

PHYTOSTABILIZATION OF ACID METALLIFEROUS MINE TAILINGS AT THE KEATING SITE IN MONTANA¹

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Abstract. The Keating Tailings site is located in Broadwater County, Montana on land administered by the Bureau of Land Management. These low pH (4 standard units) wastes resulting from historic gold and copper mining operations contain phytotoxic levels of several metals and are generally devoid of vegetation. With an estimated volume of 110,100 m³, these tailings represent unacceptable risk to the environment and human health. The objective of conducting a phytostabilization study at the Keating Tailings Site was to provide BLM managers and decision makers with site specific information and data relating to the implementation, and effectiveness of phytostabilization so that it may be applied to other similar acid metalliferous mine tailings sites administered by the Bureau. To achieve this management objective, replicated experimental plots were implemented using soil amendments, lime and organic matter, designed to ameliorate the plant inhibiting chemical characteristics of the tailings. The plots were seeded with a mix of indigenous native plant species. Vegetation performance of plants grown in the amended or phytostabilized tailings was compared to results for plants seeded into tailings that were not amended, and performance of plants seeded in an adjacent off-site, but non-impacted area. Response variables evaluated in the first growing season, 2004, included emergence and establishment, density, and canopy cover. Concentrations of metals in vegetation were evaluated in terms of plant sufficiency/excess, and in terms of maximum allowable dietary levels for cattle. Changes in soil rootzone pH, conductivity, and soluble metal concentrations before and after treatment were also determined.

Additional Key Words: phytotoxicity, mine wastes, environmental risk, reclamation

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Introduction

Using vegetation to stabilize metal mine wastes, including tailings ponds, mill wastes, and smelter impacted areas has been attempted in many locations around the world including Canada, Australia, United Kingdom, and the US. Both successes and failures have been reported in the literature. Recent reviews of the reclamation of gold heaps and metal mine wastes was provided by Munshower (2000), reclamation of lands disturbed by mining of heavy minerals by Brooks (2000), revegetation of metalliferous tailings by Richmond (2000), and reclamation of smelter-damaged lands by Winterhalder (2000).

The in-place or *in situ* treatment of mine and smelter wastes and contaminated soils in Montana has been the subject of research and demonstration since at least the late 1940s. The Anaconda Company conducted studies (from 1946 to approximately 1957) to reduce dusts from their tailings ponds using a variety of strategies including amendments and vegetation. This reclamation history was reported by RRU (1993). In the 1980s and 1990s, several phytostabilization research investigations, treatability studies, and field demonstration were conducted in Montana at abandoned mines, and at Superfund Sites within the Clark Fork River Basin. Assessments of the permanence of in situ treatment of several of these sites was reported (Munshower et al, 2003). Principles, practices and recommendations for in-place treatment of acid metalliferous mine wastes were identified in a recent report prepared for the US Environmental Protection Agency (Neuman et al. 2005).

Bureau of Land Management (BLM) managers are responsible for the cleanup of abandoned hard rock mines on land administered by their Agency. There are thousands of these sites in the western United States, and they represent a risk to the environment and to human health. The general approach to cleanup is excavating the mine wastes and depositing them in a repository followed by vegetating a cover soil cap. The BLM is investigating alternative strategies including in-place treatment of the mine wastes followed by revegetation. This report details the results of a phytostabilization study of the Keating Tailings which are located southwest of the town of Townsend in Broadwater County, Montana (Fig. 1).

Study Objectives

The objective of conducting a phytostabilization study at the Keating Tailings Site is to provide BLM managers and decision makers with site specific information and data relating to the implementation and effectiveness of this technology so that it may be applied to other similar acid metalliferous mine tailing sites administered by BLM. To achieve this management objective, the project goals were to construct replicated experimental plots using soil amendments designed to ameliorate the plant inhibiting chemical characteristics of the tailings, to seed the experimental plots with appropriate native plants that can thrive in the newly created rootzone, to monitor vegetation response variables (specifically establishment, seedling density, cover, metal levels in the established plants, and above ground biomass), and to determine tailings pH, conductivity, and soluble metal levels before and after treatment. Vegetation performance grown in the amended or phytostabilized tailings is compared to results for plants seeded into tailings that are not amended, and performance of plants seeded in an adjacent off-site, but non-impacted area.

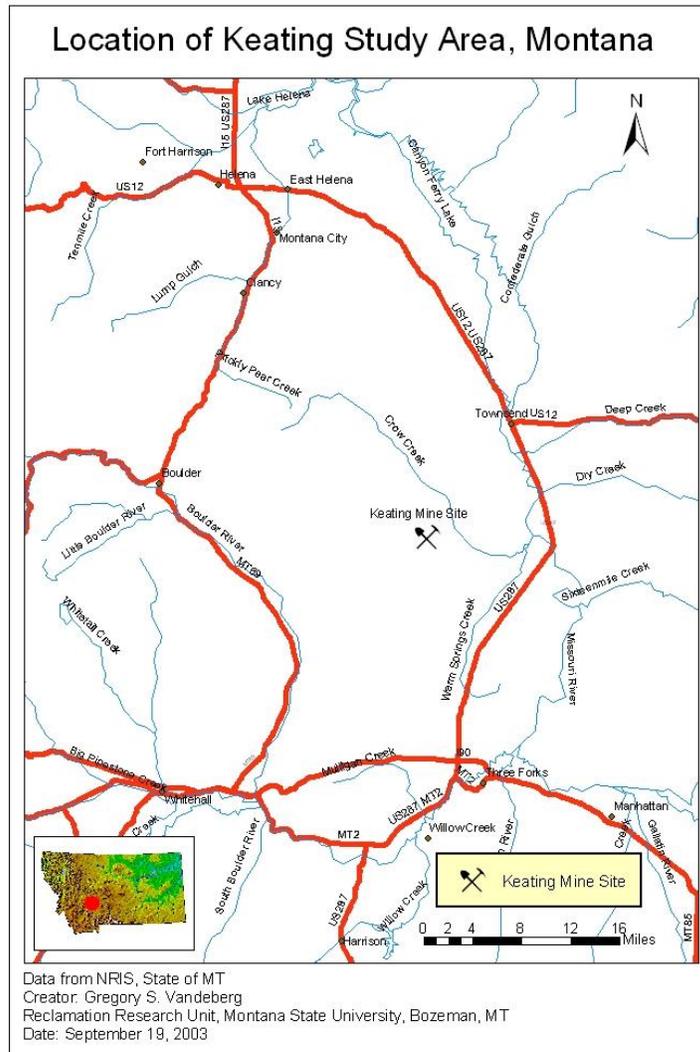


Figure 1. Location of the Keating Tailings site in Montana.

Site History

The Radersburg mining district began as a placer mining operation for gold around 1866, and a 15-stamp mill was installed in 1870. Production continued until about 1948, with ore being shipped to smelters in East Helena, Butte and Anaconda, Montana. The Keating tailings were produced by the mill and earlier mining activity. The site is estimated to contain 110,100 m³ of mine tailings from this gold and copper mining operation.

Site Characterization

Tailings and Soils

Locations for the experimental plots on the tailings and on adjacent, off-site native soils were identified, surveyed and staked. Tailings and soil samples were collected from each of the plots using a Giddings soil probe. Three subsamples from each onsite plot were collected from 0-45 cm depth and composited. Soil samples were also collected from the offsite plots using the Giddings soil probe. The samples were dried and sieved to the ≤ 2 mm fraction, and then characterized using standard methods (Table 1) for several physicochemical parameters.

Some of the chemical attributes of tailings and soils collected from the 12 experimental plots are displayed in Table 2. The pH of the tailings materials was acidic, with a mean value of 4.3. This high acidity allows increased solubility of metals as indicated by the elevated concentrations of water soluble copper (Cu), iron (Fe), manganese (Mn), and zinc (Zn). These soluble metal levels, coupled with the low pH most likely represent phytotoxic conditions (Munshower 1994, Adriano 1986) for all but the most tolerant plant species. The native soils, with a mean pH of 8.5, have very low levels of water soluble metals. Total concentrations of metals and arsenic in the tailings are approximately one order of magnitude greater than the native soils. Organic matter level in the tailings is very low, and they are relatively saline.

Vegetation

A survey of vegetation growing on native soils adjacent to the tailings was made to identify indigenous species so that an appropriate seed mix for use on the plots could be formulated. Wheat grasses (*Agropyron* species) and blue grasses (*Poa* species) dominate the adjacent plant community; forbs such as Big sagebrush (*Artemisia tridentata*) are also common in the uplands, with Rocky Mountain juniper (*Juniperus scopulorum*) found along the upper slopes of drainages. Over fifty species were identified growing on the native range. In contrast, the tailings site was nearly devoid of vegetation (Fig. 2) with a few plants growing along the margins of the tailings impoundment. Wheat grasses and Big sagebrush are common along the tailings site margin, with other grasses and forbs also present.

Methods and Materials

Experimental Design and Amendments

Eight experimental plots on the tailings pond and four plots at an adjacent off-site, native range area were identified and staked. The experimental design consisted of three treatments and four replications:

1. Lime amended profile to 45 cm depth with 2 percent (dry weight basis) organic compost (dairy cattle) added to upper 15 cm of profile;
2. An on-tailings control identical to treatment 1, but no lime or organic matter were incorporated;
3. An off-site control on an adjacent, but undisturbed area. Neither lime nor organic matter was incorporated.

Replicated plots, 3m x 6m in size, were arranged in a random block design with a 3 m buffer strip between each plot. A berm was constructed to prevent run on of surface water from the adjacent tailings. In addition, all plots were fenced to exclude grazing and off-road vehicles.

Table 1. Soil and tailings analytical methods.

CONSTITUENTS		METHOD
Soil Preparation	Separation for analysis of \leq 2mm fraction	ASTM D421-85 (ASTM, 1997)
Percent Rock Fragments	dry sieve analysis of rock fragments ($>$ 2mm) by volume and mass.	ASTM D422-63 ¹ (ASTM, 1997)
Particle Size, Soil Textural Class	Hydrometer method for soil texture, USDA classification	ASA Method 15-5 (ASA, 1986)
Saturation Percent	Saturation percent by weight of water to soil	ASA Method 21-2.2.2 (ASA, 1986)
Sodium Adsorption Ratio	Soil fraction	Method 3.2.19 (Sobek and Others, 1978)
Electrical Conductivity	Saturated Paste Extract	USDA Handbook 60, Method 3a, 4b (U.S. Salinity Lab Staff, 1969)
pH	Saturated Paste Extract	USDA Handbook 60, Method 3a, 21c (U.S. Salinity Lab Staff, 1969)
Total As and metals, Soluble As and metals	As, Cd, Cu, Pb, Hg, Co, Cr, Fe, Mn, Ni and Zn	Standard EPA- CLP methods (SOW 787, U.S. EPA) for soluble metals in saturated paste extracts. Total metals by BLM using XRF methods
K	Fertilizer requirement	Method 13-3.5 (ASA, 1982)
NO ₃ -N, NH ₄ -N	Fertilizer requirement	Method 4500 F, H (APHA, 1989)
P	Fertilizer requirement	Bray -P, Method 24-5.1 (ASA, 1982)
Total Organic Matter	Based on organic carbon	Method 29-3.5.2 (ASA, 1982)

¹ Modifications to ASTM D422-63 include volumetric determination of the percent retained on the No. 10 sieve. The set of sieves specified in ASTM D422-63 reduced to only the No. 10 sieve and any larger mesh sieves necessary for optimum laboratory efficiency.

Table 2. Mean concentrations of arsenic, selected metals, and other parameters from the 0 to 45 cm depth in tailings and native soils.

Parameter	Units	Tailings (N = 8)	Native Soils (N = 4)
Arsenic			
Total	mg/kg	309	31.2
Soluble	mg/kg	0.67	0.76
Copper			
Total	mg/kg	337	45.7
Soluble	mg/kg	44.6	0.07
Iron			
Total	mg/kg	54000	34900
Soluble	mg/kg	0.65	<0.17
Manganese			
Total	mg/kg	1184	1110
Soluble	mg/kg	424	0.22
Lead			
Total	mg/kg	260	29.6
Soluble	mg/kg	0.59	<0.28
Zinc			
Total	mg/kg	1005	110
Soluble	mg/kg	277	<0.03
pH		4.3	8.5
Conductance	mS/m	4.6	0.7
Organic matter	%	0.19	2.09
Texture		silt	sandy clay loam



Figure 2. Keating Tailings site in Southwestern Montana, summer 2003.

Acid-Base accounting of the tailings material was used to determine how much alkaline material was required to neutralize their active and potential acidity. The total lime requirement was determined by the modified Sobek method (Sobek et al., 1978, RRU, 1997) and the SMP buffer method (ASA 1982, Method 12-3). Analytical results were applied to Equation 1 to calculate the total lime requirement.

$$\begin{aligned} \text{Tons CaCO}_3 / 1000 \text{ tons soil} = & (\% \text{ HNO}_3 \text{ extractable S} + \% \text{ Residual S}) 31.25 + \\ & 23.44(\% \text{ HCl extractable S}) + \text{SMP Lime Requirement, tons CaCO}_3 / 1000 \text{ tons soil} \quad (1) \end{aligned}$$

Both Ca(OH)_2 and CaCO_3 were applied according to the application rate based on calcium carbonate equivalence (CCE) (ASA 1965, Method 91-4.2), percent of oversize (> 0.25 mm) particles determined by dry sieving, and gravimetric water content. Sufficient calcium hydroxide (Ca(OH)_2) was added to meet the requirement for the SMP active acidity, while CaCO_3 was added to satisfy the potential acidity values. A 25 percent safety factor was used with the total lime requirement determined by Equation 1. Amounts of lime added expressed as kg/plot were follows: Plot 5 (28.8 kg of Ca(OH)_2 and 167 kg CaCO_3); Plot 6 (37.3 kg of Ca(OH)_2 and 139 kg CaCO_3); Plot 9 (71.9 kg of Ca(OH)_2 and 112 kg CaCO_3); and Plot 11 (29.3 kg of Ca(OH)_2 and 117 kg CaCO_3). The lime materials were incorporated to a depth of 45 cm using standard agricultural equipment (Fig. 3).

Composted organic matter was incorporated into the upper 15 cm of the amended plots with the rototiller at a rate of 2% as organic carbon. Fertilizer (N-P-K of 34-0-0) was applied to each test plot based on medium application rate of 60 lb/acre of N. Phosphorus and potassium levels in soil and tailings were above recommended medium rates, and were not added.



Figure 3. Incorporating lime amendments into an experimental plot at the Keating Tailings, summer 2003.

The control plots on the tailings were treated identically as the treatment plots, but they did not receive neutralization lime amendments or organic matter. Plot construction was completed in early September 2003.

Vegetation

In mid October 2003, each of the twelve plots were scarified using hand rakes, and then the seed mix was broadcast applied at the rate shown in Table 3. The seeding rate is high for standard agronomic purposes, but compares well with seeding rates for amended tailings (Munshower 1994, RRU 1997). Hand rakes were then used to mix the seeds into the surface material, and the plots were rolled with a lawn roller to better adhere the seeds to the soil. Straw mulch was applied to each plot at the rate of 11 metric tons/hectare and crimped into the soil using hand shovels. This rate for straw mulch application exceeds recommendations by (Munshower, 1994) due to the fact that mechanical means for crimping the straw into the ground were limited.

Table 3. Seed mix and rate for the Keating Tailings site.

Common Name	Scientific Name	Seed Mix Kg/ha (PLS)	Variety
Cudweed sagewort	<i>Artemisia ludoviciana</i>	0.8	
Common yarrow	<i>Achillea millefolium</i>	0.8	
American vetch	<i>Vicia americana</i>	6.7	
Western wheatgrass	<i>Pascopyrum smithii</i>	20.2	Rosana
Green needle grass	<i>Stipa viridula</i>	6.7	
Indian ricegrass	<i>Achnatherum hymenoides</i>	10.1	
Big bluegrass	<i>Poa ampla</i>	1.7	Sherman
Slender wheatgrass	<i>Elymus trachycaulus</i>	13.5	Pryor
Fringed sagewort	<i>Artemisia frigida</i>	0.3	
Total		60.8	

During the first growing season in the summer of 2004, seedling density counts, canopy coverage by life form, and the determination of metal levels in the vegetation growing on the plots were made. Seedling density was determined by counting the number of stems within a 20 by 50 cm frame. Four frames were randomly placed along a diagonal transect within each of the twelve experimental plots. Mean density counts for all plots were within 200 to 400 counts/square meter, indicating acceptable germination and establishment.

Canopy coverage was estimated using the Daubenmire Method (1959). Four randomly placed 20 x 50 cm frames were located along a diagonal transect within each experimental plot (Fig. 4). Cover classes were determined by life form: perennial grasses, forbs, barley (from the mulch), litter, and bare ground.

Monitoring Results

Post-treatment Soil and Tailings pH and Soluble Metal Levels

Incremental depth samples of amended tailings and samples from the control plots were collected in the spring of 2004 and determinations of pH were made (Table 4). All pH levels in treated tailings plots were between 6.7 and 8.3, with the exception of the tailings collected from the 45 to 60 cm depth in Plot 11. Incomplete mixing or lack of placement of the neutralizing lime at this depth resulted in a pH level similar to the non treated tailings in the control plots. Water soluble As, Cd, Cu, Pb, and Zn concentrations were also determined in the collected samples from the treated and control experimental plots. Mann-Whitney Rank Sum tests (SPSS, 2003) were run to compare median concentrations of these elements within the tailings profile (Table 5).



Figure 4. Vegetation monitoring of the experimental plots, summer 2004.

Concentration of soluble Cd, Cu, Pb, and Zn were markedly reduced ($P < 0.001$) in the treated tailings compared to tailings collected from the control plots. Median reductions ranged from one order of magnitude for Cd, Cu, and Pb to three orders of magnitude for zinc. Changes in soluble As were nonsignificant. By raising the pH of the tailings through the addition of $\text{Ca}(\text{OH})_2$ and CaCO_3 , these metals are precipitated or otherwise sorbed and thereby removed from the soil solution. Other field experiments in which acid metalliferous wastes are treated with lime report similar reductions in metal solubility (Munshower et al., 2003, Brown et al., 2005).

Table 4. Levels of acidity in treated tailings experimental plots and control plots.

Plot No.	pH values of tailings collected at each depth increment					
	0 – 5 cm	5 – 15 cm	15 – 30 cm	30 – 45 cm	45 – 60 cm	
Treated tailings plots						
5	7.8	7.8	7.7	7.8	6.7	
6	8.0	8.0	7.7	7.9	7.9	
9	8.0	8.2	7.7	7.7	7.7	
11	8.3	8.2	7.8	7.8	4.1	
Control tailings plots						
7	4.2	4.3	4.0	NC ¹	NC ¹	
8	4.3	4.1	4.2	NC ¹	NC ¹	
10	4.3	4.2	4.1	NC ¹	NC ¹	
12	4.2	4.2	4.2	NC ¹	NC ¹	

¹NC indicates samples were not collected at these depth intervals.

Table 5. Median concentrations (mg/kg) of water soluble arsenic and metals in treated and untreated experimental plots.

Plots	No.					
	samples	Arsenic	Cadmium	Copper	Lead	Zinc
Treated	20	0.14	0.01	0.16	0.06	0.03
Control	12	0.11	0.70	1.35	0.11	41.0
P value		0.156	<0.001	<0.001	<0.001	<0.001

Vegetation Cover and Tissue Metal Concentrations

Canopy cover of perennial grasses and forbs are exhibited in Table 6. Kruskal-Wallis One Way ANOVA on Ranks revealed that the median cover value for perennial grasses growing on the control tailings plots was significantly less than the cover of these species growing on the native range plots and the treated tailings plots (Table 6). Mean forb canopy cover was significantly greater for the off site native soils compared to the forb cover on the tailings experimental plots.

Concentrations of metals in plant tissue (perennial grasses) collected from each experimental plot were determined (Table 7). These plant concentrations are for unwashed plant

tissue, and therefore are representative of both metal levels in the plant tissue and on the plant surface as dust.

Table 6. Comparison of perennial grass and forb canopy cover on the experimental plots.

Plots	No. of Daubenmire Cover Frames	Median Perennial	Mean Forb
		Grass Canopy Coverage (%)	Canopy Coverage (%)
Treated Tailings	20	62.5 a ¹	6.25 a
Control Tailings	20	15.0 b	1.50 b
Off-site Soils	20	62.5 a	2.25 b

¹ Values followed by same letter in columns are not significantly different ($P \leq 0.05$).

Table 7. Elemental levels (mg/kg, dry weight) in perennial grass samples from Keating Plots.

Plot No.	Arsenic	Cadmium	Copper	Lead	Manganese	Zinc
Off –Site Native Soils						
1	< 4	< 0.05	7.0	< 4	98.0	25.0
2	< 4	< 0.05	6.0	< 4	44.6	18.0
3	< 4	< 0.05	7.0	< 4	52.0	18.0
4	< 4	< 0.05	5.0	< 4	52.8	18.0
Treated Tailings Plots						
5	< 4	0.35	7.0	< 4	130	35.0
6	< 4	1.1	11.0	< 4	107	51.0
9	< 4	2.3	12.0	< 4	118	86.0
11	< 4	1.4	11.0	< 4	80.4	68.0
Control Tailings Plots						
7	< 4	4.4	14.0	< 4	202	180
8	< 4	2.9	14.0	< 4	263	150
10	< 4	3.3	14.0	< 4	411	182
12	< 4	1.8	14.0	< 4	318	134

These metal loads (concentration on and in the plant tissue) can be compared to maximum tolerable levels of dietary minerals for domestic animals (NRC, 1980). These concentrations are as follows:

Maximum tolerable dietary levels for cattle and horses: arsenic = 50 mg/kg, cadmium = 0.5 mg/kg, copper = 100 mg/kg (cattle) or 800 mg/kg (horses), lead = 30 mg/kg, manganese = 1,000 mg/kg (cattle) or 400 mg/kg (horses), and zinc = 500 mg/kg.

Most of the plant samples collected from the site revealed metal and arsenic concentration below the maximum dietary tolerance levels for cattle and horses. There were, however, exceptions. Concentrations of cadmium in these plants varied from well below the NRC suggested tolerable level to concentrations above the level. The dietary level of cadmium for domesticated animals is based on human food residue considerations (NRC, 1980), and the need to avoid increases of cadmium in the U.S. food supply. Higher residue levels (> 0.50 mg/kg) for a short period of time would not be expected to be harmful to animal health or to human food use, particularly if the animal were slaughtered at a young age (NRC, 1980).

Kabata-Pendias and Pendias (1992) provide elemental levels for generalized plant species into ranges representing deficient, sufficient or normal, excessive or toxic, and tolerable in agronomic crops. These concentrations or ranges are exhibited in Table 8.

The authors caution the use of concentration shown in Table 8 in regard to four factors: 1) concentrations or ranges are given for generalized plant species, not those that are very sensitive or those that are tolerant; 2) overall approximations can differ widely for a particular soil-plant system; 3) ranges on concentrations in plants are often very close to the contents that exert a harmful influence on plant metabolism; and 4) it is difficult to make a clear distinction between sufficient and excessive concentrations of elements in plants.

Elemental levels for perennial grasses collected for the experimental plots at the Keating Site (Table 7) are within normal range for copper, lead, and zinc. Levels of manganese for grasses collected from the off-site native soils plots and the treated tailings were within the normal range; samples from the untreated tailings were normal to elevated for this element. Levels of cadmium for plants collected from the tailings were above the normal range. The detection limit of the method used to quantify the arsenic concentration does not allow interpretation of the plant arsenic data.

Table 8. Approximate Levels (mg/kg dry weight) of Arsenic and Metals in Mature Leaf Tissue¹.

Element	Deficient	Sufficient or Normal	Excessive or Toxic	Tolerable in Agronomic Crops
Arsenic	-	1 to 1.7	5 to 20	-
Cadmium	-	0.05 to 0.2	5 to 30	3
Copper	2 to 5	5 to 30	20 to 100	50
Lead	-	5 to 10	30 to 300	10
Manganese	15 to 25	200 to 300	300 to 500	
Zinc	10 to 20	27 to 150	100 to 400	300

¹ From Kabata-Pendias and Pendias (1992).

Summary and Conclusions

Additions of Ca(OH)₂ and CaCO₃ and organic matter allowed seeded native vegetation to establish on previously barren acid metalliferous tailings. Soluble concentrations of metals in the treated rootzone were reduced one to three orders of magnitude compared to untreated tailings. Concentrations of soluble arsenic remained unchanged by treatment. Seedling density in the first growing season was acceptable. Canopy cover of perennial grasses growing on the treated tailings was statistically greater than grasses on the untreated tailings and equivalent to grasses growing on the off-site native soils. Canopy cover of the seeded forb species was greater on the native soil plots compared to the tailings plots. Elemental levels in perennial grasses were generally below the maximum tolerable concentrations suggested by the National Research Council for grazing cattle and horses. Levels of zinc, lead, and copper in the plant tissues were within normal of sufficient range for vegetation, while concentrations of cadmium and manganese were somewhat elevated. Additional monitoring of these experimental plots is planned for the 2005 field season. Canopy cover by species, species richness, identification of any invading (non-seeded) species, above ground biomass, and elemental concentrations will be determined. Amended tailing samples will also be collected to determine acid base account and pH.

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