

# USING MUNICIPAL BIOSOLIDS IN COMBINATION WITH OTHER RESIDUALS TO RESTORE A VEGETATIVE COVER ON HEAVY METAL MINE TAILINGS

by

Sally L. Brown, Charles L. Henry, Harry Compton, Rufus L. Chaney and Pammella DeVolder

**Abstract.** High metal waste materials from the Bunker Hill, ID Superfund site are being collected in a central impoundment area. The waste materials have elevated metal concentrations with total Zn, Pb and Cd ranging from 6,000 – 14,700, 2100 – 4900, and 9 – 28 mg kg<sup>-1</sup>, respectively. They also contained minimal amounts of organic matter. In June, 1997 different mixtures of biosolids, wood ash and logyard debris were surface applied to determine if these materials would enable vegetative cover to be established directly on the surface of the waste material. Surface application of biosolids in combination with other residuals was able to restore a vegetative cover to the metal contaminated materials for three years following amendment application. Plant biomass in 1999 was 0.01 Mg ha<sup>-1</sup> in the control vs 3.4 Mg ha<sup>-1</sup> in amended plots. Metal concentrations of the vegetation indicate that plants were within normal concentrations for the 3 years that data was collected. Zinc concentrations in plant tissue in the amended areas was all below 90 mg kg<sup>-1</sup> for the 1999 growing season. Surface application of amendments was also able to reduce CaNO<sub>3</sub> extractable Zn in the subsoil from 159 in the control to 10 mg kg<sup>-1</sup>. These results indicate that surface application of biosolids and wood ash +/- logyard debris are sufficient to restore a vegetative cover to high metal materials for up to 3 years following application.

**Additional Key Words:** organic residuals, biosolids, contaminated mine sites, vegetation establishment

## Introduction

Bunker Hill, ID is the second largest Superfund site in the nation. Mining and smelting of Zn and Pb ores for most of the 20th century has resulted in extensive metal contamination of the surrounding hillsides and waterways. The area that falls under the Superfund designation is well over 600 ha. As part of their clean-up efforts, EPA has been depositing contaminated tailings and dredged materials in a Central Impoundment Area (CIA). These materials generally contain high concentrations of potentially phytotoxic

metals, little to no organic matter, are moderately to severely acidic and are deficient in necessary macro and micro-nutrients. Research was conducted by the University of Washington in cooperation with the USDA-ARS and US-EPA-ERT to test the ability of a range of soil amendments to assist in the establishment of a vegetative cover on the contaminated materials in the Central Impoundment Area.

Biosolids have been used both alone and in combination with other materials to restore soils that have been disturbed by a wide range of activities including coal and gravel mining. The use of biosolids is a long-standing practice with many sites having been successfully restored for 25 years or more (Sopper, 1993). Research has consistently demonstrated that biosolids are highly effective, in many cases more so than topsoil replacement, for restoration of disturbed ecosystems. It is also possible, using biosolids, to provide sufficient organic matter and nutrients in a single application to enable "one-shot" restoration. For conventional soil restoration practices, biosolids appear to be the most effective method that is currently available.

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<sup>2</sup>S.L. Brown is Research Assistant Professor, University of Washington, Seattle, WA 98195-2100, C.L. Henry is Research Associate Professor, University of Washington, Seattle, WA 98195-2100, H. Compton is Environmental Engineer, EPA-ERT, Edison, New Jersey, R.L. Chaney is a Research Scientist, Environmental Chemistry Lab, USDA ARS, Beltsville, MD, and P. DeVolder is a Graduate Student University of Washington, Seattle, WA 98195-2100

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In recent years, there has been a growing recognition that biosolids may also be used to restore metal affected ecosystems. Initial concerns about potential negative effects to plant and human health as a consequence of application of high metal biosolids to agricultural lands prompted a great deal of research on the behavior of metals in biosolids amended soils. Scientists have consistently demonstrated that metals in biosolids are much less available than equivalent metals added to soils as salts (Brown et al, 1998a). In addition, use of high quality biosolids generally results in no observable increase in plant metals (Brown et al, 1998a). From this research came the notion that high quality biosolids could potentially be used to limit metal toxicity in metal contaminated soils. Research at Palmerton, PA and Katowice, Poland demonstrated that application of biosolids in combination with high calcium carbonate equivalent residuals to highly metal contaminated soils was sufficient to restore a vegetative cover (Sopper, 1993, Stuczynski et al., 1997). The current research was undertaken to determine if a similar remediation mixture would be effective at restoring a self-sustaining vegetative cover on the CIA. Concerns about the associated costs of using high rates of materials, as well as the logistics of applying these amendments were factors in testing both low and high rates of biosolids at this site.

#### Materials and methods

##### Applications

Field plots were installed in June of 1997 and consisted of large scale treatment plots (33 x 33 m) in a completely randomized design with 3 replicates. The surface soils in this area consist of mining waste material with little to no organic matter. These materials contain 5500 – 14,700 mg kg<sup>-1</sup> Zn, 1500 to 4900 mg kg<sup>-1</sup> Pb, and 7 to 28 mg kg<sup>-1</sup> Cd. Soil pH ranged from 4.6 to 7.0. (Brown et al., 1998b). Amendments for this study included high N (4.4-5.3%) and low N (2.8%) biosolids applied at 55 and 110 Mg ha<sup>-1</sup> dry weight. The high N biosolids were produced through anaerobic digestion. The low N biosolids were treated in a lagoon. While all of the biosolids used in this study met US EPA CFR Part 503 requirements for Class B pathogen reduction, the lower N biosolids are much more stable. Longer retention time in the treatment lagoon is associated with greater decomposition of organic matter and a more stable end product. A list of treatments used in the study is presented in Table 1.

Biosolids were mixed with 220 Mg ha<sup>-1</sup> wood ash wet weight (to provide the calcium carbonate equivalent of 55 Mg ha<sup>-1</sup>) before application. Logyard waste (20% by volume) was mixed with the low N biosolids treatments and two of the high N treatments to reduce the potential for nutrient runoff by increasing the C:N ratio of the amendments. A single high N biosolids treatment (N3) was applied without logyard waste at the high application rate (110 Mg ha<sup>-1</sup>). All amendments were mixed with a front-end loader immediately prior to application. Materials were surface applied using a side cast thrower that was mounted on a 6-wheel drive vehicle. The throw distance of the mixture was approximately 60 m. There was no attempt made to incorporate the amendments. A control treatment was also included.

##### Seeding

Seeds were hand scattered on the surface of the amendments immediately prior to application. A native seed mixture that had been used elsewhere on the site was used as the initial seed mix. Although this type of seeding technique had been successfully used at Palmerton, PA, the high pH of the wood ash (pH > 11) in combination with the high N content of the biosolids resulted in sufficient ammonia volatilized to kill of the seeds in the high N biosolids treatments. Subsequent reseeded as well as volunteer species were sufficient to establish a vegetative cover later in the first growing season. Additional studies at the site suggest that a waiting period of 48 hours is sufficient to avoid seed germination failure due to excess ammonia. Use of a less caustic lime product would also reduce the rate of ammonia volatilization.

##### Sampling

Plant samples for elemental analysis were collected in August, 1997, July, 1998 and June, 1999. In each case, a composite sample, consisting of a minimum of three subsamples, was collected from each plot for elemental analysis. For this purpose, only grass samples were collected. The samples were washed and rinsed in deionized or distilled water. Samples were ashed at 480 C°, digested with concentrated HNO<sub>3</sub>, and analyzed using a flame atomic adsorption spectrometer or an inductively coupled plasma spectrometer. Values were corrected for background variation through the use of blanks, and the inclusion of replicate samples. NIST plant standards were routinely included in the digests. Percent cover was measured in 1998 using the line transect method. Three measures, each 17 m in length were taken on each plot. Occurrence of plants within

each 20 cm increment was interpreted as being vegetated. Harvestable biomass was measured in 1999. Samples for biomass measurements were collected from 3 areas of each plot using a circular measure that was 615 cm<sup>2</sup>. All plants within the circular measure were included in the biomass measurements. For all years,

plants were randomly collected within each treatment to observe rooting depths and extent of nodulation. In addition to plant samples, soil samples were collected in the 0-15 cm horizon directly below the amendment in 1999.

Table 1. Characteristics of the amendments (values are the mean of 3 replicates, ± std dev).

	Zn	Cd	Pb	pH	Carbon	Nitrogen	Solids	Depth
	----- mg/kg -----				----- % -----			(cm)
<b>Low N</b>								
25 Mg ha	1809±409	6.1±0.7	265±95	8.4±0.4	17±2	0.5±0.1	60±5	4.1±2.5
50 Mg ha	1143±182	9.0±0.6	217±17	7.6±0.1	20±2	1.1±0.2	49±7	6.1±2.3
<b>High N</b>								
25 Mg ha	1213±438	2.7±0.1	102±15	8.7±0.2	19±3	1.0±0.2	45±3	4.6±1.8
50 Mg ha	873±103	2.7±0.1	228±34	8.5±0.1	21±4	1.8±0.6	37±3	8.1±2.3
<b>High N (2&amp;3)</b>								
50 Mg ha	554±20	2.6±0.1	168±30	8.4±0.1	24±1	2.2±0.3	30±4	5.8±1.8

These samples were analyzed for pH and extractable metals using a 0.01 M CaNO<sub>3</sub> extraction. Samples from the amended horizon were collected in all years and analyzed for carbon and nitrogen concentrations. It has been suggested that the C:N ratio of the soil over time will give an indication of the amount of microbial decomposition and therefore nutrient cycling in a soil system (Steve McGrath, IACR, Rothamstead, UK, personnel communication).

## Results and Discussion

### Soils

Soils were analyzed for CaNO<sub>3</sub> extractable metals. This extraction was designed to mimic the soil solution. In cases of contaminated soils, soil solution metals have been well correlated with plant available metals. Results from this extraction are presented in Table 2.

The reduction in CaNO<sub>3</sub> extractable Zn along with increased soil pH that was observed in the subsoil under several treatments suggests that the alkalinity added with the surface amendment was partially mobile in the soil solution. Reduced concentrations of Zn in the soil solution indicate a reduction in phytotoxicity. The proliferation of roots into the subsoil in the Phase I plots (visual observation) may be a result of the

amendment's ability to reduce bioavailable metal concentrations in the subsoil. These results also suggest that the higher N biosolids which are less stable and decompose more readily than the low N biosolids, are more effective at translocating alkalinity to the untreated subsoil. Increased subsoil pH and extractable Zn concentrations in the high rate of the low N biosolids treatment are indications that this treatment was less effective at reducing subsoil phytotoxicity and would also be less successful at maintaining a stable plant cover for an extended period. The reason for the lower subsoil extractable Zn and higher pH in the lower application rate of this treatment is not clear. Addition of logyard waste appeared to have no effect on subsoil pH concentrations or extractable metal concentrations.

Changes in the C:N ratio of the amendment horizon over the three year period of the study are presented in Figure 1. A stable C:N ratio of approximately 20:1 is indicative of a well functioning soil system. The initial amendment, at the high rate of application of the high N biosolids, had C:N ratios of under 12:1. This indicates a surplus of N in the system. As plants grow on the soil and plant tissue is deposited on the soil surface, the C:N ratio will increase. A ratio significantly over 25:1 indicates that there is not a sufficiently active soil community to decompose plant residues. If this is the case, the nutrients that have

accumulated in plant tissue will not be recycled in the soil system and so will not be available for future growing seasons. The C:N ratios in Figure I indicate that this ratio is within the healthy range for all of the high application rate of the low and high N biosolids treatments. For the low rate of both types of biosolids, the ratio either appears to be increasing (high N treatment) or remaining relatively high (low N treatment). This may suggest that there is not sufficient nutrient cycling in these systems to maintain a self-sustaining vegetative cover. The addition of logyard waste did not appear to change the nutrient cycling dynamics in the high N biosolids treatment.

Table 2. Soil pH and extractable (0.01 M CaNO<sub>3</sub>) Zn of the amendments and subsoil in 1999

Treatment	pH	Extractable Zinc (mg kg <sup>-1</sup> )
Control	5.8±0.9	150±60
Low N 110 amendment	7.1±0.5	1.9±0.3
soil	5.9±1.0	87±60
Low N 55 amendment	7.8±0.1	0.8±0.1
soil	7.3±0.4	11±1
High N3 110 amendment	7.4±0.1	1.0±0.1
soil	7.6±0.2	12±2
High N 110 amendment	7.3±0.3	1.0±0.1
soil	7.0±0.6	14±6
High N 55 amendment	7.8±0.2	1.7±0.6
soil	7.5±0.5	9±2

## Plants

**Diversity and rooting patterns.** The native mix that had been used on the hillside revegetation study was also used for the initial seeding of the Phase I plots. Germination of a wide range of grasses and legumes was observed on the low N biosolids treatments. The ammonia toxicity on the high N treatments killed all seedlings. Volunteer species began colonizing these plots before they were reseeded. These species included crab grass and bunch grasses. Subsequent reseeding of these plots was done using a wheat grass and vetch mixture. Random plants were pulled from several treatments during each sampling period. Roots for all plants pulled had penetrated the subsoil below

the amendment. Root growth into the subsoil was extensive. Low concentrations of extractable Zn may have enabled root growth into the subsoil. In addition, legumes checked showed effective nodulation. Root nodules were cut open and were bright pink, indicating that rhizobia were actively fixing N.

**Biomass.** Percent cover and harvestable biomass for the plots are presented in Table 2. Highest biomass was found in the high application rate of the low N biosolids as well as all rates of the high N biosolids. The absence of logyard waste in the amendment mix appeared to have no effect on plant cover. The high concentrations of soluble Zn in the subsoil of the high rate of the low N biosolids did not appear to have any impact on biomass. It is possible that there was sufficient moisture in the early part of the growing season (measurements were made in June, 1999) so that plants did not need to access moisture in the subsoil. In cases of limited rainfall, high Zn concentrations in the subsoil would effectively prohibit root growth into this horizon and would thereby limit access to soil water.

Table 3. Percent cover on Phase I plots in July, 1998 and harvestable biomass in June, 1999. Reported values are the means of 3 replicates (± std dev).

Treatment	Rate (Mg ha <sup>-1</sup> )	% Cover	Harvestable biomass (Mg ha <sup>-1</sup> )
Control		0%	0±0
Biosolids			
Low N	55	77%	1.6±1.0
	110	93%	2.5±1.6
High N	55	82%	4.0±1.3
	110	93%	2.8±1.3
High N - Logyard	110	95%	3.0±1.3

**Plant metal and nutrient status.** Plant metals were measured on these plots in 1997, 1998 and 1999. Results of these analyses are presented in Table 4. There was no vegetation on the control plots in 1998. The decrease in plant Zn, Cd, and Pb from 1997 to 1998 is not unusual. Metals in biosolids amended soil are generally more phytoavailable during the first growing season following application (Brown et al., 1998a). Results from this study are consistent with results of studies using biosolids on agricultural soils. Concentrations of Zn in the biosolids amended soils are similar to those expected for plants grown on uncontaminated soils. The increase in plant Zn from 1998 to 1999 is potentially the result of seasonal

variation. Additional monitoring would be necessary to confirm this observation. In all years when there was harvestable plant tissue on the control plots, plant analysis indicated severe P deficiency. This type of deficiency is relatively common for plants growing on high Zn and Pb soils. P tends to form insoluble complexes with these metals and is generally limiting

for plant growth. Phosphorus concentrations in the amended treatments were all within normal ranges, however, there was a decrease in plant P in the 1999 growing season. All of the amendments contained excess P for plant growth. This decrease may indicate that the excess P has formed insoluble precipitates and is no longer available to plants. After the first growing

Table 4. Elemental concentrations of plant tissue grown on Phase I plots ( $\pm$  std dev).

App. Rate	Plant Cd	Plant Cu	Plant P	Plant Pb	Plant Zn
----- (mg kg <sup>-1</sup> ) -----					
<b>1997</b>					
Normal Plant	0.1-1.5	5.0-15	>1500	0.1-6	20-400
Control	0.4 $\pm$ 0.1	3.9 $\pm$ 0.9	900 $\pm$ 200	8.5 $\pm$ 2	219 $\pm$ 20
Low N 55	1.4 $\pm$ 0.3	12 $\pm$ 1	3900 $\pm$ 400	11.8 $\pm$ 2	105 $\pm$ 20
110	2.3 $\pm$ 0.6	11.4 $\pm$ 0.3	2400 $\pm$ 1000	15.4 $\pm$ 5	152 $\pm$ 20
High N 55	1.2 $\pm$ 0.2	16 $\pm$ 2	4500 $\pm$ 400	16.2 $\pm$ 0.5	129 $\pm$ 1
110	0.6 $\pm$ 0.3	10 $\pm$ 3	7400 $\pm$ 3000	6.7 $\pm$ 0.6	114 $\pm$ 25
High N3 110	0.9 $\pm$ 0.7	14 $\pm$ 1	7000 $\pm$ 400	8.4 $\pm$ 0.7	70 $\pm$ 6
<b>1998</b>					
Low N 55	0.7 $\pm$ 0.1	4 $\pm$ 1.0	3100 $\pm$ 400	2.3 $\pm$ 0.6	38 $\pm$ 5
110	1.7 $\pm$ 1.2	5.8 $\pm$ 0.3	3200 $\pm$ 20	2.7 $\pm$ 0.4	61 $\pm$ 13
High N 55	0.9 $\pm$ 0.1	5.1 $\pm$ 0.5	3800 $\pm$ 400	2.7 $\pm$ 1.0	47 $\pm$ 7
110	0.9 $\pm$ 0.2	9 $\pm$ 3	5100 $\pm$ 600	3.0 $\pm$ 0.7	59 $\pm$ 12
High N3 110	0.8 $\pm$ 0.3	6 $\pm$ 2	3800 $\pm$ 200	2.8 $\pm$ 1.2	48 $\pm$ 7
<b>1999</b>					
Control	0.50 $\pm$ 0.06	5 $\pm$ 0.5	700 $\pm$ 100	27 $\pm$ 9	169 $\pm$ 10
Low N 55	0.5 $\pm$ 0.1	5 $\pm$ 0	1790 $\pm$ 60	3.5 $\pm$ 0.8	63 $\pm$ 3
110	0.6 $\pm$ 0.4	5 $\pm$ 0.5	1800 $\pm$ 100	2.6 $\pm$ 0.9	94 $\pm$ 10
High N 55	0.28 $\pm$ 0.04	3.0 $\pm$ 0.5	1700 $\pm$ 200	3.0 $\pm$ 0.4	47 $\pm$ 10
110	0.40 $\pm$ 0.08	5 $\pm$ 0.5	2100 $\pm$ 300	4.0 $\pm$ 1.0	85 $\pm$ 4
High N3 110	0.6 $\pm$ 0.1	6.0 $\pm$ 2	2500 $\pm$ 800	4.8 $\pm$ 0.8	89 $\pm$ 10

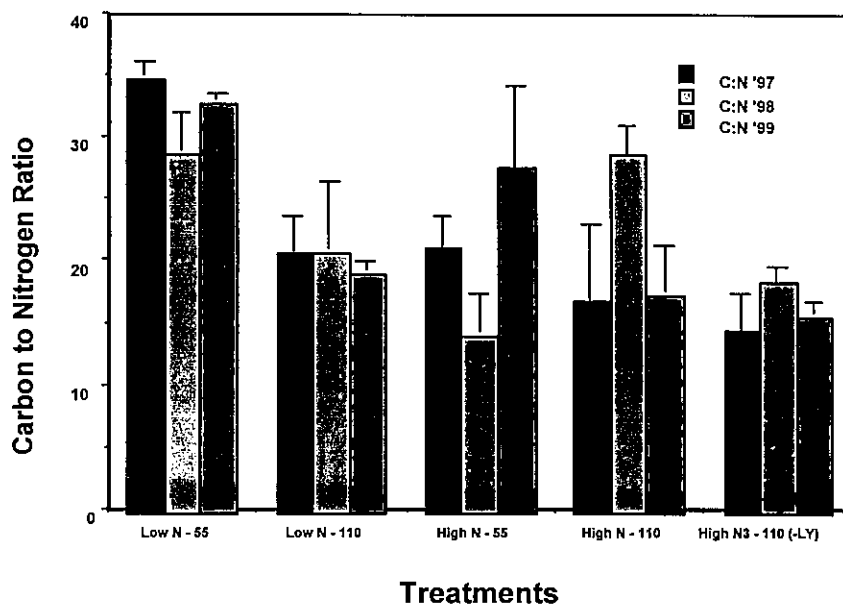


Figure 1. Changes in the C:N ratio of the amendment horizon over the three year period of the study.

season, plant concentrations of Zn and Pb were significantly lower in the amended as compared to control treatments. Cadmium concentrations for all treatments are similar for all years. Copper concentrations are comparable in all treatments across all years with the exception of the low rate of the high N biosolids + log yard waste in 1999. In this treatment, plant Cu concentrations are approaching deficiency. There was a significant amount of copper added with the biosolids and wood ash amendment and this deficiency may be the result of high soil pH and the dissolution of organo-Cu complexes that had kept Cu solubility high in the initial years of the study.

#### Conclusions

All of the biosolids treatments were effective in establishing a vigorous plant cover. The low rate of the low N biosolids did not achieve complete coverage of the plot area, indicating that the higher rate of amendment would be superior for plant growth. High extractable Zn concentrations in the subsoil of the high rate of the low N biosolids also suggest that the use of the high N materials (more chemically reactive) is a superior remedial alternative to the low N materials. Addition of logyard debris to the high N biosolids

treatments appeared to have no significant effect on plant growth.

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