

Assessing mine water pollution: from laboratory to field scale

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Abstract We use previous investigations of the waste rock deposits at the Aitik site in northern Sweden, attempting to resolve quantitatively an observed scale-dependence in mineral weathering rates, which control both contaminant loads and their natural attenuation at the site. The data considered represent a scale transition from small-scale batch experiments, via large-scale column experiments, to field investigations at the site. We identify experimental differences and quantify associated scaling factors that can to a large degree explain the observed scale-dependence in mineral dissolution rates at the Aitik site. This scale-dependence is consistent with other observations of mineral weathering in laboratory and watershed studies, suggesting that at least some of the effects identified in our analysis may be generally applicable and important when extrapolating weathering rates from laboratory to field scale.

INTRODUCTION

Uncontrolled contaminant release from abandoned and operating mines poses a major environmental hazard to freshwater resources worldwide. Current estimates indicate that 11% of the global sulphate flux from the continents to the oceans arises from mining activities alone (Nordstrom & Southam, 1997, citing Berner & Berner, 1996). The acidity and dissolved metals contamination associated with the weathering of sulphide minerals poses an immediate threat to groundwaters that interact with mine workings and to surface waters that receive contaminated discharges. Management decisions for abandoned sites and strategies for decommissioning of operating mines require quantification of potential impacts within the Source-Pathway-Target risk assessment framework. This study uses earlier investigations of the waste rock deposits at the Aitik site in northern Sweden, currently Europe's largest operating copper mine (Strömberg *et al.*, 1994; Strömberg & Banwart, 1994, 1995; Eriksson, 1996; Strömberg, 1997; Eriksson & Destouni, 1997; Eriksson *et al.*, 1997; Strömberg & Banwart, 1998a,b). The previous investigations aimed to identify dominant contaminant generation and attenuation processes and to quantify their rates across a range of physical scales, including small-scale batch experiments, large-scale column experiments, and field investigations at the site (Table 1). This paper is focused on resolving an observed scale-dependence in mineral weathering rates, which control both contaminant loads and contaminant natural attenuation at the site.

Table 1 Characteristics of the three experimental scales.

Experimental scale	M , mass of solid material (kg)	Q , water flow ($\text{m}^3 \text{s}^{-1}$)	T , temperature ($^{\circ}\text{C}$)	pH
¹ Batch	0.15	-	20–23	3.3
² Column	⁴ 1.8×10^3	9.2×10^{-9}	4–10	≈ 3.5
³ Field	⁴ 9.5×10^{10}	⁵ 1.7×10^{-1}	1–4	3.8–4.2

¹ From Strömberg & Banwart (1998a).

² From Strömberg & Banwart (1998b).

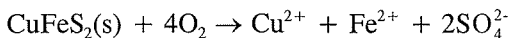
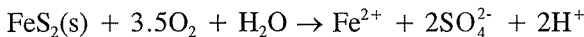
³ From Strömberg & Banwart (1994).

⁴ Calculated as: $M = HA(I - n)\rho_s$, where H is height, A is total area, n is porosity, and ρ_s is density of the solid material; in the field, the average $H = 20$ m, $A = 2.6 \times 10^6$ m², $n = 0.35$, and $\rho_s = 2.8 \times 10^3$ kg m⁻³; in the column experiments, $H = 2$ m, $A = 0.5$ m², $n = 0.35$, and $\rho_s = 2.8 \times 10^3$ kg m⁻³.

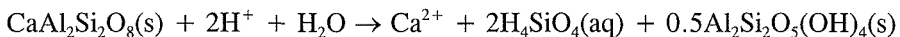
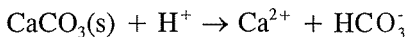
⁵ Average flow in the main drainage ditch at the Aitik site (Strömberg & Banwart, 1994).

CONTAMINANT SOURCES AND NATURAL ATTENUATION

Acidity, ferrous iron and copper are produced during oxidative weathering of pyrite ($\text{FeS}_2(\text{s})$) and chalcopyrite ($\text{CuFeS}_2(\text{s})$):



Natural attenuation of acidity is provided by weathering of calcite and aluminosilicate minerals (represented here by anorthite) associated with the waste rock:



The relative rates of these weathering reactions determine whether a mine water discharge will be net acidic or net alkaline. Compiled weathering rate data normalized to surface area (Strömberg & Banwart, 1994; Stumm & Morgan, 1996, p. 786) indicate that calcite dissolves much more rapidly than pyrite and chalcopyrite (under oxic conditions), which in turn dissolve more rapidly than silicate minerals. These relative rates suggest that if calcite is present in sufficient amount it will dissolve rapidly enough to consume acidity that is released from the sulphide minerals, and thus maintain a net-alkaline discharge with circumneutral pH. If the calcite present becomes depleted before the sulphide minerals, the slow dissolution of silicate minerals can provide additional attenuation of the acidity. The relative rates of silicate dissolution and sulphide oxidation then shift and keep the pH at a lower level. Such a transition from net alkaline to net acidic waters is critical to the evolution of contaminant loadings because of the large increase in metal ion solubility and thus mobility under acidic conditions.

SCALE-DEPENDENCE OF WEATHERING RATES AT THE AITIK SITE

Table 1 lists the characteristics of the three different experimental scales. By evaluating previously reported experimental data on relevant tracer release rates

(Strömberg & Banwart, 1994, 1998a,b) we estimate associated mineral dissolution rates, normalized by the total experimental mass, for the three experimental scales (Table 2). A comparison between the rates at different scales reveals a significant scale-dependence, with one to three orders-of-magnitude lower rates in the field than in the batch experiments and with the rates for the large column experiments in between. Scale-dependence such as we observe at the Aitik site is also commonly observed in other systems (Schnoor, 1990; White & Petersen, 1990; Swoboda-Colberg & Drever, 1993; Drever & Clow, 1996).

To resolve the cause of the scale-dependence at the Aitik site, we identified five main differences between the prevailing experimental conditions at the different scales. The identified differences are related to: (a) environmental temperature (Table 1); (b) pore water pH (Table 1); (c) particle size distribution, differing between all experimental scales and with different particle sizes exhibiting significantly different weathering rates for some minerals (Strömberg & Banwart, 1998a); (d) mineral content, e.g. sulphide content that is highly variable in the field, and on the average considerably lower than on the other experimental scales (Strömberg, 1997); and (e) water flow patterns, ranging from total mixing and no flow in the batch experiments, via homogeneous flow in the column experiments, to existence of preferential flow paths in the field (Eriksson & Destouni, 1997; Eriksson *et al.*, 1997). Based on these differences, we quantified associated scaling factors, α_i , with the index i referring to the specific experimental difference, labelled as above, for upscaling the batch dissolution rates to apply to the two larger scales, such that:

$$R_s^{C/F} = \prod \alpha_i R^B \quad (1)$$

where $R_s^{C/F}$ is the dissolution rate that is being upscaled from the corresponding batch dissolution rate R^B to column or field conditions, indicated by index C or F . The detailed description of the actual quantification of the different α_i is presented in

Table 2 Estimated mineral dissolution rates from observed tracer release rates.

Mineral/Tracer	^{1,2} R^B , mineral dissolution rate in batch experiments (mol kg ⁻¹ s ⁻¹)	^{1,3} R^C , mineral dissolution rate in column experiments (mol kg ⁻¹ s ⁻¹)	^{1,4} R^F , mineral dissolution rate in the field (mol kg ⁻¹ s ⁻¹)
Albite/Na ⁺	2.2×10^{-11}	9.3×10^{-12}	3.7×10^{-12}
Anorthite/Ca ²⁺	3.2×10^{-10}	8.4×10^{-11}	8.5×10^{-12}
Biotite/Mg ²⁺	1.9×10^{-10}	2.3×10^{-11}	2.8×10^{-12}
Chalcopyrite/Cu ²⁺	2.8×10^{-10}	1.1×10^{-11}	5.3×10^{-13}
Pyrite/ ³ SO ₄ ²⁻	1.3×10^{-9}	8.4×10^{-11}	1.2×10^{-11}

¹ Mineral dissolution rates were estimated assuming stoichiometric dissolution of the minerals with Na⁺, Ca²⁺, Mg²⁺, Cu²⁺, and SO₄²⁻ originating mainly from albite, anorthite, biotite, chalcopyrite, and pyrite and chalcopyrite, respectively.

² Tracer release rate from Strömberg & Banwart (1998a, their Table 4, average of triplicates).

³ Tracer release rate from Strömberg & Banwart (1998b, their Table 5).

⁴ Tracer release rate estimated as $S = \sum Q_i C_i / M$ where Q is water flow and C is tracer concentration in the two drainage ditches at the Aitik site (reported by Strömberg & Banwart, 1994, their Table 1) and M is the waste rock mass (Table 1).

⁵ Corrected for chalcopyrite dissolution.

Destouni et al. (1998).

Explanations other than those identified by us as important at the Aitik site, for observation of lower dissolution rates in the field compared to in laboratory experiments, have also been suggested in the literature, (cf. Brantley & Stillings, 1996, and references therein), and include e.g. incomplete wetting of reactive mineral surfaces under hydraulically unsaturated conditions, wetting and drying cycles that armour reactive surfaces, chemical inhibition due to back reactions of weathering products, chemical affinity effects as solute activities approach solubility limits for the source minerals, and accumulation of primary weathering products in secondary mineral phases or as sorbed species.

RESULTS AND CONCLUSIONS

For the upscaling from batch to field, Fig. 1 summarizes upscaled rates, R_s^f according to equation (1), for the minerals listed in Table 2. Figure 1 also shows the individual contributions of the different scaling factors α_i in equation (1). The results shown in Fig. 1, and similar results for the upscaling from the batch to the large column experiments, imply that the identified experimental differences and associated scaling factors can to a large degree explain the observed scale-dependence in mineral dissolution rates in the waste rock material from the Aitik site.

This analysis has provided an assessment of important factors that contribute to scale-dependent weathering rates at a particular mine waste deposit. The relative

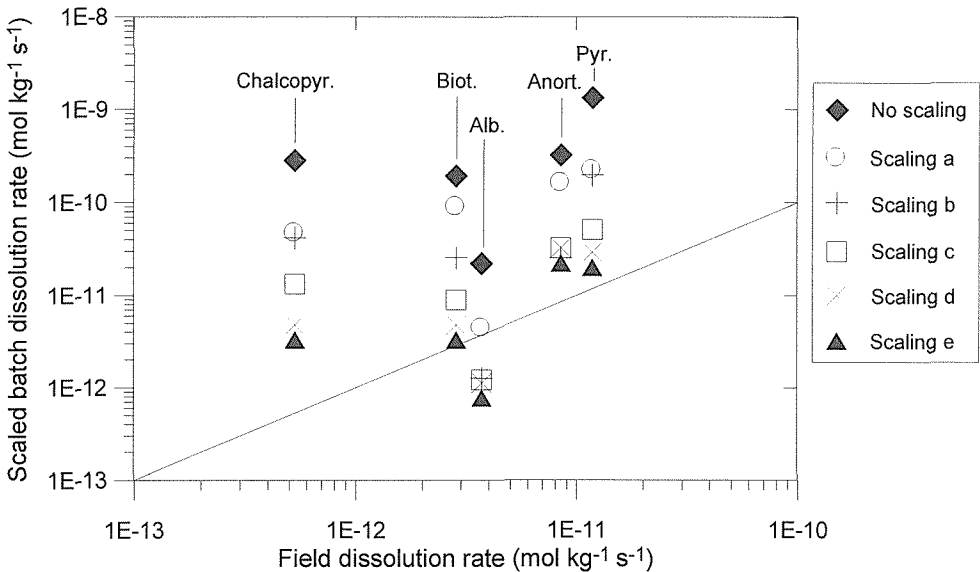


Fig. 1 Cumulative effect of upscaling dissolution rates from batch experiments to the field according to equation (1) as a function of observed dissolution rates in the field (Table 2). The solid line denotes the ideal case, “perfect prediction”, where the scaled batch rate equals the observed field rate. Individual minerals are denoted Alb. (Albite), Anort. (Anorthite), Biot. (Biotite), Chalcopyr. (Chalcopyrite), and Pyr. (Pyrite), respectively.

magnitude of the discrepancy between laboratory rates and rates at the field scale, however, is similar to previous observations of mineral weathering in laboratory and watershed studies (Schnoor, 1990; White & Peterson, 1990; Swoboda-Colberg & Drever, 1993; Velbel, 1993; Drever & Clow, 1996). This consistency suggests that at least some of the effects identified in our analysis may be generally applicable and important when extrapolating weathering rates from laboratory to field scale.

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