

METAL LEVELS IN VEGETATION GROWING ON *IN SITU* TREATED ACID METALLIFEROUS MINE WASTES IN MONTANA¹

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Abstract: The purpose of *in situ* or in-place treatment of metal mine wastes is to immobilize contaminants within the soil/plant complex, reduce metal and arsenic movement to groundwater, reduce metal and arsenic movement to receiving streams, stabilize the landscape from wind erosion, and to provide a functional ecosystem compatible with current and future land uses. In-place treatment includes the incorporation of chemical and biological amendments to provide a hospitable rootzone, and to select plant species that can thrive in the newly amended environment. During in-place treatment, metals are chemically precipitated, and/or sequestered by complexation and sorption mechanisms within the mine wastes/ contaminated soils. Metal availability to plants is minimized, and metal leaching into groundwater is reduced. Metals and arsenic that remain in soil solution are immobilized via chemical reactions at plant root surfaces. The selection of plant species for in-place treatment is based on availability of seed or seedlings, the species' relative lack of ability to translocate (or move) metals and arsenic from the roots into the above ground biomass of the plant, and land use and management considerations. In-place treatment does not remove contaminants from the soil. As a consequence, short-term and long-term effectiveness of in-place treatment has been and continues to be debated. One concern is the potential toxicity to livestock and wildlife from the contaminants in soils and/or plants. This paper provides vegetation metal data from several Superfund and AML sites at which in-place treatment has been used as a remedial strategy. Interpretations of these data based on plant toxicity and residual risk to terrestrial receptors including grazing animals are presented.

Additional Key Words: phytostabilization, metals, contaminated soils, Superfund

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Introduction

Mining companies and federal agencies, specifically the US Environmental Protection (EPA) Agency and the Bureau of Land Management (BLM) have been investing an emerging remedial technology of phytostabilization or *in situ* treatment of mine and smelter wastes and contaminated soils. EPA suggested that *in situ* soil remediation technology can be applied to Superfund and Brownfields sites, large and small mining sites, and other sites with disturbed or degraded soils (Rubin et al. 2007). Appropriate application of this technology has the potential to protect human health and the environment by reducing contaminant bioavailability and mobility at a considerably lower cost than other available options. This, in turn, allows for revitalization and reuse of these lands. The BLM published the results of in-place treatment of acid metal containing mine tailings in Montana (Neuman and Ford 2006).

Phytostabilization as defined by Salt, et al. (1995) is the use of metals-tolerant plants to inhibit the mobility of metals, thus reducing the risk of further environmental degradation by leaching into ground water or by airborne spread. In-place treatment does not remove contaminants from the soil. As a consequence, short-term and long-term effectiveness of in-place treatment has been and continues to be debated (Mendez and Maier 2008). One concern is the potential toxicity to livestock and wildlife from the contaminants in soils and/or forage. The purpose of this paper is to provide vegetation metal data from several Superfund and Abandoned Mine Lands (AML) sites in Montana at which in-place treatment has been used as a remedial strategy. Interpretations of these data based on plant toxicity and residual risk to terrestrial receptors including grazing animals are presented. The phytostabilization 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 demonstrations were conducted in Montana at abandoned mines, and at Superfund Sites within the Clark Fork River Basin. An assessment 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).

In-Place Treatment or Phytostabilization

The purpose of *in situ* treatment is to immobilize contaminants within the soil/plant complex, to reduce metal and As movement to groundwater, to reduce metal and As movement to receiving streams, to stabilize the landscape from wind erosion, and to provide a functional ecosystem compatible with current and future land uses.

The use of plants in the treatment of contaminated soils gained attention (Raskin and Ensley 2000), and the term phytoremediation has been defined in the scientific literature as the use of green plants to remove, contain, or render harmless environmental contaminants (Cunningham and Berti 1993). There are four members of the phytoremediation family that have been defined in the scientific literature:

- Phytoextraction- the use of metal accumulating plants to transport and concentrate metals from the soil into harvestable roots and above ground plant shoots (Kumar et al. 1995).
- Phytostabilization- the use of metal tolerant plants to inhibit the mobility of metals, thus reducing the risk of further environmental degradation by leaching into groundwater or by airborne spread (Salt et al. 1995).
- Phytodegradation- the use of plants and associated micro flora to convert organic pollutants into nontoxic compounds, and
- Rhizofiltration- the use of plant roots to adsorb, precipitate, and concentrate toxic metals from polluted effluents (Dushenkov et al. 1995).

Sequestration of Metals by Phytostabilization

The phytostabilization process was described schematically by Berti and Cunningham (2000) as shown Fig. 1.

During phytostabilization, metals are chemically precipitated, and/or sequestered by complexation and sorption mechanisms within the tailings/soils. Metal availability to plants is minimized, and metal leaching into groundwater is reduced. Metals and As that remain in soil solution are demobilized via chemical reactions at plant root surfaces. Plant species serve several purposes in phytostabilization. Vegetation harvests water in the root-zone and can transpire several hundred thousand liters of water per hectare during the growing season. This harvest has a significant impact on the volume of water (and metals and As) that is able to move

towards the groundwater. Plants stabilize the landscape from erosion, greatly reducing surface water runoff available to receiving streams. Plants also reduce erosion caused by wind. The selection of plant species for phytostabilization is based on availability of seed or seedlings, ability to thrive in the newly created root-zone, tolerance of elevated concentrations of metals, salinity and droughty conditions, the species' relative lack of ability to translocate (or move) metals and As from the roots into the above ground biomass of the plant, and land use and management considerations.

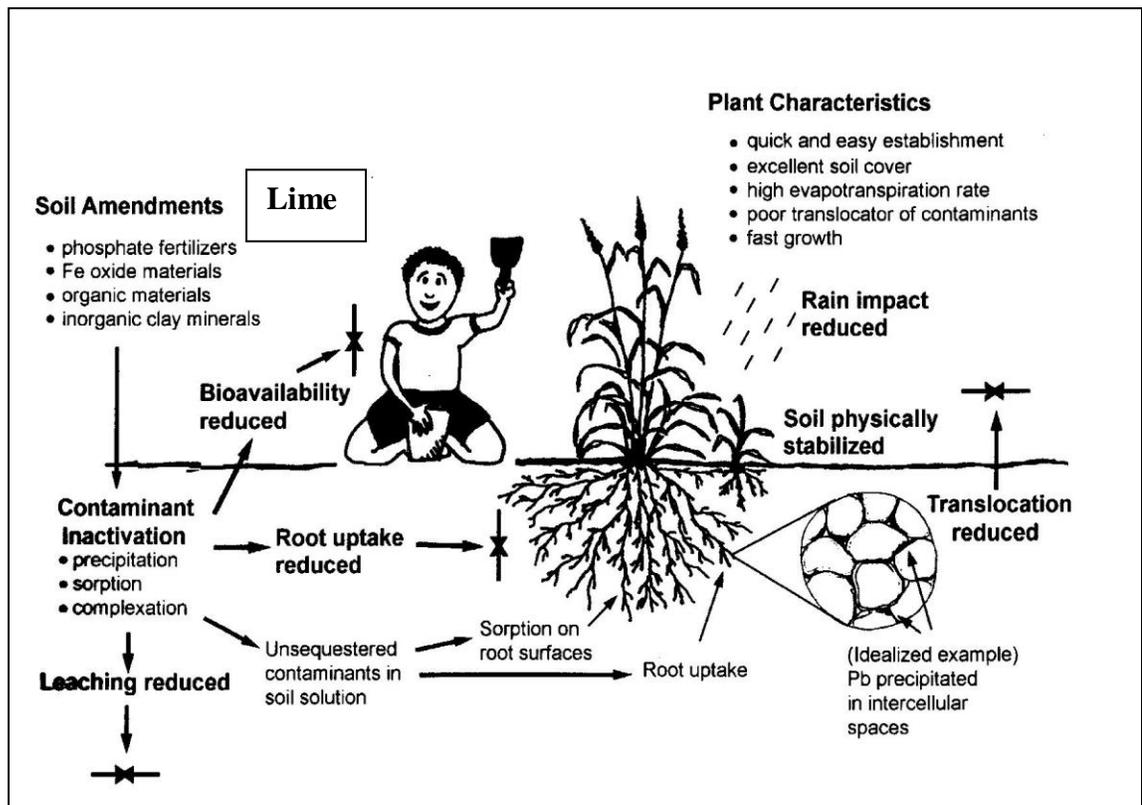


Figure 1. The Role of Soil Amendments and Plants in the Phytostabilization of Heavy-Metal-Contaminated Soil (Source: Berti and Cunningham 2000).

In a recent review of the use of phytostabilization of mine tailings, Mendez and Mair (2008), concluded (based on work by Johansson et al. 2005) that plant metal accumulation has not been well documented in the majority of field studies. They further conclude that long-term studies are lacking to evaluate the efficacy of phytostabilization in permanently reducing metal toxicity, promoting plant succession and in promoting soil development.

Phytostabilization Studies in Montana

Within the Clark Fork River Basin, two large laboratory and field phytostabilization investigations were conducted: the first using tailings from Silver Bow Creek (Schafer & Associates and Reclamation Research Unit (RRU) 1993), and the second using contaminated soils and smelter wastes near Anaconda (RRU 1993, 1996a, 1996b, 1997). These two treatability studies evaluated amendments including different sources of lime material, ferric sulfate, phospho-gypsum, Fe_2O_3 , different organic matter sources, high levels of P, and fertilizers. Plant species were evaluated for potential field use by conducting replicated greenhouse studies. Over 100 experimental plots were designed in the field and extensive monitoring of the soils, waters, and vegetation was conducted. A separate phytostabilization demonstration project was implemented along one and one half miles of land adjacent to the Clark Fork River (Schafer & Associates 2000). Six consecutive years of monitoring of soils, water, and vegetation were conducted at this demonstration of *in situ* treatment. In each of the three major investigations, data were used to determine the effectiveness of the phytostabilization treatments in terms of site specific conceptual models. Treatment-induced changes in the sources, pathways and receptors of the contamination were assessed. In addition, studies of terrestrial and avian receptors associated with phytostabilized field plots and the demonstration project were completed (Hooper 2001). These data are the foundation upon which judgment of the efficacy of phytostabilization technology is based.

The Anaconda Revegetation Treatability Study (ARTS) was a large laboratory, greenhouse, and field investigation of phytostabilization techniques applied to contaminated soils and to smelter tailings (RRU 1993, 1996a, 1996b, and 1997) in the vicinity of Anaconda, Smelter Superfund Site. This investigation was the formal Treatability Study for the Anaconda Regional Water, Waste and Soils Operable Unit of the Anaconda Smelter Superfund Site. Pre-study site characteristics, or a conceptual model, defined contaminant exposure pathways, release mechanisms, and receptors. Post-investigation monitoring data were used to quantify changes in contaminant pathways and release mechanisms. As part of Remedial Action/Remedial Design for this Operable Unit, an assessment was made of how well phytostabilization methods met the Record of Decision (ROD) defined Remedial Action Goals and Objectives (CDM and RRU 2001).

A tailings impoundment associated with an abandoned hard rock mine in southwestern Montana is another site of additional phytostabilization study. Replicated experimental plots using soil amendments, lime and organic matter, designed to ameliorate the plant inhibiting chemical characteristics of the tailings were constructed, and seeded with native plants. Yearly monitoring through 2008 has included measurements of vegetation cover, species richness, and aboveground biomass. Changes in tailings chemistry were assessed by measuring pH, acid/base account, and water soluble metal concentrations.

More recently, several areas in which in-place treatment was implemented were evaluated by a team of scientists from Montana State University (Munshower et al 2003). The purpose of this investigation was to generate sufficient data and information from areas receiving phytostabilization treatments, varying in age from 6 to 19 years, so that the permanence and self-sufficiency of the established and reconstructed ecosystem(s) can be assessed. Six different field sites were selected that represent phytostabilization implementation in different landscape positions, using slightly different equipment, and at different times. The sites are similar in that each was degraded because of impacts from the metal mine/mill/smelter processes. Soils or tailings at the sites contain acid producing materials and are elevated in metal concentrations compared to adjacent, non-impacted landscapes. At each site, neutralizing amendments were added to raise the soil or waste pH to a target level of seven, and at some sites, other amendments were also added. Vegetation response variables observed or measured at the six sites included cover, species richness, evidence of reproduction, evidence of nutrient cycling, evidence of succession, and biomass. Soil response variables measured included pH, acid base account, and soluble metal concentrations. Conclusions of this work (Munshower et al 2003) were reported:

- In situ reclamation or phytostabilization of acid waste is a valuable reclamation technique. The calcium carbonate amendment applied as ground limestone or industrial waste can be calculated to produce a non-acid root zone that will last indefinitely.
- There are indications that once vegetation is established on the waste the root mass growing into amended and non-amended materials complexes the toxic ions and thereby renders the materials less toxic. This permits further root proliferation into adjacent non-amended materials and the initiation of a cycle of growth/neutralization/growth that is self

perpetuating. This eventually permits the establishment of less tolerant vegetation on the wastes and a plant successional cycle is underway.

- Successional changes in vegetation were observed on several of the areas evaluated.

Metal Levels In And On Vegetation

Cattle grazing is a major agricultural land use on lands impacted by aerial contamination of soils from metal smelters, on abandoned hard rock mines sites, and near streams and rivers impacted by fluvially deposited mine tailings and related wastes. The protection and enhancement of this resource is a significant consideration in choosing remedial actions for contaminated land adjacent to and in close proximity to these rivers. One of the principles of phytostabilization is to select plant species that are poor translocators of contaminants (metals and As) into the above ground portions of the plant.

The following tables exhibit metal and As concentration in/on vegetation grown in acid metalliferous wastes and contaminated soils that were treated with lime, and at some sites with other amendments (organic matter, phosphorus, etc.). Table 1 displays elemental levels in vegetation grown in lime treated fluvial mine wastes deposited along the Clark Fork River. Table 2 exhibits plant elemental data for vegetation grown in several experimental in-place treatment plots constructed in fluvially deposited mine wastes along Montana's Silver Bow Creek. Metal and As concentrations in and on vegetation grown in plots constructed near Anaconda, Montana are shown in Table 3.

Perennial grasses grown on in-place treated mine tailings at the Keating Tailings site in southwestern Montana were collected during the first growing season (2004), and concentrations, (mean \pm standard deviation), determined were as follows (Neuman and Ford 2006): As (< 4 (mg kg⁻¹), Cd (1.3 ± 0.8 (mg kg⁻¹), Pb (< 4 mg kg⁻¹), Mn (108 ± 21 mg kg⁻¹), and Zn (60 ± 22 mg kg⁻¹).

Table 1. Metal and arsenic concentrations in and on vegetation collected from phytostabilized areas along the Clark Fork River (Schafer & Associates 2000).

Plant Species	N Value	Statistic	Arsenic (mg kg⁻¹)	Cadmium (mg kg⁻¹)	Copper (mg kg⁻¹)	Lead (mg kg⁻¹)	Zinc (mg kg⁻¹)
Redtop	11	Mean	3.28	0.26	13.7	1.25	112
		Min	1.80	0.09	6.7	0.40	56.4
		Max	5.40	0.63	35.5	3.7	255
Russian Wildrye	2	Mean	1.1	0.15	5.20	0.5	62.9
		Min	1.1	0.07	3.69	0.3	50.3
		Max	1.1	0.23	6.7	0.7	75.5
Alfalfa	5	Mean	2.48	0.52	10.8	0.80	83.3
		Min	1.1	0.10	6.6	0.30	36.3
		Max	5.0	0.93	15.4	2.1	152
Aster spp.	1		5.3	0.42	21.7	1.7	71.1
Alkali Cordgrass	1		3.1	0.13	10.0	1.7	31.4
Slender Wheatgrass	1		1.0	0.33	5.6	0.7	48.7
Salix spp.	4	Mean	1.48	3.03	224	0.85	505
		Min	1.0	1.66	4.5	0.50	152
		Max	2.50	4.01	874	1.7	1180
Strawberry Clover	2	Mean	1.95	0.50	10.3	0.70	198
		Min	1.1	0.23	9.9	0.50	140
		Max	2.8	0.77	10.7	0.90	255

Table 2. Metal and arsenic concentrations in and on vegetation collected from phytostabilized experimental plots Along Silver Bow Creek (Schafer and Associates and RRU 1993).

Plant Species	N Value	Statistic	Arsenic (mg kg⁻¹)	Cadmium (mg kg⁻¹)	Copper (mg kg⁻¹)	Lead (mg kg⁻¹)	Zinc (mg kg⁻¹)
Thickspike Wheatgrass	3	Mean	2.6	0.26	17.4	9.2	40.8
		Min	2.1	0.23	14.9	6.4	47.1
		Max	3.1	0.29	21.7	12.3	69.7
Altai Wildrye	6	Mean	4.5	0.54	26.3	6.5	118
		Min	2.3	0.26	12.2	3.3	44.4
		Max	7.6	0.98	46.8	14.8	236
Tall Wheatgrass	3	Mean	27.1	1.7	247	38.7	561
		Min	26.1	1.5	216	25.1	393
		Max	33.2	2.0	278	60.6	750
Intermediate Wheatgrass	3	Mean	23.5	1.9	231	32.1	552
		Min	21.7	1.6	196	28.3	491
		Max	26.6	2.1	268	38.2	589
Crested Wheatgrass	6	Mean	2.9	0.51	15.3	11.0	127
		Min	1.7	0.35	10.3	6.2	111
		Max	4.9	0.60	20.4	21.9	160
Russian Wildrye	3	Mean	3.1	0.85	17.2	11.4	130
		Min	2.6	0.60	11.7	7.2	115
		Max	3.8	1.1	20.9	15.5	154
Streambank Wheatgrass	3	Mean	3.5	0.51	38.8	5.1	87.1
		Min	3.1	0.30	31.0	4.3	74.2
		Max	4.0	0.68	50.3	6.6	103
Basin Wildrye	3	Mean	2.0	0.48	23.8	2.1	107
		Min	1.7	0.38	21.7	2.0	98.2
		Max	2.4	0.61	28.0	2.4	112

Table 3. Metal and arsenic concentrations in and on vegetation collected from phytostabilized experimental plots near Anaconda Smelter NPL Site, Montana (RRU 1997).

Location	Plant Species	Arsenic (mg kg ⁻¹)	Cadmium (mg kg ⁻¹)	Copper (mg kg ⁻¹)	Zinc (mg kg ⁻¹)
Opportunity	Intermediate wheatgrass	0.58	0.09	7.1	41.5
Tailings	Intermediate wheatgrass	0.36	0.05	6.6	28.7
Ponds	Pubescent wheatgrass	3.0	0.14	23.4	12.3
	Sheep fescue				
Anaconda	Intermediate wheatgrass	2.0	0.61	35.2	43.7
Tailings	Intermediate wheatgrass	1.3	0.20	17.5	60.6
Ponds	Pubescent wheatgrass	2.0	0.36	36.5	50.4
	Slender wheatgrass				
Smelter	Basin wildrye	6.5	0.68	18.3	25.7
Hill	Basin wildrye	16.5	1.5	13.2	57.9
Contaminated	Sheep fescue	9.9	1.0	13.4	13.6
Soils					
Red Sands	Crested wheatgrass	4.6	<0.05	11.9	16.6
Reprocessed	Crested wheatgrass				
Tailings					
Contaminated	Alfalfa	5.2	0.40	19.5	24.8
Soils	Redtop	7.1	1.3	38.5	64.3

As part of developing acid tolerant cultivars of plant species that can be used in revegetation and restoration efforts in the Clark Fork River Basin, the Bridger Plant Materials Center constructed experimental plots on lime treated metal contaminated soils within the Anaconda Smelter NPL site. Vegetation (N = 36) was collected in 2004 and plant elemental concentration ranges were as follows (Majerus 2004): Arsenic (half of the reported values were < detection limit of 5 mg kg⁻¹, maximum value was 13 mg kg⁻¹), Cd (all reported values < detection limit), Cu (50 ± 23 mg kg⁻¹), Pb (all reported values < detection limit), and Zn (71 ± 36 mg kg⁻¹).

Maximum Tolerable Metal Levels for Cattle and Horses

All plant concentrations presented in the above tables and text are for unwashed plant tissue, and therefore are representative of both metal levels in the plant tissue and on the plant surface as dust. These metal loads (concentration on and in the plant tissue) can be compared to maximum tolerable levels of dietary minerals for domestic animals as recommended by the National Research Council (NRC 2005). The maximum tolerable level is defined as the dietary level that when fed for a defined period of time, will not impair animal health or performance (NRC 2005). These concentrations for cattle and horses are as follows: As = 30 mg kg⁻¹, Cd = 10 mg kg⁻¹, Cu = 40 mg kg⁻¹ (cattle) or 250 mg kg⁻¹ (horses), Pb = 10 mg kg⁻¹ (horses) or 100 mg kg⁻¹ (cattle), and Zn = 500 mg kg⁻¹.

Most of the plant species growing in the five investigations of phytostabilization in Montana revealed metal and As concentration below the maximum dietary tolerance levels for cattle and horses. There were, however, exceptions: willow (Table 1) had elevated mean and maximum levels of Cu and Zn. Elevated concentrations of Cu, Pb, and Zn were also found in Tall and Intermediate wheatgrasses grown in treated tailings along Silver Bow Creek (Table 2).

Incidental Ingestion of Contaminated Soil

Because phytostabilization does not remove contaminants from the soil, there is a residual risk of exposure to cattle from the incidental ingestion of soil during grazing. Ingestion of soil along with forage can be a source of additional elements for grazing cattle. Mayland et al. (1975) estimated daily soil ingestion levels for cattle grazing semiarid range in Idaho to range from 100 to 1500 grams, with a median of 500 grams/animal/day. Other investigations (Healy 1974 and Thornton 1974) reported similar soil ingestion rates. Lead levels in blood from cattle residing near the East Helena Smelter Superfund Site (Neuman and Dollhopf 1992) were significantly correlated with soil concentrations of Pb, as well as vegetation concentrations, and distance (negative correlation) from the Pb smelter. It was postulated that soil concentrations may be more important than forage as a source of Pb to the cattle in the East Helena investigation.

Bioaccumulation of certain metals in domestic animals is not restricted to contaminated sites. Munshower and Neuman (1979) conducted a study of metals in tissues from mule deer and antelope collected from a pristine area in southeastern Montana. Both vegetation and soil

concentrations of metals were below most literature values and Cu in vegetation was described as marginally deficient. Bioaccumulation of Pb in antelope livers, and Cd accumulation in antelope and deer kidneys as functions of animal age was reported. The authors concluded that it was not possible to ascertain whether the Pb and Cd levels in these animal tissues and the accumulations with age were normal because of the lack of comparative data. However, they also concluded that the general condition of wildlife populations would indicate that the elemental levels were reflective of levels in healthy deer and antelope.

Edible muscle, kidney and liver tissues from six selected cattle from the Grant-Kohrs Ranch National Historic Site located within the Clark Fork River Superfund Site, were analyzed for concentrations of As, Cd, Cu, Pb and Zn (DOI 1996 and revised 1997). Ninety days prior to slaughter, three of the animals were allowed to graze within contaminated riparian areas, and three others were held in less-contaminated pastures. It was reported that riparian cows had metal tissue concentration very similar to pasture cows. Elevated diagnostic levels were reported for Cd in kidney tissue from pasture cows, Cu levels in muscle tissues were elevated in four animals, and Cu in liver tissue of one animal was reported at a toxic level. There are no site-specific data for metal levels in cattle grazing on phytostabilized lands within the Clark Fork River Basin. The assessment of residual risk to grazing animals on these treated areas is therefore a data or information gap.

White-tailed deer and cattle were selected for quantitative evaluation in the Clark Fork River Ecological Risk Assessment (ISSI 1999). Predictive analysis indicated little or no hazard of toxic effects to deer from metals or As in the terrestrial environment. A moderate hazard to range cattle was predicted from As and Cu in soils. The authors stated that results should be interpreted with caution because there is little site specific information to support the predictions.

Toxicity to Plants

Based on a review of the scientific literature, ranges of elemental levels for mature leaf tissue have been presented by Kabata-Pendias and Pendias (1992). The authors provide elemental levels (Table 4) for generalized plant species into ranges representing deficient, sufficient or normal, excessive or toxic, and tolerable in agronomic crops. These concentrations or ranges are as follows:

Table 4. Approximate levels (mg kg⁻¹) of arsenic and metals in mature leaf tissue (Kabata-Pendias and Pendias (1992).

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	20-100	50
Lead	-	5-10	30-300	10
Zinc	10 to 20	27 to 150	100 to 400	300

The authors caution the use of these 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 exerts 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.

Vegetation samples collected from lime treated soils and tailings revealed the following metal and As levels: most plant loadings were within the normal or sufficient range, with a few As concentrations in the excessive range. Willow (Table 1) and some wheatgrasses (Table 2) did reveal very high concentrations of Cu, Pb and Zn; much higher than other plants. Cadmium levels in several species were elevated, but below the approximate excessive range given by Kabata-Pendias and Pendias (1992). It is believed that the plant species growing in phytostabilized areas are generally tolerant of metal and acid. For example, Redtop (*Agrostis* spp.) are known to be able to evolve metal-resistance (Shaw 1990), and Basin Wildrye has invaded the upper portions of Smelter Hill in Anaconda, which has soils with extremely elevated metal and As concentration (RRU 1993).

Conclusions

Phytostabilization technology uses soil amendments and metals and acid tolerant plants to revegetate sites. In so doing, metals are immobilized, for the most part, and are not available for migration or exposure. Proper selection of amendments and plants is necessary to ensure growth and to prevent bioaccumulation. Metal concentrations found in several plant species grown on

treated tailings or contaminated soils revealed levels generally below, with some exceptions, the maximum tolerable limits for grazing cattle and horses. In situ reclamation or phytostabilization of acid/metal mine related wastes has been shown to be a valuable reclamation technique. This technology has been selected by EPA for remediation of certain lands within the Anaconda Smelter Superfund Site and the Clark Fork River Operable Unit of the Milltown Reservoir Superfund Site, both in Montana. Incorporation of lime and organic matter followed by seeding with selected plant species is being conducted at Operable Unit 11 (Upper Arkansas River) for the California Gulch Superfund Site near Leadville, Colorado.

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