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Project Summary

Mobility and Degradation of Residues at Hazardous Waste Land Treatment Sites at Closure

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The objectives of this study were to determine: (a) the extent to which organics remaining at hazardous waste land treatment (HWLT) sites at closure continue to degrade, (b) the migration potential of the accumulated waste residue constituents, and (c) whether available models (RITZ and MINTEQ) are appropriate for closure decisions. This was achieved by: (a) obtaining representative soil samples from the treatment zone of several HWLT sites, (b) characterizing the residual material in such samples, (c) conducting laboratory studies to determine the degradation and potential mobility of the constituents in the samples, and (d) using RITZ and MINTEQ to evaluate the migration potential of the residual constituents under different closure options. Soils used were obtained from three sites to which listed refinery or wood preserving wastes had been applied and from a site which had soil contaminated with coal tar.

In the surface soils of the HWLT sites, the concentrations of the organics and metals were greater than in background soils. At depths below 24-30" at the HWLT sites, the concentrations of PAH compounds were below detection limits and the freon-extractable concentrations were at background soil concentrations or below detection limits. At sites for which site soil concentrations could be compared to concentrations in background soils, the concentrations of chromium and nickel in site soils at depths below 24" appeared greater than background soil concentrations, whereas the concentrations of copper, lead and zinc in site soils at such depths appeared comparable to those in background soils.

Analyses for organics in TCLP extracts of the soils indicated that no PAH compounds were extracted by this procedure from either weathered or non-weathered soils. Zinc was found above detection limits in the TCLP extracts of the site soils most frequently. In most of the site soil samples analyzed, the concentrations of metals in the TCLP extracts were close to or below detection limits of 0.1 mg/L.

The PAH and metal concentrations measured in the TCLP extracts from the site soils provided no reason to conclude that these soils and accumulated residues would fail to pass a toxicity characteristic determination using the TCLP test.

The aerobic and anaerobic microcosm studies demonstrated that there was no statistically significant degradation of the PAH and freon extractables in the zone of incorporation (ZOI) samples over an eight-month period. The data suggest slow degradation of the accumulated organics following closure of an HWLT site.

MINTEQ is a predictive model that estimates aqueous equilibrium solubility concentrations for metals. However, gaps in the MINTEQ database do not allow it to identify the relative solubility of many of the metals of interest at HWLT sites.

Thus, MINTEQ appears of limited value for identifying the environmental fate of metals under different closure options.

When used with worst-case data for a petroleum refinery HWLT site, the modified RITZ model indicated major differences in mobility and persistence of PAH compounds between capped and uncapped closure scenarios. The closure options in which the sites were uncapped allowed for more rapid degradation, but movement of the constituents below the ZOI was greater. For the clay or synthetic cap option, the degradation of the organic constituents was much slower, but the movement below the ZOI diminished. This simulation indicated that the modified RITZ model can be a useful tool for assessing the impact of different closure options on accumulated residuals at HWLT sites.

This Project Summary was developed by EPA's Robert S. Kerr Environmental Research Laboratory, Ada, OK, to announce key findings of the research project that is fully documented in a separate report of the same title (see Project Report ordering information at back).

Introduction

Land treatment has been used to treat municipal wastes for centuries. In addition, over the past thirty years there has been increased use of land treatment to treat industrial wastes, such as wastes from the petroleum refining industry. With the passage of the 1984 Hazardous Solid Wastes Amendments (HSWA) to the Resource Conservation and Recovery Act (RCRA), many changes have occurred in the regulatory framework with regard to the use of land-based technologies for disposal of hazardous wastes. Due to the restrictions imposed by these regulations, a number of hazardous waste land treatment (HWLT) sites are, or soon will be, undergoing closure. The organic and inorganic waste constituents that have accumulated at these sites can be of concern due to: (a) potential mobility during the post-closure period, which can be as long as 30 years, and (b) the human and environmental exposure that might occur when the site is used after the post-closure period.

There are several major options (Table 1) for the closure of a land treatment site or a site with contaminated soils, all of which involve the careful management of the site over the post-closure period, as defined by the regulations. Both direct and indirect impacts on human health and the environment are important in the selection of the site-specific appropriate closure options.

The purpose of this study was to develop information pertaining to: (a) the characteristics of the waste constituents remaining at sites having long-term waste application, such as HWLT sites, and (b) the potential for subsequent degradation and/or migration of the constituents at such sites. Such information can help regulatory agencies and industry identify the most environmentally sound closure and post-closure conditions for a specific site and can help identify additional research needs related to the closure of HWLT sites.

Soil samples from four field sites were obtained for project use. The sites were chosen based on the following criteria: (a) waste had been applied at the site for an extended period of time, preferably at least 10 years, or the waste treatment or disposal site had been closed (unused) for an extended period of time, (b) site history and background information were available, (c) the applied waste was representative of listed refinery wastes (K-048 to K-052) or of listed wood-preserving wastes (K-001), and (d) the site was available and accessible for sampling. Four sites that most closely met the criteria were chosen.

The soils evaluated were obtained fro two operating HWLT sites, one close land treatment site and a site which ha soil contaminated with coal tar. The soi were obtained from: (a) the Conoc Ponca City, Oklahoma, closed oil refine research land treatment site, (b) th Texaco, Anacortes, Washington, HWL site which had been in operation sinc 1958; (c) an HWLT facility at a woo treating site (WTS) in Montana; and (d) Niagara Mohawk Power Corporatio (NMPC) site in New York. The NMPC sit was not an HWLT site and the result from samples obtained at this site shoul not be construed to represent condition at a managed HWLT site.

Two types of samples were obtaine from the Texaco and the Conoco site (a) surface zone of incorporation (ZO samples for a study to evaluate th spatial distribution of residu characteristics at such sites and (b) so core samples taken as a function c depth to determine whether the waste which had been applied to the surfac soils had migrated. Only core sample were taken from the other two site Although different materials have bee applied at these sites, similar constituent were in the site soils -- metals, long-chai hydrocarbons, and polynuclear aromati hydrocarbons (PAH).

Site Soil-Residue Characteristics

Results from the Texaco site indicate that volatile and acid extractabl compounds were not found in the HWL soils but that PAH compounds were detected in concentrations considerabl above background. The soils from all fou sites were evaluated for sixteen specifi PAH compounds in the laborator studies.

The Texaco site had detectable concentrations of most of the 16 PAH compounds in the top (0-6" and 12-24" soils of the site. The average concentrations of these compounds

Table 1. *Options for the Closure of an HWLT Site*

- Removal of the contaminated soil and subsequent disposal of such soil as a hazardous waste.
- Placement of an impermeable cap over the site. The cap may consist of synthetic liners or clay.
- The site is left alone with the only continued management being ground-water monitoring as well as run-on and runoff control and a vegetative cover.
- Continued managed in-situ treatment of the accumulated waste residues.
- A combination of two or more of the above.

found at increasing depths from the current surface of the site are presented in Table 2. The long-term application of wastes and residues at this site has raised the topographic surface of the soil 18-42 inches. At the plots sampled, the surface was increased about 30 inches. The absence of detectable concentrations of PAH compounds in the 36-54" layer indicated that these compounds had not migrated to this depth.

Freon extractable determinations also were done for the Anacortes samples. This analysis is a relative measure of the total oil and grease (O/G) residuals in a sample. The O/G concentration in the 0-6" samples was 61,700 mg/kg and was 56,600 mg/kg in the 6-12" sample. The O/G concentrations in soil samples taken below 24" were: 24"-36" -- less than 1000 mg/kg; 36"-54" -- 1680 mg/kg; and 54"-72" -- less than 1000 mg/kg. These concentrations below 24" were about the same as those in the site background soils.

The concentrations of PAH at the Conoco site were similar in type but much lower in concentration than those found at the Anacortes site. There has been no application of wastes at this site since 1978.

The PAH concentrations in the soil cores from the plot receiving the largest application at this site were determined. Below the ZOI, the PAH concentrations were close to or below analytical detection limits. The data indicate that there has been no migration of the PAH from the surface soils to lower soils.

The wood-treating site soils had detectable concentrations of all 16 PAH compounds except acenaphthalene and acenaphthene in the top layer (0-6") of the soil. The concentrations of PAH below the 0-6" layer were very low, indicating that little or no migration had occurred below this depth.

Sample Variability Studies

Separate surface soil samples were taken from the Anacortes and the Conoco sites to determine the variability in characteristics that may exist in the surface soils at a HWLT site. Knowledge of this variability is important to monitoring decisions, to closure conditions and to an understanding of whether the core samples taken from a site were representative of the characteristics throughout the site. The ZOI samples were analyzed for organic matter, dry weight, freon extractables, and three metals (copper, chromium and iron). These parameters represent compounds that are part of soil and of the wastes that were applied and accumulated in the surface soils. The data from these analyses were compared using standard statistical tests to determine whether there was a difference in chemical characteristics between the sampling locations.

The statistical analyses indicated that within the Texaco and Conoco plots, consistent differences in constituent concentrations at each site were not observed. The analyses also indicated that the residues that existed at the time of sampling appeared to be uniformly distributed at the site.

Based upon a comparison of core and surface soil samples at the Conoco and Texaco sites, it appeared that the core samples were representative of the soils at these sites.

Mobility and Weathering Studies

These laboratory studies identified the migration potential of constituents in the site soils. The effects of simulated weathering cycles on the quality of potential leachate were determined using the toxicity characteristic leaching procedure (TCLP). The primary variables were: (a) soil samples from six depth intervals at each site, starting at the surface and proceeding to six feet below the soil surface; (b) two weathering patterns, Freeze/Thaw and Wetting/Drying cycles, and (c) analyses of TCLP extracts from both weathered and non-weathered samples of soil.

The methods used for the simulated weathering cycles were adapted from ASTM methods for tests of soil-cement mixtures. These procedures were used with the soil samples in their as-received state, without the addition of cement.

During the gas chromatographic analysis of the approximately 220 extracts analyzed for organics, eight samples had some measurable peaks that could be quantified. These samples were analyzed on a mass spectrometer and the peaks identified by a library search of available chemicals. The results indicated that six of the samples contained phthalates, a common laboratory contaminant, probably from plastic containers used during the methylene chloride extraction procedure. The two remaining samples had peaks which were identified as long-chain hydrocarbon compounds.

The TCLP extract concentrations for metals in soils at the Conoco site as a function of depth were as indicated in Table 3. These are illustrative of the results obtained when the soils from the other sites were evaluated. The pattern was one of low concentrations, generally about 0.1 mg/L, in the extract from the surface soils and below detection limits in extracts from the deeper soils. The one exception was zinc, which in some extracts was above a concentration of 1.0 mg/L.

The results of these laboratory studies can be summarized as follows:

a) Analyses for organics in the TCLP extracts indicated that no PAH compounds were extracted by this procedure from either the weathered or non-weathered soils.

b) There was no difference in the concentration of metals in the TCLP extract of the non-weathered and the weathered samples.

c) The PAH and metal concentrations measured in the TCLP extracts from the soil/residues from these four sites provided no reason to conclude that the soils and accumulated residues would fail to pass a toxicity characteristic determination using the TCLP test and analysis of the TCLP extract.

Degradation Studies

These laboratory studies were undertaken to identify the extent to which organics remaining at closure of a HWLT or similar site will continue to degrade under closure and post-closure conditions. An impermeable cap will reduce infiltration and leaching but also will restrict oxygen transfer from the atmosphere. Anoxic or anaerobic conditions under this type of cover could occur.

Because of the expected soil conditions with these closure options, degradation under both aerobic and anaerobic conditions was evaluated and microcosm studies were conducted over an eight-month period. Only samples from the Anacortes ZOI were used. Such samples represent the accumulation of residues over 30 years. The term degradation is used to describe the results, because it was expected that microbial degradation would be the major organic compound loss mechanism. However, the actual loss mechanisms were not investigated.

Three separate composites of four different Texaco ZOI samples were prepared for two aerobic and one anaerobic degradation microcosm studies. No uncontaminated soil was added to these samples. The two aerobic studies were: (a) the ZOI soils without amendments or enhancements, and (b)

Table 2. *PAH Concentrations* at the Texaco Site as a Function of Depth*

Compound	Sample Depth** 0-6" Ave.	0-6" Range	12-24" Ave.	12-24" Range	24-36" Ave.	24-36" Range	36-54" Ave.	36-54" Range	Detection Limit +
naphthalene	1.8	1-2	2.8	2-5	1.5	<1-2.6	2.0	1.9-2.1	1.0
acenaphthalene	BDL	—	BDL	—	BDL	—	BDL	—	2.0
acenaphthene	BDL	—	BDL	—	BDL	—	BDL	—	1.0
fluorene	6.8	4-9	9.5	6-12	0.7	<0.2-2	BDL	—	0.2
phenanthrene	5.9	5-8	25	3-39	6.7	1-14	1.4	<0.1-3	0.1
anthracene	BDL	—	4.9	2-6	BDL	—	BDL	—	0.1
fluoranthene	30	26-38	75	19-100	BDL	—	BDL	—	0.2
pyrene	21	16-29	300	37-432	BDL	—	4.0	<0.1-7	0.1
benzo(a)anthracene	BDL	—	BDL	—	7.9	<0.1-20	BDL	—	0.1
chrysene	100	48-155	116	60-146	4.2	2-9	0.9	<0.1-2	0.1
benzo(b)fluoranthene	130	50-195	155	31-220	BDL	—	BDL	—	0.2
benzo(k)fluoranthene	BDL	—	BDL	—	BDL	—	BDL	—	0.1
benzo(a)pyrene	204	47-350	106	62-136	1.5	<0.1-3	BDL	—	0.1
dibenzo(a,h)anthracene	340	104-600	194	47-300	4.1	0.2-10	0.8	<0.2-2	0.2
benzo(g,h,i)perylene	82	25-160	51	8-85	BDL	—	BDL	—	0.2
indeno(1,2,3-c,d)pyrene	38	14-85	24	6-40	BDL	—	BDL	—	0.1

* *mg/kg dry weight of soil; average of three samples*
BDL below detection limits
** *samples from the 6" to 12" depth were not analyzed*
\+ *detection limit of the analytical procedures used*

Table 3. *Average Metal Concentrations + in the TCLP Extracts from the Conoco, 10% Plot Site - Non-Weathered Samples*

Depth	Cadmium	Chromium	Copper	Lead	Nickel	Zinc
0-6"	<0.1	0.2	0.1	<0.3	<0.1	2.1
6-12"	<0.1	0.2	<0.1	<0.1	<0.1	0.6
12-24"	<0.1	0.2	0.1	<0.1	<0.1	<0.1
24-36"	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1
36-54"	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1
54-72"	<0.1	<0.1	<0.1	<0.1	<0.1	<0.1

\+ *mg/L in extract; detection limit was 0.1 mg/L*

the ZOI soils with additional nitrogen and phosphorus. The nutrients were added to determine whether the addition of nutrients to HWLT soils would increase the loss of organics in these soils.

The results of these degradation studies indicated that there was no statistically significant degradation of PAH and freon extractables in the ZOI soils from the Texaco site over an eight-month period. It was in these soils that the residues from the applied wastes accumulated. The data suggest that there will be very slow degradation of the accumulated organics at closed HWLT sites.

Predictive Models for Closure

This modeling effort was conducted simultaneously with the sample characterization, spatial variability, weathering and degradation studies that were part of the project. Ideally, the evaluation and use of the predictive models should have occurred after data were available from the various laboratory studies. Such data could have been used with the models for a more focused evaluation of the models and more direct comparison of site data to model results.

This could not be done in this project because: (a) the respective models had to be evaluated and modified for use under closure and post-closure conditions, since the models originall had not been developed for that purpos (b) the modified models had to b checked to assure that results reasonabl for closure conditions were bein obtained, and (c) some of the laborato results, particularly from the degradatic studies, were not available until very la in the project.

Thus, the input parameters to th models used in this evaluation were n those determined in the laborator portion of this project. Rather, the inp parameters and site-specific scenaric used were those that were reasonable f petroleum refining and wood preservin wastes and for the land treatment of suc

wastes. The values for such parameters were obtained from peer reviewed literature and from studies in which land treatment had been used for similar wastes.

The Regulatory Investigative Treatment Zone (RITZ) model was used to evaluate the persistence and mobility of specific organic constituents. The MINTEQ model was used to evaluate the persistence and mobility of metals. The initial version of RITZ was obtained from the Robert S. Kerr Environmental Research Laboratory (RSKERL), U.S. EPA, Ada, Oklahoma 74820, and the initial version of MINTEQ was obtained from the Center for Water Quality Modeling, Environmental Research Laboratory, U.S. EPA, Athens, Georgia 30613.

The RITZ model was modified to: (a) function with no waste input, (b) consider variations in degradation rate as a function of time, soil depth and aerobic and anaerobic conditions, (c) include the existence of a separate oil phase and the separate degradation of the oil phase, and (d) utilize different chemical partition coefficients in the ZOI and LTZ. The modified model was then used to evaluate its use under conditions likely to occur at HWLT and contaminated soil sites. The sensitivity of MINTEQ was examined for the three most important parameters that are influenced by the closure option, pH, the partial pressure of carbon dioxide gas, and the redox state.

The modified versions of RITZ and MINTEQ were used to evaluate HWLT closure options. The modified models were applied to conditions at the Texaco, Anacortes refinery waste land treatment site. Considerable data for this site were available to understand and evaluate the applicability of the models and to model potential migration of constituents of the accumulated residues. In addition, conditions typical of other regions of the country also were investigated.

When used with worst-case Texaco Anacortes HWLT site data at three different hypothetical locations, the modified RITZ indicated major differences between the capped and uncapped scenarios. The closure options in which the sites were uncapped allowed for aerobic conditions, but the movement of the constituents into the LTZ was greater. For the clay or synthetic cap option, reduced conditions should occur and the movement into the LTZ was greatly diminished, due to the lack of infiltration created by the cap.

At the uncapped site, the persistence, i.e., potential residence times, of organics was estimated to be approximately 20 years for benzo(a)anthracene, and 1100 days for chrysene and pyrene. For the capped site, 8 of the 12 PAH compounds examined remained in the site for 1100 days or more, and 6 of the 12 remained in the site for 15 years or more. The sensitivity of the modified RITZ model to the differences between the various uncapped closure options was minimal, since the bulk of the degradation took place in the ZOI, and the depth constant did not alter these results. These differences are portrayed in Table 4.

The results obtained on the mobility and persistence of organics at a HWLT site under closure scenarios indicated that the modified RITZ can be a useful tool for assessing the impact of different closure options on the migration potential and persistence of residual organics at such sites.

MINTEQ is able to estimate aqueous equilibrium solubility concentrations for metals but is unable to estimate the actual mobility of metals. Gaps in the MINTEQ database do not allow it to identify the relative solubility of many of the metals of interest at HWLT sites. This evaluation indicated that MINTEQ is useful to demonstrate the potential mobility of metals as indicated by the relative solubility of metal species and to determine those most likely to migrate.

The results obtained as part of the project provided insight into the environmental effect of various closure options. The most appropriate option appears to be one in which the site is left open to the atmosphere. The option of continued treatment is beneficial because the degradation rate of the organics will be most rapid, and because the conditions used during the active site life to prevent metal migration will be continued.

This was illustrated when the Texaco Anacortes site was modeled. For the organics, no leaching was predicted for the uncapped site, and for the metals, there was no significant increase in metal solubility under oxidizing conditions. The results for the capped site indicated that the residual organics persisted for a long time.

Two important results were generated from this evaluation. One is that the potential persistence and mobility of the accumulated residue constituents are affected by the closure option chosen. Another is that RITZ and MINTEQ, when modified to represent closure conditions, can estimate the potential mobility and persistence of metals and organics for different closure options. The actual choice of a closure option will depend on such estimates as well as on operational factors and regulatory concerns.

Conclusions

The major conclusions from this project were:

1. Polynuclear Aromatic Hydrocarbons (PAH), freon extractables, and metals were determined in the site soil samples. Except for the coal tar disposal site, these chemicals accumulated in the surface soils. The difference in the coal tar site data is due to the fact that the applied residues were placed in a trench rather than being incorporated in surface soils.
2. In the surface soils of the HWLT sites, the concentration of the organics and metals were greater than in background soils. At depths below 24-30" at the HWLT sites, the concentrations of PAH compounds were below detection limits and the freon extractable concentrations were at background soil concentrations or below detection limits. At the sites for which soil concentrations could be compared to concentrations in background soils, the concentrations of chromium and nickel in site soils at depths below 24" appeared greater than background soil concentrations, whereas the concentrations of copper, lead and zinc in site soils at such depths appeared comparable to those in background soils.
3. A statistical analysis of sample variability was conducted using surface soils collected at the two land treatment sites. There were no consistent differences in chemical concentrations of the surface soils. The analysis also indicated that the residues that existed at the time of sampling appeared to be uniformly distributed in the surface soils of each site.
4. Analyses for organics in TCLP extracts of site soils indicated that no PAH compounds were extracted by this procedure from either weathered or non-weathered soils.
5. There was no difference in the concentration of metals in the TCLP extract of the weathered and the non-weathered soil samples.
6. Zinc was found above detection limits in the TCLP extracts of the site soils most frequently. In most of the site soil samples analyzed, the concentrations of other metals in the TCLP extracts were close to or below detection limits of 0.1 mg/L.

Table 4. *Estimated Persistence and Mobility of Organics Present at the Texaco, Anacortes Site as Influenced by the Closure Option*

Organic Constituent	Leave-Alone		Clay/Synthetic Cap	
	% Leach from ZOI	Time in ZOI (days)	% Leach from ZOI	Time in ZOI (days)
benzene	3.7	32	0 1	100
ethylbenzene	1.1	71	0.0	200
toluene	3.2	55	0.0	200
o-xylene	6.6	146	7.2	> 15 years
m, p-xylene	0.7	610	0.0	720
anthracene	0.3	720	0.0	15 years
benzo(a)anthracene	1.4	> 20 years	0 1	> 30 years
chrysene	0.6	1100	0.0	30 years
1-methylnaphthalene	0.0	240	0.0	1100
naphthalene	5.4	450	0.1	1100
phenanthrene	1.1	720	0 1	20 years
pyrene	1 0	1100	0.1	20 years

7. The results suggest that the type of weathering used in this study (freeze/thaw and wet/dry) is not expected to result in an increase in organic or metal mobility in soils at HWLT sites.
8. The PAH and metal concentrations measured in the TCLP extracts from the site soils provided no reason to conclude that these soils and accumulated residues would fail to pass a toxicity characteristic determination using the TCLP test.
9. The aerobic and anaerobic microcosm studies demonstrated that there was no statistically significant degradation of the PAH and freon extractables in the ZOI samples over an eight-month period. The data suggest, therefore, that there will be slow degradation of the accumulated organics at closed HWLT sites.
10. The RITZ model, as modified for HWLT site closure conditions, indicated major differences in mobility and persistence of PAH compounds between capped and uncapped closure scenarios. The closure options in which the sites were uncapped allowed for more rapid degradation, but movement of the constituents below the ZOI was greater. For the clay or synthetic cap option, the degradation of the organic constituents was much slower, but the movement below the ZOI diminished.
11. The sensitivity of MINTEQ was examined for parameters that are influenced by the closure option: pH, the partial pressure of carbon dioxide and the redox state. It was difficult to draw general conclusions about the effects of these changes, since different metals behav differently in solution. In genera most of the metals present solution as cations became les soluble as the pH increased, and the partial pressure of carbon dioxi increased. No general statemen could be made about redo conditions, as the most soluble redo state of a given metal varies fro metal to metal.
12. Based on the low migration potenti of PAH compounds, freo extractables and metals in the so at HWLT sites determined fro laboratory and model simulatio studies, it does not appear necessa to cap such sites at closure. Both th leave-alone option, or the option continue active site managemer appear to be protective of huma health and the environment.

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The complete report, entitled "Mobility and Degradation of Residues at Hazardous Waste Land Treatment Sites at Closure," (Order No. PB 90-212-564AS; Cost: $23.00, subject to change) will be available only from:
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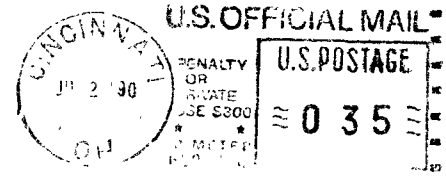

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