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Contaminant tailing in highly heterogeneous porous formations: Sensitivity on model selection and material properties

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SUMMARY

Coupled impacts of slow advection, diffusion and sorption were investigated using two heterogeneity models that differ in structure and in the mathematical framework that was used to simulate flow and transport and to quantify contaminant tailing. Both models were built using data from a highly heterogeneous exposure of the Borden Aquifer at a site located 2 km north-west of the Stanford–Waterloo experimental site at Canadian Forces Base Borden, Ontario, Canada. The inclusions-based model used a simplified representation of the different materials found at the site, while the second model was based on transitional probability geostatistics of the formation. These two models were used to investigate sensitivity of contaminant tailing on model selection and on geometric and material properties. While simulations were based on data collected at Borden, models were exercised beyond the geometric and material properties that characterize the site. Various realizations have identified very low conductive silty clay, found at volume fraction of 23.4%, as the material with dominant influence on tailing, and vertical diffusion in and out of low conductive units, affected by sorption, as the dominant transport mechanism causing tailing. The two models yielded almost identical transport results when vertical correlation lengths of silty clay were matched. Several practical implications relevant for characterization of low conductive units were identified and briefly discussed.

1. Introduction

Groundwater contamination is a serious worldwide problem – billions of dollars are spent annually on remediating contaminated sites (National Research Council, 1994, 2004, 2012). Frequently, cleanup technologies are successful in removing the bulk, but not the entire quantity of contaminants (National Research Council, 1994, 2004, 2012). Residual contamination often persists for long time periods, generating contaminant concentration above cleanup targets.

The mechanisms responsible for this phenomenon, termed "contaminant tailing", are often site specific. This paper explores the relationship between geologic heterogeneity and contaminant tailing for a well-characterized aquifer. In this work "tailing" is defined in terms of asymmetry in the travel time distribution of particles released into an aquifer under uniform source and natural gradient conditions. When particle travel time is represented graphically as a BTC (breakthrough curve or cumulative distribution function), tailing is observed if a large proportion of particles exhibit substantially longer travel times relative to the median. The shape of the BTC tail captures important aspects of contaminant transport, particularly for remediation applications where it is desired to remove a very large fraction of emplaced contaminants. For example, BTC tailing is a way of characterizing "back diffusion", which occurs when contaminants become trapped in low-permeability materials and require extended operation of remediation systems as they diffuse back out into higher-permeability material.

Contaminant tailing in porous formations is partially caused by slow advection and diffusion through low-conductive units (such as silt and clay lenses/layers) as demonstrated by various field experiments (e.g. Rasa et al., 2011; Liu and Ball, 2002; Chapman and Parker, 2005; Parker et al., 2008), laboratory tests (e.g. Levy and Berkowitz, 2003; Silliman and Simpson, 1987), and numerical simulations (e.g. Fiori et al., 2006; Jankovic et al., 2009;

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Seyedabbasi et al., 2012). Sorption processes also produce delays in contaminant transport (e.g. Roberts et al., 1986; Burr et al., 1994; Dentz and Castro, 2009; Maghrebi et al., 2014).

This manuscript examines the coupled impacts of slow advection, diffusion, and sorption on contaminant tailing. These impacts were investigated using numerical simulations that were based on two models of subsurface heterogeneity which differ in structure and in mathematical framework that was used to simulate flow and transport and to quantify contaminant tailing. The models are based on alternative geostatistical descriptions of the hydraulic conductivity K and perchloroethene (PCE) sorption distribution coefficient K_d of highly heterogeneous sediment in an exposure of the Borden Aquifer located approximately 2 km north-west of the Stanford–Waterloo experimental site at Canadian Forces Base Borden, Ontario, Canada (Weissmann et al., 2015). The main goal of the present study is to quantify the impact of model selection on tailing and to identify the impacts of different geologic materials and their geometric and material properties on tailing. Additionally, we aim to increase the general understanding of tailing in porous formations. While the models are initially built using realistic geostatistical descriptions of the formation, we are not aiming to build the best predictive model for the formation found at our site.

Heterogeneity models used for transport simulations are generally based on simplified statistical descriptions of heterogeneous formations (e.g., Gelhar, 1993; de Marsily et al., 2005). Most models, such as commonly used multi-Gaussian model, are functions of the mean, variance, and overall K correlation lengths, and do not include information derived from higher order statistical measures, such as connectivity (related to correlation lengths) of different materials. However, this simplification introduces uncertainty in the modeling process, as numerical and analytical studies have shown that the transport behavior of aquifers with the same mean, variance and overall K correlation lengths, but different higher order measures, can be different (Zinn and Harvey, 2003; Soltanian et al., 2014).

Similar to K heterogeneity, heterogeneity in sorption properties is also known to affect the transport of hydrophobic contaminants, although there is, in general, limited availability of field data to support suitable models. In this manuscript, sorption is assumed to be a linear equilibrium process characterized by a heterogeneous distribution coefficient $K_d(\boldsymbol{x})$ with an associated retardation factor of $R(\boldsymbol{x}) = 1 + \rho_b \cdot K_d(\mathbf{x})/n$, where ρ_b is the bulk density and n is the porosity. In addition to this assumption, one of two simplifications is typically used to create models of heterogeneous $K_d(\boldsymbol{x})$ fields: (1) $K_d(\boldsymbol{x})$ is modeled as a random space function independent of $K(\boldsymbol{x})$ (e.g. Burr et al., 1994; Dentz and Castro, 2009), or (2) $K_d(\boldsymbol{x})$ is obtained using a simple linear relationship between $\ln K_d(\boldsymbol{x})$ and $\ln K(\boldsymbol{x})$ (e.g. Cvetkovic and Shapiro, 1990; Bellin et al., 1993; Tompson, 1993; Burr et al., 1994; Tompson et al., 1998; Deng et al., 2013). Neither simplification is supported by the few available field data sets (Allen-King et al., 1998; Allen-King et al., 2006; Robin et al., 1991; Foster-Reid, 1994). As a result, the necessary simplification used to represent the spatial distribution of $K_d(\boldsymbol{x})$ introduces additional uncertainty in interpretation of results of the modeling process.

Recent advances in site characterization techniques have enabled very high resolution descriptions of aquifer heterogeneity that capture mm-scale variability in aquifer materials (e.g., Weissmann et al., 2015). However, incorporating such details into numerical models of flow and contaminant transport can be computationally prohibitive if the goal is to capture tailing and back diffusion processes. As an alternative, this work explores the application of indicator geostatistics coupled with an "inclusions-based" modeling approach to represent heterogeneity in both $K(\boldsymbol{x})$ and $K_d(\boldsymbol{x})$ fields. The inclusion-based approach does not require the raster-type depiction of heterogeneity needed for grid-based models. It is capable of representing the correlation structure and connectivity of heterogeneous properties through the assemblage of inclusions that represent the various aquifer materials. Such a statistical representation of the aquifer may not be suitable for generating accurate predictions of local contaminant concentrations, but can capture the macroscopic behavior in a manner that illuminates the relationship between statistical heterogeneity and important aspects of contaminant transport such as tailing.

The inclusions-based model (referred to as the inclusions model hereafter) was based on the indicator geostatistical analysis of a detailed dataset collected at our site. This 3D dataset was created using high-resolution photography and ground-based LIDAR techniques that were applied to several vertical outcrop panels exposed at the site (Weissmann et al., 2015). The $K(\boldsymbol{x})$ and $K_d(\boldsymbol{x})$ models used here were built using the geostatistical description of individual materials found in the formation (with unique K and K_d values (Allen-King et al., 2015)), instead of the overall $K(\boldsymbol{x})$ measures (mean, variance, overall correlation length) and assumptions of $K(\boldsymbol{x})$–$K_d(\boldsymbol{x})$ correlations. The description includes materials' volume fractions and correlation lengths. The model represents each individual material with a set of identical ellipsoidal inclusions that are randomly populated in the space. The size, shape, and the number of inclusions were selected based on the directional correlation lengths and the volume fraction of each material. The values for K and K_d are based on published values for the material for the Borden Aquifer (Allen-King et al., 2015).

Most solute transport simulations reported here were based on the inclusions model. This model is used to investigate transport and tailing at the spatial scale characterized by Weissmann et al. (2015) (40 m horizontal and 10 m vertical) and larger scales, and to investigate tailing sensitivity on material and geometric properties of different geologic materials. In addition to this inclusions model, another model was created based on the transitional probability geostatistics (Carle and Fogg, 1996, 1997) using TPROGS code (Carle, 1999) for comparison with the inclusions model. This model also assigns single K and K_d values for each material. This model, termed transitional probability (TP)-based model here, is limited (due to numerical constraints) to the spatial scale characterized by Weissmann et al. (2015). Sensitivity of the TP-based model on material properties is also included.

Because of generally narrow distributions of $K(\boldsymbol{x})$ and $K_d(\boldsymbol{x})$ within each material, each material is represented by a single K and a single K_d value in each realization of the models.

The rest of this manuscript is organized as follows. A brief discussion of the indicator and the overall geostatistics of the Borden formation are presented in Section 2 followed by the description of inclusions and TP-based heterogeneity models. The transport results for the heterogeneity models are compared in Section 3. This is followed by several sensitivity analyses of geometric and material properties for both models in Section 4. Finally, the concluding remarks are presented at the end.

2. Heterogeneity models

2.1. Geostatistics of field data

The contaminant transport models were based on data obtained from a sand quarry located approximately 2 km north-west of the Stanford–Waterloo (SW) experimental site at the Canadian Force Base Borden, Ontario, Canada. Several vertical outcrop panels up to 20 m long and 1.5 m in height (Fig. 1) covered an area about 40×10 m (Weissmann et al., 2015). The top of exposure was located in material that was stratigraphically equivalent to