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Uncertainty quantification of adverse human health effects from continuously released contaminant sources in groundwater systems

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ABSTRACT

We propose a computationally efficient probabilistic modeling methodology to estimate the adverse effects on humans of exposure to contaminated groundwater. Our work is aligned with the standard suggested by the regulatory agencies and allows to propagate uncertainty from hydrogeological, toxicological and behavioral parameters to the final health risk endpoint. The problem under consideration consists of a contaminated aquifer supplying water to a population. Contamination stems from a continuous source that feeds a steady plume which constitutes the hazard *source*. This scenario is particularly suited for NAPL pollutants. The erratic displacement of the contaminant plume in groundwater, due to the spatial variability of hydraulic conductivity, is characterized within the Lagrangian stochastic framework which enables the complete probabilistic characterization of the contaminant concentration at an environmentally sensitive location. Following the probabilistic characterization of flow and transport, we quantify the adverse health effects on humans. The dose response assessment involves the estimation of the uncertain effects of the exposure to a given contaminant while accounting for the exposed individual's metabolism. The model integrates groundwater transport, exposure and human metabolism in a comprehensive probabilistic framework which allows the assessment of the risk probability through a novel simple analytical solution. Aside from its computational efficiency, the analytical features of the framework allows the assessment of uncertainty arising from the hydrogeological parameters.

1. Introduction

This paper examines the impact of continuous contaminant source release in the subsurface environment on adverse human health effects. Contaminant plumes originating from continuous (or long term) release, e.g., NAPL sources, is a world wide problem (Fetter, 1993). Depending on the reactions involved, such plumes can reach steady-state conditions with a finite extent (Liedl et al., 2005; Maier and Grathwohl, 2006) and might pose risk to human health since many of the compounds within the plume are, for example, carcinogenic (Henri et al., 2015, 2016). From a remediation perspective, it is important to properly delineate the plume with the goal of providing cost-effective clean up strategies. For such reasons, there is an increasing need to develop simulation tools that predict the large scale transport behavior in realistic aquifer settings in order to improve our understanding of the risks involved and provide better environmental remediation strategies.

Predicting solutes in aquifer ecosystems is a complex task because of several factors. First, hydrogeological properties vary in space and over a multitude of scales. Second, due to limited financial resources, site characterization data are scarce. The lack of a detailed data-set leads to uncertainty in model input parameters. As a consequence, flow and transport model predictions are not deterministic and must be treated within a stochastic framework (Rubin, 2003). Stochastic models assume the hydraulic conductivity as a space random field, whose characterization should be considered in a statistical sense and based on geostatistical analyses of the measurements. Given the randomness of the hydraulic conductivity, all the other variables, like the flow velocity and the solute concentration, are random. Incorporating the effects of the spatial fluctuations of hydrogeological properties is of fundamental importance in human health risk analysis (Maxwell and Kastenberg, 1999) and for managing resources towards uncertainty reduction (de Barros and Rubin, 2008).

The impact of heterogeneity in the hydraulic conductivity on solute transport has been topic of intense research over the past decades (see Dentz et al., 2011, for an extensive review).

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Semi-analytical solution for the first two statistical moments of the resident concentration and mass fluxes are provided in the literature using Lagrangian (Selroos and Cvetkovic, 1994; Fiori and Dagan, 2000; Vanderborght, 2001) and Eulerian (Kapoor and Kitanidis, 1998; Andricevic, 1998) methods. The influence of the sampling volume on the ensemble moments of concentration has also been reported (e.g., Bellin et al., 1994; Tonina and Bellin, 2008; Schwede et al., 2008). Most of the aforementioned works focused on solute plumes originating from instantaneous release conditions. In the context of steady-state plumes in two-dimensional flows, de Barros and Nowak (2010) and Cirpka et al. (2011a) showed how focusing of streamlines, induced by fast flow conduits, led to an enhancement of dilution. Other works (e.g., Cirpka and Valocchi, 2007; Cirpka et al., 2012; Zarlenga and Fiori, 2013b) showed how the degradation of a steady-state contaminant plume is controlled by the interplay between transverse dispersion mechanisms and large scale advection.

In some cases, such as in risk analysis, it is not sufficient to characterize transport in terms of its lower order moments of the concentration at the environmental sensitive location. A statistical characterization is particularly important when estimating probabilities of extreme events (e.g., de Barros and Fiori, 2014). Several methodologies have been proposed to compute the probability density function (PDF) of the solute concentration in natural porous media. In addition to the straight forward (yet computationally demanding) brute-force numerical Monte Carlo (e.g., Schwede et al., 2008; Moslehi et al., 2015), modeling approaches based on PDF equations (e.g., Shvidler and Karasaki, 2003; Tartakovsky et al., 2009; Sanchez-Vila et al., 2008; Dentz and Tartakovsky, 2010; Boso et al., 2014), direct mapping of random variables (e.g., Cirpka et al., 2011b; de Barros and Fiori, 2014), assumed PDF models (e.g., Fiori, 2001; Bellin and Tonina, 2007; Cirpka et al., 2008; Boso et al., 2013) and probabilistic collocation methods (Zhang et al., 2010; Müller et al., 2011) are available in the literature.

The objective of this work is to develop novel semi-analytical solutions to estimate adverse human health effects due to exposure to contaminated groundwater. Our work makes use of well established stochastic methods to evaluate the uncertainty in human health risk. We present closed-form solutions for the increased lifetime cancer risk as a function of hydrogeological and public health parameters. The adoption of a stochastic contaminant transport model allows the quantification of the uncertainty arising from the hydrogeological aspects.

Motivated by real cases sites contaminated by long-term source loading (e.g., leaking landfills and chemical storage tanks or accidental spills) we derive the full PDF of the increased lifetime cancer risk for the specific case of a steady state plume originating from a continuous source zone release. In tenet with the health risk analytical methodologies of Andricevic and Cvetkovic (1996), de Barros and Rubin (2008) and de Barros and Fiori (2014), the proposed risk human health risk PDF incorporates the effects of heterogeneity and other key parameters stemming from hydrogeological and public health sciences. Furthermore, it enables to identify key parameters controlling the overall risk behavior. The analytical framework used allows decision makers to evaluate and screen different scenarios with low computational costs and to propagate the uncertainty from model input parameters. We provide a systematic theoretical analysis of the parameters controlling the uncertainty in human health risk. Finally, we illustrate the methodology using field data obtained from the contaminated Bemidji site (Minnesota, USA) (Essaid et al., 2011). Data collected during the last 30 years have been used in order to set a simplified scheme, which should be considered as an application example more than a rigorous health risk analysis for that zone.

2. Problem statement

Our methodology aims at assessing the adverse human health effects associated to exposure to contaminated groundwater. We focus on the health risks due to chronic exposure to a carcinogenic chemical emanating from a continuous releasing source and moving dissolved in the liquid phase. This section will describe the flow and transport configuration adopted and its link to the human health risk model.

In this work the environmental concentration of the hazardous substance is assumed to be a space random function, and shall be characterized through its probability density function. Our stochastic approach adopts a probabilistic description of the hydraulic conductivity K and considers K and the environmental concentration C_e as random variables. Following this approach, the risk also becomes a random variable denoted by R.

We start by considering a three dimensional (3D) fully saturated natural porous formation, with a Cartesian reference system $\mathbf{x} = (x_1, x_2, x_3)$. In this work, we assume flow to be spatially heterogeneous and at steady state with velocity field $\mathbf{v}(\mathbf{x})$. The mean velocity $\langle \mathbf{v}(\mathbf{x}) \rangle = (U, 0, 0)$ is aligned with the x_1 direction. In this work, $\langle \cdot \rangle$ represents ensemble expectation. The hydraulic log-conductivity $Y(\mathbf{x}) = \ln[K(\mathbf{x})]$ is a second order, axisymmetric stationary multi-Gaussian random variable. The spatial patterns of Y is characterized by its two-point autocorrelation function ρ_Y characterized by the mean $\langle Y \rangle$, variance σ_Y^2 and integral scales $I_{Y,i}$ (with $i = 1, 2, 3$) of Y. Due to the axisymmetry of Y, the integral scales are defined as follows: $I_{Y,1} = I_{Y,2} \equiv I_{Y,h}$ and $I_{Y,3} \equiv I_{Y,v} = f I_{Y,h}$, with f denoting the statistical anisotropy ratio, i.e. $f = I_{Y,v}/I_{Y,h}$, of the porous formation. Flow is assumed to take place far from boundary effects and governed by

$$\nabla \cdot [K(\mathbf{x}) \nabla H(\mathbf{x})] = 0 \quad \text{with} \quad \mathbf{v}(\mathbf{x}) = -\frac{K(\mathbf{x})}{n_e} \nabla H(\mathbf{x}), \tag{1}$$

with n_e denoting the effective porosity, which in the following shall be considered as constant.

The hazardous (reactive) compound is continuously released with constant concentration C_e^0 over an areal source zone $A_0 = L_2 \times L_3$ placed at longitudinal location $x_1 = 0$, normal to the mean flow. The solute plume undergoes the following physical-bio-chemical processes: advection, local scale dispersion and biodegradation. An illustrative sketch of the conceptual model along with notations is provided in Fig. 1.

Aerobic reactions, which involve the oxygen (O) as a reactor, take place along the fringe of the plume where fresh oxygenated water is mixed with the contaminant C. The chain reactions by which the contaminant C and the oxidant O produce the inert product P are sufficiently fast and irreversible (Hochstetler and Kitanidis, 2013), and shall be summarized by an effective instantaneous bimolecular reaction

$$f_C C + f_O O \longrightarrow f_P P \tag{2}$$

where f_i ($i = O,\ C$ and P) is the stoichiometric coefficient of the specie i.

Standing the instantaneous reaction assumption, the two reactants C and O cannot coexist; their concentration (C_e and C_O, respectively) are linked by the following equation

$$C_e C_O = 0 \tag{3}$$

Anaerobic reactions take place in the core of the plume, where, in anoxic conditions, micro-organisms metabolize the contaminant. Despite the slower dynamics, those reactions give a significant contribution to the contaminant degradation and have a strong impact on C_e (e.g., Essaid et al., 1995). Such reactions shall be represented by a first order kinetic