

# FORUM

## Reclamation of Wastes Contaminated by Copper, Lead, and Zinc

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**ABSTRACT** / Waste materials containing toxic levels of copper, lead, and zinc, such as mine and smelter wastes, present difficult conditions for the establishment of vegetation. This article reviews the many attempts which have been made to reclaim these wastes. Inert wastes from mining and

quarrying operations, such as slate quarry waste and certain colliery shales, seem to be good materials for reclaiming wastes contaminated by copper, lead, and zinc. Organic wastes, such as sewage sludge and domestic refuse, may provide only a temporary visual improvement and stabilization of the toxic materials.

Nontolerant plant materials may often be planted directly on modern waste materials, which are less toxic than they were in the past. However, tolerant plant materials are needed for revegetating waste materials produced by early and more primitive extraction methods.

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Second only to agriculture, mining is one of the oldest and most important industries in the world. During the past 20 years, the exploitation of mineral resources has intensified rapidly to cope with the rising demand for various minerals.

A variety of environmental problems, including air, land, and water pollution, arise from mining activities. The pollutants may be directly or indirectly hazardous to man. Other minor environmental problems resulting from mining include dust formation and noise pollution, as well as nuisance. The detrimental impact of mining upon land, air, and water has been a topic of concern by different nations for centuries. However, only in the last three decades have the problems become so acute that remedial measures have been used extensively.

Copper (Cu), lead (Pb), and zinc (Zn) are the most extensively mined metals in the world, and they very often occur in waste materials at concentrations toxic to plants. The total metal content of some typical mine waste which contains metals other than the one being mined is given in Table 1. These concentrations make the establishment of vegetation difficult, yet attempts have been made to restore mined lands, including surface amelioration using inert materials, application of soil and organic substrates to the surface layers of the toxic mineral wastes or incorporation of the substrates within them, and direct revegetation involving tolerant or nontolerant plant materials.

The present article reviews the various attempts to solve problems arising from mine and smelter wastes containing copper, lead, and zinc.

### Surface Amelioration: Inert Substrates, Topsoil, Sewage Sludge and Other Organic Substrates

Several inert materials, including colliery spoils and steel slag, were compared with domestic refuse, sewage sludge, burnt colliery spoil, and topsoil for the surface treatment of metalliferous wastes contaminated by lead and zinc (Johnson and others 1977). The steel slag amendment failed to support vegetation cover, owing to the low rate of germination and to seedling deterioration during the later phase of establishment caused by the rapid settlement at depth of water-retaining fine material. Domestic refuse, topsoil, and sewage sludge mixed with burnt colliery spoil had higher overall yields. However, swards established on these materials deteriorated within four months of sowing, exhibiting symptoms of metal toxicity caused by the accumulation of both lead and zinc. This toxicity was due to the deep-rooting nature of the developing swards caused by the high fertility of these organic substrates. The swards established on burnt or unburnt colliery spoil alone did not exhibit symptoms of metal toxicity. This was attributed to the consistent, shallow-rooting nature of the developing swards and the failure of roots to penetrate to the underlying toxic spoil.

Burnt and unburnt colliery spoils seem able to support the development of a visually acceptable sward which requires minimal long-term maintenance for amenity or recreational purposes. Furthermore, swards grown on burnt and unburnt colliery spoil contained significantly less lead and zinc than did those grown on other amendments with organic substrates (Johnson and others 1977).

At Cyprus Pima Mine, an open-pit copper mine southwest of Tucson, Arizona, USA, four soil mate-

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Table 1. Total metal content of some mine wastes.

Type of mine	Location	Metal content of waste (ppm)		
		Zn	Pb	Cu
Zinc	Trelogan, Flintshire	35,900	10,500	80
	Halkyn, Flintshire	11,700	7,400	60
	Snailbeach, Shropshire	20,500	20,900	30
Copper	Ecton, Staffordshire	20,200	29,900	15,400
Lead	Goginan, Cardiganshire	1,400	15,400	210
	Van, Montgomeryshire	1,200	40,500	90
	Glenridding, Westmorland	2,300	5,700	40

After Smith and Bradshaw (1972).

rials—desert soil, copper overburden, overburden plus copper mine tailings, and tailings—were tested for the growth of five tree species, including desert tobacco (*Nicotiana glauca*), tiny capsule (*Eucalyptus microtheca*), silver dollar gum (*E. polyanthemos*), red gum (*E. rostrata*), and velvet mesquite (*Prosopis juliflora*). It was revealed that desert soil had the highest productivity, followed by overburden, overburden plus tailings, and tailings, in decreasing order (Day and Ludeke 1980).

Subsequent experiments involved planting three legumes—blue lupine (*Lupinus hirsutus*), sesbania (*Sesbania macrocarpa*), and alfalfa (*Medicago sativa*) (Day and Ludeke 1981)—and six perennial grass species—perennial ryegrass (*Lolium perenne*), crested wheatgrass (*Agropyron cristatum*), Lehmann lovegrass (*E. superba*), and blue panicgrass (*Panicum antidotale*) (Day and Ludeke 1982)—and yielded a similar result, indicating that overburden might be a convenient source of reclamation material.

The processing of lead–zinc and copper ores at Mount Isa, North Australia, produced large quantities of concentrated tailings waste. The waste was pumped as a slurry to special dams, where the solids were allowed to settle and dry out. The waste was amended with reverberatory furnace slag, organic residue from the sewage works, straw mulch, and fly ash. Identical species, seeding rates, and fertilizer additions (2000 kg superphosphate and 400 kg ammonium nitrate/ha) were used for all treatments. It was discovered that the best amendment treatment was the incorporation of fly ash, a waste product of a coal-fired power station.

Standard soil tests showed that the addition of fly ash improved the permeability and water infiltration properties of the tailings material while soil compaction was significantly reduced (Ruschena and others 1974).

Drainage from the copper, silver, and gold smelters at the Captains Flat Mine, New South Wales, Australia,

initially caused serious water pollution. An impermeable and continuous 15-cm clay layer and soil which provided a medium for the vegetative cover were added to the drainage area. A rock layer acted as a barrier to prevent the capillary rise of polluted water above the clay layer. In addition, it also provided free lateral drainage of water infiltrating downward through the topsoil layer and prevented the erosion of the clay layer. After the addition of a topsoil layer, the area was cultivated, sown with legumes and grasses, limed and fertilized with nitrogen (N), phosphorus (P), and potassium (K) in the ratio 2:3:12. A marked reduction in pollution of the river waters occurred below the mine area, with a decrease in acidity from pH 3.8 to 6.0 and a marked reduction in concentrations of metallic salts in solution (Craze 1977).

In revegetating zinc and copper smelter waste in the Lower Swansea Valley (UK), domestic refuse, inorganic fertilizer and lime, and sewage sludge were added before planting several tolerant populations of grasses (*Agrostis tenuis*, *A. stolonifera*, *Festuca ovina*, and *Anthoxanthum odoratum*). There was no difference in response of the different species to applications of inorganic fertilizer and lime, but species differed in their response to a range of treatments involving the addition of domestic refuse or sewage sludge (Gadgil 1969). In general, the amendment of organic waste, in conjunction with the addition of fertilizer and lime, increased the growth of grasses. However, nontolerant cultivars established on organically amended waste exhibited regression after 12 months when planted on the nonferrous smelter wastes in the Lower Swansea Valley (Street and Goodman 1966, Pitcairn 1969). Sward regression occurred because of a decline in nutrient levels and gradual lowering of the organic matter status. It was also shown that roots penetrated to the boundary of the surface treatment and into the underlying toxic material and therefore were vulnerable to the toxic metals (Goodman and others 1973).

Similar problems have also been reported when using shallow dressings of soil to cover lead–zinc or copper waste in the Peak District, North Wales and North Pennines, UK (Smith 1973). Nontolerant grass species were sown on a 10-cm layer of sewage sludge at the Glenridding lead–zinc mine in the English Lake District in 1953, with considerable success at the beginning. However, erosion has now been initiated, because of the overgrazing by sheep and the dieback of vegetation caused by the spread of toxicity to the surface. The area is more susceptible to erosion, as the plants tend to root only in the sewage, which forms an outer layer more or less unattached to the underlying spoil (Smith and Bradshaw 1972).

Topsoil and organic substrates, such as sewage sludge and peat, serve as surface amendments for covering the tailings waste that normally lacks organic matter and essential plant nutrients. Furthermore, organic matter complexes heavy metals, which are therefore unavailable to plants, and it improves water holding capacity (Dean and Heavens 1973). The toxicity of metalliferous waste of zinc, nickel, copper, and sometimes chromium can therefore be combated by incorporating materials that possess a high organic matter, although it is rare for these metals to be at levels high enough to cause toxicity (Gemmell 1977). Upward movement of soluble metal salts may also cause toxicity due to evapotranspiration, regardless of amendment depth (Breeze 1973, Gemmell 1973). As a consequence, plants may deteriorate even in the absence of direct contact between roots and the toxic materials.

Owing to the gradual lowering of the organic matter and nutrient levels, regular maintenance is essential. Annual dressings of inorganic fertilizer and the application of 20–100 mm of organic matter have been suggested to maintain the vegetation cover on zinc, copper, and iron wastes in the Lower Swansea Valley (Goodman and others 1973), but the results are inconclusive.

Hay or barley straw has been suggested as a mulch on copper mine tailings and has several advantages. First, it insulates the surface from temperate extremes, thus stabilizing the soil temperature and thereby creating a better environment for seeding germination. Second, it permits the soil to absorb moisture and also creates small dams which reduce water velocity and erosion. Third, it enriches the growth of microorganisms (Ludeke 1973).

Other waste materials have also been suggested, and the idea of using one waste product to cope with the problems of another is appealing. Codisposal of wastes will therefore ease the disposal problem and at

the same time mitigate pollution. However, each waste material has its own distinct property (Table 2). Field trials should be conducted before applying any of them to toxic mine wastes on a large scale.

### Direct Seeding: Nontolerant and Tolerant Plant Materials

A comparison of the growth of commercial and metal-tolerant populations of *Festuca rubra* on recent lead–zinc mine wastes indicated that concentrations of lead and zinc were not so toxic as to prevent growth and development of commercial varieties of grasses. The major limitation was found to be the very low level of nitrogen and phosphorus (Johnson and others 1976). The lower concentrations of metals were probably due to the more efficient extraction methods employed recently.

Seeds of a commercial origin (grasses: *Festuca rubra* subsp. *rubra*, *F. rubra* subsp. *commutata*, *F. ovina*, *F. longifolia*, *Agrostis tenuis*, *A. stolonifera*, *Poa pratensis*; legumes: *Lotus corniculatus*, *Trifolium dubium*, *Medicago lupulina*) were hydraulically seeded along with fertilizer and wood pulp on Pb and Zn dumps (total concentration of Pb 1000–9900 ppm, Zn 2000–63,000 ppm) at Tynagh, County Galway, Ireland. The areas were previously covered with limestone chips (5 cm) and nontoxic, calcareous glacial drift (4 cm) to dilute the toxic surface and provide a microbial inoculum and improved cation-exchange properties. The vegetation cover was complete, and a well-developed root mat had been formed 20 months after the seeds were sown (Jeffrey and others 1974).

Commercial grass seed mixture was also used in the reclamation of a nickel tailings area at Port Colborne and a copper tailings area near Sudbury, Ontario, Canada. Feed grade lime (7.5 tonnes/ha) and fertilizer of 5–20–20 (N–P–K, 480 kg/ha) were applied, followed by seeding and another application of 5–20–20 fertilizer (N–P–K, 336 kg/ha). Fall rye, harvested from the last year's seeding, was sown as a companion crop at the rate of 336 kg/ha. The grass seed mixture consisted of 550 kg of Canada bluegrass and 5.7 kg of mixed seeds (containing 1 part timothy, 2 parts redtop, 1 part Kentucky bluegrass, 1 part crested wheatgrass, and 1 part creeping red fescue). Bromegrass was also seeded at the rate of 12 kg/ha afterward. Once grass was established, the annual maintenance consisted of (a) 90 kg of 5–20–20 fertilizer (N–P–K) applied immediately after cutting, and (b) urea applied at the rate of 120 kg/ha in late fall. On longer-established areas, both fertilizer and lime were applied as indicated by a soil analysis. In some areas, grasses

Table 2. Properties of waste materials which can be used as amendments for toxic mine wastes.

	Composition (%)				Possible effects
	N	P	K	Organic matter	
Farmyard manure	0.6	0.1	0.5	24	Can be toxic direct on plants
Pig slurry	0.2	0.1	0.2	3	High water content
Poultry manure					
Broiler	2.3	0.9	1.6	68	High levels of ammonia
Battery	1.5	0.5	0.6	34	High levels of ammonia
Sewage sludge (air dried)	2.0	0.3	0.2	45	Possible toxic metals and can have high water content
Peat (partly dried)	0.1	0.005	0.002	50	Variable, especially calcium content
Mushroom compost (dried)	2.8	0.2	0.8	95	None except high lime content
Domestic refuse (municipal)	0.5	0.2	0.3	65	Miscellaneous objects
Straw	0.5	0.1	0.8	95	Adverse C/N ratio
Building rubble	0.05	1.8	2.0	0.5	Brick and pieces of masonry
Colliery spoil	0.03	0.04	0.4	0	Possible high levels of pyrite causing acidity
Pulverized fuel ash	0	0.05	2.2	0	High boron

After Bradshaw and Chadwick (1980).

were harvested and marketed as hay for livestock (Peters 1970).

A wide range of commercially available nontolerant plant materials, including trees, shrubs, cacti and other desert plants, grasses, forbs and legumes, were hydro-seeded and used for reclaiming copper tailings at the Pima Mine in Arizona. To insure germination and provide a better growth medium, sewage effluent was applied to the tailing slopes at a rate of 11,233 liters/ha along with 12.6 tonnes/ha of compost manure. It was concluded that the ultimate objective of maintaining the natural splendor of a low desert environment in the disposal area had been achieved by the success of the vegetative stabilization at a reasonable cost (Ludeke 1973).

Bougainville is the site of the first very large mining development in Papua New Guinea. Attempts have been made to reclaim copper tailings containing 0.069% copper by distributing inoculated seeds of *Desmodium intortum* into the tailings that had been cultivated to a depth of about 10 cm. Germination was inhibited by inclement weather followed by a series of storms. Seeds were then germinated in small peat pots and transplanted on the sites but, as in the previous procedure, plants were not established because the peat containers dried out, thereby preventing root penetration.

Attention was then focused on the possibility of using tree species to stabilize the tailings surfaces and provide organic enrichment through leaf falls. It was confirmed that *Eucalyptus tereticornis* would be used to pioneer growth on the tailings, provided there were sufficient nutrients. The absence of naturally occurring nitrogen and severe deficiencies of phosphorus and boron made artificial fertilizers necessary for seedling establishment. For planting at a 5 × 5 m density, the fertilizer application was equivalent to 80 kg nitrogen, 40 kg phosphorus, and 0.8 kg boron/ha annually. Preliminary results suggested that *Rhizophora apiculata* and *Bruguiera gynanorhiza* seedlings developed stilt roots after four months on the soft sediment at the mouth of Jaba River, which receives tailing discharges from the Bougainville copper mine (Hartley 1976).

Mine wastes often contain excessive concentrations of metals that are toxic to plants, and hence these wastes are often devoid of vegetation. Studies of *Agrostis tenuis* populations from different habitats indicated that plants collected from an old lead mine site in Wales grew on mine wastes, whereas plants from uncontaminated sites died (Bradshaw 1952). It has also been proved that several native species of mine tailings required only minimal amendments to the Cu, Pb, and Zn tailings in Canada and had a good chance

of long-term survival. *Arctagrostis latifolia* was the most successful, and the others included *Artemisia tilesii*, *Carex bigelowii*, *Hierochloa alpina*, *Deschampsia caespitosa*, and *Hordeum jubatum* (Kuja and Hutchinson 1979).

The existence of tolerant races on metal mines is the result of natural selection rather than innate physiological tolerance (Bradshaw 1976). A comparison of plant roots of mine and nonmine populations grown in a solution of metal salts suggested that a limited range of species existed on various metal mine wastes because they have evolved tolerant populations (Smith and Bradshaw 1972). Some tolerant cultivars derived from three different mine sites have multiplied successfully on a commercial scale as follows:

Merlin: red fescue (*Festuca rubra*) for neutral and calcareous lead/zinc wastes

Goginan: bent grass (*Agrostis tenuis*) for neutral and acidic lead/zinc wastes

Parys: bent grass (*Agrostis tenuis*) for neutral and acidic copper wastes.

The practical value of using tolerant varieties on tailings with high levels of copper, lead, and zinc has been demonstrated by Smith and Bradshaw (1972). On the majority of sites, tolerant varieties were considerably superior to their nontolerant equivalents. Furthermore, tolerant plant materials can withstand adverse site conditions other than those associated with toxic metals. For example, it has been noted that tolerant plants survive drought on the coarser grades of mine soil (Smith and Bradshaw 1972) and even on nontoxic soils (Humphrey and Bradshaw 1976) better than nontolerant plants. Tolerant *Agrostis tenuis* plants from an acidic lead mine were found to be adapted to low levels of both calcium and phosphate (Jowett 1959).

Regular applications of fertilizer are necessary to ensure a continuous supply of nutrients and to prevent any deterioration of the established vegetation cover. However, it has been shown that metal-tolerant populations have the capacity to grow on toxic waste without the addition of organic matter or lime, unless the metal levels are too high (Gemmell 1973).

The importance of fertilizer treatments and the use of tolerant plant populations have been demonstrated by Goodman and others (1973). It was indicated that, in the presence of fertilizer (N, P, and K), copper-tolerant *Agrostis tenuis* thrived in pure copper waste. However, zinc tolerance would not mitigate the dominant toxicity of pure zinc wastes unless a considerable dilution was made with ash and peat. It was concluded, therefore, that the two logical alternatives for

Table 3. Some native species growing on toxic mine wastes in temperate and tropical regions.

Temperate	Tropical
<i>Agrostis stolonifera</i>	<i>Andropogon gavanus</i>
<i>Agrostis tenuis</i>	<i>Becium homblei</i>
<i>Anthoxanthum odoratum</i>	<i>Celozia trigyna</i>
<i>Armeria maritima</i>	<i>Combretum molle</i>
<i>Festuca rubra</i>	<i>Cynodon dactylon</i>
<i>Festuca ovina</i>	<i>Dicoma macrocephala</i>
<i>Holcus lanatus</i>	<i>Eragrostis racemosa</i>
<i>Mimulus guttatus</i>	<i>Fimbristylis exilis</i>
<i>Minuartia verna</i>	<i>Gomphrena canescens</i>
<i>Plantago lanceolata</i>	<i>Pogonarthria squarrosa</i>
<i>Silene vulgaris</i>	<i>Polycarpea glabra</i>
<i>Thlaspi alpestre</i>	<i>Tephrosia longipes</i>

After Bradshaw and Chadwick (1980).

reclamation of these toxic spoils would be (a) organic matter plus a commercial grass, and (b) a tolerant grass without organic matter. The latter alternative is likely to provide a more stable community on a long-term basis.

Mine wastes often contain high levels of metals other than the ones being mined. For example, copper mine waste may contain elevated levels of zinc and lead (Smith and Bradshaw 1972). It would be useful, therefore, to produce varieties that are tolerant to the three common heavy metals—copper, lead, and zinc (Jeffrey and others 1974). Recently, it has been discovered that *Festuca rubra* growing on a copper mine waste at Ecton (Staffordshire, UK) was tolerant to all three metals (Wong 1982), although no attempt has been made to determine its actual application value. Furthermore, it is desirable to explore other potential tolerant temperate and tropical species varieties to suit local climatic conditions. This is possible as a wide range of species has been recorded as indicators of the presence of metals (Antonovics and others 1971), and a variety of species has been noted as occurring on metal anomalies (Nicolls and others 1964). It seems certain, although it has not been proved, that many of these species possess metal-tolerant populations (Table 3). Some of the species listed in Table 3 have been used for reclamation purposes, but investigations should be undertaken on other species to discover their practical usage.

A simple screening technique has been developed by Walley and others (1974). It involves sowing seeds on a culture medium that contains heavy metals and retaining the plants that survive and grow well over a period of four to six months. A medium of mine soil, ameliorated with a small portion of potting compost, has the advantage of selecting tolerant plants adapted

to other mine soil factors, although another artificial medium may be used, such as sand or vermiculite soaked in a nutrient solution containing heavy metals.

## Conclusions

Stabilization of toxic materials by vegetation establishment is generally superior to alternative techniques (Williamson and Johnson 1984). Inert wastes from mining and quarrying operations, for example limestone chippings, slate quarry waste, and certain colliery shales, have several advantages for the surface treatment of metalliferous wastes: they are usually available in large quantities and at low cost in the vicinity of derelict metal mines; their coarse granular nature deters upward movement of metal salts by capillary action due to the physical isolation; and their inherent low nutrient status allows the productivity, species composition, and rooting characteristics to be regulated by initial seeding and the type of fertilizer application (Johnson and others 1977).

Input of organic matter with satisfactory nutrient content encourages the growth of microorganisms, organic matter decomposition, and the release of nutrients. Nevertheless, organic and topsoil amendments may only provide a temporary visual improvement and stabilization of the toxic material, and the use of commercial grass varieties tends to accelerate regression of plant growth. By contrast, tolerant varieties ensure persistence of the vegetation cover (Gadgil 1969, Goodman and others 1973) and should be used in seed mixtures, especially if only a shallow surface dressing of the ameliorant is applied.

Some of the modern waste materials are less toxic than in the past, and the total toxic metal level rarely exceeds 1% weight by weight, because of improvements in mining procedures and processing methods. Consequently, plant material may often be sown or planted directly. A wide range of nontolerant plant materials have been used for the purpose of revegetating different tailings. Most of them have been grasses or other herbs, especially legumes, although shrubs and trees are also used. They are often able to thrive in these waste materials provided that the appropriate nutrients are supplied, together with proper long-term maintenance.

On the other hand, waste materials produced by early and more primitive extraction methods usually require metal-tolerant varieties of plant species. Further exploration of other potential varieties that thrive on waste materials and artificial screening are desired to produce tolerant varieties suited to local climatic conditions.

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