MODELING FIELD-SCALE TRANSPORT OF WEATHERING PRODUCTS IN MINING WASTE ROCK DUMPS[1]

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Abstract: A stochastic-advective approach to modeling field-scale transport of weathering products in mining waste rock dumps is presented for irreversible dissolution of primary minerals in the waste rock. The effect of flow heterogeneity is investigated for a classical unimodal distribution of water residence time and for bimodal distributions that reflect the existence of preferential flow paths. The relationship between the characteristic time scale of the dissolution process and the characteristic hydrological time scales expressed by the water residence times in the dump plays an important role for the effect of flow heterogeneity on the field-scale solute transport. In particular, flow heterogeneity manifested by the existence of preferential flow paths will reduce the maximum value of the field-scale solute flux and increase the temporal spreading of the solute breakthrough curve at the lower boundary of the dump.

Additional Key Words: mining waste, flow heterogeneity, solute flux, water residence time distribution, stochastic modeling, preferential flow paths.

Introduction

When uncovering the ore body in an open pit, large quantities of waste rock are produced and deposited on the ground surface. Oxidative weathering of sulfide minerals produces free acidity, sulfate and dissolved metal species. Heavy metals may then leach through the deposit into surrounding drainage ditches or underlying aquifers. To evaluate the need for protective measures and to optimize such measures, the transport of environmentally disturbing weathering products through waste rock dumps needs to be quantified. The required output of such modeling is the field-scale mass arrival of solute to and through the lower boundary of the waste rock dump, to be used as input for predicting the subsequent solute movement in the drainage ditches or the aquifer (Destouni and Eriksson 1994).

Because waste rock dumps constitute highly heterogeneous formations with regard to their texture and structure, realistic models of field-scale solute transport in mining waste rock should be able to handle large and irregular variability in their model parameters. A stochastic-advective approach to modeling reactive solute transport in heterogeneous porous media (Shapiro and Cvetkovic 1988, Cvetkovic and Shapiro 1990, Destouni and Cvetkovic 1991, Dagan et al 1992) will be extended to include irreversible dissolution of primary minerals in this paper. For weathering products that are soluble in water, the weathering process will be considered as a spatially and temporally distributed source of solute. Weathering of chalcopyrite will thereby be used as an illustrative study case, based on field data and simulation results that were reported by Strömberg and Banwart (1994).

For the considered study case, the effects of spatial variability in hydraulic properties of the waste rock and of the temporal variation of the weathering rate on the field-scale solute transport through the dump will be investigated. A

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mathematical model for handling the concept of preferential flow paths in the stochastic-advective framework will be discussed (Destouni 1993; Destouni et al. 1993, Sassner et al 1994). In particular, the importance of the relationship between the characteristic time scale of the dissolution reaction and the characteristic water residence times in the dump will be elucidated. Preferential flow paths, channeling, or piping has been observed in mining waste rock (Harries and Ritchie 1983), however, the existence and influence of such preferential flow paths has not been quantified. At the Aitik site in northern Sweden, which is the site investigated by Strömberg and Banwart (1994), field observations and modeling results indicate that only 20% of the waste material contributes to the observed release of sulfate and copper (Gibson et al 1992), which may be a preferential flow path effect.

Solute Transport in Weathering Waste Rock

In waste rock dumps, interacting physical, chemical, and biochemical processes weather (dissolve) the waste rock continuously. In this paper, weathering of primary mineral is considered as an irreversible, heterogeneous reaction. Under oxic conditions, the weathering process will take place throughout the total height of the dump and the source of solute will be distributed along the streamlines that define the mean flow direction. When water infiltrates the waste rock, weathering products that are soluble in water will be transported through the dump into the underlying aquifer or into surrounding drainage ditches. To quantify the long-term release of solute at the lower boundary of the waste rock dump we consider advection-dominated transport where the flow of water is steady and the mean flow direction is downward.

The advective approach is based on the assumption that field-scale spreading of solute is primarily an effect of variability in solute advection between different stream tubes such that local dispersive mechanisms can be neglected (Dagan1989). Under these conditions the mass balance equation for an arbitrary stream tube can be formulated as:

$$\theta\frac{\partial c}{\partial t}+\frac{\partial(qc)}{\partial z}=\theta\left[\frac{\partial c}{\partial t}\right]_{chem}, \qquad (1)$$

where c is the solute concentration, t is time, θ is the volumetric water content, z is the vertical space coordinate with origin at the dump surface and positive downward, and q is the flow of water per unit cross-sectional area normal to z.

The right-hand side of the equation expresses the change in concentration due to the prevailing weathering processes such that

$$\left[\frac{\partial c}{\partial t}\right]_{chem}=\frac{1}{\theta}r\alpha=-\frac{1}{\theta}\frac{\partial c^{*}}{\partial t}, \qquad (2)$$

where r is the reaction rate defined as the produced amount of the considered aqueous phase species per unit time and unit area of reacting interface, and α is the specific reactive surface area of the primary mineral defined per unit bulk volume. Furthermore, $\partial c^{*}/\partial t$ expresses the rate of dissolution of the solid phase species with $c^{*}\equiv\rho_{m}\eta$, in which ρ_{m} is the considered mineral content per unit bulk volume and η is the amount of produced aqueous phase species per unit amount of dissolved mineral.

Combining equations 1 and 2, inserting the mean porewater velocity $v = q/\theta$, and introducing the new variable $d\tau \equiv dz/v(z)$, equation 1 can be rewritten as:

$$\theta\frac{\partial c}{\partial t} + \theta\frac{\partial c}{\partial \tau} = -\frac{\partial c^*}{\partial t}. \qquad (3)$$

In equation 3, $\tau(z) = \int_0^z d\zeta / v(\zeta)$ expresses the water residence time in the steam tube, or the travel time for an ideal tracer from the surface of the dump to the depth z (Shapiro and Cvetkovic 1988).

The specific surface area α in equation 2 will be a function of the primary mineral content of the deposit and may thus vary with time due to the weathering process. The reaction rate, r, is dependent on the rate determining step of the reaction and may also be time-dependent, for instance, due to changes in pH or temperature. For chalcopyrite oxidation ($CuFeS_{2(s)} + 4O_{2(aq)} \rightarrow Cu^{2+} + Fe^{2+} + 2SO_4^{2-}$) of relatively fresh waste rock (deposited 5 years on the average) Strömberg and Banwart (1994) reported an r value in the order of 10^{-11} moles/(m^2s); this value was the result of initial geochemical modeling of an oxic waste rock dump with the mineral content, ρ_m, approximately equal to 10 moles/m^3. The specific surface area of chalcopyrite was estimated to about 2,000 m^2/m^3, however, comparison with field data indicated that only a fraction of the area is reactive. Because neither field data nor the geochemical modeling of Strömberg and Banwart (1994) provide information on the long-term dynamics of the dissolution rate, different functions $c^*(t)$ are conceivable for the dissolution process, two of which will be presented here for illustrative purposes.

The first dissolution process, in the following referred to as case 1, is obtained by assuming a constant rate of dissolution until all the primary mineral has been dissolved, and can be expressed as:

$$c^* = c_0^*(1 - kt)\,, \qquad (4)$$

in which k is a constant rate coefficient and c_0^* is defined from the initial mineral content ρ_{m0}. Assuming initial conditions consistent with those reported by Strömberg and Banwart (1994), a relevant k value for chalcopyrite dissolution can be estimated by differentiating equation 4 and inserting in equation 2, which results in $k = \alpha r / c_0^*$. With a reactive surface area of 10% of the specific surface area, $k = 0.1 \cdot 2000 \cdot 10^{-11}/10 = 2 \cdot 10^{-10}$ s^{-1}, which corresponds to a turnover time $t_s = 1/k$ of approximately 150 years.

Inserting equation 4 in equation 3 and solving for the solute mass flux $s(t,Z) = c(t,Z)q(Z)$, where Z is the total height of the waste rock dump, yields:

$$\frac{s(t,Z)}{c_0^* Z} = \begin{cases} \dfrac{t}{Tt_s} & \text{for } t < \mathrm{T} \\ \dfrac{1}{t_s} & \text{for } \mathrm{T} \le t < t_s \\ \dfrac{1}{t_s} - \dfrac{t - t_s}{\mathrm{T}t_s} & \text{for } t_s \le t \le t_s + \mathrm{T} \\ 0 & \text{for } t > t_s + \mathrm{T} \end{cases} \qquad \mathrm{T} \le t_s \tag{5}$$

$$\frac{s(t,Z)}{c_0^* Z} = \begin{cases} \dfrac{t}{\mathrm{T}t_s} & \text{for } t < t_s \\ \dfrac{1}{\mathrm{T}} & \text{for } t_s \le t < \mathrm{T} \\ \dfrac{1}{\mathrm{T}} - \dfrac{t - \mathrm{T}}{\mathrm{T}t_s} & \text{for } \mathrm{T} \le t \le t_s + \mathrm{T} \\ 0 & \text{for } t > t_s + \mathrm{T} \end{cases} \qquad \mathrm{T} > t_s$$

The solute flux $s(t,Z)$ quantifies the mass flow rate produced in a stream tube per unit cross-sectional area of the dump. In equation 5, $\mathrm{T} = \tau(Z)$ is the water residence time in an arbitrary stream tube spanning the vertical extent of the dump. For Swedish hydrological conditions (average annual rainfall: 0.7m/yr), an average infiltration rate of 0.5 m/yr can be expected, implying an average water residence time of 4 yr for a 20-m-high waste rock dump with an average volumetric water content of 10%.

The second dissolution process, in the following referred to as case 2, describes an exponentially decreasing mineral content and can be expressed as:

$$c^* = c_0^* e^{-kt} . \tag{6}$$

Inserting equation 6 in equation 3 and solving for $s(t,Z)$, yields:

$$\begin{aligned} \frac{s(t,Z)}{c_0^* Z} &= \frac{1}{\mathrm{T}}(1 - e^{-kt}) && \text{for } t < \mathrm{T} \\ \frac{s(t,Z)}{c_0^* Z} &= \frac{1}{\mathrm{T}}(e^{k\mathrm{T}} - 1)\, e^{-kt} && \text{for } t \ge \mathrm{T} \end{aligned} \tag{7}$$

In figure 1, the solutions 5 and 7 for case 1 (linearly decreasing mineral content, solid line) and case 2 (exponentially decreasing mineral content, dashed line), respectively, are illustrated; these solutions are relevant for an individual stream tube or for a homogeneous dump. The maximal solute flux is reached when water that has traveled in the considered stream tube throughout the total height of the dump reaches the lower boundary of the dump at $z = Z$. For case 1 dissolution, the solute flux will remain constant at its maximum value until all mineral has been dissolved, after which the solute mass flux will drop linearly to zero during the time period of one water residence time T. For case 2 dissolution, the solute flux will decrease exponentially in time after reaching its maximum value at $t = \mathrm{T}$. The maximum level of the solute flux will be the same in both cases for equal water residence time, T. The case 2 dissolution will continue over a longer period of time, but at a lower solute flux than case 1.

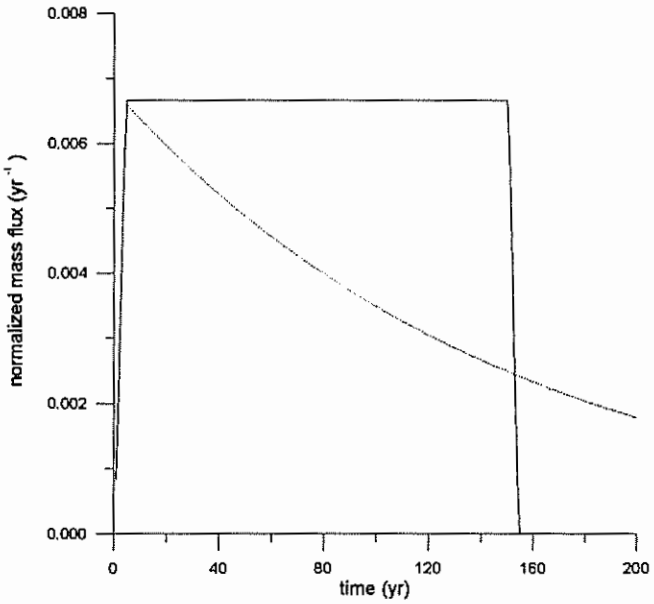

Figure 1. The normalized solute flux $s(t,Z)/c_0^*Z$ for Z=20m and T=4yr and k=1/150 yr^{-1} is illustrated for case 1 (linearly decreasing mineral content, solid line) and case 2 (exponentially decreasing mineral content, dashed line) dissolution. The solute flux peak is reached at the same time (t = T) for both dissolution processes. For case 2 dissolution, however, the solute flux is distributed over a longer period of time and continues at a lower level than for the case 1 dissolution.

The two illustrated dissolution cases (figure 1) represent two examples of possible dissolution dynamics. Because for our study case, i.e. the Aitik site, neither field nor laboratory data have yet been obtained for evaluating any potential changes in the dissolution rate, we will in the following illustrate the modeling methodology by use of case 1. This particular case, i.e. constant dissolution rate, was also used by Gibson et al (1992) to predict copper leaching at the Aitik site. Wherever applicable, however, the case 2 dissolution, or other dissolution dynamics, can readily be incorporated in the modeling approach that will be discussed in the following.

Transport Through a Heterogeneous Waste Rock Dump

Because waste rock dumps are large and heterogeneous porous formations, large and irregular variability in flow and transport parameters is conceivable. Such heterogeneity implies an unevenly distributed waterflow within the dump, such that both q and θ vary between different stream tubes. Because of the significant size of a typical waste rock dump, it is not likely that field measurments will provide sufficient data for deterministic knowledge of the fluctuations in v throughout the dump. A rational way to deal with this uncertainty is to regard v as a random variable (Dagan 1989). In the following, we will therefore regard the water residence time $\mathrm{T} = Z/v$ as a random variable, quantified by a probability density function (pdf), $f(\mathrm{T})$.

We regard each stream tube in the actual dump as a realization of an ensemble of all possible stream tubes according to $f(\mathrm{T})$, which are statistically equivalent on the field scale but locally different. As a consequence, the solute mass flux becomes a random function of the random variable T, depending deterministically on k, c_0^*, Z, and t. The pdf $f(\mathrm{T})$ is assumed stationary, i.e., the moments of $f(\mathrm{T})$ do not depend on spatial position. Under ergodic conditions, i.e., if the considered waste rock dump is sufficiently large relative to the correlation scale of T, ensemble averaging and space averaging at the lower boundary of the dump are equivalent (Cvetkovic et al. 1992; Destouni 1992). The expected solute flux can then be expressed as:

$$\bar{s}(t,Z) = \int_0^\infty s(t,Z;\mathrm{T}) f(\mathrm{T}) d\mathrm{T} \qquad (8)$$

The choice of $f(T)$ in equation 8 will in each field situation depend on the available field data and the sensitivity of the solution to the chosen pdf. The function $f(T)$ may thus be an ordinary unimodal pdf, or a bi- or multimodal distribution to reflect potential preferential flow paths. An often used unimodal pdf of T is the lognormal distribution (e.g., Simmons 1982; Cvetkovic et al. 1992)

$$f(T) = \frac{1}{T\sqrt{2\pi\sigma^2}} \exp\left[-\frac{1}{2} \frac{\left(\ln T - \ln T^G \right)^2}{\sigma^2} \right]. \qquad (9)$$

In equation 9, T^G is the geometric mean of the water residence time and σ^2 is the variance of $\ln T$.
Figure 2 illustrates the effect of flow heterogeneity on the expected solute flux for case 1 dissolution (linearly decreasing mineral content), by comparison of solution 8 for a lognormally distributed water residence time, T, to the homogeneous solution 5. The comparison shows that for realistic parameter values and for the considered unimodal distribution of T, flow heterogeneity within the waste rock dump does not have a significant effect on the expected solute flux through the dump. The reason for this insensitivity is that the characteristic hydrological time scale, i.e., the average water residence time of 4 years, is much smaller than the characteristic chemical time scale of 150 years. As a consequence, the transport process becomes chemically dominated.

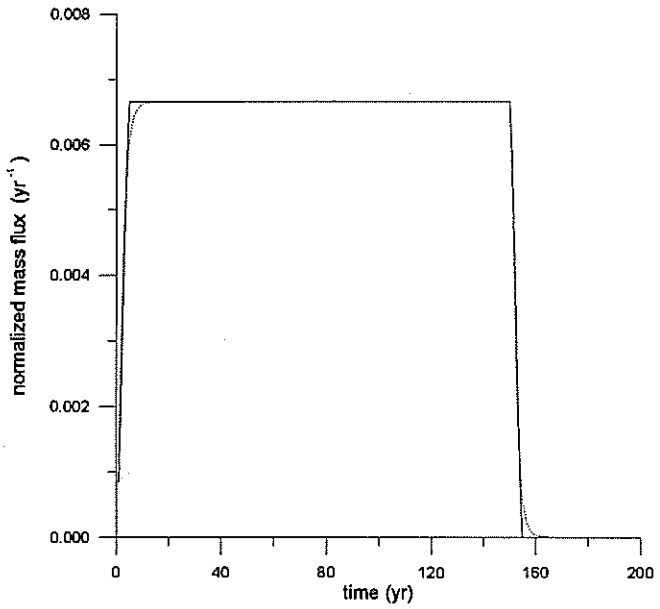

Figure 2. The normalized expected solute flux, $\bar{s}(t,Z)/c_0^* Z$ for a lognormally distributed water residence time with T^G=3.44 yr and σ^2=0.3 ($\bar{T}$=4yr; dashed line,) compared with the homogeneous solution 5 with T=4yr (solid line). The comparison shows that for realistic variance in the water residence time, flow heterogeneity does not have significant effect on the expected solute flux.

Transport Through a Waste Rock Dump With Preferential Flow Paths

In many cases, high-permeability channels or preferential flow paths may be the primary controls of water movement through the dump. Although confirmation through field observations is lacking (Morin et al. 1991),

double- or multipeaked solute plumes or breakthrough curves have been reported for field soils (Jury et al. 1986, Butters et al. 1989, Roth et al. 1991, Sassner et al. 1994). The concept of preferential flow paths implies that a large part of the waterflow takes place in a small part of the cross-sectional area of the dump. This concept does not exclude flow in the regions of less mobile water; it only implies water residence times that are significantly greater than in the preferential flow paths. The concept of preferential flow paths can therefore be appropriately described by a bi- or multimodal distribution of the water residence time (Destouni 1993, Sassner et al 1994, Destouni et al. 1994).

A bimodal distribution quantifies the existence of two populations of water residence times, which are related to each other by use of water flow continuity formulated as

$$\overline{q} = \upsilon\overline{q_1} + (1-\upsilon)\overline{q_2} \ . \tag{10}$$

In equation 10, $\overline{q}$ is the mean water flow per unit cross-sectional area of the dump, and $\overline{q_1}$ and $\overline{q_2}$ are the mean water fluxes in the relatively slow flow paths (SFP) and in the preferential, or fast flow paths (FFP), respectively. Furthermore, $\upsilon\overline{q_1}/\overline{q}$ quantifies the volumetric flow fraction through the SFP, and υ is the probability for the local water flux being dominated by SFP. For illustrative purposes, we will in the following assume that the water flux through 20% of the cross-sectional area of the dump is influenced by FFP, and that the water flux through the remaining 80% is dominated by SFP, i.e., υ= 0.8.

For bimodally distributed q, a relevant bimodal pdf of the water residence time T can be expressed as (Destouni et al. 1994)

$$f(\mathrm{T}) = \frac{\upsilon}{\mathrm{T}\sqrt{2\pi\sigma_1^2}} \exp\left[-\frac{1}{2}\frac{\left(\ln \mathrm{T} - \ln \mathrm{T}_1^{\mathrm{G}}\right)^2}{\sigma_1^2}\right] + \frac{1-\upsilon}{\mathrm{T}\sqrt{2\pi\sigma_2^2}} \exp\left[-\frac{1}{2}\frac{\left(\ln \mathrm{T} - \ln \mathrm{T}_2^{\mathrm{G}}\right)^2}{\sigma_2^2}\right], \tag{11}$$

which is a combination of two lognormal distributions. In equation 11, $\mathrm{T}_i^{\mathrm{G}}$ is the geometric mean of the water residence time and σ_i^2 is the variance of $\ln \mathrm{T}$, in which i =1 refers to the SFP and i =2 to the FFP. The geometric mean of the water residence time is related to the geometric mean of the water flux through $\mathrm{T}_i^{\mathrm{G}} = Z\theta/q_i^{\mathrm{G}}$, in which $q_i^G = \exp(\ln\overline{q_i} - \sigma_i^2/2)$.

Equation 11 quanitifies the residence time distribution of an ideal tracer (non-reactive, non-diffusive). This distribution is combined with the prevailing local (bio)chemical reactions and diffusive mechanisms through equation 8. In this paper, we have only considered irreversible chemical dissolution, which is quantified by the term $s(t,Z;\mathrm{T})$ in equation 8; this term is in turn quantified by equation 5 for case 1 dissolution, and by equation 7 for case 2 dissolution. Other types of local mass transfer mechanisms, such as equilibrium and/or non-equilibrium adsorption-desorption (Destouni and Cvetkovic 1991), degradation (van der Zee and Destouni 1992), or diffusive mass transfer between mobile and immobile water zones (Destouni 1993, Destouni et al. 1994) can also be incorporated in this modeling approach. For the case of preferential flow paths, incorporation of diffusive mass transfer between mobile and immobile water could be relevant. However, the studies of Destouni (1993) and Destouni et al. (1994) showed that the influence of this mechanism on field-scale solute transport is small; for simplicity, we shall therefore neglect diffusive mass transfer in the following.

In figure 3, the effect of the existence of preferential flow paths on the solution 8 is illustrated for case 1 dissolution and for two different bimodal pdfs (equation 11) by comparison with the homogeneous solution 5. For both bimodal pdfs the mean water flux $\overline{q}$=0.5m/yr; the parameters T_i^G, however, differ between the two pdfs. The comparison shows that the expected solute flux is sensitive to even small changes in the mean water residence time in the preferential flow paths. The effect of preferential flow paths is that the peak value of the solute flux decreases and a tail develops in the expected breakthrough curve. When $\overline{T_2}$=1.09yr (T_2^G=0.94yr), equation 10 implies that $\overline{T_1}$=1,000yr (T_1^G=600yr), which leads to a significant decrease in the peak value of the solute flux and a considerable tailing effect. The reason for these effects is that the characteristic hydrological time scale in the SFP is greater than the characteristic time scale of the weathering process. This indicates that preferential flow paths may affect the field scale solute transport of weathering products significantly. Moreover, the existence of preferential flow paths may also be the reason for the field observations of solute concentration being considerably lower than expected from geochemical modeling with reaction rates based on literature values (Strömberg and Banwart 1994).

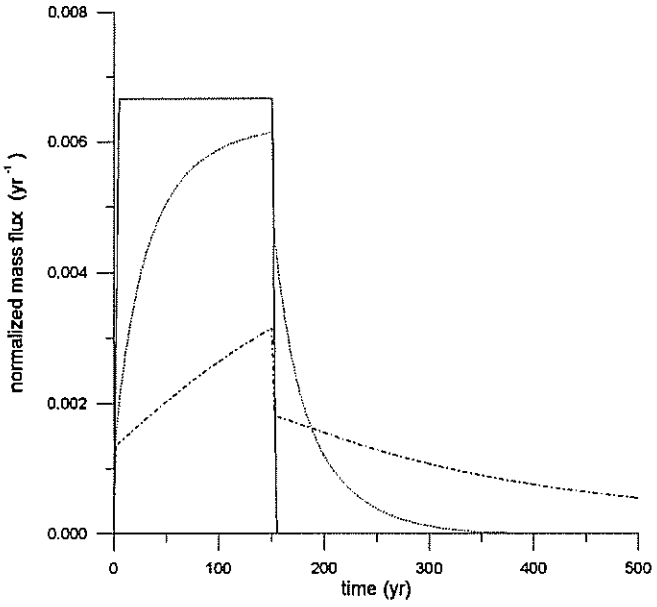

Figure 3. The homogeneous solution 5 with T=4yr (q=0.5m/yr) (solid line) and the normalized heterogeneous solutions 8 for υ= 0.8 and for two bimodal pdfs: T_1^G=60yr, T_2^G=1.02yr, σ_1^2=0.3, σ_2^2=1.0 (dashed line) and T_1^G=600yr, T_2^G =0.94yr, σ_1^2=0.3, σ_2^2=1.0 (dash-dotted line). The comparison shows that the existence of preferential flow paths may considerably affect both the maximum value and the temporal distribution of the solute flux.

Conclusions

The effect of flow heterogeneity on the field scale mass flux of dissolved weathering products from a waste rock dump has been investigated for a classical unimodal distribution of water residence time and for bimodal distributions that reflect the existence of preferential flow paths. Results indicate that for realistic parameter values, flow heterogeneity reflected by a unimodal distribution of water residence time does not affect the solute flux significantly. The reason for this insensitivity is the difference in characteristic time scales between the water residence time in the dump and the dissolution process. Flow heterogeneity in the form of preferential flow paths, however, is better described by bi- or multimodal distributions and may have a considerable impact on field-scale solute flux in terms of both the peak value and the temporal spreading of solute. Hence, flow heterogeneity should be taken into account when estimating field-scale solute transport in mining waste rock dumps by coupling spatially distributed hydrological modeling with local or average geochemical conditions.

For simplicity, we have in this paper only regarded the water residence time as a random variable. The stochastic-advective modeling approach that was presented here, however, can readily be extended to include randomness also in the rate coefficient, k, or any other parameter of interest. Furthermore, additional chemical reactions and mass transfer mechanisms, such as equilibrium and/or non-equilibrium adsorption-desorption (Destouni and Cvetkovic

1991), degradation (van der Zee and Destouni 1992), or diffusive mass transfer between mobile and immobile water zones (Destouni 1993, Destouni et al. 1994) or precipitation/dissolution of secondary minerals (Eriksson and Destouni 1994, paper in preparation) can also be incorporated in the stochastic-advective modeling approach. In addition, the transport process in the underlying aquifer, can be coupled with the solute flux from the waste rock dump(Destouni and Eriksson 1994).

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