

# Cottongrass Effects on Trace Elements in Submersed Mine Tailings

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## ABSTRACT

Phytostabilization may limit the leakage of metals and As from submersed mine tailings, thus treatment of acid mine drainage with lime could be reduced. Tall cottongrass (*Eriophorum angustifolium* Honckeney) and white cottongrass (*E. scheuchzeri* Hoppe) were planted in pots with unlimed (pH 5.0) and limed (pH 10.9) tailings (containing sulfides) amended with sewage sludge (SS) or a bioash-sewage sludge mixture (ASM). Effects of the amendments on plant growth and plant element uptake were studied. Also, effects of plant growth on elements (Cd, Cu, Pb, Zn, and As), pH, electrical conductivity (EC), and concentrations of  $\text{SO}_4^{2-}$  in the drainage water as well as dissolved oxygen in tailings, were measured. Both plant species grew better and the shoot element concentrations of white cottongrass were lower in SS than in ASM. Metal concentrations were lowest in drainage water from limed tailings, and plant establishment had little effect on metal release, except for an increase in Zn levels, even though  $\text{SO}_4^{2-}$  levels were increased. In unlimed tailings, plant growth increased  $\text{SO}_4^{2-}$  levels slightly; however, pH was increased and metal concentrations were low. Thus, metals were stabilized by plant uptake and high pH. Amendments or plants did not affect As levels in the drainage water from unlimed tailings. Thus, to reduce the use of lime for stabilizing metals, phytostabilization with tall cottongrass and white cottongrass on tailings is a sound possibility.

WASTE FROM MINES with elevated heavy metal concentrations is a major environmental problem all over the world. Many ores that are rich in metals consist of sulfide minerals that may form stable sulfide complexes with heavy metals (Schnoor, 1996). However, if sulfides (e.g., pyrite) interact with atmospheric oxygen and water, sulfuric acid is formed. In addition, if other sulfides are oxidized, metals and As may be released (Holmström, 2000). Thus, acid elements drain from the site and spread into the environment. At many mine sites in Sweden, the water passes through a row of tailing ponds and in the last pond, the acid mine drainage is limed before discharge into the environment. The lime raises the pH and prevents release of metals due to their precipitation.

To prevent the process of sulfide weathering, mine tailings can be covered with a high water table (approximately 2 m) to create an anaerobic environment. A high water table, if not in low-lying areas, requires high impoundment walls that are expensive to build. Additionally, the pressure from the water may reduce the stability of the walls and may eventually cause them to collapse (Grimalt et al., 1999). Submersing the tailings under shallow water tables (<1 m) is another option. The placement of organic layers and plants over tailings reduces oxygen intrusion into the tailings, consumes oxygen (due to chemical and biological processes), and

will probably prevent the weathering of sulfides. Plants also reduce wind and wave erosion. This would reduce the use of the lime treatment and might be a more sustainable way to treat tailings.

Wetland plants are adapted to the anaerobic environment in the substrate. These plants cope with such situations by transporting oxygen within the tissue from the atmosphere through a lacunar system of intercellular airspaces or through aerenchyma to underground organs (Armstrong et al., 1992; Brix, 1993) for root respiration. The roots may release some oxygen into the medium surrounding the roots, and thereby alter the geochemistry of the rhizosphere (Chen and Barko, 1988). The released oxygen could induce weathering of sulfides. Wright and Otte (1999) studied the effects of common cattail (*Typha latifolia* L.) and manna grass [*Glyceria fluitans* (L.) R. Br.] on pH, redox potential (Eh), and solubility of Fe, Zn, and As of the pore water in tailings. They found that cattail caused a small decrease in pH and increased soluble Zn concentrations near the roots, while manna grass did not show any effect on the metals in the tailings. Plant roots can take up released elements from the tailings and thereby the dispersion of elements might be reduced.

The properties of mine tailings are not optimal for plant growth. Mine tailings are often acidic (when weathered), low in macronutrients, and high in heavy metals (McCabe and Otte, 2000). Smith and Bradshaw (1979) concluded that the best way to establish plants on mine tailings is to amend the tailings with fertilizers. Instead of commercial fertilizers, amendments like nutrient-containing waste products may be used. Whitbread-Aburatat (1997) found that tree establishment on metalliferous mine tailings was most successful with cake sludge and diatomite, mineral phosphate, or calcareous sand, depending on the species. Another alternative might be bioashes, which have a liming effect and contain nutrients (Greger et al., 1998). Furthermore, sewage sludge may be used to create an organic layer rich in N for vigorous plant growth (Borgegård and Rydin, 1989; Greger et al., 1998).

The aims of this study were to (i) identify effects of amendments and plants on the metal and As concentrations in the drainage water from both limed and unlimed mine tailings containing sulfide minerals and (ii) identify how plant establishment and element uptake and translocation by plants are affected by different amendments added on top of submersed tailings. The first hypothesis was that in unlimed tailings, addition of amendments and plant establishment will increase metal and As leakage due to root oxygen release. The second hypothesis was that metal concentrations in the drainage water

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from limed tailings will not be affected by plants, since the high pH will stabilize the metals, but not As. The third hypothesis was that plants can be established on mine tailings if nutrients are supplied and that different amendments will affect plant element uptake. Tailings and plant seeds were collected at the Kristineberg mine area in northern Sweden, where both unlimed and limed tailing ponds are present. Differences in Cd, Zn, Cu, Pb, and As concentrations in the drainage water of the two tailings with various plant and amendment treatments were studied. Also, EC, pH, and  $\text{SO}_4^{2-}$  in the drainage water and the dissolved  $\text{O}_2$  level in the pore water of the mine tailings were measured to find out the processes in the tailings of the different treatments. Furthermore, plant growth and plant metal uptake were studied in the different treatments.

## MATERIALS AND METHODS

### Tailings and Seeds

Submersed mine tailings rich in sulfides were collected from Impoundment 3 and Impoundment 4 at the Kristineberg mine area, northern Sweden (65°04' N, 18°44' E). Copper and Zn are the main mined metals at the Kristineberg mine site. Some chemical composition (inductively coupled plasma atomic emission spectroscopy [ICP–AES] analyses by SGAB Analytica, Sweden) characteristics of the tailings are shown in Table 1. The major minerals in both tailings were chlorite, talc, quartz, and mica. Also, amphiboles, pyrite, and feldspar were common (X-ray diffraction [XRD] analyses by the Swedish Museum of Natural History). The mine tailings in Impoundments 3 and 4 are believed to be similar to the tailings in Impoundment 1, which have a sulfide content between 10 and 30% (Holmström et al., 2001), and more than 80% of the material has a particle size less than 0.2 mm (Carlsson, 2000). In Impoundment 4, the tailings had been treated with lime (here called limed) at the mine site and had a pH of approximately 10.9; Impoundment 3 tailings were not limed (here called unlimed) and had a pH of approximately 5.0. The unlimed tailings were dark gray at the time of collection and the limed tailings were light gray. No visual signs of weathering were observed in any of the two tailings. Seeds of tall cottongrass and white cottongrass were collected from plants growing on the edges of the impoundments, which otherwise had no vegetation. Tall cottongrass was growing around both Impoundments 3 and 4, whereas white cottongrass was only found around Impoundment 3. The seeds were stored wet in darkness at 4°C for four months and then treated with diurnal fluctuations in both temperature and light (a 12-h dark period at 6°C and a 12-h light period at 19°C) for three days, which,

according to Thompson et al. (1977), increases the viability of wetland plant seeds. After the diurnal treatment, the seeds were left to germinate on wet filter paper for two days before the onset of the experiment. The choice of plant species was a result of tall cottongrass being the most common species in the edges of both the limed and unlimed tailing ponds at the mine site of Kristineberg. Tall cottongrass is stress tolerant and mostly confined to acidic soils ( $\text{pH} < 5$ ), although it can also occur in neutral and calcareous soils with high pH of approximately 8 (Grime et al., 1988, Phillips, 1954). Since white cottongrass also was common on the edges of the unlimed tailings and belonged to the same genus as tall cottongrass, it was likely that this species would have similar characteristics as tall cottongrass, and would survive in limed tailings.

### Experimental Setup

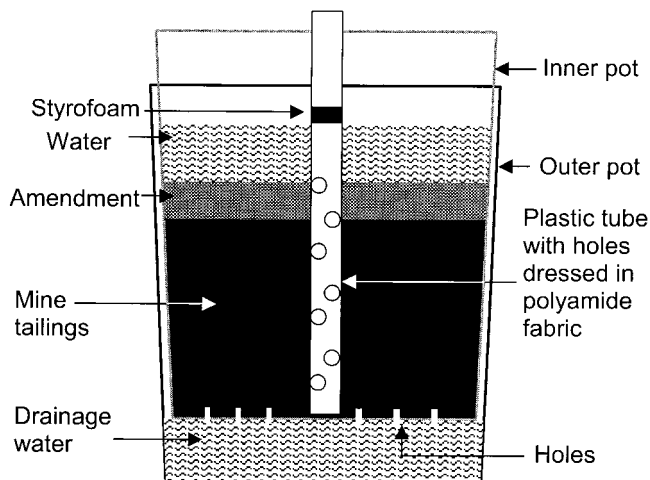
One-liter plastic pots with six holes (diameter = 1 mm) in the bottom were filled to two-thirds with water-saturated tailings (Fig. 1). The pots were placed into similar pots but without holes for drainage water sampling. A 0.1-m-long plastic tube (diameter = 10 mm) with 36 evenly distributed holes (diameter = 5 mm) was placed in the center of the pots. The plastic tubes were dressed in polyamide fabric (pore size 25  $\mu\text{m}$ ; Sintab, Malmö, Sweden) to keep the tailings out. To prevent atmospheric oxygen penetrating the plastic tubes, pieces of styrofoam were placed inside the tubes at water level.

After one week, when the tailings had settled, a 20- to 30-mm-thick layer of amendments was added to the pots on top of the tailings. Control pots did not receive any amendments or plants. The amendments used were sewage sludge (SS) and a mixture (1:2 v/v) of ash and sewage sludge (ASM). The SS was collected at a sewage-treatment plant in Skellefteå, northern Sweden, and the ash, which was fly ash from wood combustion, was collected from a power plant also situated in Skellefteå. Water was added throughout the experiment to keep the tailings water saturated.

After another week, germinated seeds of either tall cottongrass or white cottongrass were placed in the pots with amendments. Thus, pots had the same amount of germinative seeds. Seeds of tall cottongrass, collected from Impoundment 3, were used in the pots with unlimed tailings from Impoundment 3 and tall cottongrass from Impoundment 4 was used in limed tailings from Impoundment 4. Seeds of white cottongrass from Impoundment 3 were used in both limed and unlimed tailings. There were 25 to 30 or 35 to 40 seeds per

**Table 1. Chemical composition of the mine tailings used in the experiment ( $n = 4$ ).**

Element	Impoundment 3	Impoundment 4
	% dry wt. $\pm$ SE	
$\text{SiO}_2$	41.5 $\pm$ 0.05	38.8 $\pm$ 0.05
$\text{Al}_2\text{O}_3$	8.91 $\pm$ 0.005	8.03 $\pm$ 0.005
$\text{CaO}$	1.55 $\pm$ 0.005	10.4 $\pm$ 0.05
$\text{Fe}_2\text{O}_3$	20.0 $\pm$ 0.05	13.6 $\pm$ 0.05
$\text{K}_2\text{O}$	0.786 $\pm$ 0.002	1.17 $\pm$ 0.005
$\text{MgO}$	11.0 $\pm$ 0.05	9.68 $\pm$ 0.005
$\text{MnO}_2$	0.129 $\pm$ 0.0005	0.188 $\pm$ 0.0005
$\text{Na}_2\text{O}$	0.345 $\pm$ 0.0005	0.473 $\pm$ 0.0005
$\text{P}_2\text{O}_5$	0.084 $\pm$ 0.001	0.118 $\pm$ 0.0005
$\text{TiO}_2$	0.235 $\pm$ 0.0005	0.204 $\pm$ 0.0005
S	11.0 $\pm$ 0.025	7.38 $\pm$ 0.015



**Fig. 1. Sketch showing how the pots were assembled.**

**Table 2. Number of replicates at the end of the experiment in limed and unlimed tailings with different treatments of amendments and cottongrass species. At the start the number of replicates was six in all treatments except the unstudied treatments (–). ASM, mixture of ash and sewage sludge; SS, sewage sludge.**

	Amendments					
	ASM		SS		Control	
	Limed	Unlimed	Limed	Unlimed	Limed	Unlimed
Tall cottongrass	0	3	5	6	–	–
White cottongrass	0	5	0	6	–	–
No plants	6	6	6	6	6	6

pot of tall cottongrass and white cottongrass, respectively. No plants were planted in control pots (i.e., pots without amendments), since a pilot investigation showed that plants did not survive in unamended tailings. The experiment was set up equally for the two different tailings (Table 2). The pots were kept in a greenhouse (set for  $18 \pm 1^\circ\text{C}$ , relative humidity approximately 70%) equipped with supplementary lamps (Osram [Munich, Germany] Daylight lights, HQI-BT 400W) and a 12-h light period. The water level was kept about 20 to 30 mm above the amendments. Pots without amendments had a water level of approximately 50 mm above the tailings. Thus, the total covering level on top of the tailings was the same in all pots. There were six pots with each plant species and six pots without plants in each amendment treatment (Table 2). Due to lack of plant growth some treatments had a lower number of replicates (Table 2).

### Drainage Water Analyses

Five months after the seeds were spread in the pots, the drainage water in the outer pots was discarded. The outer pots were washed and rinsed in redistilled water and replaced. About one month after the outer pots had been rinsed, the dissolved  $\text{O}_2$  concentrations of the water inside the plastic tubes were measured (oxi-196; WTW, Weilheim, Germany). The  $\text{O}_2$  electrode was placed 10 mm below the surface of the mine tailings and was stabilized for 45 s. Four measurements were made during one day with 4 h in between each of them. Since no difference in  $\text{O}_2$  levels was found over one day the mean value of those four measurements was used. Six months after the seedlings had been transplanted in the pots, the roots had established throughout the amendments and, additionally, roots could be seen through the plastic pots further down in the tailings.

One month after the  $\text{O}_2$  was measured, 100-mL samples were taken from the drainage water in the outer pots. Fifty milliliters of the sample was used to measure pH (Metrohm [Herisau, Switzerland] 744 pH meter) and the electrolytic conductivity (EC) (LF 196; WTW). The rest of the sample was treated with 5  $\mu\text{L}$  concentrated  $\text{HNO}_3$  per milliliter sample and used for metal analysis with an atomic absorption spectrophotometer (Varian [Mulgrave, Australia] SpectrAA-100), flame technique for Zn and Cu, graphite furnace technique (GTA-97) for Cd and Pb, and hydride vapor generation technique (VGA-77) for As.

One week after the initial water samples were taken, more water from the outer pots was sampled for  $\text{SO}_4^{2-}$  measurements. A spectrophotometric method modified from Vogel (1961) was used. Five milliliters of NaCl and HCl (60 g NaCl in 200 mL redistilled water, 5 mL of concentrated HCl were added and diluted to 250 mL with redistilled water) and 1 mL of gummi arabicum Dobb solution (1 g gummi arabicum dissolved in 200 mL hot redistilled water) were added to a 50-mL flask. The samples were diluted with redistilled water and together with 0.3 g  $\text{Ba}(\text{NO}_3)_2$  (40–30 mesh, i.e., the radius of salt crystals was 0.59–0.42 mm) was added to the 50-mL flask,

which was filled up with redistilled water and shaken. After 10 min the absorbance was measured in an UV/VIS spectrophotometer (Jasco [Tokyo, Japan] 7800). The  $\text{SO}_4^{2-}$  content was calculated from a standard curve made up from  $\text{K}_2\text{SO}_4$ .

### Analyses of Elements in Plant Tissue and Tailings

One week after the samples for the  $\text{SO}_4^{2-}$  analysis had been taken, all plants were taken from the pots and rinsed in redistilled water, divided into roots and shoots, and dried at  $80^\circ\text{C}$  and thereafter weighed. Plant material was wet-digested in  $\text{HNO}_3$  and  $\text{HClO}_4$  (7:3, v/v) and analyzed for Cd, Zn, Cu, and Pb with an atomic absorption spectrophotometer (Varian SpectrAA-100) with the flame technique. For As the hydride vapor generation technique (VGA-77) was used. Mine tailings were dried in the same way as plant material and wet-digested for 30 min in 7 mol  $\text{L}^{-1}$   $\text{HNO}_3$  at  $120^\circ\text{C}$  to obtain the “total” fraction of heavy metals. The bioavailable fraction was obtained when dried tailings were extracted for 16 h in 1 mol  $\text{L}^{-1}$   $\text{NH}_4\text{OAc}$ , having a pH that was one unit lower than that of the tailing sample (Andersson, 1976). Ammoniumacetate was also used to extract As, since the same extractant as for the metals is commonly used for As (e.g., Kalbitz and Wennrich, 1998). The tailings were analyzed in the same manner as plant tissue.

### Calculations

Statistical analyses of the data were performed with simple regression, a *t* test with separate variance estimates, and analysis of variance (ANOVA) followed by post hoc comparisons with Tukey's honest significant difference (HSD) test to identify differences between the different treatments. The software STATISTICA for Windows (StatSoft, 1995) was used for all statistical analyses.

## RESULTS

The results will be presented in four parts. The first two parts are the results of plant establishment and the measurements of the drainage water of the two different tailings. The third part gives results of the comparison between the effects of the two tailings. The fourth part gives results of the plant element uptake in the different treatments.

### Unlimed Tailings

The biomass of both plant species was highest when growing in SS (Table 3). Furthermore, tall cottongrass became established in all pots with the addition of SS, but only in three of the pots with ASM (Table 3). For white cottongrass, plant growth was established in all unlimed pots where seeds had been spread, except for one of the pots containing ASM. In comparison with



**Table 3.** Mean dry weight ( $\pm$ SE) of plants per pot, electrical conductivity (EC), pH, and  $\text{SO}_4^{2-}$  in the drainage water and  $\text{O}_2$  of water inside the tailings of pots with unlimed and limed mine tailings, supplied with different amendments with or without tall cottongrass (TC) or white cottongrass (WC). ASM; mixture of ash and sewage sludge (1:2 v/v); Control, no amendments and plants; SS, sewage sludge ( $n = 3$  to 6; Table 2).

	Dry weight		EC	pH	SO <sub>4</sub> <sup>2-</sup>	O <sub>2</sub>
	Shoot	Root				
	g					
Unlimed tailings						
Control			305 ± 10a***†	2.61 ± 0.01c***	1137 ± 47a	2.23 ± 0.09a
ASM			219 ± 19ab	3.15 ± 0.09c***	744 ± 51bc**	0.97 ± 0.10d**
SS			148 ± 32b	2.75 ± 0.32c***	394 ± 32d***	0.74 ± 0.08d
ASM + TC	0.33 ± 0.13c	0.43 ± 0.18c	216 ± 30ab	4.37 ± 0.51b	900 ± 98ac	1.77 ± 0.14b
SS + WC	2.32 ± 0.15b	3.02 ± 0.19b	159 ± 12b	5.59 ± 0.30a	761 ± 83bc	1.48 ± 0.10bc
ASM + TC	1.36 ± 0.02bc	1.95 ± 0.15bc	212 ± 32ab	6.46 ± 0.23a	1019 ± 150ab	1.77 ± 0.11ab
SS + TC	4.93 ± 0.62a	6.59 ± 0.87a**	150 ± 25 b**	5.85 ± 0.18a***	608 ± 37cd***	1.13 ± 0.12cd*
Limed tailings						
Control			205 ± 18y***	10.02 ± 0.55x***	1092 ± 29y	2.16 ± 0.07x
ASM			196 ± 04y	10.79 ± 0.14x***	988 ± 24y**	0.67 ± 0.05y**
SS			115 ± 06z	8.09 ± 0.16y***	634 ± 26z***	0.61 ± 0.07y
SS + TC	3.25 ± 0.42	2.69 ± 0.40**	266 ± 11x**	7.23 ± 0.11y***	1961 ± 160x***	0.77 ± 0.07y*

\* Significant at the 0.05 probability level.

\*\* Significant at the 0.01 probability level.

\*\*\* Significant at the 0.001 probability level.

† Different letters (a, b, c, and d for unlimed tailings and x, y, and z for limed tailings) in the same column indicate a significant difference at  $p < 0.05$  (Tukey's honest significant difference [HSD] test).

the control, the SS treatment (in the absence or presence of plants) decreased both EC and  $\text{SO}_4^{2-}$  levels of the drainage water (Table 3). The pH of the drainage water was higher (4.4–6.5) in the presence of plants, and similar to the pH at the onset of the experiment (approximately 5.0) compared with the pH in the absence of plants (2.6–3.2). Among the pots containing plants, the lowest pH value was found in pots with white cottongrass in ASM. This species seemed to have resulted in a lower pH compared with pots containing tall cottongrass (Fig. 2). The dissolved oxygen was significantly higher in the controls compared with all other treatments apart from ASM + tall cottongrass (Table 3).

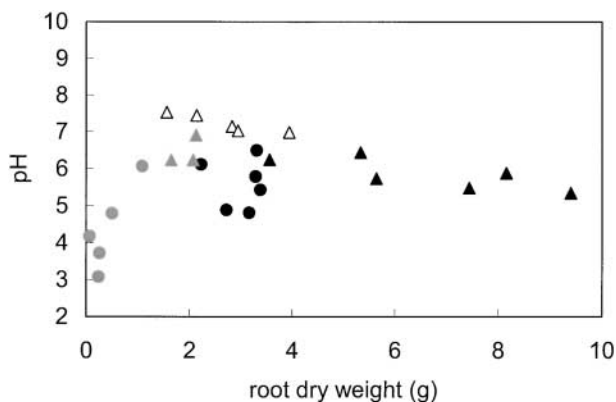
The metal concentrations of the drainage water were highest in the controls (Table 4). Furthermore, there was a tendency that the Cd, Cu, and Pb concentrations in the drainage water from treatments with plants and amendments were lower compared with only amendments. For As, no differences were observed in concen-

trations of the drainage water between the various treatments.

### Limed Tailings

In pots with limed tailings, tall cottongrass was established in five of the six pots with the addition of SS but there was no plant establishment in pots with ASM, and none of the white cottongrass plants were established (Table 3). The highest EC was found in drainage water from pots with both plants and SS, and the lowest were found in pots with only SS (Table 3). The EC level in the drainage water from controls and pots with ASM were similar and in between the other two treatments. The highest pH (10.0–10.8) was found in the drainage water of controls and with the addition of ASM in the absence of plants. This was a similar pH as that at the onset of the experiment, 10.9 (not shown). The addition of SS with and without plants reduced the pH of the drainage water (Table 3). The  $\text{SO}_4^{2-}$  levels of the drainage water were, in relation to controls, not affected by ASM while the addition of SS decreased and SS + tall cottongrass increased the  $\text{SO}_4^{2-}$  values. The dissolved oxygen level was significantly higher in the controls compared with the rest of the treatments.

No significant differences were shown for Cu and Pb concentrations of the drainage water (Table 4). Zinc concentrations were significantly higher in drainage water from SS + plant-treated tailings compared with only SS treatment. For Cd, the concentration of the water from treatment with SS was significantly lower compared with the ASM treatments. The As concentrations were lowest in the drainage water from the controls. Addition of ASM and SS gave a five-times-higher As concentration compared with the control, but in the presence of tall cottongrass the As concentration decreased (Table 4).



**Fig. 2.** Relation between pH of drainage water from pots and root dry weight per pot of white cottongrass (circles) and tall cottongrass (triangles). The two plant species were growing in unlimed (filled symbols) and limed (open symbols) tailings treated with ash + sewage sludge (gray) or sewage sludge as an amendment (black).

**Table 4.** Mean metal concentrations ( $\pm$ SE) in drainage water from pots with unlimed and lime-treated mine tailings with additions of different amendments with or without tall cottongrass (TC) or white cottongrass (WC). ASM, mixture of ash and sewage sludge (1:2 v/v); Control, no amendments and plants; SS, sewage sludge ( $n = 3$  to 6; Table 2).

	Cd	Zn	Cu	Pb	As
	$\mu\text{g L}^{-1}$	$\text{mg L}^{-1}$		$\mu\text{g L}^{-1}$	
<b>Unlimed tailings</b>					
Control	434.74 $\pm$ 49.89a***†	122.96 $\pm$ 14.03a***	3.21 $\pm$ 0.37a***	8134 $\pm$ 253a***	18.78 $\pm$ 3.62a
ASM	26.81 $\pm$ 6.58b**	6.23 $\pm$ 1.38b**	0.33 $\pm$ 0.11b*	1050 $\pm$ 308b*	14.61 $\pm$ 1.69a***
SS	29.15 $\pm$ 8.41b*	5.12 $\pm$ 1.07b**	0.30 $\pm$ 0.11b	5864 $\pm$ 1431a*	15.57 $\pm$ 3.41a***
ASM + TC	6.54 $\pm$ 0.47b	3.99 $\pm$ 1.07b	0.10 $\pm$ 0.05b	340 $\pm$ 36b	11.51 $\pm$ 1.24a
SS + WC	9.05 $\pm$ 1.21b	8.27 $\pm$ 1.72b	0.06 $\pm$ 0.02b	149 $\pm$ 49b	11.67 $\pm$ 1.65a
ASM + TC	5.72 $\pm$ 1.60b	1.99 $\pm$ 0.60b	0.02 $\pm$ 0.002b	57 $\pm$ 14b	13.49 $\pm$ 0.74a
SS + TC	6.83 $\pm$ 0.76b***	5.64 $\pm$ 0.88b**	0.04 $\pm$ 0.02b	92 $\pm$ 24b*	17.68 $\pm$ 2.20a*
<b>Limed tailings</b>					
Control	0.58 $\pm$ 0.20xy***	0.07 $\pm$ 0.02y***	0.10 $\pm$ 0.04x***	34 $\pm$ 8x***	19.14 $\pm$ 2.76z
ASM	0.04 $\pm$ 0.01y**	0.03 $\pm$ 0.004y**	0.06 $\pm$ 0.01x*	19 $\pm$ 3x*	117.03 $\pm$ 6.43x***
SS	0.74 $\pm$ 0.19x*	0.07 $\pm$ 0.04y**	0.05 $\pm$ 0.01x	9 $\pm$ 3x*	137.21 $\pm$ 15.48x***
SS + TC	0.61 $\pm$ 0.16xy***	0.28 $\pm$ 0.06x**	0.06 $\pm$ 0.01x	29 $\pm$ 5x*	64.10 $\pm$ 12.01y*

\* Significant at the 0.05 probability level.

\*\* Significant at the 0.01 probability level.

\*\*\* Significant at the 0.001 probability level.

† Different letters (a, b, and c for unlimed tailings and x, y, and z for limed tailings) in the same column indicate a significant difference at  $p < 0.05$  (Tukey's honest significant difference [HSD] test).

### Comparison of Drainage Water from Limed and Unlimed Tailings

The highest EC level of the drainage water was found in control pots with unlimed tailings (Table 3). Similar changes as in the EC due to the treatments were found in  $\text{SO}_4^{2-}$ , thus a positive correlation between these two parameters was found in both of the tailings (simple regression,  $p < 0.05$ ). All treatments in limed tailings had significantly higher pH and  $\text{SO}_4^{2-}$  (except the control) in the drainage water than in the same treatments of unlimed tailings (Table 3). In limed tailings, the  $\text{O}_2$  levels were significantly lower in ASM and SS + tall cottongrass treatment compared with unlimed tailings. The metal content of the drainage water from pots with

unlimed tailings was higher than in pots with limed tailings with the same treatment (Table 4). One exception was Cu, where SS treatment (with and without plants) did not show any significant difference between the two tailings. Furthermore, the As concentration was significantly higher in the drainage water from limed tailings compared with the same treatment with unlimed tailings, except in controls where the levels were similar.

### Plant Metal Content and Effect on pH

The concentration level of the measured elements, both in plants and in tailings, were  $\text{Zn} > \text{Pb} = \text{Cu} > \text{As} > \text{Cd}$  (Table 5). The concentration was always higher in roots than in shoots of all measured elements. There

**Table 5.** Mean ( $\pm$ SE) element concentrations in limed and unlimed tailings and in shoot and root of tall cottongrass (TC) and white cottongrass (WC) growing in the tailings with additions of different amendments. ASM, mixture of ash and sewage sludge (1:2 v/v); SS, sewage sludge ( $n = 3$  to 6; Table 2).

	Cd	Zn	Cu	Pb	As
	$\text{mg kg}^{-1}$ dry wt.				
<b>Shoots</b>					
Unlimed tailings					
ASM + WC	1.65 $\pm$ 0.53a†	2576 $\pm$ 691a	54.8 $\pm$ 9.0a	61.9 $\pm$ 9.6a	13.1 $\pm$ 3.2a
SS + WC	0.54 $\pm$ 0.08b	600 $\pm$ 133b	50.1 $\pm$ 10.2a	23.9 $\pm$ 6.7b	5.6 $\pm$ 1.1b
ASM + TC	0.96 $\pm$ 0.17ab	1018 $\pm$ 81ab	31.8 $\pm$ 2.3a	16.9 $\pm$ 3.3b	6.8 $\pm$ 2.1ab
SS + TC	0.57 $\pm$ 0.07b	779 $\pm$ 115b	53.3 $\pm$ 12.7a	14.9 $\pm$ 0.9b	3.4 $\pm$ 0.3b
Limed tailings					
SS + TC	0.52 $\pm$ 0.13b	211 $\pm$ 109b	20.9 $\pm$ 1.9a	24.7 $\pm$ 1.4b	4.3 $\pm$ 0.9b
<b>Roots</b>					
Unlimed tailings					
ASM + WC	5.73 $\pm$ 1.32x	3391 $\pm$ 667x	245.0 $\pm$ 30.7y	417.6 $\pm$ 91.5xy	146.2 $\pm$ 19.4x
SS + WC	6.23 $\pm$ 0.62x	1633 $\pm$ 313y	466.4 $\pm$ 55.3x	500.3 $\pm$ 34.4x	111.4 $\pm$ 29.3x
ASM + TC	5.71 $\pm$ 1.29x	1838 $\pm$ 605xy	329.0 $\pm$ 96.3xy	456.0 $\pm$ 62.3xy	121.4 $\pm$ 11.0x
SS + TC	4.56 $\pm$ 0.42x	1137 $\pm$ 158y	237.2 $\pm$ 34.4y	279.2 $\pm$ 36.9y	83.4 $\pm$ 11.5x
Limed tailings					
SS + TC	6.31 $\pm$ 0.85x	738 $\pm$ 198y	270.7 $\pm$ 46.4xy	268.3 $\pm$ 38.1y	85.3 $\pm$ 23.8x
<b>Tailings</b>					
Unlimed tailings					
Total	21.5 $\pm$ 0.8	6853 $\pm$ 295*	1069.5 $\pm$ 8.9***	1062.9 $\pm$ 21.8*	440.3 $\pm$ 11.1*
NH <sub>4</sub> OAc-extractable	2.5 $\pm$ 0.1***	810 $\pm$ 48	269.22 $\pm$ 1.5***	56.4 $\pm$ 0.6***	3.2 $\pm$ 0.3***
Limed tailings					
Total	20.3 $\pm$ 0.3	5800 $\pm$ 199*	913.9 $\pm$ 24.1***	1132.5 $\pm$ 16.5*	493.7 $\pm$ 15.7*
NH <sub>4</sub> OAc-extractable	4.7 $\pm$ 0.1***	886 $\pm$ 87	509.5 $\pm$ 16.1***	1.1 $\pm$ 0.8***	<0.001***

\* Significant at the 0.05 probability level.

\*\* Significant at the 0.01 probability level.

\*\*\* Significant at the 0.001 probability level.

† Different letters (a, b, and c for shoots and x, y, and z for roots) in the same column indicate a significant difference at  $p < 0.05$  (Tukey's honest significant difference [HSD] test).

was no significant difference in metal and As concentration between the two plant species. Neither was there any clear difference in the element concentrations of roots or shoots between treatments, or between unlimed and limed tailings, even though there were some differences in both total and  $\text{NH}_4\text{OAc}$ -extractable element concentrations (Table 5). In unlimed tailings, the shoot concentrations of Cd, Zn, Pb, and As in white cottongrass were significantly higher in treatments with ASM than with SS.

The correlation between pH and the root weight showed that the pH was between 5 and 7 when root dry weight was higher than 1 g (Fig. 2). The only significant correlation was found in limed tailings where the pH decreased with increasing root weight ( $r = -0.93$ ,  $p < 0.05$ ).

## DISCUSSION

The results were not found to be in accordance with the first hypothesis, that is, that the metal levels would increase due to plant establishment. Even though the  $\text{O}_2$  and  $\text{SO}_4^{2-}$  levels in most cases were higher in the drainage water where plants and amendments were added, compared with only amendments, the metal release had a tendency to be lower (only significant for Pb in SS treatment). This was not the case for Zn, which had similar levels of the two treatments (Tables 2 and 3). These results indicate that the weathering process of sulfides was not reduced but that the metals were stabilized by the high pH and plant uptake. The plants seemed to prefer a pH of between 5 and 6 and plants with higher root biomass (tall cottongrass) achieve this pH more easily than plants with lower root biomass (white cottongrass) (Fig. 2). A lower weathering process of sulfides might explain the low metal concentrations in the drainage water of amendment-treated tailings compared with controls (Table 4), since oxygen levels were low (Table 3). However, since the low pH indicates a sulfide weathering process, the low metal levels might have been due to adsorption and complex bonding by organic compounds in the amendments (Theodoratos et al., 2000). In addition, the metals can bind to inorganic compounds such as P (Cotter-Howells and Caporn, 1996; Schnoor, 1996) in sewage sludge. The metal concentrations were reduced in the drainage water. Thus, establishment of the two cottongrass species on untreated mine tailings reduces the use of lime to precipitate metals.

The second hypothesis, that plants will not affect the metal concentrations in the drainage water from limed tailings, was supported by the results, except in the case of Zn. Even though plant growth seemed to decrease the pH (Fig. 2) and increase the weathering, since the  $\text{SO}_4^{2-}$  levels were high (Table 3), the pH was still high enough (approximately 7.2) to stabilize the metals of the drainage water. The high As concentrations when amendments were added were probably due to the combination of high pH and low oxygen levels (Tables 2 and 3) (Masscheleyn et al., 1991).

The results were found to be in conformity with the

third hypothesis, that is, that plants can be established on submersed mine tailings amended with nutrients, and that different amendments might affect the metal uptake. As an amendment, SS was the most efficient nutrient supplier for plant establishment in the two tailings for both plant species in comparison with ASM (Table 3). Similarly, Borgegård and Rydin (1989) found that SS was an efficient nutrient supplier for plant growth. This result might be due to the fact that ash initially might have increased the pH (Greger et al., 1998) to a level that is unfavorable for seedling growth. Moreover, the amount of N was probably lower in the ASM since a release of N might have occurred when ash and sewage were mixed (Greger et al., 1998). Additionally, the ASM treatment only contained two-thirds of the SS (containing N) compared with the treatment with only SS. Thus, the reduced amounts of plant-available N in combination with a high pH might have caused the reduced plant growth in pots with ASM. Furthermore, the lower growth might also be a result of the higher shoot concentrations of Cd, Zn, Pb, and As in plants grown in ASM compared with SS (Table 5). The high shoot concentrations might also have affected the growth.

Tall cottongrass showed a great ability to adjust to different pH levels since the shoot biomass was equal in unlimed and limed tailings treated with SS (Table 3). However, the failure of tall cottongrass establishment in limed tailings with the addition of ASM might have been due to an excessively high pH level. White cottongrass did not survive at all in limed tailings, which might also be due to the high pH, which might have been expected since no white cottongrass plants were found in connection with the limed tailings in the field.

We conclude that plant establishment has an influence on both unlimed and limed tailings. Plants and amendments stabilize Cd, Cu, Pb, and Zn of the drainage water from submersed unlimed mine tailings. The use of lime to reduce metal mobility is an efficient method, but if acid is continuously generated the effect of lime will be reduced and the lime treatment has to be continuously repeated. Phytostabilization of metals in submersed mine tailings might be a cost-efficient method to maintain a high pH and thereby reduce the metal mobility. At least then, the need of lime treatment will decrease. Unfortunately, the As concentration is still a problem since it was not affected by plant establishment in unlimed tailings. However, in limed tailings, plant growth reduced As levels compared with amendment-treated limed tailings.

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