### Revegetation of historic acidic mine waste with Agrostis capillaris – Remediation strategy

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### Abstract

To establish vegetation on historical acidic mine waste and metal polluted soil has several environmental, physical and chemical advantages on metal release. Pot experiments with material from the Ljusnarsbergs mine waste deposit in Sweden showed that different kinds of refuse can be used to sustain germination and improve growth of *Agrostis capillaris* where it otherwise is impossible. Bark compost increased the water holding capacity and an increase of pH in the top 50 mm by addition of water works granules made it possible for the grass to survive. An even better growth was obtained if those systems were also inoculated with mycorrhiza.

Keywords: historic mine waste, acidic, revegetation, amendments.

# Introduction

Historic mine waste with a high sulfidic/pyritic content is one of the major sources for the release of heavy metals to the Swedish environment. Many of these waste sites were produced from medieval times until the Second World War, after which the technological development allowed for a more complete recovery of metals from the ore. Hence the metal content of these wastes often exceeds what is considered as an ore today. Some of these sites are quite small, although rich in metals, and found in remote regions with neither power nor road access. Well established techniques such as dry or wet covers are, therefore, often impossible to use because of these constraints (c.f. Nathanail and Bardos 2004; Johnson and Hallberg 2005). Hence, there is a need to develop efficient and economically rational strategies for remediation.

If a vegetation cover can be established it is a cheap, and often efficient, first action strategy to lower the amounts of metals that are released from this type of waste site. In principle, the reduction is achieved by reducing the infiltration of rain water, mainly through increased evapotranspiration. The vegetation cover also conducts rainwater along slopes, minimizes dust formation and possibly also lowers the oxygen diffusion by heterotrophic activity in the root zone.

The ideal plant cover would contain plant species with a good coverage of the soil/waste and a high tolerance towards acidic conditions and high concentrations of (heavy) metals. In addition, such species should cope with harsh physical conditions since many waste piles are exposed to sunlight and winds that result in large variations of the temperature, leading to severe water deficiency. Depending on the size distribution of the substrate an additional problem is expansion/contraction of the substrate as a result of diurnal freezing and thawing cycles in early winter and spring. Unfortunately, such properties are not characteristic for many plant genera, with possible exception of grasses. In an

initial study of the possibilities of establishing vegetation on the unremediated mine waste from the Ljusnarsberg site, several different plant species were tested. The results were conclusive, and disappointing, since none of the dicotyledonous plants survived once the nutrient supplies in the seeds were depleted. Also the tested grasses continued to grow for a couple of weeks but then wilted, most likely as a response to high metal concentrations, low pH and lack of nutrients.

It is known, however, that among the grasses several genera and species are rather tolerant to the harsh environment on historical mine waste sites. Unfortunately, few of them tolerate the combination of several different physical and chemical stressors, including the shortage of nutrients. The latter, and perhaps also toxicity, can possibly be circumvented by novel uses of arbuscular mycorrhiza (Turnau *et al.* 2008). Another serious problem of mine waste generally is its high porosity and hence a very low water holding capacity. Fortunately, on many sites the rapid oxidation of exposed sulphides in combination with acidic weathering of parent rocks has resulted in a rather fine grained material that is beneficial to plants.

Here, we report some results from an ongoing study that attempts to overcome, or at least lower, the impact of some of the stress factors that have limited the success of revegetating historical mine waste using various different methods. These approaches include physical amelioration (porosity and water holding capacity), lowered heavy metal availability (pH, complexing agents) and improved nutrient availability (pH, mycorrhiza). The additives used for this amelioration consisted of refuse from several different industries, including water works granules, as well as mycorrhiza. The latter was isolated from the site and proliferated under controlled conditions to suit the ecology of the grass *Agrostis capillaris* that was chosen for the experiment.

# Methods

The effect of the different treatments was evaluated in pot experiments using commercial plastic flower pots (180 mm outer diameter. x 160 mm height). Sieved (< 5 mm) mine waste from the Ljusnarsberg mine waste deposit in Sweden was used as substrate. This composition of this material has previously been described in detail (Sartz 2010) but in summary it consists of a complex mixture of carbonate (Ca, Mg) and sulfide (chalcopyrite, galena, pyrite, sphaelerite) minerals in a matrix of pegmatite, granite and aplite as well as skarn minerals. Biotite, phlogopite and muscovite are also commonly found in the waste along with fluorite, and related fluoride minerals. The mine waste was produced from the early 17th century until the end of the 1970s so it is well oxidized. The size fraction less than 5 mm was chosen and mixed with commercial bark compost (bought at Bauhaus) at 30% volume to improve the water holding capacity. All other additives, except the Aspen wood shavings, were manually mixed into the top 50 mm of the substrate. The pots were then left outdoors at Listregården (50°22'28.95"N 15°17'37.43"E) on a corrugated painted metal slate to allow for free drainage of rain water.

The following treatments were evaluated in triplicates:

- System 1 Reference: Mine waste (70%) and bark compost (30%).
- System 2 Water works granules (wwg): Solid (1-3 mm) spheres of mainly Ca/Mg carbonates left from softening of water in municipal water works. Here they were used to locally increase pH, at a dosage of 7.6 mL per pot.
- System 3 Mycorrhiza (dead): Served as a reference to the active mycorrhiza evaluated in system 4.
- System 4 Mycorrhiza (living): These mycorrhiza species had been isolated from the site and the proliferated.
- System 5 Mycorrhiza (dead) and water works granules. This combination should reflect any effect of the carbon added as mycorrhiza at the higher pH.
- System 6 Mycorrhiza (living) and water works granules: The combination served to elucidate if the pH had any impact on the performance of the mycorrhiza.
- System 7 Aspen (*Populus tremula*) wood shavings: The shavings (200 mL per pot) served as a proxy for easily mineralised wood. After three weeks a single dose of commercial inorganic fertilizer was added.
- System 8 Commercial garden compost.

The growth was followed weekly as shoot length and areal coverage by image analysis of photographs taken at fixed distances and angles. At the end of the growth season the lengths of the shoots and roots were measured. In addition, weekly sampling of the mobile water phase was conducted by adding 10 mm of rain water that had been collected. The water that left the pots was analysed for major hydrochemical parameters, transition elements and trace elements as well as DOC (dissolved organic carbon). Sample preparation and analytical procedures are presented elsewhere (Karlsson *et al.* 2012). Temperature, rain-fall, humidity, wind speed and direction were recorded every fifteen minutes during the growth period.

Field experiments using the same additives were carried out on a clearing of the test field (Sartz, 2010) on the Ljusnarsbergs deposit in Kopparberg. All coarse material, larger than 50 mm, was removed manually from the site. Sieved mine waste (diam < 5 mm), the same as for the pot experiments, was placed on top of the clearing to a depth of 100 - 150 mm. To this layer 30% (volume) bark compost (commercial from Bauhaus) was added and thoroughly mixed (manually with garden forks). Quadratic seedbeds with sides 0.85 m were prepared with the surface lying approximately 150 mm above the waste rock. A random number generator was employed to assign the treatments for the individual sub-plots. The field test began on June 16, 2011.

### **Results and Discussion**

#### Substrate solution composition

The composition of the solution phase that drained from the pots is discussed in detail elsewhere (Karlsson et al. 2011) with just a brief summary provided here. The Ljusnarsbergs historical waste generates an acidic water that is rich in sulfate, aluminium, calcium and magnesium but is quite low in iron. The composition reflects the presence of carbonate minerals and long-term weathering (c.f. Sartz 2010). The presence of fluorite in the waste is reflected by the rather high concentrations of dissolved fluoride, which has an impact on the speciation of several elements including Al, Ca and Fe.

In Table 1 the mean composition of all systems is provided. Week 1 represents the time of sowing, when the large standard deviations indicated that the systems were rather heterogeneous. The high concentrations of DOC are entirely attributed to the bark compost used to improve the water holding capacity of the waste.

During the experiment the systems responded in a similar manner with slightly increasing pH and lowered concentrations of most other principal and trace ions, as well as DOC. Two elements deviated from this general time tendency; both calcium and lead had increasing concentrations. The extent to which this behaviour is related to physical, chemical or biological mechanisms is discussed elsewhere (Karlsson *et al.* 2012).

The general conclusions from these short time series during one single growth season would be that the most stressful chemical conditions were found at the time of sowing, i.e. during germination of the seeds and first growth of the seedlings. As time progressed, the concentrations of toxic elements declined and pH increased somewhat which would increase the chance for continued growth of *Agrostis capillaries*. These findings indicate that only optimising the chemical conditions during early growth of the plants might be a cheap strategy in order to establish a grass cover. Monitoring of the systems performance during the next years will determine if such a low maintenance approach is possible.

### Weather conditions

A late snow melt in the end of April was followed by a dry and cold period in May that delayed the sowing to mid June when the weather improved. During the experiment rain-fall was rather high, with almost daily showers (Figure 1). On two occasions thunderstorms resulted in intense precipitation events with some 50 mm falling during 2-4 hours. The sum of precipitation and the watering at sampling gave a final L/S ratio (liquid to solid ratio) slightly higher than 5 at the end of the experiment.

Variable	Week 1		Week 3		Week 8	
	Mean	S.D.	Mean	S.D.	Mean	S.D.
pН	3.84	0.114	3.98	0.09	4.38	0.15
SO42- (mg/L)	990	539	920	441	638	126
F <sup>-</sup> (mg/L)	62.9	45.7	45.1	17.5	15.3	3.8
Cl <sup>-</sup> (mg/L)	9.0	14.4	4.2	1.7	1.1	0.1
Al (mg/L)	26.38	14.77	20.39	5.11	8.68	2.47
Ca (mg/L)	99.8	45.7	119.0	20.7	220.0	22.9
Fe (mg/L)	0.400	0.221	0.232	0.080	0.126	0.011
K (mg/L)	29.81	14.74	24.00	4.92	3.86	0.64
Mg (mg/L)	58.15	32.12	44.05	11.19	2.64	0.65
Mn (mg/L)	35.24	20.53	26.84	7.40	1.27	0.55
Na (mg/L)	6.95	3.15	5.66	1.17	1.42	0.13
DOC (mg/L)	48.8	25.1	33.7	7.11	9.47	3.76
Cd (µg/L)	157	94	120	35	35	13
Cu (µg/L)	1109	577	668	177	355	96
Pb (µg/L)	36	19	66	18	108	37
Zn (μg/L)	58205	35806	44107	12951	8623	3203

**Table 1** Concentrations of selected constituents in the solution phase draining from the pots at three occasions. Mean values for all systems (n = 22) and standard deviation (S.D.).

The germination of the seeds in the different treatments was not different (T-test, p 0.05) from the control, i.e. the fraction of seeds that germinated on wet filter paper (Figure 2). During the first three weeks of growth there was no difference in shoot length among the treatments (Figure 3). There was possibly a slight depression of the growth rate in system 4 (living mycorrhiza) but it was not different (T-test, p 0.05) from the control. Hence, it seems that the substrate conditions did not interfere with the initial growth phase of the plants. With increasing time, several differences became obvious. After the growth season, the shoot lengths were lowest in treatment 1 (control), 3 (dead mycorrhiza) and 4 (living mycorrhiza) (Figure 3).

These treatmens also had the shortest roots, being 12 to 30 mm. According to these results, addition of the bark compost to the mine waste made it possible for the plants to survive. Only the addition of mycorrhiza to the mixture, without any pH adjustment, was insufficient to improve the growth of the grass.



Figure 1 Temperature (°C) and daily precipitation (mm) during the experiment.

The poor function of the mycorrhiza was unexpected but most likely related either to a lack of a suitable carbon source for the fungi or to the low pH in the root zone. The additions of wwg indicate that pH is crucial since growth was promoted in treatments (systems 2, 5 and 6) where wwg was added. The pH in the water that left the pots containing wwg were not different from the others, why any positive effect on growth must have been localized in the root zone. In fact, the longest shoots were found in series 6 where both wwg and living mycorrhiza had been added. Since no such positive effect was found in series 5 (wwg and dead mycorrhiza) it seems reasonable to conclude that a higher pH is necessary in order for the mycorrhiza to function effectively.



Figure 2 The number of germinated seeds in the treatment. (Treatments: 1-Control; 2-Water works granules (wwg); 3-Dead mycorrhiza; 4-Mycorrhiza; 5-Dead mycorrhiza and wwg; 6-Mycorrhiza and wwg; 7-Aspen wood shavings; 9-Wet filter paper)



Figure 3 The length of the straws after three weeks and after the growth season, respectively. (Treatments: 1-Control; 2-Water works granules (wwg); 3-Dead mycorrhiza; 4-Mycorrhiza; 5-Dead mycorrhiza and wwg; 6-Mycorrhiza and wwg; 7-Aspen wood shavings)

From Figure 3 it is also evident that the longest shoots were found in system 7, where Aspen wood shavings and the commercial inorganic fertilizer had been added. These findings indicate that the poor nutrient status of the original waste contributed to the limited growth. On the other hand, a slight increase of pH in the presence of mycorrhiza maintained growth (system 4), although at a lower rate. It is therefore not immediately evident which strategy to apply for practical remediation. Although the addition of the inorganic fertilizers promoted growth it

might not be a suitable treatment in the long term, particularly not if mycorrhiza is used. Inorganic fertilizers might induce a suppression in the mycorrhizal infestation and activity even at nitrate levels in rainwater (Hofland-Zijstra and Berendse 2009) and if this is the case a severely lowered growth would be expected once the added nutrients have been consumed. From a sustainable, and also practical, point of view the use of wwg and mycorrhiza seems more attractive but there is need for further studies before any conclusions can be reached.

The increase in shoot length was also reflected in a similar growth of the roots. In treatment 1 (mine waste and bark compost) the shoot length was on average 12 mm. In those treatments with addition of wwg they were up to 60 mm and were not only found in the upper 50 mm but some roots had penetrated some 20 mm further below. Where both wwg and living mycorrhiza had been added the roots extended even further, some 10% reaching down to 100 mm below the surface. The reason for this observation is not clear but it is unlikely that the positive effect of wwg that was mixed into the upper 50 mm would have extended below this level. Most likely, the neutralizing capacity of the wwg would be balance acid production by the waste only in the proximity of the granules. Wwg additions, however, appear to improve the function of the mycorrhiza in the top 50 mm, enabling a deeper penetration of the roots. This hypothesis is strengthened further by the presence of Aspen wood shavings (treatments 19-21) where the roots extended to 100-150 mm and also had a more complete lateral infiltration into the mine waste. Most of the roots were found close to, or overgrowing the decaying wood shavings why it is possible that the mere physical support from the shavings was the cause of this positive response. It is also possible that the decomposition processes of the shavings improved the chemical conditions in this microenvironment, making it more favourable for root growth. There was no evidence that bark compost had any such positive effect though addition of the inorganic fertilizer may have increased root length and this warrants further investigations.

The results from the field experiment were similar to those from the pots although the length of the shoots was some 10% shorter on average. Addition of the bark compost had a general positive impact and the longest shoots were found in the plots where both wwg and mycorrhiza had been added. It should be noted, however, that the field experiment did not evaluate the impact of aspen wood chips or addition of inorganic fertilizer.

# Conclusions

The pot experiment has shown that it is potentially feasible to establish a grass cover on historical acidic mine waste by treatments with alkaline refuse, provided that the water holding capacity is high sufficient. The addition of bark compost to the waste and of water works granules in the top 50 mm made it possible for the grass *Agrostis capillaris* to germinate and maintain growth during a growing season. Improved growth was obtained if this treatment was inoculated with mycorrhiza. These results were confirmed by field experiments. The combination of bark compost and Aspen wood shavings also had a positive effect on grass growth, but at this point this treatment has not yet been validated by field trials.

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