Technical Communication

Reclamation of Abandoned Coal Mine Waste in Korea Using Lime Cake By-Products

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Abstract. In Korea, there are hundreds of abandoned and closed coal mines in the steep mountain valleys due to economic downturns in the mining industry. Enormous amounts of coal waste from these mines were dumped on the slopes, which cause sediment and acid mine drainage to be discharged directly into streams. A limestone slurry by-product (lime cake), which is produced during the manufacture of soda ash, was investigated for its potential use in reclaiming the coal waste. The lime cake is fine grained, has low hydraulic conductivities (10^{-8} to 10^{-9} cm sec^{-1}), high pH, high electrical conductivity due to the presence of CaO, MgO, and $CaCl_2$ as major components, and trace amounts of heavy metals. A field experiment was conducted; each plot was 20 x 5 m in size on a 56% slope. Treatments included a control (waste only), lime ($CaCO_3$), and lime cake. The lime requirement (LR) of the coal waste to pH 7.0 was determined, and treatments consisted of adding 100%, 50%, and 25% of the LR. The lime cake and lime were applied in either a layer between the coal waste and topsoil or mixed into the topsoil and coal waste. Each plot was hydroseeded with grasses, and planted with trees. In each plot, soils, surface runoff, and subsurface water were collected and analyzed, and plant cover was measured. The lime cake treatments increased the pH of the coal waste from 3.5 to 6, and neutralized the pH of the runoff and leachate of the coal waste from 4.3 to 6.7. Moreover, the surface cover of seeded species was significantly increased; sufficient acidity in the coal waste was neutralized in the 25% LR plots to allow seed germination.

Key words: abandoned mine land; acid mine drainage; acid runoff; acid soils; coal refuse; lime cake; reclamation; revegetation

Introduction

In Korea, over 300 coal mines have been closed or abandoned due to economic difficulties in the mining industry since the late 1980s (Coal Industry Promotion Board 2000). Many of these mines are located in steep mountain valleys. Enormous amounts of associated mine waste and acid mine drainage (AMD) are discharged into streams, degrading soil and water quality. The environmental disruptions caused by these mines are very serious (Jung and Thornton 1997; Lee et al. 2001). The wastewater from the portals of the closed mines and the leachate from the waste piles are low in pH and contain high concentrations of Fe, Al, and Mn (Yang et al. 2000).

The Coal Industry Promotion Board in Korea has spent over $15 million dollars (U.S.) annually to remediate mine-related damages and to improve the environment. Most of the costs are directed to waste water treatment, such as installing passive AMD treatment systems, and forest restoration. However, given the large number of waste piles and AMD sources, there has been little environmental improvement (Yang 2004). Very little money has been spent reclaiming coal waste piles due to liming costs and the scarcity of soil cover materials.

A lime by-product (lime cake) is produced during the manufacture of soda ash as part of the Solvay process. In Korea, over 3x 10^6 Mg of lime cake are stockpiled. This material has been used sparingly in the past to reclaim disturbed lands, and due to concerns from environmental groups, remains in stockpiles with no plan for proper disposal. The lime cake is very fine grained, and has low hydraulic conductivities (10^{-8} to 10^{-9} cm sec^{-1}), high pH, and high electrical conductivity (EC) due to the presence of CaO, MgO, $CaCl_2$, and NaCl as major components (Min et al. 2004). Due to these physical and chemical properties, the lime cake has the potential to be used as a neutralizer and an amendment for acid-producing materials.

We evaluated water quality, soil properties, and ground cover after either layering or mixing the cake into the coal waste with varying amounts of lime cake. If the lime cake application proves successful, this research will help in properly disposing of this material and reclaiming abandoned lands.

Materials and Methods

Ten treatments were installed on a large, abandoned coal waste pile to test the application of lime cake for reclamation of these piles. The slope of the coal waste site was 29.2° (56%). Plots measured 20 x 5 m (L x W) in size (Figure 1) and were separated by plastic boundaries. Treatments included the control (coal waste alone), agricultural lime ($CaCO_3$) as a reference, and lime cake (Table 1). X-ray fluorescence (Philips PW 2400, Amsterdam, the Netherlands) and X-ray diffraction (Rigaku Model D/Max-2400, Tokyo, Japan) were conducted on pulverized samples of lime cake and coal waste to determine major elements and crystalline minerals. Concentrations of As, Cd, Cr, Cu, Hg, and Pb were determined by 0.1N HCl extraction and the supernatants were analyzed using inductively coupled plasma atomic emission spectrometry (ICP-AES) (Perkin Elmer Optima 3100XL, Wellesley, MA) The lime requirement (LR) for the coal waste to pH 7.0 was determined to be 16.5 Mg/ha, and this was used to determine the amount of lime cake application. Lime cake treatments consisted of 25%, 50% and 100% of the LR (as $CaCO_3$) (Jones 2001). The lime cake and lime were either layered between the coal waste and topsoil or mixed with coal waste and topsoil. Each plot was hydroseeded with grasses and planted with trees. Surface coverage by grasses was determined by computer image analysis using Win RHIZO v. 5.0A software (Regent Instruments, Quebec, Canada) (Cheng and Bledsoe 2004).

Three pipes, 5 cm in diameter, were buried in each plot to collect the leachate into a reservoir. A flume and gutter at the base were connected to the reservoir to collect all the runoff from each plot (Figure 2).

Chemical properties such as pH and ion concentrations of the runoff and leachate were analyzed periodically (Yang 2004).

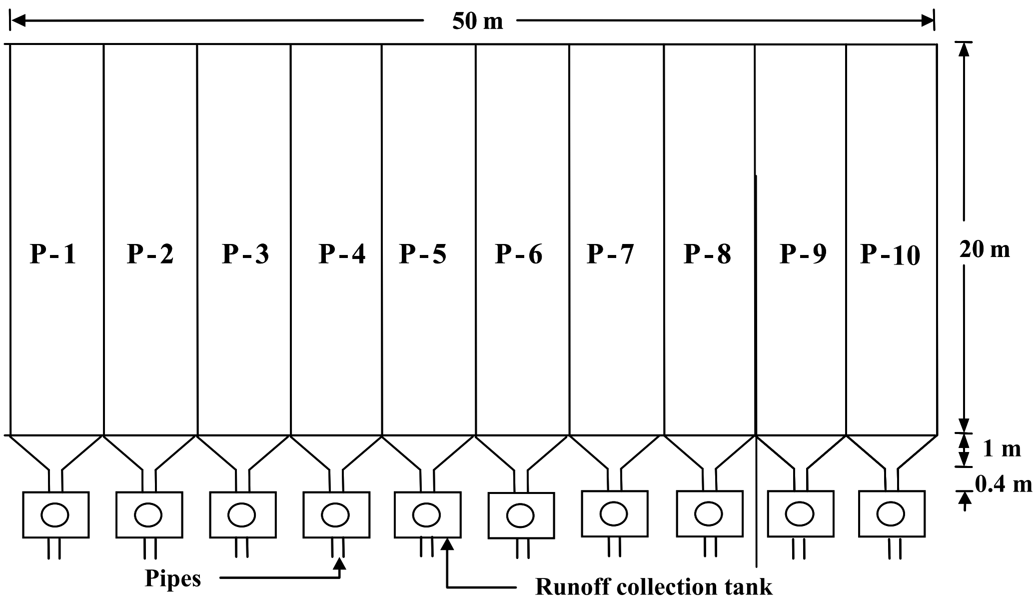

Figure 1. Layout of the coal waste plots with runoff and leachate collection reservoirs

Table 1. Treatment design and revegetation of the coal waste; LR represents lime requirement (as $CaCO_3$)

Plot number	Treatments	Lime treatment methods	Vegetation†
1	Coal waste only		Grass and trees
2	Coal waste + Lime cake (LR 100%)	Mixed	Grass and trees
3	Coal waste + $CaCO_3$ + topsoil	Layered	Grass and trees
4	Coal waste + $CaCO_3$ + topsoil	Mixed	Grass and trees
5	Coal waste + Lime cake (LR 100%) + topsoil	Layered	Grass and trees
6	Coal waste + Lime cake (LR 100%) + topsoil	Mixed	Grass and trees
7	Coal waste + Lime cake (LR 50%) + topsoil	Layered	Grass and trees
8	Coal waste + Lime cake (LR 50%) + topsoil	Mixed	Grass and trees
9	Coal waste + Lime cake (LR 25%) + topsoil	Layered	Grass and trees
10	Coal waste + Lime cake (LR 25%) + topsoil	Mixed	Grass and trees

† Grasses: Orchard grass (*Dactylis glomerata* L.), Kentucky Bluegrass (*Poa pratensis* L.), and Eulalia (*Miscanthus sinensis* Anderss); Trees: Pine (*Pinus densiflora* S. et Z.), White birch (*Betula platyphylla* var. *japonica*), and Alder (*Alnus firma* S. et Z)

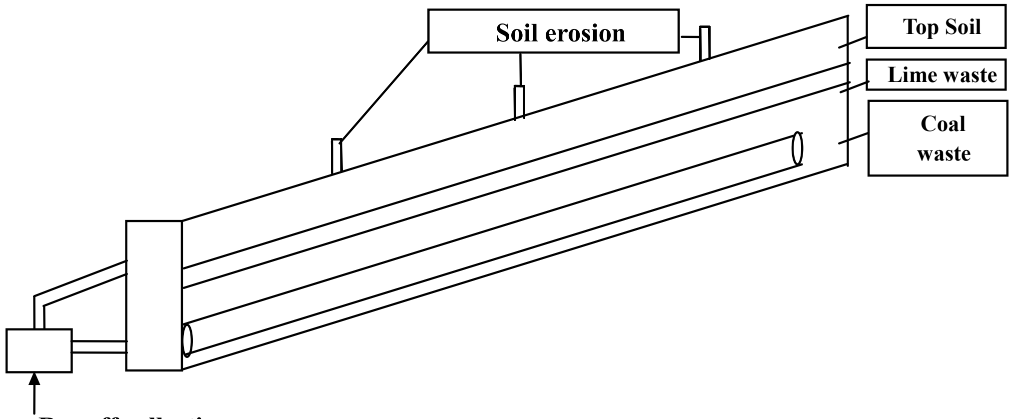

Figure 2. Side view of the experimental coal waste plots and location of water collection systems

Soil analyses were conducted on <2mm, air-dried soil samples. Soil particle size distribution was measured by pipette. Soil pH and EC in water were measured using standard methods (Jones 2001). Soil organic matter was estimated by loss-on-ignition (LOI) (Ben-Dor and Banin 1989). Exchangeable cations were extracted by 1M NH_4OAc at pH 7.0 and analyzed by a Shimadzu (Japan) AA-6900 AAS for Ca, Mg, K, and Na. Phosphorus was determined by the Bray P1 method. The pH of runoff and leachate sample was measured by a pH meter (Orion 920A, Pulse Instruments, Van Nuys, CA, USA). Water samples were filtered with pre-rinsed cellulose nitrate Sartorious filters, 0.45 μm pore diameter, stored at 5º C, and analyzed within 5 days of collection (Boudot et al. 2000). Sulfate was determined by ion chromatography (IC), and Fe and Al by ICP-AES.

Results and Discussion

Chemical Properties of the Coal Waste, Lime Cake, and Topsoil

The pH of the coal waste was 3.5; 16.5 Mg of $CaCO_3$ per ha were needed to adjust the pH to 7.0 (Table 2). The lime cake was high in bases such as Ca, Mg, and Na with a pH of 11.2 and a high EC of 19.6 dS m^{-1}. The topsoil was obtained from a nearby road cut and had an optimal pH of 6.5, but was low in fertility. Particle size analysis indicated a silt loam texture for the topsoil (percentages of clay, silt, and sand were 8, 53 and 39, respectively). The coal waste was composed mainly of SiO_2, Al_2O_3, and carbon, while the lime cake contained CaO, MgO, Na_2O, SiO_2, Cl, and other carbonaceous compounds (Table 3). X-ray diffraction analysis indicated that major minerals in the lime cake were calcite, quartz and plagioclase (Min et al. 2004). The heavy metal contents based on the 0.1M HCl extraction in lime cake and coal waste were undetectable for As, Cu, Hg, Cr, and very low for Cd (<0.20 mg/kg). Coal waste Pb concentrations were 6.9 mg/kg; no Pb was detected in the lime cake. Based on Tables 2 and 3, it appeared that the lime cake had the potential to neutralize the acidic coal waste, and had no obvious detrimental properties to limit its use in reclaiming coal wastes.

Effects of Lime Cake on the pH of Coal Waste

The pH of coal waste alone (plot 1) was 3.5, but increased to 7.5 when mixed with lime cake without topsoil (plot 2). The soil pH of all the other plots with topsoil were about the same (pH 6.0) regardless of the amount of lime cake applied and irrespective of layering or mixing (Figure 3). The neutralizing effects of lime cake were equivalent to that of agricultural lime. No treatment plot assessed coal waste and topsoil without neutralization, so the possible neutralizing effect of the soil alone cannot be judged. However, it was clear that the combined treatment of

Table 2. Chemical characteristics (pH, electrical conductivity (EC), soil organic matter (OM), phosphate, lime requirement (LR) as $CaCO_3$, and exchangeable cation content of the lime cake, coal waste, and topsoil

Sample	**pH** (1:5)	**EC**(1:5)	**OM**†	P_2O_5	**LR**	**Exchangeable cations** ($cmol_c$ kg^{-1})			
		dS m^{-1}	g kg^{-1}	mg kg^{-1}	Mg ha^{-1}	**Ca**	**Mg**	**K**	**Na**
Lime cake	11.2	19.6	8.3	7.9	-	233.8	50.5	2.3	77.9
Coal waste	3.5	0.2	165.5	9.1	16.5	3.9	0.3	0.1	0.1
Topsoil	6.5	0.1	80.8	15.7	0.37	4.5	0.5	0.1	0.1

† Organic matter based on loss on ignition (LOI)

Table 3. Elemental compositions (as %) of the lime cake and coal waste, as determined by X-ray fluorescence

	Lime cake	**Coal waste**
Na_2O	2.3	0.2
MgO	11.5	0.3
CaO	39.3	0.2
SiO_2	2.8	37.1
P_2O_5	0.1	0.2
K_2O	0.2	3.5
Cl	10.2	ND
Fe_2O_3	1.0	4.8
Al_2O_3	1.3	21.2
Cr_2O_3	ND	ND
MnO	ND	ND
SrO	0.1	0.1
Water Content and LOI†	31.1	33.4

† Loss on ignition

lime cake with topsoil effectively neutralized the acidic coal waste.

Effects of Lime Cake on Water Quality

The runoff pH was 4.3 but increased significantly (to 6.7-7.1) with treatments of agricultural lime and lime cake with topsoil (Figure 4A). There were no significant differences in pH among treatments. The rise in runoff pH is due to the combined effects of lime cake and buffering capacity of the topsoil.

Sulfate concentrations in runoff were highest in the control (plot 1) and decreased significantly with the lime cake treatment (plot 2) (Figure 4B). When coal waste was treated in combinations of lime cake and topsoil, the sulfate concentrations in the runoff were slightly decreased. Sulfate concentrations in runoff from abandoned coal waste and mines in Korea are shown to be up to several thousand mg/L (Yang et al., 2000; Yang et al, 2002). The decreases in sulfate with lime cake and lime treatments could be due to less pyrite oxidation with higher pH thereby generating less sulfate, but the lower sulfate concentrations may also be due to precipitation of $CaSO_4$ in the treated soil with added calcium.

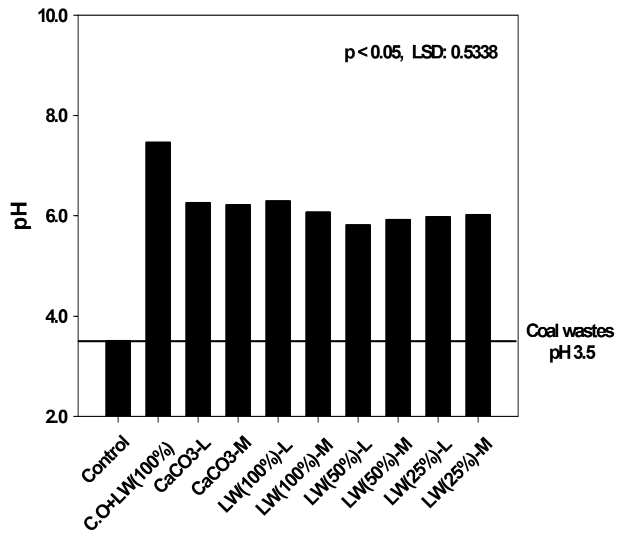

Figure 3. Soil pH in each treatment plot: coal waste (CO); lime cake (LW); layered (L); and mixed (M)

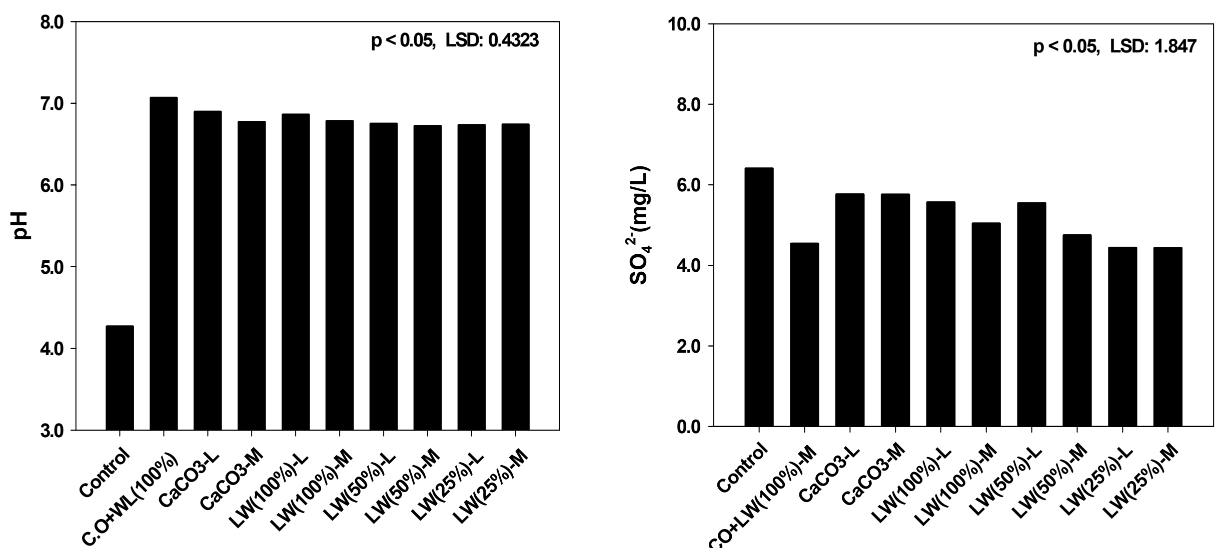

Figure 4. Runoff and leachate pH (4A) and sulfate concentrations (4B) of each treatment plot: coal waste (CO); lime cake (LW); layered (L); and mixed (M)

Aluminum and Fe from coal waste are the major ions deteriorating the water quality in abandoned coal mine areas of Korea. The initial Al concentrations in runoff ranged from 30 to 60 mg/L, but those sharply decreased with time (Figure 5A). There were no significant interactions among treatment plots, dates and precipitation on the Al concentrations in the runoff. Concentrations of Fe in the runoff, however, fluctuated with date and precipitation (Figure 5B). It is not clear why Fe concentrations increased with rainfall 2 to 4 weeks after treatment. No soluble iron should have been available in the topsoil or lime cake, yet all the treatments except the lime cake alone showed a temporary increase in runoff Fe concentrations. At the experimental site, soils surrounding the coal waste pile and a nearby stream both exhibited the effects of Fe coatings on soils and stream sediments prior to lime cake treatment; these coatings disappeared with the lime cake treatment of the coal waste.

Effects of Lime Cake on Revegetation of the Coal Waste

Due to the climate in Korea, grasses such as orchard grass (*Dactylis glomerata* L), Kentucky bluegrass (*Poa pratensis* L.) and eulalia (*Miscanthus sinensis* Anderss) were hydroseeded at the end of May. The grasses covered only 15.5% of the coal waste plot in June but the cover had increased to 33% by August (Table 4, Figure 6). Growth of grasses was enhanced with the combined treatments of lime cake and topsoil (plots 5 to 10), resulting in increased plant cover. The plant cover was highest with the 25% lime cake treatment. Bioassay tests in the greenhouse confirmed that seed germination of these grasses was highest when lime cake was applied at 25% of the LR, while germination was significantly suppressed at the 50% and 100% lime cake treatments (Yang 2004). The results suggest that the high salt content of the lime cake at higher application rates may be a limiting factor in immediate revegetation of lime cake-treated coal waste. Many researchers have shown a similar detrimental effect of salts on plant germination and suggest a leaching period of several weeks to a year for flushing of these salts from materials high in soluble salts (Korcak 1995; Martens and Beahm 1976).

Conclusions

A large coal waste pile in Korea was treated with lime and lime cake in various treatments to examine the chemical qualities of soil and water (runoff and leachate), and to assess the surface cover of grasses. Lime cake treatments increased the pH of the coal waste from 3.5 to 6, and raised the pH of runoff and leachate from 4.3 to 6.7. Concentrations of sulfate, Al and Fe in the runoff and leachate were significantly decreased with lime cake. Surface cover by grasses on coal waste was significantly increased with lime cake treatment. Application of lime cake at 25% of the lime requirement was sufficient to neutralize the acidic coal waste and to allow germination of grasses. Either layering the lime cake between the coal waste and topsoil or mixing with coal waste and topsoil could be adopted as reclamation methods. Results demonstrated that the lime cake from soda ash production has good potential for reclaiming abandoned coal waste piles and for alleviating the environmental problems associated with acid soils and water from coal waste.

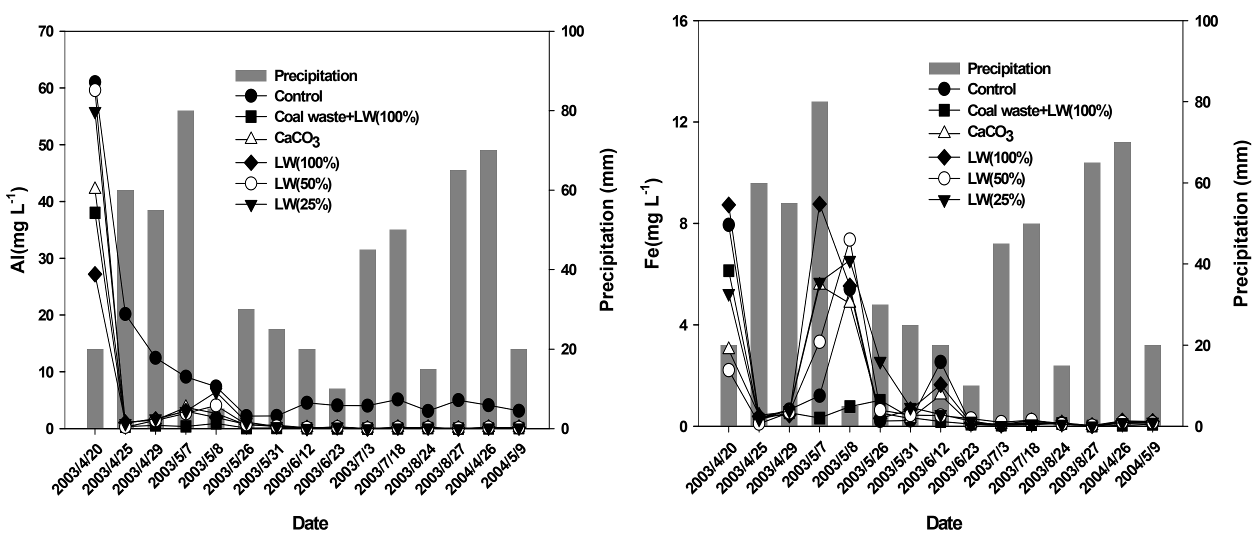

Figure 5. Aluminum (5A) and iron (5B) concentrations in runoff and leachate as affected by lime cake treatment and precipitation

Table 4. Vegetation cover percent and surface coverage (cm^2) of each treatment plot from June through August

Month		Treatment Plots†									
		1	**2**	**3**	**4**	**5**	**6**	**7**	**8**	**9**	**10**
June	**%**†	15.5	13.2	14.5	14.6	15.8	25.6	22.4	30.3	25.5	25.6
	cm^2‡	310.9	246.9	256.0	259.0	287.7	509.3	471.6	537.9	494.0	510.8
July	**%**	25.9	23.2	26.1	22.8	21.0	30.9	29.3	37.9	36.3	37.2
	cm^2	531.9	529.6	553.7	537.0	477.7	604.8	575.2	758.9	739.6	752.9
August	**%**	33.4	27.5	46.3	45.7	40.4	37.5	36.9	45.6	52.6	61.2
	cm^2	680.3	547.4	920.1	913.8	797.0	783.2	760.1	908.2	1016.3	1175.8

† Refer to Table 1 for the treatment combination; ‡ Percentages and surface coverage were averaged over 5 measurements

Figure 6. Pictures of the plots: A. treatments established; B. after hydroseeding; C. vegetation cover in June; D. vegetation cover in August

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