Science of the Total Environment 512–513 (2015) 428–443

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

An integrated Markov chain Monte Carlo algorithm for upscaling hydrological and geochemical parameters from column to field scale

Bhavna Arora [a,b,*], Binayak P. Mohanty [a], Jennifer T. McGuire [c]

[a] Water Management & Hydrologic Science Program, Texas A&M University, USA
[b] Earth Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, CA, USA
[c] Department of Biology, University of St. Thomas, St. Paul, MN, USA

HIGHLIGHTS

- This study evaluates the effect of lithologic heterogeneity on upscaled geochemical concentrations
- This study highlights the use of Bayesian methods in obtaining improved parameter estimates
- We recommend the use of local scale geochemical data and hydrologic parameters in upscaling solute concentrations

ARTICLE INFO

Article history:
Received 9 September 2014
Received in revised form 13 January 2015
Accepted 18 January 2015
Available online 30 January 2015

Editor: Simon Pollard

Index terms:
Landfill
Environmental chemistry
Modeling

Keywords:
Scale
Landfill site
Sulfate reduction
Spatial variability

ABSTRACT

Predicting and controlling the concentrations of redox-sensitive elements are primary concerns for environmental remediation of contaminated sites. These predictions are complicated by dynamic flow processes as hydrologic variability is a governing control on conservative and reactive chemical concentrations. Subsurface heterogeneity in the form of layers and lenses further complicates the flow dynamics of the system impacting chemical concentrations including redox-sensitive elements. In response to these complexities, this study investigates the role of heterogeneity and hydrologic processes in an effective parameter upscaling scheme from the column to the landfill scale. We used a Markov chain Monte Carlo (MCMC) algorithm to derive upscaling coefficients for hydrological and geochemical parameters, which were tested for variations across heterogeneous systems (layers and lenses) and interaction of flow processes based on the output uncertainty of dominant biogeochemical concentrations at the Norman Landfill site, a closed municipal landfill with prevalent organic and trace metal contamination. The results from MCMC analysis indicated that geochemical upscaling coefficients based on effective concentration ratios incorporating local heterogeneity across layered and lensed systems produced better estimates of redox-sensitive biogeochemistry at the field scale. MCMC analysis also suggested that inclusion of hydrological parameters in the upscaling scheme reduced the output uncertainty of effective mean geochemical concentrations by orders of magnitude at the Norman Landfill site. This was further confirmed by posterior density plots of the scaling coefficients that revealed unimodal characteristics when only geochemical processes were involved, but produced multimodal distributions when hydrological parameters were included. The multimodality again suggests the effect of heterogeneity and lithologic variability on the distribution of redox-sensitive elements at the Norman Landfill site.

1. Introduction

Knowledge about effective hydrologic and geochemical properties at field scales is necessary in predicting and managing the fate and transport of reactive contaminants from landfill and waste management sites. However, the transition of biogeochemical processes across scales is not well understood. Therefore, the challenge is to acquire detailed knowledge of key processes at individual scales and identify the dominant linkages to predict geochemical dynamics from one scale to the other.

Reactive transport is strongly influenced by hydrological processes across different spatial scales (Kimball et al., 1994; Vogel and Roth, 2003; Jardine, 2008). Temporal hydrologic variations such as seasonality and direction of groundwater flow, water table dynamics, and precipitation events also strongly influence reactive transport processes (Prommer et al., 1998; McGuire et al., 2000; Cozzarelli et al., 2011; Arora et al., 2013). For example, Fendorf et al. (2010) suggested that the patterns of groundwater recharge and discharge, especially

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* Corresponding author at: Earth Sciences Division, Lawrence Berkeley National Laboratory, 1 Cyclotron Rd., MS 74-327R, Berkeley, CA 94720, USA.
E-mail address: barora@lbl.gov (B. Arora).

http://dx.doi.org/10.1016/j.scitotenv.2015.01.048

groundwater pumping and time since recharge, were important factors influencing arsenic concentrations in South and Southeast Asia. Furthermore, hydrological parameters or process interactions that are applicable at one scale may not necessarily be applicable to other scales (van Grinsven and van Riemsdijk, 1992; White and Brantley, 2003). For example, variations in hydraulic conductivity, which are known to affect contaminant transport, have to be evaluated based on the scale of study (Hunt, 2003; Schulze-Makuch and Cherkauer, 2004). Hydrologic processes themselves exhibit scale variability (Bloschl and Sivapalan, 1995) and are affected by a number of physical attributes such as topography, vegetation, and other characteristics of the porous media (Sharma et al., 2006; Das et al., 2008; Jana and Mohanty, 2012a). Therefore, it is crucial to isolate and understand the contribution of hydrological processes to geochemical concentrations across scales.

Apart from hydrologic variations, understanding the natural variability of geochemical processes is difficult from the standpoint of heterogeneity in the subsurface. Structural heterogeneity resulting from the presence of macropores and fractures leads to preferential flow movement and faster pathways for contaminants to reach groundwater (Mohanty et al., 1998; Jarvis et al., 2007; Arora et al., 2011). Heterogeneity in the form of textural interfaces and lithological variations is known to intensify biogeochemical activity and affect the distribution of chemical concentrations. In their study, Hansen et al. (2011) clearly demonstrated that heightened redox activity was observed at small scale interfaces of a layered soil column as compared to two texturally homogeneous soil columns. Similarly, Schilling and Jacobson (2012) indicated that variations in nutrient concentrations were closely related to lithologic variations within the Cedar River floodplain in Iowa. They demonstrated that water beneath sand-dominated ridges was aerobic, had higher concentrations of NO_3-N, and lower concentrations of dissolved organic carbon (DOC) as compared to the anaerobic groundwater beneath shales that had lower NO_3-N and higher DOC. While the influence of physical, chemical, and biological heterogeneities on reactive transport processes is recognized (Dagan, 1984; Cushman and Ginn, 1993; Werth et al., 2006; Liu et al., 2014), an upscaling approach that incorporates the influence of subsurface heterogeneity from fine (e.g., column) to coarse (e.g., field) scales is lacking.

Upscaling is the process of replacing such heterogeneous systems with effective mean properties that capture the key field scale behavior, such as by matching hydrologic fluxes and geochemistry data from the field site (Rubin, 2003; Zhu and Mohanty, 2002, 2003, 2004; Vereecken et al., 2007). Most upscaling schemes for soil hydrologic and reactive transport parameters homogenize the effect of heterogeneity in their derivation of effective parameter values (Zhu and Mohanty, 2006; Mohanty and Zhu, 2007; Vereecken et al., 2007; Dentz et al., 2011). These include approaches such as volume averaging, stochastic averaging, and homogenization, among others (e.g., Gelhar and Axness, 1983; Dagan, 1984; Whitaker, 1999; Lunati et al., 2002). However, real-world applications of solute scaling schemes require that the effect of small-scale heterogeneity on redox activity and geochemical parameters be incorporated into these schemes. For example, Onsoy et al. (2005) concluded that the mismatch between effective mean concentrations and nitrate observations at the field scale was a result of the heterogeneous flux conditions that were not accounted for by the mass-balance approach used in their study. In the same way, Khaleel et al. (2002) indicated that dispersivity values at the field scale were dependent on geologic formations, and averaged concentration profiles for flow parallel to bedding were highly skewed and affected by geologic layering.

Certain upscaling techniques such as the multi-continuum approach and moment equations have also been developed to describe heterogeneity in porous media (Haggerty and Gorelick, 1995; Oates, 2007; Neuman and Tartakovsky, 2008; Deng et al., 2010). However, most of these process-based upscaling approaches suffer from an increasingly greater number of mechanistic details, while parameter-based upscaling approaches target only a single or a couple of flow and transport parameters like hydraulic conductivity, reactive surface area, reaction rate parameters, retardation factor, or macrodispersion coefficients (Dai et al., 2009; Dentz et al., 2011; Soltanian et al., 2015). It is also widely known that the scale dependence of these parameters is usually a result of concentration gradients across physical, chemical, or biological heterogeneities (Valocchi, 1985; Steefel et al., 2005; Li et al., 2006; Scheibe et al., 2006; Ritzi et al., 2013). In contrast, the novelty of this study is that it deals with subsurface heterogeneities by directly employing local measurements of solute concentrations in the upscaling algorithm.

In this study, Bayesian methods are used to develop an upscaling algorithm that identifies scale-appropriate hydrological and geochemical parameters to represent the transition of chemical concentrations across lithological heterogeneities. Because flow and transport in porous media pose a nonlinear inverse problem and can potentially lead to non-unique solutions for the unknown parameters (Ginn and Cushman, 1990; McLaughlin and Townley, 1996), Bayesian methods have the advantage of treating these hydrologic and geochemical parameters in a probabilistic manner. Bayesian methods, and particularly Markov chain Monte Carlo (MCMC) techniques, can thus explore parameter space efficiently and reduce uncertainty associated with parameter values (Vrugt and Dane, 2005; Vrugt et al., 2008; Smith and Marshall, 2008). Given that Bayesian methods have the ability to combine prior information with direct observations, these methods have been increasingly used to upscale soil hydrologic properties and parameters (Efendiev et al., 2005; Das et al., 2008; Sams and Saussus, 2011; Jana and Mohanty, 2012b). However, upscaling reactive transport parameters or properties using Bayesian methods has been limited at best (Chen et al., 2009, 2012; Deng et al., 2010). As suggested above, most of these studies target a single or a few parameters (e.g., sorption coefficients) pertaining to a dominant reactive transport process (e.g., mineral dissolution or precipitation reactions), or else suffer from model uncertainty issues stemming from linking geochemical concentrations to indirect observations (e.g., petrophysical relationships, pseudo models). To our knowledge, this is the first study that presents an integrated upscaling framework that accounts for both hydrological and geochemical parameters and uses direct fine scale geochemical datasets to predict effective upscaled concentrations across heterogeneous formations using Bayesian methods.

The objectives of this study are to isolate and quantify the influence of (i) lithologic heterogeneity (lenses, layers) and (ii) hydrological parameters on effective upscaled geochemical concentrations at the coarse scale. The remainder of this paper is organized as follows: Section 2 introduces the integrated upscaling framework featured in this research and presents a brief description and overview of Bayesian methods; Section 3 presents the heterogeneous system considered in this work; Section 4 presents results on two cases: one for validation and another for application of the upscaling algorithm; Section 5 describes the limitations of this work, and Section 6 offers relevant conclusions obtained from this work and its applicability beyond the current study.

2. Approach

In this section, the development of an integrated upscaling algorithm using Bayesian methods is described. Fig. 1 illustrates the framework for developing such an algorithm that examines the scale dependency of reactive transport processes as a result of (i) subsurface heterogeneity and (ii) hydrological parameters.

For verifying the effect of heterogeneity on upscaling coefficients, two different mathematical structures, i.e. with and without heterogeneous formulations, are proposed. For verifying the effect of hydrologic processes, two different sets of input parameters, i.e. with and without upscaling soil water retention parameters, are considered. As Fig. 1 illustrates, the upscaling algorithm requires the selection of the mathematical structure of the model (with or without considering heterogeneous formulations). Next, prior probabilities of parameters are established based on the choice of the parameter set (with or without upscaling