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Evaluation of single- and dual-porosity models for reproducing the release of external and internal tracers from heterogeneous waste-rock piles

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ABSTRACT

Accurate predictions of solute release from waste-rock piles (WRPs) are paramount for decision making in mining-related environmental processes. Tracers provide information that can be used to estimate effective transport parameters and understand mechanisms controlling the hydraulic and geochemical behavior of WRPs. It is shown that internal tracers (i.e. initially present) together with external (i.e. applied) tracers provide complementary and quantitative information to identify transport mechanisms. The analysis focuses on two experimental WRPs, Piles 4 and Pile 5 at the Antamina Mine site (Peru), where both an internal chloride tracer and externally applied bromide tracer were monitored in discharge over three years. The results suggest that external tracers provide insight into transport associated with relatively fast flow regions that are activated during higher-rate recharge events. In contrast, internal tracers provide insight into mechanisms controlling solutes release from lower-permeability zones within the piles. Rate-limited diffusive processes, which can be mimicked by nonlocal mass-transfer models, affect both internal and external tracers. The sensitivity of the mass-transfer parameters to heterogeneity is higher for external tracers than for internal tracers, as indicated by the different mean residence times characterizing the flow paths associated with each tracer. The joint use of internal and external tracers provides a more comprehensive understanding of the transport mechanisms in WRPs. In particular, the tracer tests support the notion that a multi-porosity conceptualization of WRPs is more adequate for capturing key mechanisms than a dual-porosity conceptualization.

1. Introduction

Accurate predictions of solute release from waste-rock piles (WRPs) are paramount for decision making in mining-related environmental processes. Such predictions rely on robust conceptual and mathematical models of transport, which are informed by experimental observations in field and laboratory environments (e.g. Amos et al., 2015; Jodeiri Shokri et al., 2016; Lahmira et al., 2017; Lefebvre et al., 2001; Lorca et al., 2016; Pedretti et al., 2015, 2017; Smith and Beckie, 2003). Tracer tests are experimental tools widely used nowadays for the identification and quantification of dominant transport mechanisms in the subsurface (e.g. Cvetkovic et al., 2016; Liang et al., 2016; Molinari et al., 2015; Wilson et al., 2016; Xie et al., 2016; Zaramella et al., 2016). In WRPs, tracer tests can be used to characterize complex unsaturated flow and transport patterns, which are controlled by physical and chemical heterogeneities (e.g. Blackmore et al., 2014; Eriksson et al., 1997; Marcoline, 2008; Neuner et al., 2013; Nichol et al., 2005; Peterson, 2014; Silva et al., 2014). Although our work mainly focuses on conservative tracer tests, reactive compounds can also be used to identify geochemical processes within the waste rock at both laboratory and field scales (e.g. Prasad and Kumar, 2015; Ramírez-Pérez et al., 2013; Strömberg and Banwart, 1999).

External tracers are used in the majority of hydrogeological applications (e.g. Molinari et al., 2015; Pedretti and Bianchi, 2018) and can be applied either at one location (e.g. an injection well) or on a surface, such as in the case of WRPs through an artificial rain event. In the context of WRPs, Blackmore et al. (2014) analyzed the results of an external tracer test performed on waste rock in laboratory columns and experimental piles in the field. The results showed that the physical heterogeneity of these materials generates non-symmetric breakthrough curves (BTCs) characterized by persistent concentrations at times longer than the time of BTC peaks, a behavior defined in the literature as BTC tailing (e.g. Pedretti and Bianchi, 2018). Using a combination of mathematical techniques, Blackmore et al. (2014) found that a one-dimensional (1D) dual-porosity-based mobile-immobile (MIM) model was able to simulate observed BTCs, while a 1D

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advection-dispersion equation (ADE) solution could not reproduce BTC tailing. The MIM model is based on a 1D advection-dispersion equation (e.g., Domenico and Schwartz, 1990), which embeds a nonlocal operator to simulate the non-equilibrium storage of the tracer in an "immobile" domain and allows for fitting non-Fickian tailing on experimental BTCs (e.g. van Genuchten and Wierenga, 1976; Simunek et al., 2003).

Internal tracers are defined as those compounds that already exist in the subsurface and that are mobilized in response to external stresses, such as a natural or artificial recharge event or a change in geochemical conditions of the systems. Internal tracers offer an alternative to understand and parameterize the formation and persistence of drainage generated from in-situ geochemical processes. In WRPs, soluble by-products from chemicals used in blasting operations at mine sites are potential internal tracers present on the surface of waste-rock particles throughout the entire pile. These internal tracers are likely associated with the full spectrum of grain sizes, including the finest-grained matrix materials. Examples of these compounds commonly associated with mine sites include nitrogenous compounds (ammonia and nitrate) and perchloride (Aziz and Hatzinger, 2008; Bailey et al., 2013; Chlot et al., 2015; Jermakka et al., 2014; Karlsson and Kauppila, 2016; Mahmood et al., 2017; Revey, 1996). In other contexts, these types of tracers have been associated with blasting occurring during road construction (e.g. Degnan et al., 2016).

There is limited evidence or literature documenting the use of internal tracers to provide quantitative insights into WRP flow and transport processes. In Bailey et al. (2013), concentrations of total sodium and chloride were used as conservative tracers to identify the first flush of water through the waste rock. Bailey et al. (2013) concluded that blasting agents proved effective for quantifying, for instance, the small portion of WRPs that has been flushed compared with its total drainage system (< 10% in their case). At present, however, we know of no attempts to interpret internal tracers using a process-based model, such as a dual-porosity model.

This study analyzes external bromide and internal chloride tracer data from two well-characterized experimental WRPs at the Antamina Mine site in Peru. Bromide and chloride BTCs were observed in the piles over 1000 days, providing a sufficiently long data record to evaluate transport mechanisms occurring in the piles. The interpretation of the tests is carried out using a mathematical approach similar to the one adopted by Blackmore et al. (2014) and includes temporal-moments analysis and MIM-based modeling. We analyze and discuss the results, focusing on the information that each tracer is able to provide based on the resulting parameterization obtained from the fitting of the experimental curves.

Based on the results and analysis from the Antamina data set, the specific goals of this work are: (1) to present an approach to quantify transport processes occurring within WRPs using both external and internal tracers, and (2) to discuss the advantages and disadvantages of each tracer to assess specific mechanisms occurring within heterogeneous waste-rock piles. The ultimate purpose of this analysis is to evaluate whether the combined use of internal and external tracers can provide a more complete assessment of the mechanisms and processes controlling solute transport in WRPs compared to the assessment performed by the analysis of an individual tracer. In particular, the use of different tracers suggests a modeling approach able to more effectively describe transport in heterogeneous WRPs (e.g., through a multi-porosity conceptualization of the system).

2. Materials

2.1. Site description

The experimental WRPs are located at the Antamina Mine, about 270 km north of Lima (Peru) in the Peruvian Andes (Fig. 1a). The mine has a typical Andean climate with two distinct seasons; a wet season from October to April and a dry season for the remainder of the year. The area receives between 1200 mm and 1500 mm of precipitation per year, of which approximately 80% falls in the wet season as rain. Over the 1000 day study period (from June 2009 to May 2012), the Antamina site received a total of 3900 mm of precipitation. Median air temperatures (T) at Antamina are around $T = 5.5\,^{\circ}\mathrm{C}$, with limited seasonal and daily fluctuations (e.g. Lorca et al., 2016).

Waste rock produced at Antamina is subdivided into three classes named A, B and C, based on their expected reactivity and potential to generate poor-quality drainage (i.e., lower pH values and higher metal loads). Class A is expected to have the greatest potential for producing poor-quality drainage. This material represents rocks close to the highly mineralized zones of the mine with larger amount of sulfides, lower amount of carbonates and finer grain size than Class B and C. Class C has the lowest potential to generate poor-quality drainage, representing coarse carbonate-rich rocks with low sulfide content. Class B material is described as having an intermediate potential between Class A and C. Fig. 2 shows the grain size distributions (GSD) and soil-water characteristic curves (SWCC) of the two classes A and C, obtained from previous studies (e.g., Bay et al., 2009; Peterson, 2014; Speidel, 2011). Note that about 90% of the Class C material has a particle size > 10 mm, while Class A material has a much higher component of material finer than 1 mm (approximately 20%).

There are five experimental WRPs at Antamina, each with the same geometry of 36 m (l) × 36 m (w) × 10 m (h) (Fig. 1b) and waste-rock mass (M_{WR}) of about $M_{WR} = 19000 - 25000$ t. This study focuses primarily on Piles 4 and 5. Pile 4 contains > 80% Class C material and < 20% Class B material. Pile 5 contains about 50% of Class A and 50% Class C waste rock (Fig. 1d). All experimental WRPs were constructed using an end-dumping approach, resulting in a sequence of depositional units or "tipping phases" (TPs) with an angle of repose close to 37°. In a cross-sectional view, all planar contact surfaces between TPs dip parallel to the external slope (or batter).

At the base of each WRP is one large 36 m (l) × 36 m (w) lysimeter, D, to collect the exfiltrating drainage. Three 4 m × 4 m sublysimeters, A, B and C (Fig. 1c) are embedded within lysimeter D along the centerline of the WRP to collect drainage from specific sections of the WRPs, similar to the approach described by Nichol et al. (2005). Fig. 1d presents a vertical projection showing the classes of waste rock intersected by the drainage flow paths. Because flow in WRPs is generally sub-vertical (e.g. Amos et al., 2015), sublysimeter C is expected to mainly reflect the flow starting as recharge from the pile batter, which has shorter flow paths than the center of the pile. On the contrary, the batter is expected to have the smallest influence on sublysimeter A and B and be more representative of flow occurring in the core of the pile, with average flow-path lengths comparable to the pile's vertical height (~10 m). Outflow drainage from all lysimeters is directed to an instrumentation hut for continuous recording of flow rates and volume via calibrated tipping buckets and for bi-weekly geochemical sampling. This includes measurements of concentration of total bromide and chloride, which are used as primary tracers in this study. For both compounds, the detection limit was established at 0.01 mg/l. Additional details about the construction of all five WRPs, calibration of the instrumentation and other in-situ equipment can be found elsewhere (Bay et al., 2009; Corazao-Gallegos, 2007; Lorca et al., 2016; Peterson, 2014).

2.2. Tracer tests

External tracer tests were performed on Piles 4 and 5 using a similar approach. Bromide (Br^-), applied as a well-mixed solution of lithium bromide, was used as a conservative tracer because of its very low reactivity, relative absence in the mineral composition of the waste rock, and low natural background concentrations of 0.11 mg/l and 0.18 mg/l for Piles 4 and 5, respectively. Rainwater contains negligible bromide in this region, with concentrations near to or below 0.01 mg/l (Flury and

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