chartered Gas, Light and Coke Company, although the first record of experimental manufactured gas production from coal dates back to seventeenth century England (*1*). North America's first manufactured gas plants were built in Baltimore in 1816, in Boston in 1822, and in New York in 1825 (*2*). The early processes involved the "carbonization," or destructive distillation of bituminous coal at temperatures of 600–800°C in small cast iron retorts (*3*), producing "retort gas" or "coal gas." Over the next hundred years, a variety of gas manufacturing processes were developed, with different fuels and processes used under the varying circumstances of geography, demography, transportation, and fuel availability (*4*). These processes included coal carbonization, carburetted water gas, oil gas, coke oven gas, and product and blast furnace gas.

Manufactured Gas Plant Waste Characteristics

Manufactured gas plants produced large quantities of tar and related byproducts. Although there are some similarities among these sites, the sites often vary significantly in the specific types and quantities of wastes present, depending on the types of processes used (coal, water, or oil gas) and the era in which the plant operated. Polynuclear aromatic hydrocarbons (PAHs) are components of all kinds of tars, and tars are a byproduct of all of these plants. The physical and chemical characteristics of these tars vary, however, according to the process employed (*3–5*). Oxide wastes from purifier boxes may contain ferrocyanide and varying amounts of arsenic, chromium, copper, lead, nickel, and zinc (all of which appear on the Environmental Protection Agency priority pollutants list). Lamp black, clinker, cinders, and ash may also be present (*1,5,6*).

The MGP wastes can be categorized into five major types:

1. Free tars, oils, and lamp black;
2. Organic waste or tar-contaminated soils;
3. Organic waste or tar/oil-contaminated waters;
4. Purifier box (or spent oxide) wastes;
5. Mixed wastes and fills.

Specifically, the contaminants of interest present at MGP sites can also be divided into five categories: inorganics, metals, volatile aromatics, phenolics, and polynuclear aromatic hydrocarbons.

The organic-contaminated or PAH-containing soils represent the largest waste type at most sites. Some of the PAHs are suspected or potential carcinogens and, therefore, must be removed or treated to reduce their concentrations so that they do not pose a risk to humans and/or animals. These risks may be caused by the direct contact with soils (ingestion, dermal contact, inhalation, and so on) as well as direct and/or indirect effect associated with the groundwater and surface water.

The Institute of Gas Technology (IGT) has been developing processes to treat soils and water at former MGP sites that are contaminated with wastes such as PAHs, cyanides, and metals (*7–11*). Remediation technologies are also being developed for other gas industry wastes such as halogenated hydrocarbons, including polychlorinated biphenyls (PCBs), and perchloroethylene (PCE). The ultimate goal of this technology development program is to provide a cost-effective waste treatment technology alternative to conventional options that include containment, land filling, chemical fixation, soil washing, thermal treatment, conventional bioremediation, and so on. The limitations of the conventional options include either limited/incomplete/insufficient waste degradation, considerable expense, or both. IGT's approach has been to identify the treatment-limiting steps and then develop approaches to overcome those limiting steps. As a result of extensive bench-scale studies conducted since 1987, IGT has developed and demonstrated two processes for PAH-contaminated soils that are a combination of biological treatment and physical/chemical treatment: The integrated Chemical/Biological Treatment (CBT) or MGP-REM Process, and The Fluid-Extraction/Biological Degradation (FEB) process.

The MGP-REM or Chemical/Biological Treatment (CBT) process combines two complementary remedial techniques: chemical oxidation as the pretreatment for hard-to-degrade contaminants, and biological treatment using an aerobic biosystem. The CBT process uses mild chemical treatment with Fenton's reagent ($H_2O_2 + Fe^{2+}$) that produces hydroxyl radicals that start the chain reaction with the organic contaminants, resulting in modification and degradation of organics to biodegradable and environmentally benign products. These products and other organics are later degraded in the biological step. Results from bench-scale studies conducted with approx 25 MGP soil samples show that the CBT process is capable of enhancing the rate as well as the extent of PAH degradation. The results indicate that when a soil is dominated by 4–6-ring PAHs, chemical treatment is best performed as a pretreatment step. When initial soil screening indicates that a soil contains 2- and 3-ring PAHs in significant amounts, biological treatment is recommended as the initial step to remove as many of these compounds as possible. Chemical treatment using Fenton's reaction is then used to reduce the remaining persistent PAHs.

METHODS AND MATERIALS

Bench-Scale Soil Treatability Protocol

A treatability protocol developed for the Gas Research Institute (GRI) and IGT's Sustaining Membership Program (SMP) by IGT scientists and other GRI contractors has been successfully evaluated using coal tar-laden soils. Some of the soil was used as a source of inoculum in a basal mineral salts nutrient medium and is incubated aerobically to determine if indigenous microorganisms exist that can grow at the expense of organic contaminants. Following this soil-characterization phase, more of the same soil was evaluated for contaminant mass transfer using a method called "Solids Desorption," which was performed to estimate to what extent, if any, adsorbed wastes would passively desorb from the soil.

The next step of the treatability study evaluated biodegradation of the organic wastes. The rates and extents of biodegradation were determined in liquid-culture experiments and soil slurry reactor studies that are conducted for 30–60 d. Microbial cultures obtained from site enrichment or cultures maintained in IGT's stock culture collection was used as inoculum if needed. The results of these studies provide an indication of how effective a bioremediation process is likely to be for a given soil sample.

Simulated land treatment options are evaluated using soil-pan microcosm. Typically, 1–2 kg of contaminated soil is placed into Pyrex glass pans and tested under a variety of treatment conditions. A typical aerobic soil-microcosm study takes a minimum of 4–6 mo to perform even for aerobic conditions. Soil microcosm treatability studies are usually incubated under optimal conditions of pH, moisture, tilling, and temperature. Usually the most degradable pollutants are removed within a period of one growing season, the length of which depends on geographical location and climate conditions, normally 80–120 d.

Chemical Treatment Protocol

Typically, chemical treatment consists of 1% Fe^{2+} (16.87 g of ferrous sulfate/kg of soil) and 1% hydrogen peroxide (31.4 mL of 30% technical grade H_2O_2/kg of soil). Because of the exothermic nature of the chemical reactions, the temperature is maintained by slow addition of the hydrogen peroxide to the soil or soil slurry containing the Fe^{2+}.

Field Design and Operating Protocol

Based upon the treatability results on the soil from the site in Dubuque, IA, it was decided to demonstrate this technology in the field. Several experimental designs were evaluated. One treatment plot design included four treatment cells with dimensions of 4′ × 12′ each. Each plot was to employ a different land treatment technology with one of them acting as a control. Another design included eight treatment plots of the same dimensions. The purpose of this design was to provide repetition in treatment and analysis, and thus reduce random error. The third design included a total of sixteen treatment plots of the same dimensions. Four plots were to be dedicated to one of four land treatment technologies. The sixteen plots, however, were to be loaded in a random sequence, thus minimizing errors associated with loading, treatment, and analysis.

A liner system was established under each of the treatment blocks to facilitate water management and to prevent contamination of the underlying soil. Above this liner was a layer of rock or gravel, then clean sand, and finally 15–20 cm of the contaminated soil. Each of the treatment plots were bounded by similar boards. This made it possible to till the various plots using a garden tractor fitted with a rototiller on a weekly or biweekly basis. Decontamination of the equipment was performed between treatment blocks. A water management system was established to hold water derived from each of the treatment blocks and the decontamination zone in an onsite impoundment. Collected water was used for irrigation during the experiment. Meteorological data were collected for ambient temperature, wind speed and direction, and precipitation, at the site. Soil samples were collected at the beginning of the experiment and after 1 d, 1 wk, 2, 4, 6, 8, and 10 wk of treatment. Parameters measured included PAHs, nutrient levels, microbial population levels, pH, and moisture.

In the first field study the main conditions evaluated were Treatment 1—unamended control; Treatment 2—amendment with nutrients, tilling, and irrigation; Treatment 3—amendment with nutrients, tilling, moisture, and Fenton's reagents at the beginning of the study and on a periodic basis by adding hydrogen peroxide directly to the soil during irrigation.

A second field trial for land treatment of PAH-contaminated town gas soil (designated TGS-18) was conducted from August–November 1992. The contaminated soil was collected from a site held by Midwest Gas Company. The treatments examined were:

1. Control, conventional bioremediation (nutrients, water, and tilling);
2. Light CBT (0.5% hydrogen peroxide followed by bioremediation);
3. Regular CBT (1.0% hydrogen peroxide followed by bioremediation); and
4. Heavy CBT (2% hydrogen peroxide followed by bioremediation).

Table 1
Summary of Field Weather Data

Parameter	Field test no. 1, 1991, 8/14-9/18	Field test no. 2, 1992, 8/7-9/18
Air temperature, °F		
Average	73.6 + 12.2	68.1 + 11.4
Median	72.6	67.0
Soil temperature, °F		
Average	81.9 + 10.3	80.2 + 11.1
Median	82.5	79.0
Relative humidity, %		
Average	74.9 + 19.2	89.0 + 10.7
Median	79.6	96.2
Total rainfall, in.	3.11	4.66

Table 2
Comparison of Field Test Soils Analytical Data

	1992	1991
Soil pH	4.5[a]	7.9
Organic matter, %	2.5	3.0
Phosphorus, ppm	17	9
Potassium, ppm,	55	80
Nitrate-N, ppm	<1	12
TPH, ppm	3400	N.D.[b]
Oil and grease	9275	N.D.[b]
PAHs, ppm	3000	1000
Sand, %	96	64
Silt, %	1	30
Clay, %	3	6
Density, g/cu cm	1.35	N.D.[b]

[a] Very low buffering capacity causing low retention of lime.

[b] N.D., not determined.

During both field studies, analytical measurements monitored include PAH concentration, soil pH, and total heterotrophic bacterial counts. Other data collected included air and soil temperature, rainfall, and relative humidity. The environmental data for both field studies are reported in Table 1. A comparison of analysis data for the field soil are reported in Table 2.

Sample Collection, Preservation, and Transportation for Analysis

Random samples were collected from each of the test plots over the duration of the experiment. Composite samples were collected for each sampling event using a soil corer or auger. Eight to ten cores were taken from each plot and mixed together to form a composite sample of about 500 g. On occasions where samples were sent to laboratories for validation, 1 kg was collected as a composite. This 1-kg composite was divided, and 500 g were sent to the laboratory for analysis. Because test methods for Soxhlet extraction require only 10 g of soil, this provided a sufficient sample for dry soil, PAH analysis, and other tests.

All soil composites were placed in clean glass containers (widemouth jars with Teflon-lined caps) for transport to the analytical laboratory. All samples were stored on ice in a cooler for preservation and shipped by overnight express to the analytical laboratories. On receipt at the laboratory, samples were stored refrigerated (4°C), Soxhlet extracted within 7 d, and extracts analyzed within 40 d. The pH determinations were performed immediately, and microbial enumeration were performed within 48 h.

PAH Determinations

PAHs were determined in the composite sample from the site using a modified EPA SW-846 Method 8100 analysis of Soxhlet extracts (*12–15*). Soxhlet extraction was performed by EPA SW-846 Method 3540 using a 1:1 mixture of acetone and hexane as the extraction solvent. The modifications of Method 8100 involved use of mesitylene (1, 3, 5-trimethylbenzene) for the treatability test and 2-fluorobiphenyl for the field test as an internal standard and detection using ion-trap detecting gas chromatography/mass spectrometry (Perkin-Elmer Model 8320/800).

A suite of PAHs was analyzed in this treatability study and is summarized in Table 3 for IGT Total PAHs. These 23 compounds are subdivided into two groups: IGT 2 and 3-ring PAHs and IGT 4 to 6-ring PAHs, based on the number of rings represented by the various constituent PAHs. IGT's previous experience has revealed that 2- and 3-ring PAHs are most readily biodegraded, whereas the 4- to 6-ring PAHs are more persistent.

RESULTS AND DISCUSSION

Bench-Scale Feasibility

The effectiveness of the MGP-REM treatment process was demonstrated using the highly contaminated soil. In an example of this effectiveness, MGP soil was used in experiments involving Fenton's reaction

Table 3
Polynuclear Aromatic Hydrocarbons Assayed by IGT Chromatography and EPA SW-846 Method

PAH analyte	Ring number	IGT total PAHs	IGT 2 and 3 ring PAHs	IGT 4 to 6 ring PAHs	EPA total PAHs	EPA noncarcinogenic PAHs	EPA carcinogenic PAHs
Naphthalene	2	Naphthalene	Naphthalene		Naphthalene	Naphthalene	
1-Methylnaphthalene	2	1-Methylnaphthalene	1-Methylnaphthalene				
2-Methylnaphthalene^a	2				2-Methylnaphthalene^a	2-Methylnaphthalene^a	
Biphenyl	2	Biphenyl	Biphenyl				
Acenaphthylene	3	Acenaphthylene	Acenaphthylene		Acenaphthylene	Acenaphthylene	
Acenaphthene	3	Acenaphthene	Acenaphthene		Acenaphthene	Acenaphthene	
Dibenzofuran^a	3	Dibenzofuran	Dibenzofuran		Dibenzofuran^a	Dibenzofuran^a	
Fluorene	3	Fluorene	Fluorene		Fluorene	Fluorene	
Phenanthrene	3	Phenanthrene	Phenanthrene		Phenanthrene	Phenanthrene	
Anthracene	3	Anthracene	Anthracene		Anthracene	Anthracene	
1-Phenylnaphthalene	3	1-Phenylnaphthalene	1-Phenylnaphthalene				
2-Methylphenanthrene	3	2-Methylphenanthrene	2-Methylphenanthrene				
1-Methylphenanthrene	3	1-Methylphenanthrene	1-Methylphenanthrene				
Fluoranthene	4	Fluoranthene		Fluoranthene	Fluroanthene	Fluoranthene	
Pyrene	4	Pyrene		Pyrene	Pyrene	Pyrene	
Benz(a)anthracene	4	Benz(a)anthracene		Benz(a)anthracene	Benz(a)anthracene		Benz(a)anthracene
Chrysene^b	4	Chrysene^b		Chrysene^b	Chrysene^b		Chrysene^b
Triphenylene^b	4	Triphenylene^b		Triphenylene^b			
Benzo(b)fluoranthene^c	5	Benzo(b)fluoranthene^c		Benzo(b)fluoranthene^c	Benzo(b)fluoranthene^c		Benzo(b)fluoranthene^c
Benzo(k)fluoranthene^c	5	Benzo(k)fluoranthene^c		Benzo(k)fluoranthene^c	Benzo(k)fluoranthene^c		Benzo(k)fluoranthene^c
Benzo(a)pyrene	5	Benzo(a)pyrene		Benzo(a)pyrene	Benzo(a)pyrene		Benzo(a)pyrene
Indeno(1,2,3-cd)pyrene	5	Indeno(1,2,3-cd)pyrene		Indeno(1,2,3-cd)pyrene	Indeno(1,2,3-cd)pyrene		Indeno(1,2,3-cd)pyrene
Dibenzo(a,h)anthracene	5	Dibenzo(a,h)anthracene		Dibenzo(a,h)anthracene	Dibenzo(a,h)anthracene		Dibenzo(a,h)anthracene
Benzo(g,h,i)perylene	6	Benzo(g,h,i)perylene		Benzo(g,h,i)perylene	Benzo(g,h,i)perylene	Benzo(g,h,i)perylene	

[a] 2-Methylnaphthalene and dibenzofuran are PAHs listed as EPA PAHs for consistency with previous testing performed by Midwest Gas. These PAHs are not normally analyzed by EPA SW-846 Method 8100 but had been selected from the suite of compounds listed in EPA SW-846 Method 8270. Also 2-methylnaphthalene is not an analyte included in the IGT chromatography protocol. All other PAHs listed as EPA PAHs were analyzed for this treatability study.

[b] Chrysene and triphenylene coelute during chromatography.

[c] Benzo(b)fluoranthene and Benzo(k)fluoranthene coelute during chromatography.

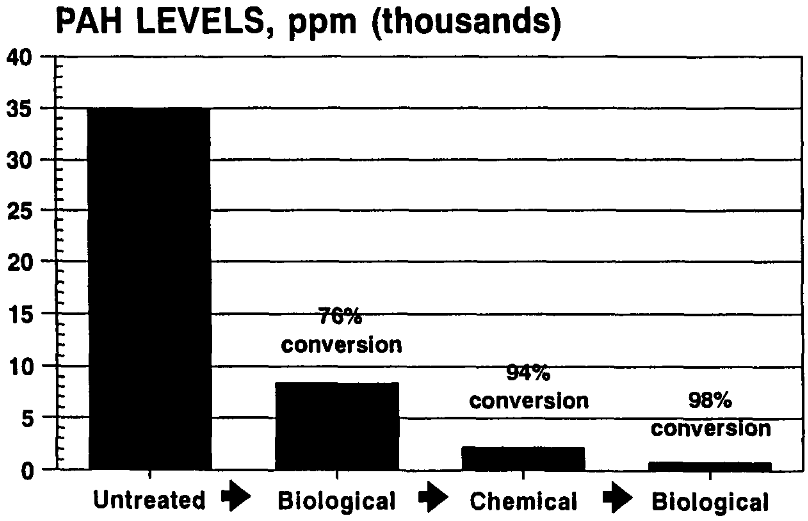

Fig. 1. Total PAHs in TGS-7 soil after sequential biological-chemical-biological treatment.

applied after an initial period of aerobic biological treatment. Figure 1 shows that the initial period of aerobic biological treatment reduced total PAHs by 76% from 35,000 to about 8400 ppm. When Fenton's reaction was used as a posttreatment, PAH levels dropped by an additional 18% to 2200 ppm; and when coupled with a second round of biological treatment, the total PAH reduction approached 98%. Added cycles of combined chemical and biological treatments could be implemented as deemed necessary to reduce the end point further. The aerobic biological treatment alone could reduce PAHs to only just under 80% after a long treatment duration.

Field Evaluation Data

The merits of the MGP-REM (or CBT) process were verified in a field experiment conducted with the soils from an MGP site that is also a Superfund site in the state of Iowa (*2,4,11,16–20*). The primary objective of this field experiment was to compare PAH degradation as a direct consequence of microorganisms, or from chemical treatment using Fenton's reaction as a pretreatment or as a cotreatment in conjunction with biological processes. Figure 2 presents the results of the field test conducted in the landfarming mode of the CBT process. In this figure, the residual PAH concentrations in the soil are compared with control plots that did not receive any nutrients, conventional bioremediation plots that received nutrients, and CBT treatment plots. Because this particular soil naturally contained nutrients, the control plots also exhibited some degradation of

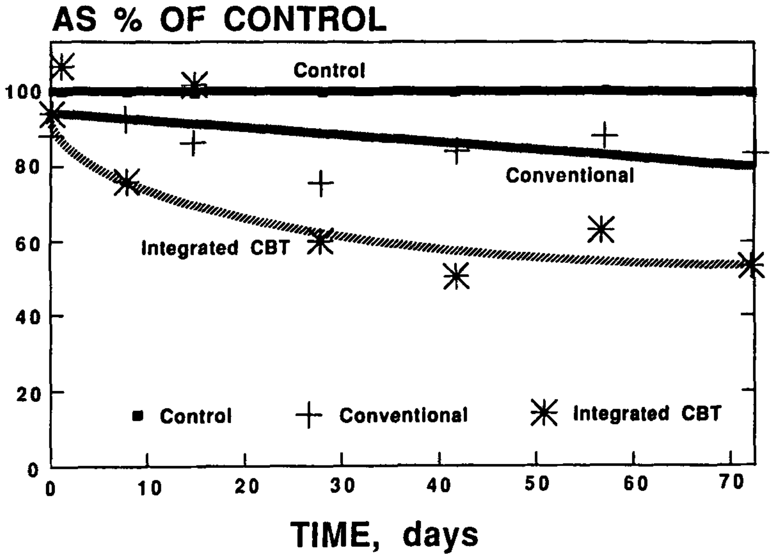

Fig. 2. PAH Concentrations in midwest gas soil during field test in 1991.

Table 4
Total PAH Reduction After 42 D of Treatment

Treatment	Explanation	PAH reduction, percent	
		Total PAHs	Carcinogenic PAHs
1	Control, not managed	41 ± 7	29 ± 5
2	Conventional bioremediation, addition of N + P at d 21	49 ± 6	29 ± 3
3	Integrated chemical, Biological treatment Chemical addition as cotreatment (d 1 and d 29)	72 ± 6	56 ± 4

PAHs. Therefore, this figure is intended to show the PAH reductions over and above those observed in control plots. Table 4 shows percent degradation of PAHs in each treatment plot groups as total EPA PAHs and carcinogenic EPA PAHs (as defined by US EPA and presented in Table 3) after 42 d. Initial land treatment of 1% chemical treatment on d 0 and additional 1% chemical treatment on d 29 appears to give the best results with higher PAH degradation and at faster rate of degradation. The integrated treatment reduced PAHs at a higher rate and to a greater extent than the conventional bioremediation. The treatment goals for this soil were met within the first 28 d when using the CBT process.

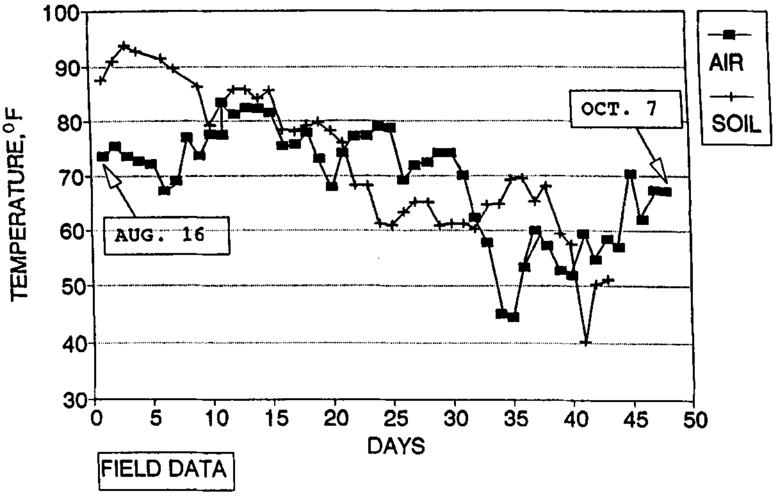

Fig. 3. Temperature profiles for soil and air during the field demonstration period.

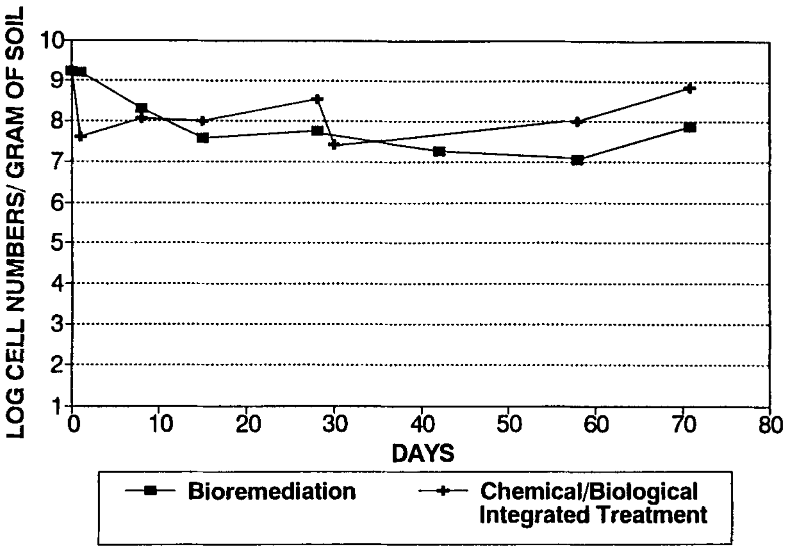

Fig. 4. Bacterial soil titer for each treatment during the field demonstration.

Meteorological data were collected so that ambient temperature (both air and soil), wind speed and direction, precipitation, and relative humidity, were known at the site. Nighttime temperatures below 32°F were first seen after 31 d of operation, and soil temperatures were below 45°F after 40 d of operation (Fig. 3). Table 3 summarizes the field weather data.

Bacterial-soil titer for all the treatment remained relatively constant at 10^8 cell/g of soil (Fig. 4). An apparent loss in bacterial titer, of approx one

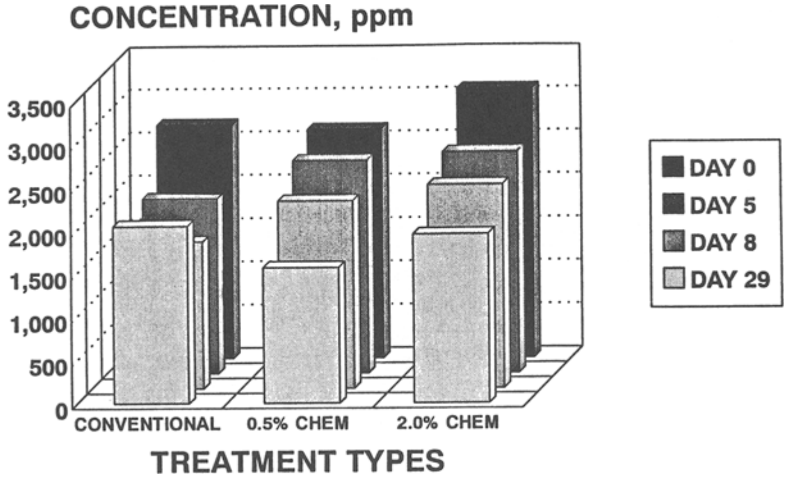

Fig. 5. 1992 MGP-REM field test, concentration of EPA total PAHs during the first 29 d of treatment.

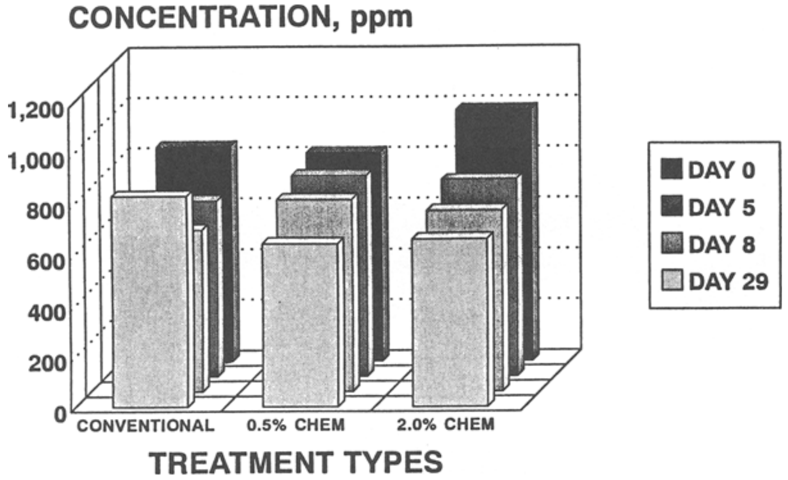

Fig. 6. 1992 MGP-REM field test, concentration of EPA carcinogenic PAHs during the first 29 d of treatment.

log coincides with the chemical treatments; however, the significance of this decline appears to be inconsequential.

A second field trial was conducted with a different soil (Table 2). The objective of the first field experiment was expanded to compare not only conventional bioremediation with the CBT process, but also to compare several strengths of the chemical treatment. Concentrations of EPA Total PAHs and EPA Carcinogenic PAHs are presented in Figs. 5 and 6, respectively. The data from the conventional treatment, light treatment, and heavy treatment are shown. Total microbial counts and pH levels are given in Figs. 7 and 8, respectively. The control plot, conventional CBT,

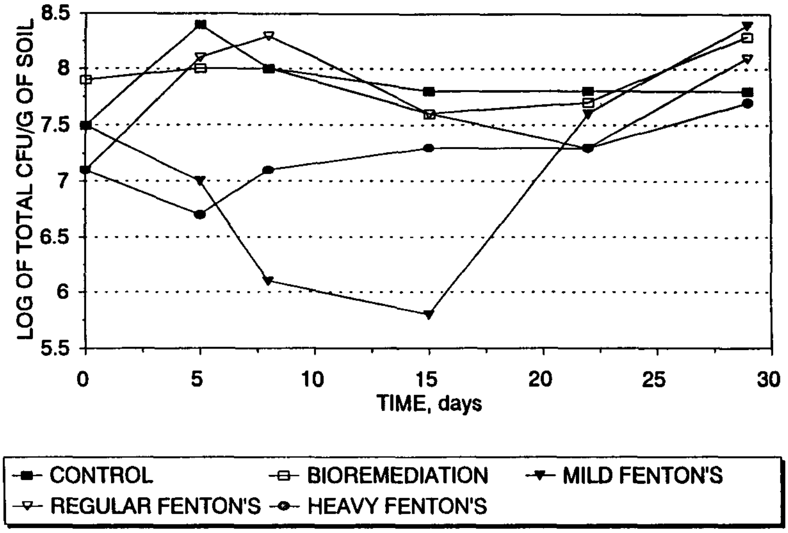

Fig. 7. Microbial counts during 1992 land treatment trial.

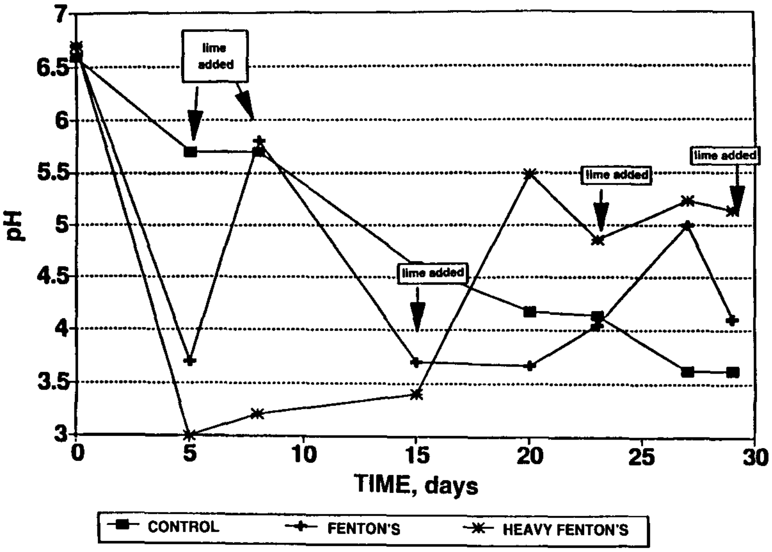

Fig. 8. The pH levels during 1992 land treatment trials with pH adjustments indicated.

and heavy CBT are compared in these figures. These preliminary data indicate a removal of both EPA Total PAHs and EPA Carcinogenic PAHs at a slow but steady rate. There does not appear to be a great deal of difference between the treatment regimes in relation to the amount of chemical treatment. This may be a result of the inhibition of the microogranisms by the extreme drop in pH after chemical treatment. This drop in pH was caused by a lack of buffering capacity in this soil. Lime additions appear to be increasing, but not completely neutralizing, the pH. This could

Table 5
Preliminary Economic Analysis:
Landfarming at Iowa Site 1991[a]

Chemical concentration, wt%	Treatment cost, $ per cu yd
0.5	48
1.0	60
1.5	72
2.0	85
3.0	110

[a] A 1% chemical addition was the minimum dosage that resulted in the successful attainment of the treatment goals. The 2% and 3% additions also reached the goals (*16*).

cause a decrease in total and PAH-degrading microorganisms. This hypothesis is supported by the microbial counts reported in Fig. 7.

These difficulties may be alleviated by a more aggressive lime-addition procedure to rapidly return the soil to neutrality and/or bioaugmentation with competent microbial cultures. The environmental conditions may also limit the rates of removal. First, the average temperature was much milder in 1992. Second, the rainfall during this test period has been greater than the same period during the 1991 field test (Table 1). Saturated soil becomes rapidly anaerobic, which would decrease or arrest the biodegradation activities of the microbial community. These difficulties may be controlled by covering the plots during inclement weather or tilling after the soil drains to increase air concentration in the soil column.

Table 5 summarizes the preliminary economic evaluations for this site (*16*). Results show that this site can be cleaned at $60/cu yd or less when using the landfarming mode of soil treatment using the CBT process. Process optimization results as well as the initial PAH concentration and final treatment goals would affect the soil remediation costs for other sites.

SUMMARY

The manufactured gas plant sites are nonhomogeneous in soil and waste types and concentrations of contaminants (*6*). PAH-contaminated soils represent the largest fraction of the contamination at the MGP sites (*6*). Studies conducted by IGT show that:

- Bioremediation is effective in removing PAHs from MGP soils.
- Integrated chemical/biological treatment improves the rate as well as the extent of PAH removal.
- MGP soils can be effectively cleaned in the landfarming mode of the CBT process.

- Soils with high sand content are easier to clean. Also, soil-slurry reactors are an effective and efficient system for contaminated soil bioremediation.
- Integrated chemical/biological treatment is also effective for soils with high silts and clay contents.

Finally, IGT also continues to develop, optimize, and demonstrate innovative and accelerated contaminated-soil treatment technologies. A field evaluation of the integrated treatment in the soil-slurry treatment system is being planned.

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