COST AND PERFORMANCE REPORT

Phytoremediation at the Argonne National Laboratory — West
Waste Area Group 9, Operable Unit 9-04
Idaho Falls, Idaho

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Background Information

The Idaho National Engineering and Environmental Laboratory (INEEL), located in Idaho Falls, Idaho, is a government facility managed by the U.S. DOE. The ANL-W is located in the southeastern portion of the INEEL. The INEEL is divided into ten Waste Area Groups (WAGs), and each is further divided into Operable Units (OUs) which focus on specific areas of interest. Various sites at ANL-W are contaminated with wastes generated from the scientific and engineering research at ANL-W and contain various levels of petroleum products, acids, bases, PCBs, radionuclides, and heavy metals.

In November 1989, the Environmental Protection Agency issued a final ruling that listed the INEEL as a National Priorities List (NPL) site. The Record of Decision (ROD) for WAG 9 was signed on September 29, 1998, and identifies seven areas that will undergo remediation to levels established in the ROD. To meet the remediation goals, DOE has selected phytoremediation as the remedy, with a contingent remedy of excavation and disposal.

In support of the ROD, and as a pre-condition for implementing phytoremediation in these areas, bench scale tests were performed. The bench-scale tests were used to evaluate the applicability of phytoremediation as well as to determine operating parameters and time frames for full-scale implementation. This case study discusses both the bench scale experiments and the remedial designs for full-scale implementation. The bench scale tests conducted during 1998 and the remediation plan for several sites at the ANL-W are discussed below.

The ANL-W is a relatively flat, semi-arid, sagebrush desert. The general climate characteristics and soil type is shown in Table 1. Bench scale experiments were conducted using soil from the Main Cooling Tower Blowdown Ditch. The soil was contaminated with mercury, zinc, and chromium (Table 1).

Table 1. Characteristics of Climate and Soil Matrix at the Main Cooling Tower Blowdown Ditch Soil

Parameter	**Value**
Climate conditions	• Temperature range: 7.9°F - 84.8°F • Growing season: April – mid-October • Annual average precipitation: 8.7 inches
Texture	Loam
Particle size distribution	47% sand, 34.6% silt, 18.4% clay
pH	8.57
Fertility	1.59% organic matter, 5.41% lime, 5310 mg/kg extractable Ca, 510 mg/kg extractable Mg, 76 mg/kg extractable Na, 438 mg/kg extractable K, 48 mg/kg extractable P, 71 mg/kg soluble SO_4, 76 mg/kg soluble Na
Metal contaminants	<1.5 mg/kg total Hg, 56.32 mg/kg total Zn, 44.85 mg/kg total Cr

Source: Negri et al., 1998

Bench Scale Technology Description and System Design

Prior to acceptance of phytoremediation as a remedial alternative at WAG 9, bench scale tests (laboratory and greenhouse) were performed. The goals of the bench scale testing were to: (1) determine uptake rates and metal concentration factors for each plant species; (2) determine the most effective, non-hazardous chelating agent to increase the availability of the metals from the impacted soils; and (3) evaluate potential maximum uptake of metals by candidate plant species under conditions of total bioavailability (sand experiments) compared to uptake from site soils.

Laboratory tests were conducted to identify effective chelating or extracting agents to make the metals in the soil more soluble and bioavailable. Options evaluated included the addition of EDTA and citric acid. Based on the agronomic and leaching testing results, a combination of both EDTA and citric acid was selected for testing in the greenhouse experiment to enhance metal availability to plants.

Based on literature findings, site-specific data for the ANL-W site, and agronomic testing, three candidate plant species were selected from plants growing naturally or planted as crops suitable for growing at ANL-W. The candidate species were: Prairie Cascade hybrid willow (*Salix x*), a fast growing phreatophytic tree; canola (*Brassica napus*) a sterile rapseed plant; and kochia (*Kochia scoparia*) an introduced weedy plant that grows naturally in the area. Contaminants of concern included chromium, zinc, mercury, silver, and selenium.

The greenhouse evaluation included two experiments: (1) the three selected plants were grown in soil taken from the Main Cooling Tower Blowdown Ditch, and (2) the three selected plants were also grown in clean sand. The sand was spiked with a solution containing soluble forms of Cr, Zn, Hg, Ag, and Se. The sand experiment was performed to determine the maximum potential uptake capabilities of the test species.

For the soil experiment, the species were planted in May 1998, the soil was spiked with EDTA and citric acid (chelants) in August 1998, and the plants were harvested in October. For the sand experiment, the species were planted in May 1998, the soil was spiked with metals in August 1998, and the plants were harvested in October. (Negri et al., 1998)

Bench Scale Technology Performance

Results from the bench scale tests are summarized below:

1. The optimum formulation of chelating agents for the solubilization of the contaminants of concern was determined to be a 0.05 molar solution of 40% EDTA and 60% citric acid. This formulation should not expose the plants to excessive EDTA toxicity.

2. In the sand experiment, the best recovery levels for zinc, chromium, mercury, and silver were found in the willow with 96%, 38%, 42%, and 24% recovery, respectively. The zinc was found in the root and the tops while the chromium, mercury, and silver was almost exclusively found in the root.

3. Testing using actual soils yielded significantly lower removals than with the sand experiment. The amount of zinc and chromium removed was 4-5% and 2%, respectively. The willow roots had better removal of the metals than either kochia or canola.

Based on the bench scale studies, it was concluded that willows would be used in field because they provided the greatest removal rate, and the roots would be harvested. Possible removal rates of up to 14% of Zn and 3 - 4% Cr per year were predicted, which could result in cleanup times between 6 - 7 years for Zn and 9 years for Cr. (Negri, et al., 1998)

Summary of Observations and Lessons Learned From Bench Scale Tests

Since the results of the field implementation are not complete, lessons learned are derived from the bench-scale tests.

- Metal uptake is highly dependent on the metals' solubility and bioavailability. Metal uptake was considerably higher in the sand experiments as compared to the ANL-W soil studies.
- Partitioning of the metals into plant organs (tops and roots) was similar for all of the plants, with considerably more metal remaining in the roots than was translocated to the tops.
- Root harvesting is an important component of plant-based cleanup systems and can be used to maximize contaminant removal from the soil. Willows provide the best solution because of both their higher total removal rates, and by the fact that their root system makes them most suitable for root harvesting.

Remediation Plan

There are seven areas that are targeted to undergo remedial activities in accordance with the WAG 9 ROD. Two of these areas will not undergo remedial activities until their current use is concluded (i.e. Industrial Waste Pond - ANL-01, and Sewage Lagoons - ANL-04). The five areas that will be treated by the selected remedy, i.e., phytoremediation, include the Ditch A (ANL-01), Ditch B (ANL-01, Main Cooling Tower Blowdown Ditch (ANL-01A), Interceptor Canal (ANL-09), and the Industrial Waste Lift Station Discharge Ditch (ANL-35). At these areas, phytoextraction will be conducted *in situ* to remove the metals and the radionuclides from the soils via normal uptake mechanisms of the selected plants. Table 2 identifies the contaminants requiring remediation at each of the seven areas, the contaminant concentration (95% upper confidence intervals), and the contaminant remediation goal.

Based on the best uptake removal rates of the plant species evaluated, ANL-W calculated the number of years of phytoremediation that would be required to meet the remediation goals for several site areas (Table 3). Those number of years required is estimated to be worst case and does not include redistribution of nonhomogeneous contaminants into a more homogeneous matrix. These redistribution effects will reduce the years to reach the remediation goals. Based on the results from this study, and a

Table 4-11. Remediation Goals for Areas at ANL-W

Area	Contaminant	95% UCL Concentration[a]	Remediation Goal Concentration[a]
Industrial Waste Pond (ANL-01)	Cesium-137	29.2	23.3
	Chromium III	1,030	500
	Mercury	2.62	0.74
	Selenium	8.41	3.4
	Zinc	5,012	2,200
Ditch A (ANL-01)	Mercury	3.94	0.74
Ditch B (ANL-01)	Chromium III	1,306	500
	Zinc	3,020	2,200
Main Cooling Tower Blowdown Ditch (ANL-01A)	Chromium III	709	500
	Mercury	8.83	0.74
Interceptor Canal- Mound (ANL-09)	Cesium-137	30.53	23.3
Sewage Lagoons (ANL-04)	Mercury	3.2	0.74
Industrial Lift Station Discharge Ditch (ANL-35)	Silver	352	112

a. Concentration in mg/kg for metals, pCi/g for Cesium-137
Source: Negri et al., 1998

Table 3. Number of Years of Phytoremediation Required to Meet Remediation Goals[a]

Area	Contaminant	Plant Uptake %	Phytoremediation Years Needed
Industrial Waste Pond (ANL-01)	Chromium III	2	36
	Mercury	2	63
	Selenium	4.5	20
	Zinc	4.5	28
Ditch A (ANL-01)	Mercury	2	82
Ditch B (ANL-01)	Chromium III	2	47
	Zinc	4.5	7
Interceptor Canal-Mound (ANL-09)	Cesium-137	4.5	6
Main Cooling Tower Blowdown Ditch (ANL-01A)	Chromium III	2	17
	Mercury	2	122
Sewage Lagoons (ANL-04)	Mercury	2	72
Industrial Lift Station Discharge Ditch (ANL-35)	Silver	2	56

a. Refer to Table 4-11 for Remediation Goals.

similar companion study for Cs-137, the following sites were selected as candidates for phytoremediation; Ditch A (ANL-01), Main Cooling Tower Blowdown Ditch (ANL-01A), Interceptor Canal-Mound (ANL-09), and the Industrial Lift Station Discharge Ditch (ANL-35).

Each of the five sites (Ditch A, Ditch B, ANL-09, MCTBD, and ANL-35) will be treated using phytoremediation during a two-year field test. Each site will be planted with three-foot tall bare-root willow trees in a grid pattern. The bare-root willow will be spaced approximately eighteen inches on center to optimize the biomass of the plant at the end of the field season. Supplemental irrigation will be installed to keep the soil moisture between 40 to 50 percent. Soil moisture detectors will be installed with the ability to turn off the irrigation system if the soil moisture exceeds the optimal range. Two moisture detectors will be stacked vertically at depths of 0.5 and 1.5 feet below land surface. The automatic watering system will be installed at the 1.0-foot depth and will "train" the willow plant roots to stay within the area of contamination. The lower moisture detector (1.5-foot depth) will be used to show that no increase in soil moisture has occurred beyond the plant roots. The irrigation system will also be used to distribute fertilizers and amendments (EDTA and citric acid). The use and control of the chemical injection system will only be operated after the root zone fully covers the contaminated area.

Whole tree harvesting (roots and above ground) will occur at the end of each growing season. To minimize the carryover of contaminated soil during root harvesting the irrigation will be shut off until soil moisture reaches approximately 30%. Harvesting of the whole willows will be performed using a specially designed implement mounted on a front-end loader. The front-end loader will be attached to a tractor. The tractor will drive down the ditch and as the trunks of the trees get wedged into the attachment the loader can be raised to remove the root from the ground. The excavated trees will be chipped and transported to an on-site incineration facility for disposal.

Remediation Cost

In the Draft Remedial Design for ANL-W (DOE, 1999), the cost for conducting phytoremediation of five sites (Ditch A, Ditch B, Main Cooling Tower Blowdown Ditch, Interceptor Canal-Mound, and the Industrial Waste Lift Station Discharge Ditch) was estimated (Table 4). Construction and ongoing operation of the initial 2-year field test is estimated to be about $300,000. A contingency of $542,000 is included for having to continue phytoremediation for five more years. The largest cost is $780,000 for 20 years of post-remediation operations and maintenance.

Table 4-13. Summary of Cost Estimate for Phytoremediation of Four Sites at ANL-W

Cost Element	Cost (1999 $)
Management	528,000
Documentation (surveying, sampling, design, permits)	98,000
Construction:	
• 2-year field test	299,000
• Additional 5 years of phytoremediation	542,000
Operations and Maintenance (20 years)	780,000
Total	$2,247,000

Source: DOE, 1999

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