

UPWARD MIGRATION OF CONSTITUENTS IN SOIL COVERS AT SEMI-ARID MINE SITES¹

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Abstract. Upward migration of either acidity or sodium is often cited as a concern when soil covers are placed over mine wastes. Regulatory concerns associated with upward migration are commonly used to justify capillary barriers or increased cover thickness. Upward migration of soluble salts is known to occur in soils with permanent or seasonal water tables near the soil surface and is a common problem in agricultural contexts. Short-term studies conducted at mine reclamation sites where changes in pH or salinity occurred at the contact between the soil cover and waste have been used to infer that upward migration would ultimately degrade cover quality. Thus, we investigated the potential for upward migration in soil covers overlying acid mill tailing and coal spoils 20 to 25 years after cover placement and revegetation. The soils were sampled and analyzed at discreet intervals above the soil-waste contact to assess upward migration. Changes in soil pH and/or extractable constituents were inconsistent among sites and varied from about 2 to 10 cm above the soil cover-waste contact. Field observations suggest that physical mixing of the cover and tailing during construction, rather than advection, accounted for acidification of the basal soil cover. Based on evidence from our investigations and an understanding of soil developmental processes in arid-regions, we conclude that upward migration of constituents from wastes is not a time-transgressive process that will result in cover degradation in arid and semi-arid regions where permanent or seasonal water tables occur well below the soil surface. Finally, we consider the changes in chemistry at the soil cover above the waste interface to be biologically insignificant.

Additional Key Words: Acid migration; soil covers; reclamation soil development; soil suitability.

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Introduction

The translocation of chemical constituents upward from mine spoils into overlying cover soils was identified as a potential concern in the reclamation of western mines based on early studies (Sandoval and Gould, 1978; Merrill et al., 1983; Dollhopf et al., 1985; Carlstrom et al.; 1987). Reference to upward migration was codified in federal coal mining mine regulations (30 CFR 715.14). The overriding concern was that chemical changes in the soil cover associated with the upward migration of salts would reduce its ability to support plant growth. Under extreme projections, upward migration of salts would eventually render the soil cover unsuitable for plant growth leading to a cycle of erosion and eventual failure of cover function.

Upward migration of soluble constituents by convective flow is a concern in situations where a permanent or seasonal water table occurs near the soil surface. Soil salinization, which results from capillary rise of water and evapoconcentration of salts in the upper soil profile, has long been recognized as a problem in irrigated situations where drainage is inadequate (Israelsen and Hansen, 1950). Soil salinization is typically not a concern when the water table is at depths greater than 1 to 2 m below ground surface. Upward movement of salts is not considered to be an important soil developmental mechanism in well-drained soils where leaching is the predominant process (Buol et al., 1980). Well drained conditions are promoted by avoiding the creation of closed depressions as required by most reclamation designs. Nonetheless, these concerns still exist, even though evidence from long-term studies is generally lacking that this phenomenon occurs in properly constructed mine land reclamation applications (Dollhopf, 2001).

The intent of this paper is to evaluate the chemical characteristics of cover soils 20 years after placement over acidic copper mill tailing. In addition, we assess the biological implications of changes in the cover with respect to reclamation performance.

Background

Chemical redistribution in soils results from a combination of diffusive and convective processes (Kemper, 1986). Diffusion is restricted to the movement of dissolved constituents within the water lattice, whereas convection refers to the movement of dissolved constituents with soil water. The magnitude and direction of diffusion in soils is dependent on ion concentration gradients, soil matrix-ion interactions, ion-ion interactions, soil-water content, temperature, and tortuosity of flow paths. Furthermore, diffusion is a self-limiting process and the rate of diffusion decreases as the concentration gradient decreases away from the primary source. Diffusion is generally considered to be a slow and inefficient transport mechanism compared to convective flow. Diffusion operates at a small scale, and manifestations of diffusion may be obliterated by the potentially overwhelming magnitude of convective flow.

Upward Migration of Sodium

The potential for upward migration of sodium was originally investigated at coal mines. Merrill et al. (1983) indicated that sodication of soil covers may occur based on laboratory column studies and field investigations conducted within a few years after cover placement. They concluded that diffusion was the primary mechanism for sodium redistribution from the sodic spoils to nonsodic soils and that leaching may have eliminated evidence of the upward (diffusional) migration of sodium at two of four field test sites.

The potential for sodication of cover soils overlying sodic spoils was studied in test plots at the Decker Mine in Montana (Dollhopf et al., 1985 and 1992). Cover soils were sampled in three equal depth increments (each 23-cm thick) and analyzed on an annual or semiannual basis over an 11-year period. Dollhopf et al. (1985 and 1992) concluded that sodication of the cover soil resulted primarily from sodium diffusion from the spoil to the soil. In the 1985 report, using a linear regression model, they predicted that the topsoil could become sodic in 18 to 36 years (Dollhopf et al., 1985). Later, using a negative exponential growth model, they indicated that only the lower part of the cover soil profile would be affected, with a maximum sodium adsorption ratio (SAR) near 7 (Dollhopf et al., 1992).

The Decker test plot results revealed that the SAR in the cover soil profile decreased in the upper third (0 to 23 cm), remained nearly constant in the middle interval (23 to 46 cm), and increased in the lower soil layer (46 to 90 cm). The potential for leaching (downward translocation) of sodium in the upper soil layers was recognized in the 1992 report, but was not accounted for in the empirical predictive model (Dollhopf et al., 1992). The decrease in SAR in the upper soil layer, coupled with zero net change in SAR in the middle layer and an increase in SAR in the lower layer, is consistent with the expected sodium distribution that would occur under a leaching regime dominated by the downward translocation of sodium. Thus, we interpret the time-transgressive SAR distribution in the cover soil profiles as resulting from the translocation of sodium from the upper soil layers to the lower soil layers. Sampling and analysis of the cover soils and spoils on the non-irrigated test plots at Decker in 1999 (22 years after covering) revealed no evidence of progressive salinization or sodication of the basal topsoil layer (Munk et al., 2000).

Upward Migration of Acidity

Chammas et al. (1999) concluded that upward migration of acidity was occurring from acidic and neutral (non-acidic) copper mill tailings to the overlying Gila Conglomerate cover soil based on sampling and analysis conducted 1 year after cover placement. The primary evidence for upward migration was increased diethylenetriaminepentaacetic acid (DTPA) extractable Fe levels in the lower 2 inches of the soil covers. Elevated DTPA-iron concentrations occurred at the base of the soil profiles regardless of the occurrence of lime- or capillary-barrier treatments. The DTPA-iron results are incongruous as indicators of upward migration as there was no accompanying change in electrical conductivity, which might be expected if the Fe was accompanied by a charge balancing anion.

We believe that the increased DTPA-iron may have been associated with fertilizer and irrigation treatments rather than diffusion from the underlying wastes. Chammas et al. (1999) indicated that all plots were treated with an acid-forming fertilizer and humic acid soil conditioner, and then irrigated. These treatments could have resulted in the release and chelation of Fe originating from the Gila Conglomerate cover soil and downward translocation of organo-iron complexes to the lower profile, rather than upward migration.

Dollhopf (2001) evaluated long-term changes of a cover soil overlying a lime reject layer and acidic tailing in Anaconda, Montana. He studied acidic (pH 2.0 to 2.3) Cu tailing that was covered with an 8-cm thick lime layer and 41-cm thick cover soil in 1974. The lime reject layer was an alkaline waste product that contained As, Fe, Cu, and Zn. After 26 years, chemical changes in the lime layer were restricted to a 0.7-cm zone at the tailing contact. He concluded that acid from the tailing has the potential to migrate upward into earthen covers, but this phenomenon was confined to a thin layer at the base of the cover.

Although not discussed, the data from the Anaconda tailing indicated no enrichment of the soil cover by As, Fe, Cu, or Zn, even though these constituents were elevated in the reject lime material (Dollhopf, 2001). The lack of evidence for As enrichment in the basal soil cover layer is significant as the mobility of As should not be inhibited by alkaline conditions to the same extent as cationic metals, like Cu, Fe, or Zn. These data suggest that diffusion is not operating as an important transport mechanism that could affect the cover soils.

Methods

Study Area

The study site comprises a 60 ha (150 acre) tailing repository covered and revegetated in 1981 (Fig. 1). The project area sits along the Continental Divide in southwestern New Mexico at an elevation of about 1,900 m above mean sea level. The climate is warm and dry with a mean annual precipitation of 410 mm and mean annual temperature near 10°C. Precipitation falls mainly in short, intense, thunderstorms between July and October; lower intensity rain storms and snow occur from November to March. Native soils are loamy-skeletal, clayey-skeletal, and fine families of Aridic Haplustalfs formed in residuum from late Tertiary and Quaternary conglomerates (Gila Conglomerate Formation). The surrounding vegetation is characteristic of the transition zone between desert grassland and the mixed evergreen woodland. In 2004, vegetation cover on the repository averaged 38 percent and was dominated by warm season grasses, forbs, and shrubs (Romig et al., 2006).



Figure 1. Vegetation condition on the reclaimed tailing repository in October, 2004.

Field Methods

We chose five sampling sites with convex slope configurations (0.5 to 3 percent slopes) to emulate the conditions that would exist on future reclamation areas in this region. Four samples of cover soil were collected from each profile using the method described below. Three discrete 5-cm interval samples were collected starting about one inch above the contact with tailing. The 2.5-cm zone immediately above the soil-tailing contact was not sampled to avoid mixing of tailing and soil. A single sample was collected from the remainder of the soil profile above the highest 5-cm interval sample. Two samples of underlying tailing were collected from each excavation. One sample represented the 7.5-cm interval immediately below the contact and the other tailing sample represented the next 13- to 15-cm interval.

Lab Methods

The cover soil and tailing samples were analyzed by Energy Laboratories in Billings, Montana. The soils were air-dried and passed through a 2-mm sieve. The analyses were conducted on the fine-earth fraction and included for particle size distribution (Gee and Bauder, 1986); CaCO_3 equivalent, saturated paste pH, paste extract EC (Salinity Laboratory Staff, 1954); and paste extract Al (EPA 200.7; ICP), Fe (EPA 200.7; ICP), and SO_4^{-2} (EPA 300.0; IC).

Results

Cover Soil Characteristics

The cover soils varied in texture from loamy sand to silty clay loam, ranging from 6 to 30 percent clay (Table 1). The rock fragment contents ranged from 2 to almost 30 percent by mass. The cover soils were placed in lifts using scrapers and the initial character of the materials was likely to have varied. The intra-profile variation of the cover soils is not precisely known because sampling was not conducted at the time of placement. Thus, the original character of the materials is inferred from the data represented by the upper sampling intervals of the test pits recognizing that the basal contact zone may have been affected by diffusion, convection, or construction related mixing.

Excluding data from the lowest cover soil interval, the paste pH ranged from 6.0 to 7.7; extract EC ranged from 0.45 to 3.20 dS/m; paste extract Al and Fe were not detected at levels above 0.5 mg/L; paste extract SO_4^{-2} ranged from 40 to 2,290 mg/L; and lime range from 0.9 to 3.7 percent. Based on the analyses of 78 soil samples from undisturbed areas, the paste pH, extract EC, and CaCO_3 data are consistent with data for the surrounding soils (pH ranged from 5.0 to 7.9; EC ranged from 0.18 to 3.74 dS/m; and the CaCO_3 content ranged from 0.4 to 6.0 percent).

Tailing Characteristics

The chemistry of the tailing indicates various degrees of oxidation and weathering occurred prior to deposition and covering (Table 1). Paste pH ranged from 2.5 to 7.3; extract EC varied from 2.94 to 14.2 dS/m; paste extract Al ranged from less than 0.5 mg/L in the circumneutral pH tailing to 1,850 mg/L in the lowest pH materials; similarly paste extract Fe ranged from non-detect to 965 mg/L; extract SO_4^{-2} ranged from 1,890 to 32,000 mg/L; and measurable CaCO_3 equivalent was detected in a few samples.

Table 1. Physical and chemical properties of cover soils and underlying tailing.

Site	Depth	Material	Sand	Silt	Clay	USDA Texture	Coarse Fragments	pH	EC	Al	Fe	SO ₄	CaCO ₃
	Cm		----- % -----				-- % wt --		dS/m		----- mg/l -----		%
TRT-1	0-43	Soil	75	15	10	SL	12	6.0	3.20	<0.5	<0.5	2290	0.9
	43-48	Soil	86	8	3	LS	20	7.1	3.27	<0.5	<0.5	2330	0.9
	48-53	Soil	82	11	7	LS	23	7.1	3.87	<0.5	<0.5	2830	1.1
	53-58	Soil	82	11	7	LS	25	4.8	3.73	1.5	<0.5	2770	1.2
	61-69	Tailing	46	35	19	L	4	3.5	4.03	15.5	3.1	3880	0.4
	69-84	Tailing	42	43	15	L	2	6.6	3.20	<0.5	<0.5	2100	0.4
TRT-2	0-23	Soil	68	14	18	SL	11	7.4	1.07	<0.5	<0.5	336	1.4
	23-28	Soil	72	12	16	SL	5	7.5	1.07	<0.5	<0.5	375	1.2
	28-33	Soil	74	12	14	SL	9	7.5	1.13	<0.5	<0.5	425	1.1
	33-38	Soil	52	32	16	L	12	6.4	3.36	<0.5	<0.5	2150	0.9
	41-48	Tailing	41	41	18	L	4	3.6	3.63	36.3	18.1	3380	0.2
	48-56	Tailing	38	45	17	L	3	7.3	2.94	<0.5	<0.5	1890	0.7
TRT-3	0-43	Soil	75	10	15	SL	25	7.4	0.45	<0.5	<0.5	40	3.7
	43-48	Soil	73	8	19	SL	28	7.5	0.83	<0.5	<0.5	196	1.6
	48-53	Soil	76	6	18	SL	27	7.5	1.13	<0.5	<0.5	418	1.6
	53-58	Soil	78	8	14	SL	24	5.2	2.72	<0.5	<0.5	1790	0.9
	61-69	Tailing	41	40	19	L	2	2.6	4.45	102	127	2680	<0.1
	69-84	Tailing	58	28	14	SL	<2	2.5	6.21	208	463	4750	<0.1
TRT-4	0-43	Soil	32	44	24	L	6	7.7	0.65	<0.5	<0.5	131	2.5
	43-48	Soil	75	13	12	SL	20	7.4	2.59	<0.5	<0.5	1530	1.2
	48-53	Soil	76	10	14	SL	23	7.0	3.79	<0.5	<0.5	2660	1.1
	53-58	Soil	72	13	15	SL	24	3.8	8.50	342	<0.5	11500	0.6
	61-69	Tailing	30	47	23	L	6	2.5	13.6	1850	691	29600	<0.1
	69-84	Tailing	32	47	21	L	5	2.6	14.2	1910	965	32000	<0.1
TRT-5	0-36	Soil	22	50	28	CL	8	7.6	0.57	<0.5	<0.5	72	3.7
	36-41	Soil	20	52	28	SiCL	4	7.6	1.13	<0.5	<0.5	408	3.0
	41-46	Soil	18	52	30	SiCL	3	7.6	1.95	<0.5	<0.5	1010	2.7
	46-51	Soil	22	50	28	CL	2	6.7	4.24	<0.5	<0.5	2760	2.5
	53-61	Tailing	42	36	22	L	7	2.9	7.28	642	22.2	12000	<0.1
	61-76	Tailing	43	36	21	L	7	3.1	5.41	271	91.5	7010	<0.1

Abbreviations: SL = sandy loam; LS = loamy sand; L = loam; CL = clay loam; SiCL = silty clay loam

Profile Distribution of Constituents

Paste pH in the basal contact zone of the cover decreased by less than 1 to slightly more than 3 units when compared to the overlying soil layer; otherwise no definitive changes in pH were noted in the cover soils (Fig. 2). Sulfate is the predominant anion in solution in these soils and the SO_4^{-2} and EC profiles show similar patterns (Fig. 2). For each of the five profiles, SO_4^{-2} concentrations generally increased regularly with depth to the tailing contact. Extractable Fe occurred below the detection limits in all cases and extractable Al was measured in the basal contact zone in only two profiles (Fig. 2). CaCO_3 equivalent generally, but not always, decreased with depth.

The lower 7.5 cm of the cover soils (basal contact zone) exhibit clear decreases in soil pH and increases in salinity and SO_4^{-2} concentrations. However, it is difficult to conclude that changes in the soil chemistry in the basal contact zone were associated with upward migration as discussed below.

Discussion

Acidification of the Basal Contact Zone

Hydrolysis of Al and Fe are the primary reactions that lead to acidification of soils (Thomas and Hargrove, 1984; Evangelou, 1998). Oxidation of sulfide minerals (e.g., pyrite) in tailing, waste rock, and spoils results in the generation of acidity and the release of Fe and SO_4^{-2} . If not buffered by reactive neutralizing minerals, the acidic conditions associated with pyrite oxidation accelerate weathering of primary and secondary aluminosilicate minerals releasing Al and other weathering products. These weathering products may react with charged mineral surfaces, form new minerals and sesquioxides, or dissolve in the soil solution. Weathering products that are not partitioned in solid phases or the exchange complex are available for transport. Following the oxidation of sulfide-bearing wastes, the major weathering products expected to be available for transport are Fe, Al, and SO_4^{-2} . Thus, the fate (movement and reactions) of Fe and Al are of primary concern with respect to understanding cover soil acidification. Because SO_4^{-2} is relatively conservative and unaffected by secondary reactions, it is important in detecting the movement of constituents.

The reduction in pH in the basal contact zone indicates that some process has caused acidification in the lower part of the cover. Oxidation of tailing physically mixed into the lower part of the cover during construction and diffusion are the two primary mechanisms identified that could contribute to acidification of the basal contact zone. Physical mixing is identified as a potential mechanism based on field observations of tonguing and incorporation of tailing in the cover soils (Fig. 3). Although none of the profiles sampled showed gross manifestations of mixing, and a 2.5 cm zone immediately above the tailing contact was excluded, it is impossible to conclude that mixing during construction did not occur at the interface. Distinguishing the causative mechanism is important because the effects of physical mixing are constrained, whereas the diffusion may proceed into the future, albeit at ever diminishing rates.

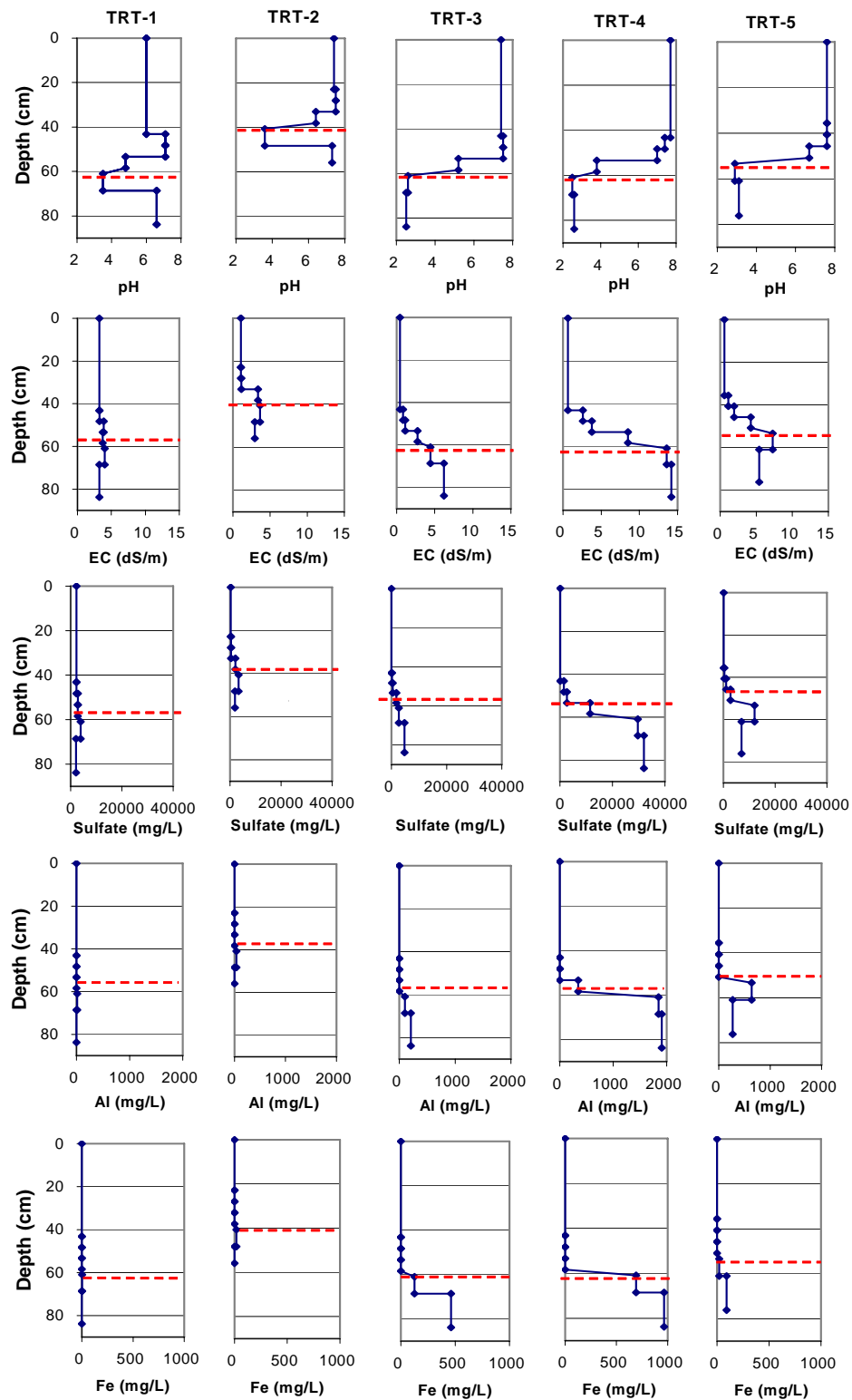


Figure 2. Property depth curves for cover and tailing profiles. Dashed lines represent the cover-tailing interface.

The chemical data collected from these profiles do not provide conclusive evidence that upward migration is the causative factor for acidification in the basal contact zone. Incorporation of tailing probably explains at least part of the observed decrease in pH. The chemical data are discussed below with respect to the potential for upward migration. Sulfate is more conservative with respect to mobility than Fe or Al, which are strongly affected by adsorption reactions and secondary mineral formation. Sulfate solubility is likely to be controlled primarily by gypsum where free Ca occurs in solution. The regular increase in SO_4^{-2} concentrations with depth in the cover soils suggests downward translocation (leaching) of SO_4^{-2} from the surface layers. Diffusion from the underlying tailing, or SO_4^{-2} generated through oxidation of tailing mixed in the basal cover during construction are other possible causes. It is not possible to distinguish the relative contributions of these processes with respect the SO_4^{-2} distribution. Thus, the SO_4^{-2} distribution is inconclusive with respect to demonstrating upward migration as an active process in these cover-waste systems.



Figure 3. Tonguing of tailing in the basal contact zone of a soil cover overlying tailing. Wire mesh grid spacing is 10 cm.

The extractable Fe and Al data are incongruous with respect to demonstrating upward migration from the tailing. The extractable Fe and Al concentrations in the tailing are indicative of systems controlled by SO_4^{-2} salts. In contrast, extractable Fe was not detected in any of the cover soils, and extractable Al was measured in the basal contact zone in only two profiles, indicating that Fe and Al in the basal contact zone were generally not associated with more soluble SO_4^{-2} salts. The strong tendency of Fe to hydrolyze and form oxyhydroxides of low

solubility at pH <3 may account for the apparent lack of enrichment of Fe above the tailing contact. Similarly, Al tends to form relatively insoluble oxyhydroxides at pH greater than about 4.

The presence of SO_4^{-2} and other complexing ligands are expected to increase the solubility of Fe and Al relative to the concentrations that would be predicted solely on the basis of pH. The extractable Al concentration in the basal contact zone of TRT-4 is indicative of control by an Al SO_4^{-2} salt, suggesting upward migration of Al and SO_4^{-2} . However, the lack of measurable Fe in this sample suggests that Fe was not co-transported, arguing that the underlying tailing was not the source for the Al and SO_4^{-2} in the basal contact zone. Assuming that Fe and Al did migrate from the underlying tailing, we hypothesize that they must first undergo hydrolysis (lowering the pH of the cover soil) and then be partitioned into insoluble solid phases and/or adsorbed on the exchange complex. A similar chemical signature would be expected in association with weathering of tailing incorporated in the basal contact zone of the cover. Thus, differentiating the causative mechanism is problematic on the basis of these data.

Regardless of the mechanism (mixing or diffusion), partitioning of Fe and Al into insoluble forms reduces the potential for Al toxicity and limits the concentration gradient relative to the overlying layers, thereby restricting the driving force behind diffusion and the potential for future upward migration. Over the long-term, leaching in the upper part of the cover will redistribute salts into the accumulation zone near the basal contact, which lies at 18 to 24 inches below the soil surface based the typical depth of CaCO_3 accumulation in the native soils. Infrequent, episodic leaching events are expected to move salts below this depth in association with above normal winter precipitation. Significant leaching events are expected to occur 2 to 3 time per century based on long-term water balance modeling and soil development relations.

Implications for Revegetation and Cover Function

Modest changes in the chemistry of the cover soils occurred over the 20-year period since construction. These changes were restricted to the basal contact zone. The chemical conditions in the basal contact zone are generally deemed non-limiting for plant growth, especially considering that they occur below the primary depth of rooting for vegetation in this region (Peace et al., 2004; Romig et al., 2006). Specifically, 66 percent of the roots occur in the upper 20 cm (8 inches) and almost 90 percent occur in the upper 50 cm (20 inches) of the soils based on measurements made in both native and reclaimed sites in the project area (Romig et al., 2006).

Salinity levels in the basal contact zone are typically in the 2 to 4 dS/m range, which even sensitive plants can tolerate and the highest salinity level (8.5 dS/m) is within the acceptable range for more tolerant plants (Rhoades et al., 1992).

Extractable Al, which is typically the primary constituent of concern in acid soils, generally occurred at low levels. Inorganic Al is toxic to plants, with the degree of toxicity best correlated with the Al^{3+} ion (Adams and Lund, 1966; Pavan et al., 1982; and Parker et al., 1987). The mechanism of toxicity is not well understood, but steric or size considerations may account for the reduced toxicity of the mono- and polynuclear-aluminum hydroxide complexes (Parker et al., 1989). Similarly, complexation by SO_4^{-2} , F^- , and organic acid ligands reduces the phytotoxicity of Al (Hue et al., 1986 and Parker et al., 1989). In addition, solution Ca and Na are attributed with decreasing Al toxicity through reductions in Al activity (ionic strength effect) and direct physiological effects (Adams and Hathcock, 1984; Alva et al., 1986; Kinraide and Parker, 1987;

and Parker et al., 1987). Gypsum has been used as an acid soil amendment partially because of the reduced phytotoxicity of the $\text{Al}_2(\text{SO}_4)_3$ complexes and because it may reduce induced Ca deficiencies (Oates and Caldwell, 1985 and Sumner et al., 1986). The use of gypsum as an amendment to ameliorate Al toxicity is significant because most sulfidic mine wastes generate gypsum. The elevated levels of soluble Al measured in the basal contact zone of profile TRT-4 are of limited concern considering the high ionic strength of the solutions and the occurrence of SO_4^{2-} and other complexing ligands (e.g., fluoride and phosphate).

Soil cover function in the project area is dependant on having adequate vegetation cover to provide erosion control and transpiration demand. Vegetation canopy cover is sufficient to control erosion on the tailing repositories. Mean maximum leaf area index (LAI) measured on the tailing repository was 0.29, which provided adequate transpiration demand based on unsaturated flow water balance modeling (Romig et al., 2006). Thus, soil covers similar to those evaluated here support sufficient levels of vegetation to maintain cover functions.

Summary

The results of this investigation are consistent with studies conducted at other mine sites that involving upward migration of sodium and acidity. In particular, preliminary concerns of deleterious affects on cover performance based on short term measurements prove to be inconsequential over the long-term. Evidence for time-transgressive upward redistribution of constituents of sufficient magnitude to disrupt biological processes has not been demonstrated and the changes in the chemistry of the cover soil are restricted to the relatively thin zone at the interface between the cover and underlying waste material. The mechanisms leading to acidification of the basal contact zone are difficult to determine with certainty, but probably include physical mixing of tailing during construction and/or a component of upward migration. Regardless of the mechanism, the degree and magnitude of change in chemistry in the cover soils are considered practically insignificant with respect to plant growth and cover function in this ecosystem.

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