Stochastic analysis of the efficiency of coupled hydraulic-physical barriers to contain solute plumes in highly heterogeneous aquifers

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Abstract:

The expected long-term efficiency of vertical cutoff walls coupled to pump-and-treat technologies to contain solute plumes in highly heterogeneous aquifers was analyzed. A well-characterized case study in Italy, with a hydrogeological database of 471 results from hydraulic tests performed on the aquifer and the surrounding 2-km-long cement-bentonite (CB) walls, was used to build a conceptual model and assess a representative remediation site adopting coupled technologies. In the studied area, the aquifer hydraulic conductivity K_a [m/d] is log-normally distributed with mean $E(Y_a) = 0.32$, variance $\sigma^2_{Y_a} = 6.36$ ($Y_a = \ln K_a$) and spatial correlation well described by an exponential isotropic variogram with integral scale less than 1/12 the domain size. The hardened CB wall's hydraulic conductivity, K_w [m/d], displayed strong scaling effects and a lognormal distribution with mean $E(Y_w) = -3.43$ and $\sigma^2_{Y_w} = 0.53$ ($Y_w = \log_{10} K_w$). No spatial correlation of K_w was detected. Using this information, conservative transport was simulated across a CB wall in spatially correlated 1-D random Y_a fields within a numerical Monte Carlo framework. Multiple scenarios representing different K_w values were tested. A continuous solute source with known concentration and deterministic drains' discharge rates were assumed. The efficiency of the confining system was measured by the probability of exceedance of concentration over a threshold (C^*) at a control section 10 years after the initial solute release. It was found that the stronger the aquifer heterogeneity, the higher the expected efficiency of the confinement system and the lower the likelihood of aquifer pollution. This behavior can be explained because, for the analyzed aquifer conditions, a lower K_a generates more pronounced drawdown in the water table in the proximity of the drain and consequently a higher advective flux towards the confined area, which counteracts diffusive fluxes across the walls. Thus, a higher $\sigma^2_{Y_a}$ results in a larger amount of low K_a values in the proximity of the drain, and a higher probability of not exceeding C^*.

Keywords: cutoff walls, aquifer heterogeneity, solute transport, risk, uncertainty, pump-and-treat

1. Introduction

The combination of hydraulic and physical barriers has been adopted for decades for the remediation of solute polluted aquifers. A well-known approach is for instance the use of pump-and-treat technologies combined with vertical cutoff walls (e.g. Bayer and Finkel, 2006; Beretta, 2015; Pedretti et al., 2013a). The accurate hydraulic design of combined pump-and-treat systems (CP&Ts) requires a reliable parameterization of the hydrogeological properties of both the physical barriers and the surrounding aquifer. These issues have been documented in multiple analyses presented in the past (e.g. Bayer and Finkel, 2006; Beretta, 2015; Britton et al., 2005; Kaleris and Ziogas, 2013; Khandelwal et al., 1997; Pedretti et al., 2011; Russell and Rabideau, 2000; Wu et al., 2016; Xanthakos, 1979).

The hydraulic conductivity of the aquifer (K_a) and the hydraulic conductivity of the vertical cutoff walls (K_w) are two of the key parameters for the hydrogeological design of CP&Ts. These parameters control the net balance between inward and outward advective and diffusive solute fluxes across the walls. Outwards refers here to solute fluxes from the cutoff-wall- confined area towards the clean aquifer. Inwards indicates the opposite flux direction. These concepts are well explained for instance by Devlin and Parker (1996) and Neville and Andrews (2006). In short, when the sum of outward fluxes exceeds the sum of inward fluxes, some solute particles are able to escape the confined system, creating potential environmental, social and economic risks. For instance, a concern is created when the concentrations exceed a predefined threshold established by local regulations. On the other hand, if the total inward flux is equal or larger than the total outward fluxes, no solute particles escape the confined system and no risks exist.

Because of the limited accessibility to the subsurface and the high cost of explorations, the ubiquitous hydraulic heterogeneity of geological sites is never adequately characterized. This complicates the reliability of predictions regarding the expected behavior of the aquifers and the CP&T systems, when present. Lack of complete characterization of hydrogeological heterogeneity generates an epistemic type of randomness and uncertainty in the decision-making process, such as for risk assessment and aquifer remediation. In this context, stochastic-based modeling analysis provides a suitable approach tool to support decision makers when making predictions with a quantitative estimation of the expected model behavior and associated uncertainty (e.g. Tartakovsky, 2007). Unfortunately, stochastic modeling has not been yet routinely adopted by practitioners (e.g. Renard, 2007; Sanchez-Vila and Fernàndez-Garcia, 2016) and more efforts are required to researchers in order to illustrate the actual benefits of stochastic modeling against traditional deterministic approaches to quantify flow and transport under uncertain aquifer conditions.

The majority of documented stochastic analyses in hydrogeology have mainly targeted K_a as a primary source of randomness and uncertainty (e.g. Dai et al., 2004; Kawas and Karakas, 1996; Kitanidis, 1988; Sanchez-Vila et al., 1996). Yet, the presence of cutoff walls can add further variability to the behavior of the aquifers (e.g. Elder et al. 2002, Hemsi and Shackelford 2006). This occurs mainly since (1) the hydraulic characterization of vertical cutoff walls may be complex and expensive as much as for (or more than) the characterization of aquifers, and (2) the strong scaling effects of K_w measurements, especially when comparing laboratory-based and in-situ estimations (Britton et al., 2005; Daniel, 1984; Joshi et al., 2009; Manassero, 1994). In the case of cement-bentonite (CB) walls, the latter issue poses severe difficulties for the accurate design of the physical confinement. The synthetic mixture placed in the ground to form the CB walls after cement curing or hardening is designed in laboratory. The resulting hydraulic conductivity of this perfectly mixed hardened liquid is firstly estimated under laboratory-controlled conditions, which are generally not representative of field conditions. Indeed, laboratory-based conductivities are expected to be of the order of K_w=10^{-8} m/s (e.g. Turchan et al., 1989) or lower. When released in a soil trench or similar excavation, undetected hydrogeological heterogeneities in the surrounding aquifer or inhomogeneous mixing of chemical solutions can result in undesired cracks and fissures following the CB wall's hardening. In-situ K_w is therefore generally larger than laboratory-based K_w, by some orders or magnitude (e.g. Britton et al., 2005; Daniel, 1984; Joshi et al., 2009; Manassero, 1994). Making *a priori* estimations of the magnitude of this scale-dependent difference remains however challenging in the practice.

The implication of the uncertainty related to the hydraulic properties of physical walls (variability of K_a) in presence of heterogeneous aquifer conditions (variability of K_w) has not been studied exhaustively. Britton et al. (2005) used a lognormal distribution of K_w to study contaminant flux through a cutoff-wall with idealized initial and boundary conditions and surrounded by a homogeneous aquifer. They showed that an increase in variability of the wall's hydraulic conductivity determines an increase in solute flux across the walls, since a larger K_w increase the flux escaping the wall compared to the flux evaluated using only the average hydraulic conductivity of the walls. They concluded that estimates of contaminant flux may be incorrect if the variability of K_w is not taken into account.

These issues motivate this paper, in which we developed a stochastic model based on numerical Monte Carlo (MC) simulations of conservative transport to examine the implication of combined aquifer and CB wall's variability on the effectiveness of a CP&T configuration to contain a solute plume. Our goal is to provide answers to questions such as:

1. How much does the lack of complete knowledge of K_a and K_w affect the expected efficiency of the CP&T system?
2. How uncertain is this estimation, for different degree of heterogeneity of the aquifer and for different combination of CB walls?

To this end, we operated within a classic stochastic framework based on the solution of ensembles of MC results from the solution of transport in randomly varying correlated fields (realizations) of aquifer's hydraulic conductivity and according to different testing scenarios of CB wall conductivity. In each realization, we simulated flow and transport using a conceptual model and boundary conditions that resemble the CP&T and aquifer configuration observed from a former Italian SIN site (acronym for Site of National Interest, comparable to a US superfund site). We chose this site as a representative well-characterized working example to apply the stochastic model. In particular, we had access to a hydrogeological database with 414 results from hydraulic conductivity tests performed on the local CB walls at different scales and 64 results from pumping tests performed at different locations (boreholes) of the surrounding unconfined sedimentary aquifer. These unique results allowed for an exhaustive geostatistical assessment of the variability of K_a and K_w and provided the basis to formulate a realistic stochastic assessment. Our framework allows providing quantitative estimation of the expected efficiency of the CP&T system and the associated uncertainty around the mean. Such an efficiency is measured through the probability that solute concentrations exceed a predefined concentration threshold at a control section outside the polluted area over time. We are then able to provide an answer to our initial questions, (1) by directly relating the random variability of K_a and K_w to the expected likelihood of pollution of the clean aquifer surrounding the confined site, and (2) by evaluating the uncertainty affecting the mean behavior of the aquifer, quantitatively associated with the variability of hydrogeological properties of the site. To ensure that results are exportable to other field conditions, our analysis covers K_w values ranging from the entire spectrum of typical hydraulic conductivities reported in operations with CB walls. In particular, we studied walls with progressive increase of walls' hydraulic conductivities from $K_w = 10^{-9}$ m/s to values approaching the hydraulic conductivities of the aquifer ($K_w \rightarrow K_a$).

The paper is structured as follows. Section 2 provides an overview of the site, including some historical information regarding the source of pollution and the chronology of the plume confining activities. Section 3 introduces the hydrogeological database. We describe the type of hydraulic tests that were performed on the aquifer and the CB walls, the solution adopted to estimate K_a and K_w at the different location, and the results of the geostatistical analysis. Section

4 presents the methodology and results from the stochastic analysis, including the model implementation, the adopted governing partial differential equations and related mathematical solutions. Here, the key results from the analysis are presented and discussed. Limitations of the study and future developments are also addressed in Section 4. The manuscript ends with the main conclusions drawn from this work.

2. Case study overview

The analyzed system (Figure 1) is located underneath a decommissioned chemical facility nearby Cengio (Italy). Since 1882, a large portion of this land (approximately 122 ha) was used to dispose chemical wastes and refuse materials by dumping and burying them in the ground without hydrogeological control (Domenico et al., 1992). The prevalent soil contaminants were metals (As, Cr, Hg, Nu, Cu, Pb, Zn), hexachlorobenzene, aromatic halogenated compounds, phenols, aromatic amines, nitrobenzenes, dioxins, furan, PAH and PCB. This resulted in a multicomponent solute plume which has persistently damaged the local ecosystem and severely affected the watersheds downstream the facility. The site has received a great attention at the end of the 1900s due to the high concentration of heavy metals and aromatic hydrocarbons observed in the neighboring Bormida di Millesimo river (Baldi et al., 2007). Detected contaminants were Al, As, Fe, Mn, Pb and naphthalenesulfonic compounds. Water discharges over a century are believed to have affected an area of over 22000 ha in Liguria and Piemonte regions. In the late 1990s, multiple remediation activities were started, most of which were funded by the Italian Government. They included the installation of multiple CB walls, aiming to confine the pollution within the source and prevent the direct contact of the solute plume with the river (De Paoli and Marcellino, 1992; Maione, 1995) and the construction of landfills to store chemical wastes and part of the contaminated soils. Since then, several advanced remediation activities have been documented related to this site (e.g. D’Annibale et al., 2005; Domenico et al., 1992; Falasco et al., 2009). Nowadays, the contamination is controlled and the Bormida di Millesimo river water has been recently classified as of good quality (Durante and Grasso, 2010).

[FIGURE 1: HERE]

The former chemical site lies on an unconfined heterogeneous 1-to-20-m-thick aquifer composed of a mixture of natural soil (alluvium), chemical waste, building debris and other byproducts. Below the aquifer, the marl bedrock creates an impervious aquiclude, with thickness over 100 m in the local area. A series of Lugeon tests indicate the good quality of the bedrock all over the area, giving maximum hydraulic conductivity values of about 10^{-9} m/s. During the remediation activities, only the main point sources of contamination had been physically removed from the aquifer by dig and dump operations, but almost all the rest of area remains affected by the presence of contamination. Both anthropic and natural alluvial soils act as a diffuse source of multicomponent solute contamination.

The aquifer is recharged by direct rainfall infiltration, hill slope runoff and lateral underground influx. No significant groundwater flow occurs along the uphill contact between unconsolidated deposit and the marl formation, due to the extremely limited groundwater circulation in the marl. The distance from the farthest recharge location to the discharging drain is about 1000m, although the main sparsely distributed sources of contamination can be located at closer distances from the drains in the center of the river bend (in correspondence of the label “former chemical facilities” in Figure 1a). The aquifer used to discharge directly to the Bormida di Millesimo River, although today multiple vertical cutoff walls and drains create an impervious confinement to isolate the source of pollution within the river bend. The hydraulic pumping system is designed to pump out all the groundwater flux and treat it *ex-situ*, before being redirected to the river.

The results presented in this work refer to the first 2-km-long CB wall created in the early 2000s to isolate the contaminated soil (Figure 1a). The wall was nailed into the impervious marl bedrock and combined with an initial draining system, which collected the polluted water and created an inward head gradient to contrast outward diffusive solute fluxes, as conceptually shown in Figure 1b. A large characterization activity was performed to measure the variability of K_a and K_w at different points of the aquifer and locations (zones) along the wall at different CB hardening times. Part of the results was collected and available for this study. No raw data existed, however, such that we could not test the actual reliability and accuracy of the pre-interpreted results. We note that this issue does not directly affect our conclusions. Indeed, the case study and this database were analyzed with the primary purpose of providing representative and realistic preliminary information for our stochastic analysis and obtain general conclusions regarding the ability of CP&T sites to contain solutes under hydrogeological uncertainty.

3. Analysis of the hydrogeological database

3.1. Aquifer hydraulic conductivity

The database contained 64 measurements of K_a, which represent the aquifer hydraulic conductivity estimated from an equivalent number of boreholes sparsely located in the site after a series of conventional pumping tests. The tests were pre-interpreted and no raw data were available to ensure the actual correctness of the interpretation, which is assumed as valid. The location of the boreholes is schematically shown in Figure 2a. This figure also highlights the mean direction of groundwater flux in the aquifer, which is conditioned by the presence of the continuous drain in the proximity of the CB walls.

We used these results to perform a geostatistical analysis to estimate the spatial and global variability of K_a in the aquifer. Figure 2b shows the experimental variogram of $Y_a = \ln(K_a)$ at different lags, overlapped by the exponential model used to parameterize the spatial variability of Y_a. The exponential model for unit variance has form (e.g. Gooverts, 1997, p.90)

$$\gamma(r) = 1 - \exp\left(-\frac{3r}{d}\right) \qquad (1)$$

where r is the distance between two points [m] and d is the range [m]. The exponential variogram (1) has a characteristic integral scale I [m], defined as $I = d/3$. Figure 2c and Figure 2d illustrate the empirical histogram and cumulative density of Y_a, respectively.

We found that the aquifer is characterized by a univariate log-normally distributed hydraulic conductivity distribution with mean $E(Y_a) = 0.32$ and variance $\sigma^2_{Y_a} = 6.36$. After testing isotropic and anisotropic variograms, an isotropic exponential variogram with range d ≈150 m ($I \approx 50$m) was able to satisfactory describe the spatial variability of Y_a. These results have direct implications for the selection of stochastic modeling framework to evaluate the implication of aquifer and wall's variable hydraulic properties. The high variance of $\ln(K)$ suggests a very strong aquifer hydraulic heterogeneity. Using a conservative approach and referring to the geological trace on Figure 1a, let us consider a solute source located at A', which is approximately the farthest point of the river bend at A (with L ≈600m) and use I=50m. Under these circumstances, a plume forming from the solute sources would only travel L/I ≈12 integral scales before entering the river, which may be insufficient to ensure ergodic solute transport within the studied domain. Indeed, under such high aquifer variances and uniform flow conditions, solute particles should sample a much larger amount of heterogeneity (i.e. several I) to become ergodic (e.g. Jankovic et al. 2006). When considering source locations closer to the drains solute particles would travel for even fewer integral scales, magnifying the effects of non-ergodicity. The presence of drains renders it even more difficult to define transport ergodicity, for instance because flow is by definition not stationary under non-uniform conditions (e.g. Matheron, 1967).

[FIGURE 2: HERE]

3.2. CB walls hydraulic conductivity

For the characterization of the CB wall, 407 K_w measurements collected at four unevenly distributed zones of the CB wall (A, B, C, D in Figure 2a) were available from the historic database. Zone D was the most characterized, while zone A was the least characterized. Laboratory tests and three in-situ methods (CPTU, Geon-BAT piezometers and slug tests) were performed using standard approaches and analytical method for the interpretation of the hydraulic tests. The complete list of values by location or zone in the site, the type of measurement and known analytical methods for their interpretation are reported in the Supplementary Material.

Laboratory tests were performed at different hardening (i.e., curing) times from samples collected on site and assessed using small-scale permeameters. Due to their relative simplicity and low execution cost, laboratory tests represented the majority of the experimental database (about 200 samples). The laboratory testing methodology followed standard procedures (ASTM D5084-03).

In-situ tests were also performed at different curing times. Slug tests were performed on micropiezometers, and K_w was estimated from the interpretation of hydraulic tests using the method by Bouwer and Rice (1976). This type of tests is widely adopted for the analysis of in-situ K_w, (e.g., Britton et al., 2002; Choi and Daniel, 2006; Choi et al., 2014; Lim et al., 2014; Nguyen et al., 2010). CPTU tests were performed following the methodology by Manassero (1994). The piezocone probe is characterized by a water-pressure transducer activated by a grease fluid that fills a slot located just behind the cone shoulder. In the CPTU method, K_w is estimated from the consolidation coefficient of CPTU dissipations through simple analytical expressions. No direct information regarding the execution of the CPTU in the site (e.g. point resistance and sleeve friction) was available for this study. The CPTU is able to provide a profile of K_w along the vertical direction of piezocone penetration. However, for this study only averaged K_w over a vertical section of the walls were available for different points of the barrier. BAT piezometers (Torstensson, 1984) with pressure sensors were adopted at approximately 110 curing days. This technology is also based on an in-situ pore-pressure dissipation approach, as the CPTU. In this case, a porous cell is introduced into the ground and isolated through a rubber membrane. A vacuum sampler and a pressure transducer (connected to a data logger on the surface) are introduced into the porous cell, and measure the relative change in pressure with the aquifer. Similar to the CPTU, K_w is estimated from a set of precalibrated empirical relationships. To the best of our knowledge, the Geon-BAT method has not been presented elsewhere for the characterization of K_w.

Figure 3 illustrates the comparison of estimated K_w from laboratory and combined in-situ tests, which emphasizes the impact of support scale on the measurements, at different curing times (t_c). The dotted lines represent a best-fit regression curves obtained from a power-law model of form $K_w \propto t_c^{-m}$. A first visual inspection confirms the existence of strong scaling effects in the estimated hydraulic conductivity of the CB wall. K_w was found to be smaller for laboratory-scale measurements than for in-situ measurement, consistent with previous investigations on vertical cutoff walls (e.g. Britton et al., 2005; Joshi et al., 2009). Our analysis also suggests that the characteristic hardening factor (m) is larger for in-situ measurements than for laboratory samples. This may be related to the different behavior of CB mixture in the field compared to laboratory conditions, and could be a possible reason of the resulting scaling effects in the estimated conductivities (in addition to the other mechanisms previously described).

[FIGURE 3: HERE]

Figure 4a reports four boxplots summarizing the statistical distribution of in-situ K_w calculated for an equivalent curing time t_c=280 days. This equivalent value is obtained by extrapolating each in-situ measurement using the fitted power-law regression function, and useful to obtain a larger dataset for statistical inference of the clustered data. These clusters are obtained by subdividing the dataset into the four major zones (A,B,C,D) where the tests were performed, in an attempt to obtain an indication of spatial structure or correlation in the distribution of K_w. Let us now observe the boxplots, reminding that the width of each box is proportional to the number of samples (n_s) used for the calculation of the empirical distributions. It can be observed that, while n_s is smaller for zone A than for zone D, the median values are quite similar among all locations, and 1st-to-3rd interquartile distance is also comparable between the four zones and ranging in an interval comprised between 5×10^{-2} m/d and 5×10^{-3} m/d (i.e. about one order of variability of K_w). In zone D, the extremes of the distribution seem to display a larger variability than in zone A.

[FIGURE 4: HERE]

Figure 4b reproduces the statistical distribution of the combined K_w measurements from the four zones. We found that the K_w of the entire CB wall is nicely described by a univariate log-normal distribution with the mean $E(Y_w) = -3.43$, where $Y_w = \log_{10}(K_w)$. The resulting variance $\sigma^2_{Yw} = 0.53$ is however much lower than that of the aquifer ($\sigma^2_{Yw} = 6.5$). The relative homogeneity of the CB wall does not surprise, since all points of the cement-bentonite wall are made up of the same chemical mixture, and the presence of defects associated to localized cracks, fracture or in-situ inhomogeneity of the CB mixture can be considered as minor along the wall from a hydraulic perspective. This also implies that, although we lack of sufficient information for the spatial analysis of K_w, poor or no correlation of K_w is expected in space along the wall. Indeed, CB walls are artificial structures created without a specific spatial-dependent process, such as alluvial and glacial deposition or tectonic and post-tectonic deformation as in the case of sedimentary or fractured formations, respectively.

4. Stochastic analysis

The experimental geostatistics obtained from the analysis of the historic database and the aquifer configuration of the ACNA site provided a representative real-life example to develop the stochastic modeling analysis targeting the efficiency of the CP&T system in heterogeneous systems. The analysis is based on the solution of an ensemble of random synthetic realizations within a Monte Carlo (MC) framework. The likely lack of transport ergodicity conditions implies the use of MC-based numerical simulations under bounded flow domains (e.g. Pedretti et al., 2014) to adequately evaluate the stochastic problem analyzed in this work. These aspects may impede for instance the direct applications of more computationally efficient stochastic methods such as effective analytical solutions (e.g. Dagan, 1989) which are formally valid for specific conditions not applicable to our case study, such as unbounded domains and weak heterogeneity ($\sigma^2_{Y_a} \leq 1$).

4.1.Model framework and setup

We conceptualized the flow and transport model as graphically depicted in Figure 5. Because of the presence of continuous drain pumping in the analyzed aquifer, the mean direction on the groundwater flux was perpendicular to the CB wall at all points along the wall. In addition, no well-defined point source of pollution existed in the domain, but rather the entire confined aquifer played the role as a macroscopic sparsely distributed source, which threatened the pollution of the nearby Bormida di Millesimo river.

[FIGURE 5: HERE]

Based on this knowledge, we idealized a one-dimensional (1D) unconfined heterogeneous aquifer, which was discretized and solved using the finite-difference code MODFLOW-96 (Harbaugh and McDonald, 1996). Each cell had a top elevation of 15 m and a bottom elevation of 0 m. The total length of the domain was 1000 m. The domain was oriented along the mean groundwater flow direction and parallel to the x coordinate of a Cartesian reference grid. The system was discretized with a telescopic refinement towards the central region of the domain. In the middle of the domain (i.e. nearby the source of pollution, the drain and the CB wall) the cell size was 0.1 m and increased with a logarithmic-based increment towards the boundaries, with a maximum cell size of 1 m. The flow boundary conditions were imposed to create, under unpumped conditions, a natural gradient equal to 0.001. The model was recharged by prescribed-head boundary conditions at one side of the domain and discharges at the other side, as in Figure

5. To create pumped conditions, we imposed a drain condition in one cell of the domain, with a constant deterministic pumping rate Q_w. A set of cells with different hydraulic conductivity than the remaining cells of the domain formed the CB wall, with thickness 1m and adjacent to the drain.

We generated two sets of n_{MC}=1000 random fields of Y_a. Both sets were generated using a variogram-based Sequential Gaussian Simulation (SGS) algorithm coded within SGEMS (Remy et al., 2009). The first set was generated using an exponential variogram (Eq. 1) similar to the one obtained from the hydrogeological database, with range d=150m and sill (i.e. variance) $\sigma_{Y_a}^2 = 6.5$. The second set was developed to study an aquifer with a milder heterogeneity; we imposed the same variogram structure as for the first set of simulations, but with sill $\sigma_{Y_a}^2 = 1$. The Y_a values from the two sets of SGS simulations were transformed into K_a and used as hydraulic conductivity parameters in MODFLOW-96.

Because of 1-D nature of our models and the lack of experimental information regarding the vertical variability of K_w values, we could not adopt the approach of Britton et al. (2005). Instead, we simulated three scenarios, each of which embedding a different homogeneous CB wall hydraulic conductivity. Each homogeneous K_w value can be seen as an effective (or upscaled) CB wall's hydraulic conductivity which lumps together the variability of K_w due to local cracks and fissures that can be found along the vertical direction of the walls.

For each realization in each set of SGS simulations and for each K_w scenario, we obtained an ensemble of flow fields from MODFLOW-96 which were used as input for an analogous number of 1-D transport simulations based on the advection-dispersion-equation (ADE) model. We simulated transport of an idealized conservative species using the code MT99 (Zheng and Wang, 1999), which is efficiently coupled to MODFLOW-96. A similar approach was already successfully used for the analysis of transport across low-permeable barriers in homogeneous settings (e.g. Devlin and Parker, 1996; Hudak, 2004; Neville and Andrews, 2006). The ADE is defined upon the groundwater flux q [m/d] calculated using MODFLOW-96, an effective kinematic porosity ϕ [-] and a hydromechanical effective scale-invariant local dispersion term $D = D^* + \alpha_L|v|$, where $D^*$is the effective diffusion coefficient [m^2/d], α_L is the longitudinal dispersion [m] and $v = q/\phi$. The solution of the ADE, obtained using a TVD algorithm, is a concentration C [mg/L] of the solute species in space x [m] and time t [d]. We arbitrarily assumed homogeneous $D^* = 10^{-5}$ m^2/d, $\alpha_L = 0.1$ m and $\phi = 0.1$, such that the entire variability of the solution is uniquely associated to the variability of K_a and K_w (our primary targets).

Transport was calculated for each realization in both sets of SGS realizations and each of the three K_w scenarios. In all realizations, we assumed $C(x, t_0) = 0$, i.e. a pristine aquifer at initial time, except at one cell in which we imposed a constant concentration (C_0) throughout the entire simulation time. This cell simulates a continuous source of contaminant spilling from the upgradient side of drain (Figure 5) and occurring immediately at t_0.

4.2. Analysis and discussion

We calculated the efficiency of the CP&T from the probability that C exceeds an arbitrary predefined threshold C^* at t=10 years at a specific model cell (Figure 5), acting as an observation piezometer (passive borehole), at which we tracked the evolution of concentration as a resident breakthrough curve (BTC). For illustrative purposes, we selected an arbitrary realistic dimensionless maximum concentration threshold $C_{max} = C^*/C_0 = 10^{-4}$, or $\log_{10}(C_{max}) = -4$. This threshold represents for instance a maximum concentration threshold of 0.1 mg/L occurring for a spill with concentration of 100 mg/L. The targeted piezometer is located downgradient of the solute source in order to track the concentration at a piezometer located outside the confined area within the pristine part of the aquifer potentially threatened by the solute plume. We chose to monitor the first cell adjacent to the vertical walls in order to minimize the effects of concentrations dilution in other cells downgradient of the source.

We first recall the expected theoretical effect of the drains and the barrier on the system, and their consequences on solute transport. In principle, the drain should lower the water table within the unconfined aquifer, as conceptually shown in Figure 1b. If it does, the lowered water table creates an artificial hydraulic head gradient between the two sides of the barrier. This gradient should create an inward advective flux that contrasts the outwards diffusive flux. As such, the efficiency of the CP&T system depends directly on the amount of inward advective flux versus the outwards diffusive flux, which in turn depend on the hydraulic conductivities of aquifer (which in our work are randomly varied in each simulation) and CB walls (which are varied by scenarios).

We now evaluate how likely this theoretical behavior applies under hydrogeological randomness. All the results from this analysis are summarized in Figure 6. Each panel reports the statistical distribution of the ensemble of concentrations measured at the targeted piezometer at 10 years from the initial spill and obtained from each specific set of simulation and K_w scenario. The results are presented in three forms: the histogram of concentrations (blue bars), the cumulative density function of the concentrations (red curve) and the ensemble-averaged concentration (green dotted line). The concentrations are expressed as $\log_{10}(C/C_0)$, to be directly compared to $\log_{10} C_{max}$. The ensemble mean expresses the expected behavior of each set of simulations and scenario, and in turn the expected efficiency of the tested CP&T configuration. The top panels show the results from the less heterogeneous set of SGS simulations ($\sigma_{Y_a}^2 = 1$), while the bottom row shows the results for the high heterogeneous set of simulations ($\sigma_{Y_a}^2 = 6.5$). Each column represents a different K_w value, which increases from left to right.

[FIGURE 6: HERE]

We first analyzed the set of simulations with lower heterogeneity (top rows of Figure 6). We note from the green line that the ensemble-averaged concentration increases as K_w decreases. This occurs since a higher mean hydraulic conductivity of the CB walls ensures a higher mean inward advective flux that more efficiently contrasts the outwards diffusion. More specifically, we found that for $K_w = 10^{-3}$ m/d, the normalized concentration is of the order of $\log_{10}(C/C_0)$ = -42. This extremely low value has to be assumed as virtually zero, since from a practical perspective such concentration would likely fall much below to the detection limit of typical analytical approaches (considering C as expressed in mg/L). In addition, numerical stability may affect the actual reliability of this number. For $K_w = 10^{-4}$ m/d, the expected concentrations is slightly smaller than $\log_{10}(C/C_0)$=-4, suggesting that this CP&T configuration is in the limit of providing an effective approach for the confinement and remediation of the contamination plume, when compared to the selected maximum concentration threshold. For $K_w = 10^{-5}$ m/d, the mean concentration exceeded C^* by several orders of magnitude and the system is clearly not effective for the purpose of its application.

The analysis of the histograms and the corresponding CDFs reveals that the variability of the concentrations around the mean value (which is directly associated to the variability of K_a fields and directly measurable by $\sigma_{Y_a}^2$) affects the reliability of the ensemble mean as an informative indicator about the actual efficiency of the CP&T. For $K_w = 10^{-3}$ m/d, the concentrations were observed to fluctuate over 5 orders of magnitude around the mean. None of the realizations detected a concentration above C^*, suggesting that the likelihood of pollution is negligible for this scenario, even from a very conservative perspective. For the scenario with $K_w = 10^{-5}$ m/d, the likelihood of aquifer pollution is maximum, since C^* is always exceeded by all the realizations. In the intermediate case with $K_w = 10^{-4}$ m/d, however we observed that the distribution is quite symmetric around the mean. In this specific scenario, it occurred by chance that the mean also corresponds to the maximum concentration threshold. Because of that, and observing the CDF, it is easy to infer that C^* is exceeded by about 50% of the total realizations. In other words, the probability that one individual aquifer out of the polluted site will be polluted after 10 years is 50%. When generalized to other field studies with boundary conditions similar to the one analyzed in this work, the result can be also read as if one out of two aquifers embedding this CP&T configuration and having $\sigma_{Ya}^2 = 1$ will be polluted after 10 years.

The set of simulations with higher heterogeneity (bottom row of Figure 6) provides additional important insights from this analysis. In this set of SGS simulations, the scenario with $K_w = 10^{-3}$ m/d still ensures a great solution to contrast the downstream solute migration. Note however that despite the normalized concentrations being very low, the mean has increased by some 10 orders of magnitude compared to the same K_w in the set of SGS simulations with lower variance. Moreover, although numerical stability may still play a role for such very low numbers, the range of concentrations around the mean is now observed being >20 orders of magnitude larger than for the simulations with lower σ_{Ya}^2. For the case with $K_w = 10^{-5}$ m/d, the expected concentration remains remarkably higher than the threshold C^*. However, in this case the expected concentration is lower than in same K_w scenario for the low heterogeneity settings, suggesting that an increase in heterogeneity generates (on average) a better general efficiency of the CP&T under the same CB wall conditions. It is noted that the range of variability of the concentrations has significantly increased even for this scenario, an effect once again associated with the increase in $\sigma^2_{Y_a}$.

It is critical to observe and analyze the behavior of the CP&T embedding $K_w = 10^{-4}$ m/d of the set of simulations with higher heterogeneity. Here, we found that the average concentrations have dropped to $\log_{10}(C/C_0) \approx$-6, which is two orders of magnitude lower than the same result for lower heterogeneity, and also than the threshold C^*. This issue confirms that (on average) the more heterogeneous system is expected to better contain the solute plume than the more homogeneous system. In addition, the analysis of the histograms and CDFs reveals that for $\sigma^2_{Ya} = 6.5$ there is a significant fluctuation of measured concentrations around the mean (spanning over about 8 orders of magnitude). However, about 20% of the simulations exceeded the critical threshold C^*, or in other terms that the probability of aquifer pollution is one out of five aquifers. This means that for $\sigma^2_{Ya} = 6.5$ the estimated likelihood of aquifer pollution is less than one half the likelihood of aquifer pollution estimated for $\sigma^2_{Ya} = 1$.

Apart of the scenario with $K_w = 10^{-3}$ m/d, which may be affected by numerical instability, the results suggest that simulations based on $\sigma^2_{Y_a} = 6.5$ show a higher mean efficiency than those based on $\sigma^2_{Y_a} = 1$. To explain why this is occurring, we recall the behavior of these systems under homogeneous conditions and considering the selected type of boundary conditions and aquifer parameterization. For mass balance and under the same Q_w, an aquifer with a lower K_a experiences more pronounced drawdown in the water table than an aquifer with a higher K_a to maintain the same flux (e.g. Devlin and Parker, 1996; Neville and Andrews, 2006). This creates a more pronounced drawdown head gradient across the barrier and a higher advective flux. For the same K_w, a lower K_a value reduces diffusive fluxes more efficiently across the walls than for a higher K_a value. Unlike homogeneous systems, however, our simulations embed a range of correlated K_a values drawn from lognormal distributions. By definition, this distribution, which is typically observed in nature (e.g. Freeze, 1975) and not merely valid for the site-specific conditions analyzed here, generates more K_a values below the $E(Y_a)$ than values above the $E(Y_a)$. Thus, a heterogeneous correlated field with mean $E(Y_a)$ =0 and $\sigma^2_{Y_a} = 6.5$ has a much larger probability of generating low K_a value than a heterogeneous field with mean $E(Y_a)$ =0 and $\sigma^2_{Y_a} = 1$, and thus a higher probability of more pronounced drawdown induced by the drain. To quantitatively support this interpretation, Figure 7 reports the statistical distribution of maximum drawdown observed nearby the drains for the six scenarios analyzed in this work. It is noted that the scenarios embedding $\sigma^2_{Y_a} = 6.5$ (right-hand boxplots) generate more drawdown than those embedding $\sigma^2_{Y_a} = 1$ (left-hand boxplots). It is important to note that this is true independently from the selection of K_w.

[Figure 7 : HERE]

4.3. Limitation and future developments

Our analysis aims to target primarily the spatial variability of K_a and K_w as the key source of uncertainty. Some important aspects that can potentially control the long-term estimation of the effectiveness of the CP&T system have been left outside this study. They include, for instance, the temporal change in CB walls conditions associated with the durability of the cement-bentonite mixtures (e.g. Inyang and Tomassoni, 1992), such as the chemical attack of aquifer contaminants with the soil-bentonite mixtures (e.g., Carreto et al., 2015; Malusis and McKeehan, 2013). These aspects require a non-trivial modeling approach (e.g. nonlinear time-dependent K_w parameterization and reactive transport modeling analysis). Another aspect to be considered is that the strong heterogeneity of the system could result in preferential flow paths, which may require a different type of modeling analyses to be properly understood. Indeed, preferential flow paths in bounded systems could be either reproduced via multidimensional flow modeling, such as 3-D simulations (e.g. Pedretti et al., 2013b) or via effective 1-D models in which preferential flow is associated with connected, highly mobile paths and embedded into a less mobile matrix (e.g. Molinari et al., 2015). The implication of these aspects for the effectiveness of coupled systems will be investigated in future developments.

Summary and conclusion

The combination of pump-and-treat technologies and vertical cutoff walls is a common solution for the remediation of solute plumes in polluted aquifers. The design of these systems requires a correct parameterization of the hydrogeological properties of both the physical barriers and the surrounding aquifer. The presence of heterogeneity complicates the assessment of these parameters, generating uncertainty in the reliability of model predictions and requiring a stochastic modeling approach to be properly evaluated.

We adopted a stochastic modeling approach to study the expected long-term efficiency of the remediation system to contain solute plumes in highly heterogeneous aquifers, using a hydrogeological database from a well-characterized case study in Italy. The hydrogeological setup of this site, commonly found in application of plume confinement and remediation in alluvial aquifers is used to draw the working conceptual site model. The database contained 471 results from pre-elaborated in-situ and laboratory tests performed to characterize the hydraulic conductivity of the aquifer and the 2-km-long cement-bentonite (CB) walls. Based on a geostatistical analysis of this database and site, we developed a one-dimensional (1D) stochastic-based modeling framework evaluating different scenarios of randomly variable aquifer heterogeneity (K_a) and effective hydraulic conductivity of the barrier (K_w).

From the analysis of the results presented in the paper, we conclude that (1) the presence of aquifer heterogeneities, (2) the difficulties to select an adequate K_w and (3) the uncertain scaling-up of laboratory-estimated K_w into field-scale counterparts render the management of polluted aquifer difficulties, with potential risk to exceed predefined thresholds of contaminant concentrations at targeted vulnerable zone. More specifically,

- heterogeneity was seen to play a major role for the long-term assessment of the effectiveness of the confinement system;
- for a given effective K_w, the increase of aquifer heterogeneities (here condensed into a single indicator $\sigma^2_{Y_a}$, i.e. the variance of the univariate $\ln(K_a)$ distribution) determines, on average, an increase in efficiency of the confinement system for the analyzed 1D system;
- the increase of aquifer heterogeneities adds more uncertainty regarding the optimal hydraulic design of the system, adding uncertainty in the optimal strategies for the effective management of solute plumes over long time scales.
- a very low value of K_w may not be necessarily beneficial for an effective aquifer remediation.

The last conclusion is drawn considering that a low K_w requires higher drawdowns in the polluted aquifer to maintain sufficiently steep hydraulic gradients across the barrier to generate advective groundwater flux counterbalancing outwards solute diffusive flux (higher drawdowns may not be easily overcome by increasing the pumping rates, for instance due to limited costs or aquifer transmissivity).

In light of the well-known misconceptions regarding stochastic analysis, which limit its routine use among practitioners (e.g. Renard, 2007; Sanchez-Vila and Fernàndez-Garcia, 2016), our study showed that it is indeed possible to bridge the gap between stochastic theory and application by means of efficient calculation tools and using traditional measurements from physically and hydraulic barriers. It is acknowledged that our conclusions are valid, provided the conditions of applicability of the selected model analysis are fulfilled. For instance, ADE-based 1D models may not be able to effectively describe preferential flow and channeled transport, which may occur in heterogeneous media. Moreover, conservative transport cannot reproduce a possible time-dependent change in the walls' hydraulic properties due to the interaction between solute plume and cement-bentonite matrix. The extension of our modeling analysis to multidimensional and reactive systems is left open for future developments.

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Figures and captions

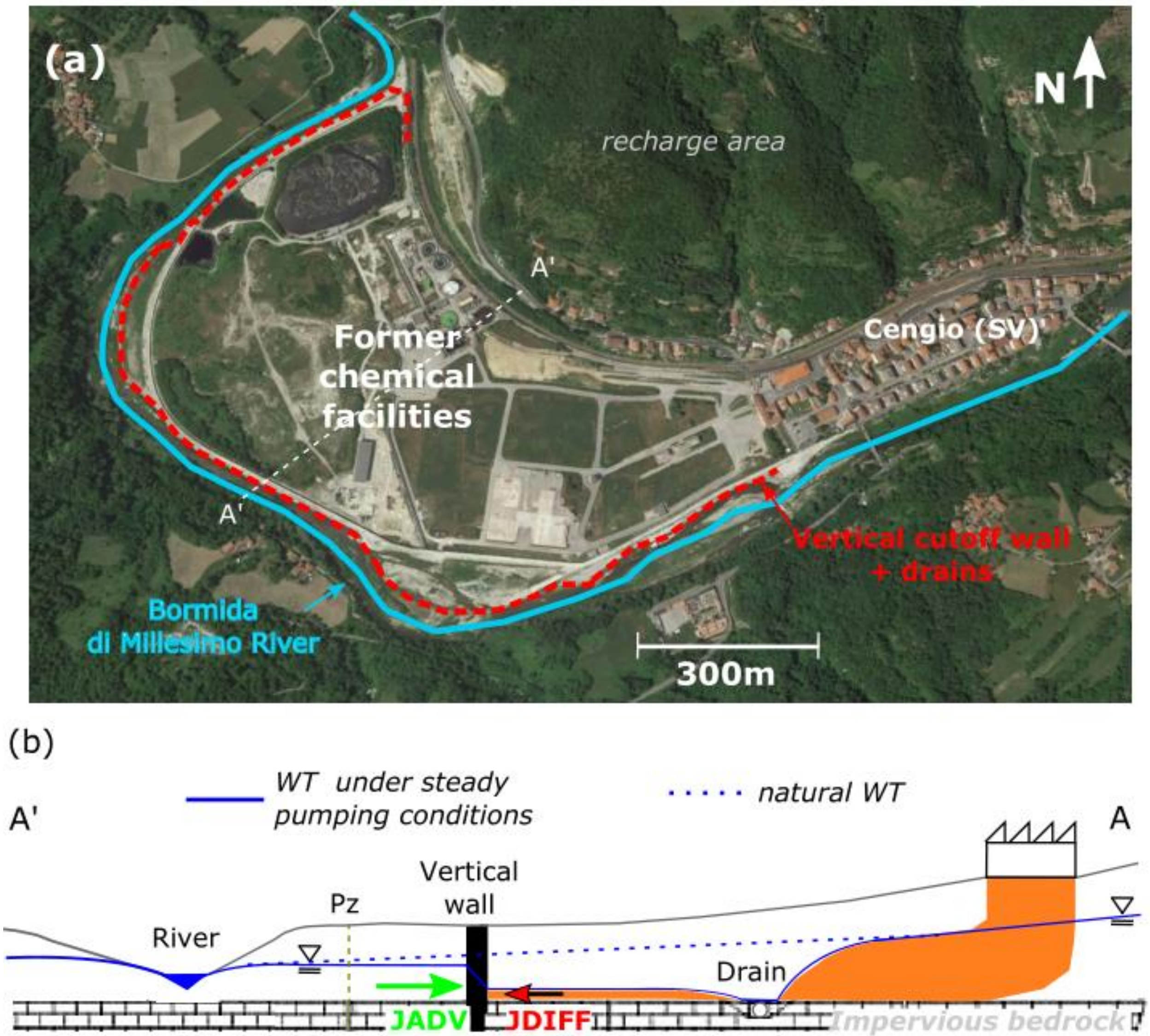

Figure 1 (a) Aerial view of the former chemical facilities. (b) Conceptual site model. JADV=inwards advective flux (under pumped conditions), JDIFF= diffusive flux, Pz=piezometer; *WT* = water table. The dotted line indicates the trace of the vertical section.

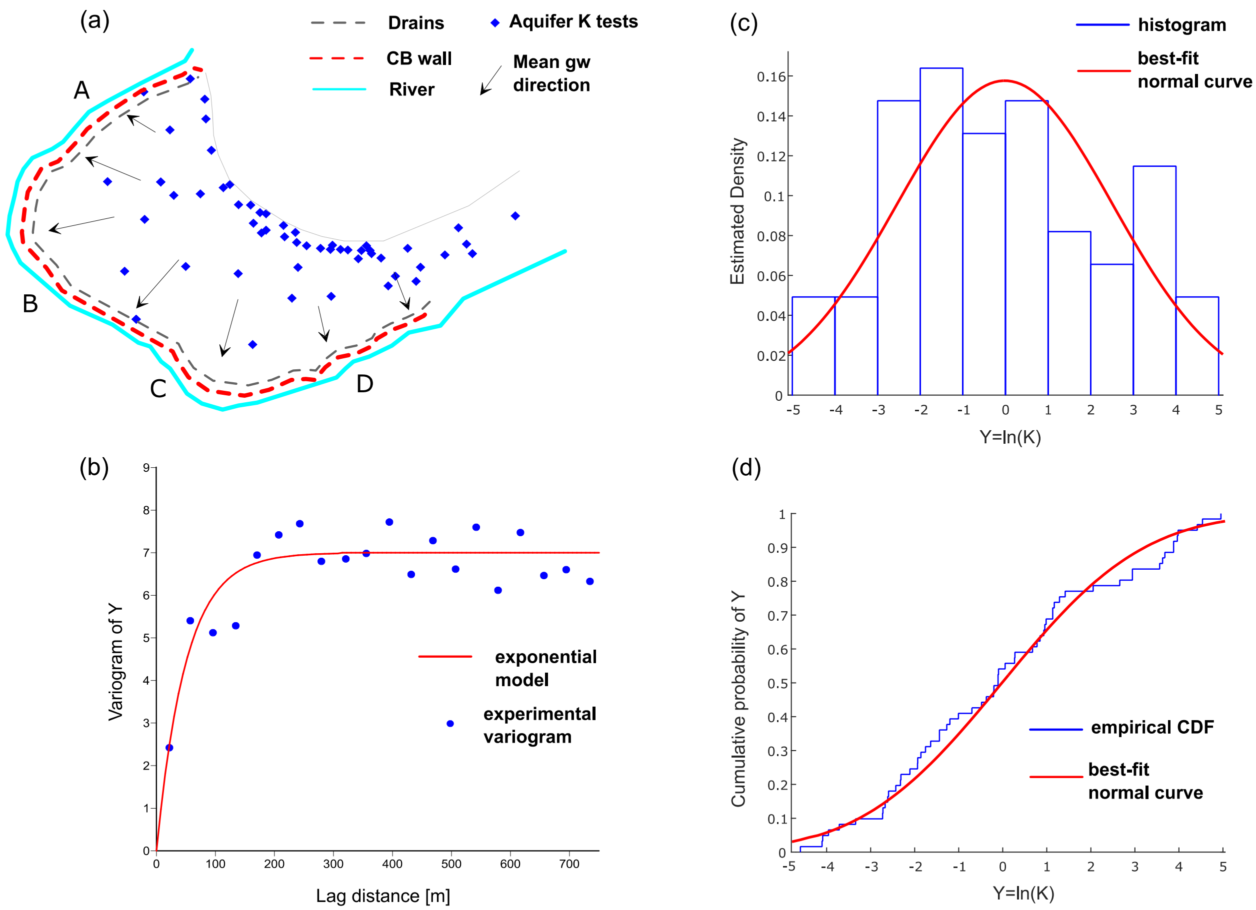

Figure 2 (a) Distribution of aquifer hydraulic testing locations (diamonds), the mean direction of the groundwater flow and geometry of the confinement area at the time the aquifer characterization. A,B,C,D are the zone labels for the CB walls hydraulic characterization. (b) Experimental and exponential variogram of Y_a . (c) Histogram and (d) cumulative empirical distributions of Y_a, respectively, along with the best-fir normal model.

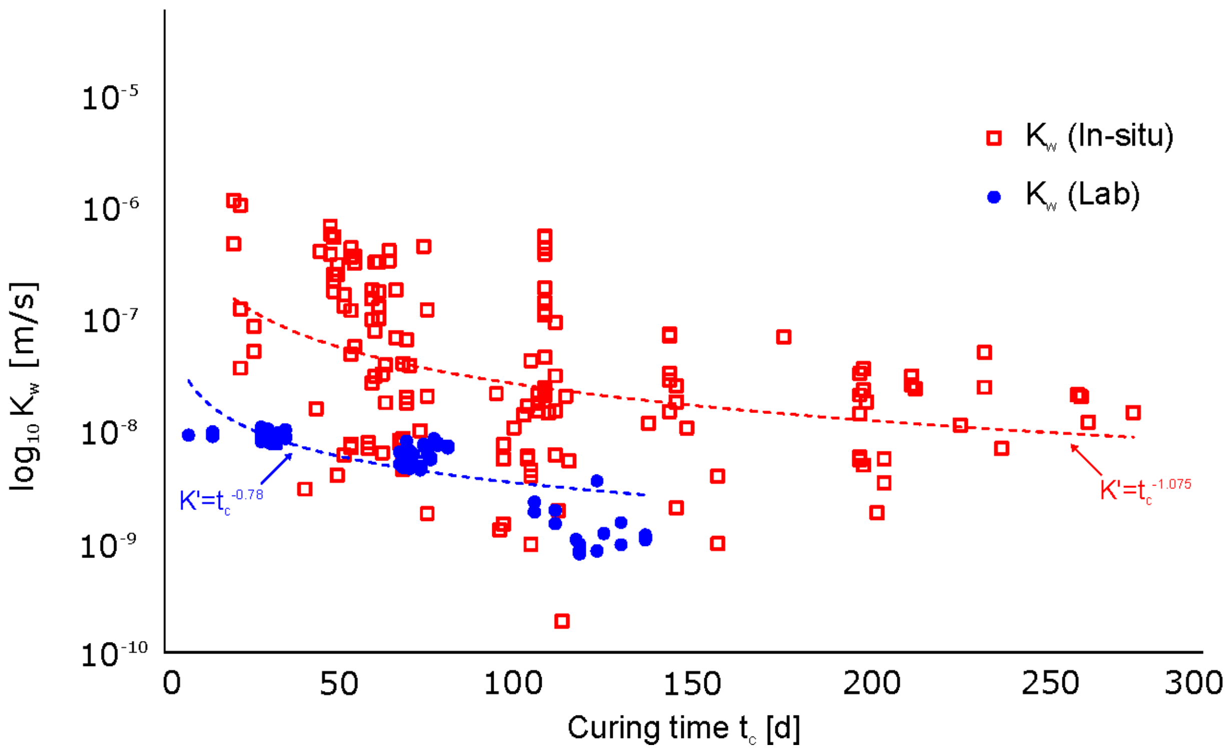

Figure 3 Evolution of measured K_w obtained from laboratory tests (blue) and from in-situ tests (red) at difference curing times (t_c). Here, $K' = K/K_0$, where K_0 is estimated from a best-fit regression curve of form $K = K_0 K_w^{-m}$ and m is the characteristic (scale-dependent) hardening factor.

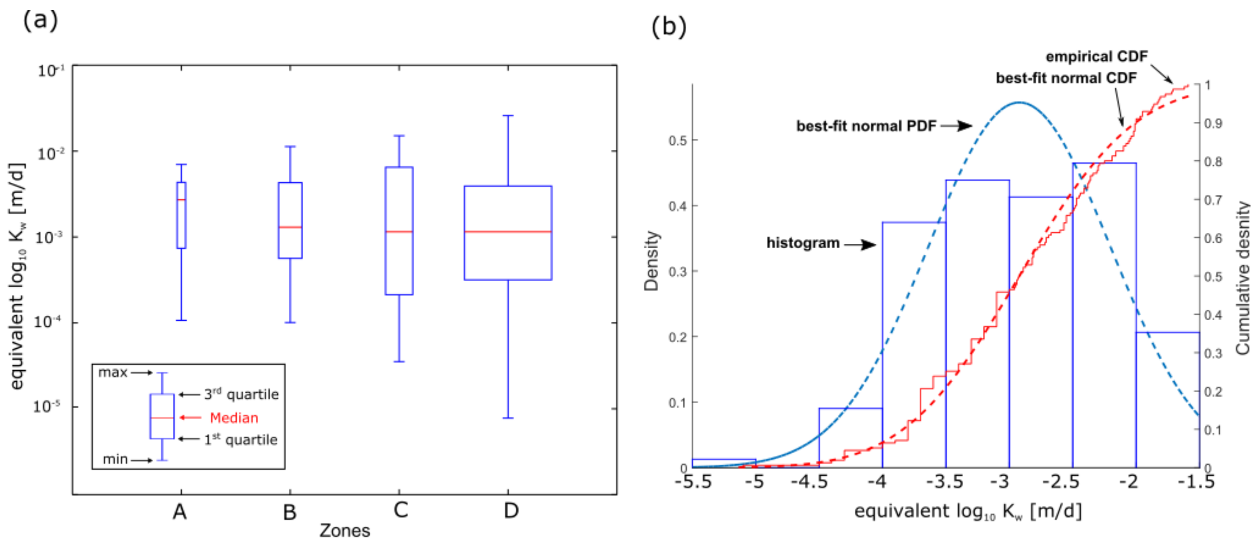

Figure 4 (a) Boxplots representing the statistical distribution of the equivalent $\log_{10} K_w$ estimated from in-situ tests and extrapolated at 280 days for the four different tested zones along the CB wall. The width of the boxes is proportional to the number of samples (n_s) used to calculate the empirical distributions. (b) Empirical histograms (blue) and cumulative density functions (red) of the equivalent $\log_{10} K_w$ at 280 days merging the four zones. Straight lines are the best-fit normal curves fitting the $\log_{10}$-transformed hydraulic conductivity values.

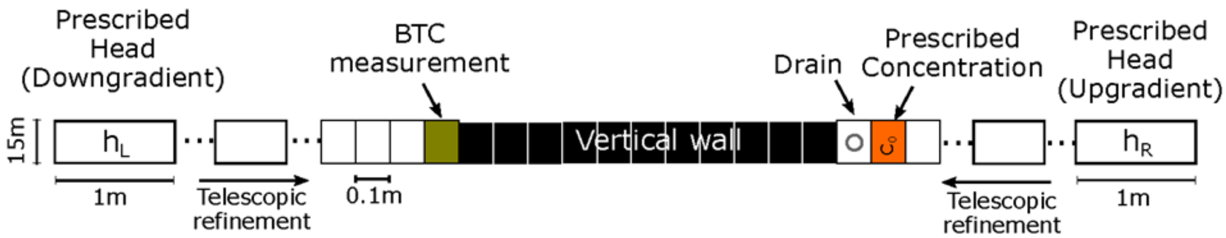

Figure 5 Conceptual model and numerical discretization of the domain for the stochastic solution of flow and transport in random domains. The size of the cells decrease telescopically from the sides towards the center of the domain. h_L and h_R are the downgradient and upgradient prescribed head boundary conditions, respectively. C_0 is the prescribed concentration at the continuous source nearby the drain, which has a steady discharge rate. The vertical walls are located between the drain and the cell where (resident) concentrations are measured in the form of breakthrough curve (BTC).

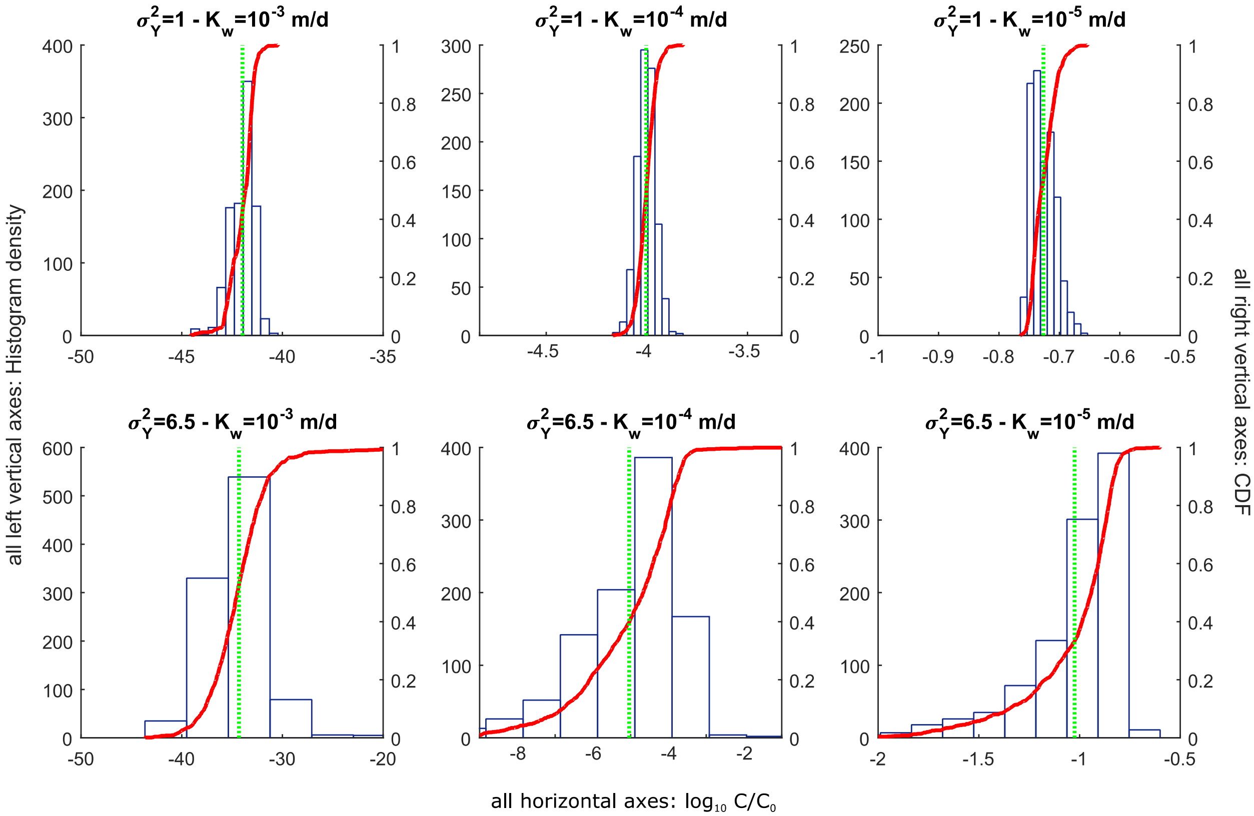

Figure 6 Summary of the stochastic analysis. Each panel shows the histogram of the distributions of resident concentrations monitored at the piezometer after 10 years from the initial spill from the ensemble of simulations, and the corresponding cumulative density function. The dotted green line is the ensemble average value, which suggests that the mean efficiency of the system and the variability around the mean are larger for the more heterogeneous set of simulations.

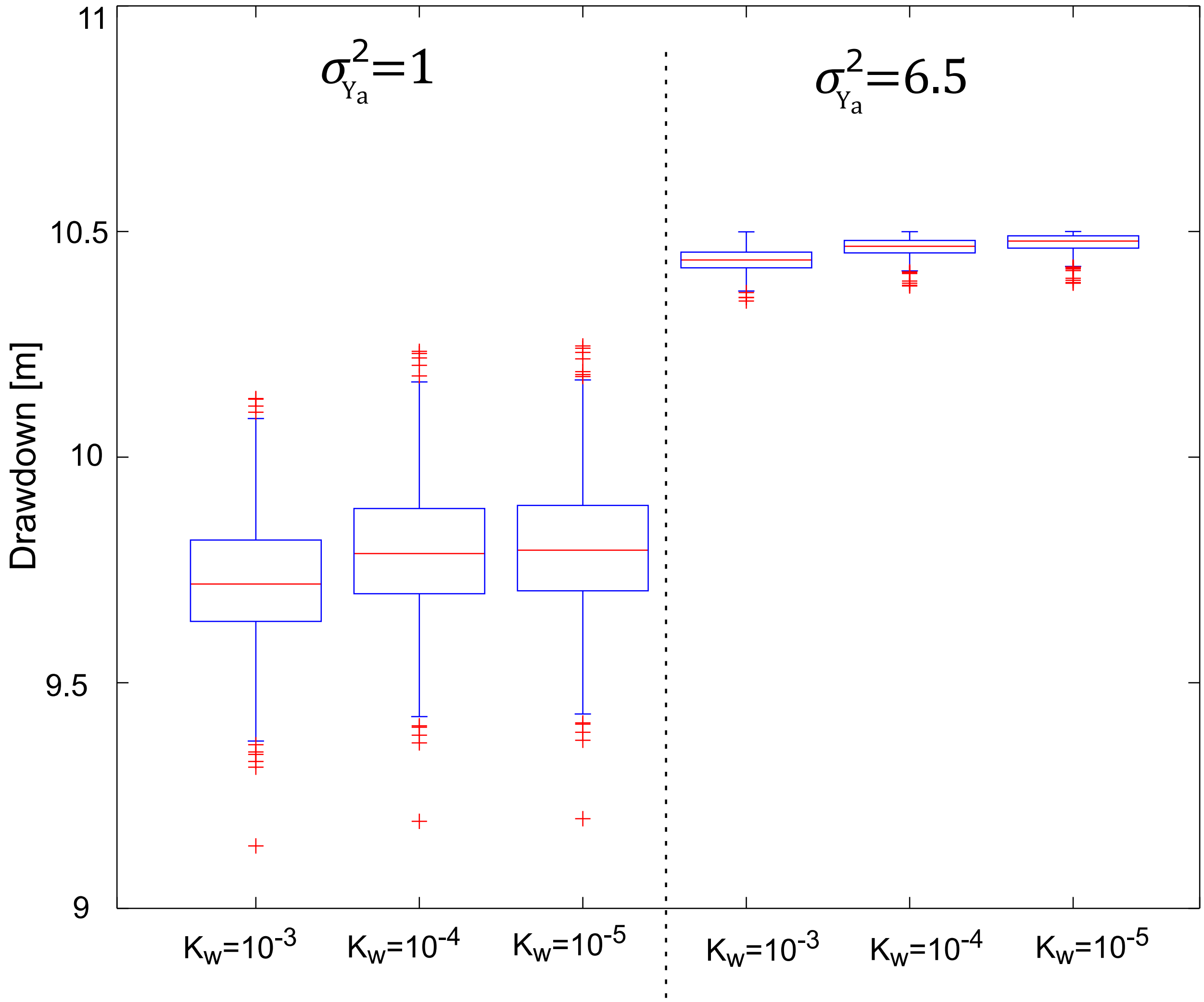

Figure 7 Boxplots representing the statistical distribution of maximum drawdown observed nearby the drain cell from the six analyzed scenarios. Note that greater drawdown is generated when the heterogeneity is higher (i.e. for higher variance of the log-transformed aquifer hydraulic conductivity, $\sigma^2_{Y_a}$). The units of the wall's hydraulic conductivity, K_w, are [m/d].

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