

USING SPARSE POLYNOMIAL CHAOS EXPANSIONS FOR
THE GLOBAL SENSITIVITY ANALYSIS OF GROUNDWATER
LIFETIME EXPECTANCY IN A MULTI-LAYERED
HYDROGEOLOGICAL MODEL

G. Deman, K. Konakli, B. Sudret, J. Kerrou, P. Perrochet, H. Benabderrahmane



Data Sheet

ETH Project: Sensitivity analysis in computational hydrogeology

Project ID: 34971

Journal: -

Report Ref.: RSUQ-2015-001

Arxiv Ref.: <http://arxiv.org/abs/1502.01477> - [stat.CO]

DOI: -

Date submitted: January 20th, 2015

Date accepted: -

Using sparse polynomial chaos expansions for the global sensitivity analysis of groundwater lifetime expectancy in a multi-layered hydrogeological model

G. Deman¹, K. Konakli^{*2}, B. Sudret², J. Kerrou¹, P. Perrochet¹, and H. Benabderrahmane³

¹*The Centre for Hydrogeology & Geothermics (CHYN), University of Neuchâtel, Rue Emile Argand 11, CH-2000 Neuchâtel, Switzerland*

²*ETH Zürich, Institute of Structural Engineering, Chair of Risk, Safety & Uncertainty Quantification, Stefano-Franscini-Platz 5, CH-8093 Zürich, Switzerland*

³*Andra, 1-7 rue Jean Monnet, 92298 Châtenay-Malabry Cedex, France*

Abstract

The present study makes use of polynomial chaos expansions to compute Sobol' indices in the frame of a global sensitivity analysis of hydro-dispersive parameters in a synthetic multi-layered hydrogeological model. The model introduced in this paper is a simplified vertical cross-section of a segment of the subsurface of the Paris Basin. This 15-layer numerical model is aimed at exploring the behavior of groundwater and solute fluxes when accounting for uncertain advective and dispersive parameters. Applying conservative ranges, the uncertainty in the input variables is propagated upon the mean lifetime expectancy of water molecules departing from a specific location within a highly confining layer situated in the middle of the numerical model. The sensitivity analysis indicates that the variability in the mean lifetime expectancy can be sufficiently explained by the uncertainty in the petrofacies, *i.e.* the sets of porosity and hydraulic conductivity, of only a few layers of the model.

^{*}Corresponding author, e-mail: konakli@ibk.baug.ethz.ch

1 Introduction

With the improvement of computing power, numerical modeling has become a popular tool for understanding and predicting various kinds of subsurface processes addressed in the fields of geology and hydrogeology. However, the incomplete/imprecise knowledge of the underground system frequently compels the modeller to make a number of approximations and assumptions with regard to the geometry of geological structures, the presence of discontinuities and/or the spatial distribution of hydro-dispersive parameters in their models Renard (2007). These uncertainties can possibly lead to large variabilities in the predictive modeling of subsurface processes and thus, it becomes of major importance to account for the aforementioned assumptions in the frame of uncertainty and sensitivity analyses. Uncertainty analysis (UA) aims at quantifying the variability of a given response of interest as a function of uncertain input factors, whereas sensitivity analysis (SA) has the purpose to identify the input factors responsible for this variability. Hence, SA determines the key variables to be described in further detail in order to reduce the uncertainty on the predictions of a model.

Methods of SA are typically classified in two categories: local SA and global SA methods. The former investigate effects of variations of the input factors in the vicinity of nominal values, whereas the latter aim at quantifying the output uncertainty due to variations in the input factors in their entire domain. Among several global SA methods proposed in the literature, of interest herein is SA with *Sobol' sensitivity indices*, which belongs to the broader class of variance-based methods Saltelli et al. (2008). These methods rely upon the decomposition of the response variance as a sum of contributions of each input factor or combinations thereof; unlike regression-based methods, they do not assume any kind of linearity or monotonicity of the model.

Various methods have been investigated for computing the Sobol' indices that were first defined in Sobol' (1993), see *e.g.* Archer et al. (1997); Sobol' (2001); Saltelli (2002); Sobol' and Kucherenko (2005); Saltelli et al. (2010). In these papers, Monte Carlo simulation is used as a tool to estimate these sensitivity indices. This has revealed extremely costly, although more efficient estimators have been recently proposed Sobol et al. (2007); Janon et al. (2013). In the last few years, new approaches using surrogate models have been introduced in the field of global sensitivity analysis Storlie et al. (2009); Zuniga et al. (2013). A popular method to compute the Sobol' indices, originally introduced by Sudret (2008), is by post-processing the coefficients of the polynomial chaos expansion (PCE) of the response quantity of interest. PCE constitute an efficient UA method in which the key concept is to expand

the model response onto a basis made of orthogonal polynomials in the input variables. Once a PCE representation is available, the Sobol' indices can be calculated analytically with elementary operations at almost no additional computational cost. Sparse PCE make the approach even more efficient, as shown in Blatman and Sudret (2010a).

In the frame of the stochastic modeling of subsurface flow and mass transport, PCE meta-models have proven to be robust and comprehensive tools in performing SA at low computational cost. As an example, applying a PCE-based global SA upon a fine-grid numerical model of flow and mass transport in a heterogeneous porous medium, Fajraoui et al. (2011) and Younes et al. (2013) established the transient effect of uncertain flow boundary conditions, hydraulic conductivities and dispersivities on solute concentrations at given observation points. Sochala and Le Maître (2013) propagated uncertain soil parameters upon three different physical models of subsurface unsaturated flow. Their study proved the higher efficiency of PCE meta-models, in comparison to a classical Monte-Carlo method, for representing the variability of the output quantity at low computational cost. In the frame of radionuclide transport simulation in aquifers, Ciriello et al. (2013) analyzed the statistical moments of the peak solute concentration measured at a specific location as a function of the conductivity field, the dispersivity coefficients and the partition coefficients associated to the heterogeneous media. The comparison of the Sobol' indices obtained for various degree of PCE meta-models showed that low-degree PCE models can yield reliable indices while considerably reducing the computational burden. Formaggia et al. (2013) used PCE-based sensitivity indices to investigate effects of uncertainty in hydrogeological variables on the evolution of a basin-scale sedimentation process. However, the various aforementioned contributions consider simplified models for the description of subsurface flow and mass transport. A more realistic representation of these processes is employed in the present study by using a large-scale simulator.

In the scope of the deep geological storage of radioactive wastes, ANDRA (French National Radioactive Waste Management Agency) has conducted many studies to assess the potentiality of a clay-rich layer for establishing a mid to long-lived radioactive waste disposal in the subsurface of the Paris Basin. The thick impermeable layer from Callovo-Oxfordian (COX) age has been extensively studied (Delay et al., 2006; Distinguin and Lavanchy, 2007; Enssle et al., 2011) together with the two major limestone aquifers, in place of the Dogger and the Oxfordian sequences (Brigaud et al., 2010; Linard et al., 2011; Landrein et al., 2013), encompassing the claystone formation. A recent study (Deman et al., 2015) used a high-resolution integrated Meuse/Haute-Marne hydrogeological model (AND, 2012) to compute the average time for

water molecules departing from a given area in the COX to reach the limits of the numerical model. SA over hydro-dispersive parameters in 14 hydrogeological layers proved that the Dogger and Oxfordian limestone sequences have a large influence on the residence time of groundwater. Indeed, advection processes occurring in permeable layers strongly influence the water transit in the subsurface of the Paris Basin, in contrast to the slow-motion diffusive processes taking place in impermeable rocks.

However, the analysis of the effect of uncertainties related to other advective-dispersive parameters, such as boundary conditions, orientations and anisotropies of hydraulic conductivity tensors or magnitudes of dispersion parameters, represents a great effort that cannot be carried out with the integrated model at reasonable computational costs. Addressing the issue of performing UA with the use of high-resolution numerical models of geological reservoirs, Castellini and co-workers (Castellini et al., 2003) established that numerical models built at the coarse scale, but covering a reasonable number of geological and geostatistical features, can be particularly informative in capturing the main subsurface processes at low computational costs.

The present study introduces a vertical two-dimensional multi-layered hydrogeological model representing a simplification of the underground media of the Paris Basin in the vicinity of the site of Bure and does not integrate the complex geometry of the layers, neither does it include the numerous discontinuities or heterogeneities observed in the field. It must be emphasized that the use of this model is focused on numerical issues, sensitivity analysis and calibration purposes and the results cannot be considered with respect to the real situation.

The main objective of the present work is to assess the effect of multiple advective-dispersive parameter on the mean lifetime expectancy (MLE) of water molecules departing from a target zone in the central layer. The MLE corresponds to the average time required for a given solute, taken at a specific location, to reach any outlet of the model. Conservative uncertainty ranges are defined for the input factors analyzed in the frame of a SA relying on the estimation of PCE-based Sobol' indices. This study provides a preliminary assessment on the relative effect of factors governing the MLE in the subsurface of the Paris Basin and recommendations are made for the application on the high-resolution integrated Meuse/Haute-Marne hydrogeological numerical model of the Paris Basin.

2 The numerical model

2.1 Geometry and finite element mesh

Originally inspired by the COUPLEX numerical model from Bourgeat et al. (2004), the present model stands as a vertical two-dimensional (x - z) cross-section of $25,000 \times 1,040$ meters representing a segment of the Paris Basin subsurface. The mesh is discretized into 5×5 meters square elements for a total of 1,040,000 elements. In order to subdivide the domain into entities related to geological formations, the main features of the subsurface were extracted from the lithostratigraphic log of the deep EST433 borehole (Landrein et al., 2013) in the vicinity of the experimental site of Bure (Haute-Marne, France). Therefore, the model consists of 15 hydrogeological layers characterized by tabular geometries, uniform thicknesses and homogeneous parameters. Figure 1 summarizes the geometry of the model and gives an overview on the succession and thicknesses of layers.

The bottom layer stands as a 110 m thick low-permeability layer attributed to the Toarcian marl formation (T). Overlying the latter, the succession of carbonate formations from the Dogger sequence is subdivided into 5 layers of which the total thickness attains 250 m in the numerical model. The sequence encompasses the Bajocian (D1) and Bathonian limestones (D3 and D4) representing the main aquifer formations of the Dogger, a clastic dominated interval ("*Marnes de Longwy*", D2) separating the two. The Dogger sequence is topped with a thin oolitic limestone from Lower Callovian ("*Dalle Nacrée*", C1), implemented as a 15 m thick layer in the model. The latter marks the transition with the thick, highly impermeable, claystone formation of Callovo-Oxfordian age (C2) of which the thickness reaches 150 m in the model. In the numerical simulations, a target zone (TZ) located in the middle of layer C2 (Figure 1) represents the location for the computation of the output quantity of interest.

The low-permeability COX layer is overlaid by a limestone sequence of the Oxfordian age. The latter is incorporated as a 260 m thick formation subdivided into 6 hydrogeological entities. A relatively confining layer from the Upper Argovian (C3ab) rests directly on the COX and is followed by permeable formations of the Rauracian-Sequanian sequence (L1a to L2c). A thick interval of marls and argillaceous limestones from Kimmeridgian age (K1-K2) covers the whole and is implemented as a 160 m thick low-permeability layer. The top layer is a 120 m thick confining formation attributed to the Tithonian (K3). The latter outcrops in the vicinity of Bure.

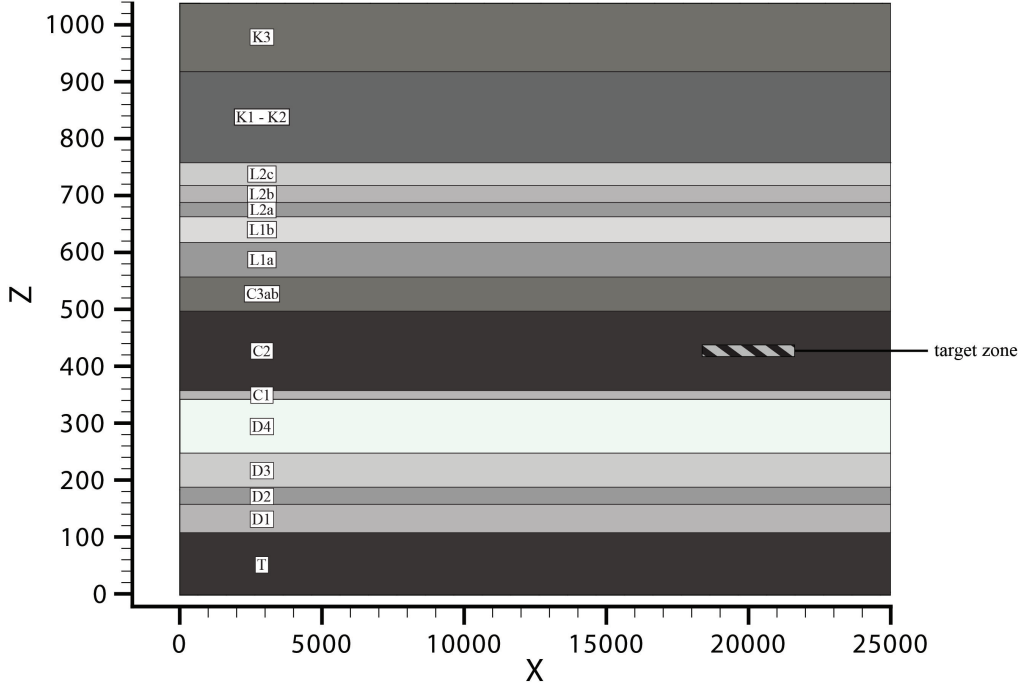


Figure 1: Geometry and geological layers with the localization of the target zone (vertical exaggeration: 20).

2.2 Governing equations and model outputs

In the numerical simulations the flow is governed by the steady-state equation

$$\nabla \cdot \mathbf{q} = 0, \quad (1)$$

where $\mathbf{q} = -\mathbf{K} \nabla H$, is the Darcian flux vector [$L T^{-1}$], \mathbf{K} is the tensor of hydraulic conductivity [$L T^{-1}$] and H is the hydraulic head [L]. The anisotropy A_K in the components of the tensor of hydraulic conductivity is defined as the ratio between the hydraulic conductivities in the two principal directions: $A_K = K_z/K_x$.

Here, it is assumed that \mathbf{K} has orthotropic properties. Considering a hydraulic conductivity tensor \mathbf{K}_p of which the components are mapped into the Cartesian system and given along their principal direction, X_p , the tensor \mathbf{K} in the global Cartesian space is retrieved by means of the rotation matrix \mathbf{R} with the expression

$$\mathbf{K} = \mathbf{R}^T \mathbf{K}_p \mathbf{R}. \quad (2)$$

For the two-dimensional problem considered, the rotation matrix \mathbf{R} is defined

in terms of the Euler angle θ (in degree) as

$$\mathbf{R} = \begin{pmatrix} \cos \theta & \sin \theta \\ -\sin \theta & \cos \theta \end{pmatrix}. \quad (3)$$

In the present study, steady-state flow simulations are carried out together with the computation of the lifetime expectancy probability density function (PDF) at any point x in the domain. Under stationary conditions (i.e. steady-state flow), the lifetime expectancy PDF addresses the probability distribution of the time required for a solute, taken at any position x , to leave the domain. In its formulation, the lifetime expectancy PDF assimilates the forward advective-diffusive transport equation (ADE) to the Fokker-Planck (forward Kolmogorov) equation measuring the random motion of solute particles (Uffink, 1989). For more details on the computation of the lifetime expectancy PDF the reader is referred to Cornaton and Perrochet (2006a,b) and Kazemi et al. (2006).

Based on the ADE, the lifetime expectancy PDF is computed using the backward transport equation requiring reversed flow directions ($\mathbf{q} := -\mathbf{q}$) as well as adjusted downstream boundary conditions. The lifetime expectancy PDF $g_E(x, t)$ at any point x in the domain is then governed by

$$\phi \frac{\partial g_E}{\partial t} = \nabla \cdot (\mathbf{q} g_E + \mathbf{D} \nabla g_E), \quad (4)$$

where ϕ is the effective porosity [-] and where \mathbf{D} is the dispersion tensor

$$\phi \mathbf{D} = (\alpha_L - \alpha_T) \frac{\mathbf{q} \otimes \mathbf{q}}{\|\mathbf{q}\|} + \alpha_T \|\mathbf{q}\| \mathbf{I} + \phi D_m \mathbf{I}, \quad (5)$$

where \mathbf{I} is the identity matrix, D_m is the coefficient of molecular diffusion [$L^2 T^{-1}$], α_L and α_T are the longitudinal and transverse components of the macro-dispersion tensor [L] respectively. In the present study, the anisotropy in the macro-dispersion tensor is determined with the coefficient $A_\alpha = \alpha_T/\alpha_L$.

The straightforward computation of the first moment of the lifetime expectancy PDF is the so-called *mean lifetime expectancy* (MLE) $E(x)$ [T] at any position x , governed by

$$-\nabla \cdot (\mathbf{q} E + \mathbf{D} \nabla E) = \phi, \quad (6)$$

where it can be seen that the porosity ϕ [-] acts as the sink term in the aging process.

The target zone (TZ) comprises a set of 1,947 nodes in layer C2, covered by a rectangle which lateral and vertical extensions are $x = [18,440 ; 21,680]$, z

= [425 ; 435] (Figure 1). In the present study, the arithmetic mean of $E(x, z)$ calculated at each of these 1,947 nodes stands for the output response of interest and is used in the subsequent analysis. It can be seen as the average time for a solute originating from the TZ to reach any outlet of the model.

The finite element simulator *GroundWater* (Cornaton, 2007) was employed to solve Eq. (1)-(6) using the finite element technique. A single run of steady-state flow and MLE computation takes about 120 seconds using a parallel solver with 6 CPU.

The reader should note that the use of a 2D vertical model to solve for the hydro-dispersive processes cannot capture correctly the real behavior of the Paris Basin subsurface because, apart from being a simplified model, it omits the lateral flow and dispersion along the third dimension. This has the effect to underestimate the magnitude of the modeled processes Kerrou and Renard (2010). It is however recognized that this bias is equivalent for all the layers considered, thus the interpretation of the SA results obtained with the 2D cross-section may be generalized to a synthetic 3D case employing the same settings.

2.3 Flow boundary conditions

The fully saturated model considers stationary flow conditions in a confined aquifer which are implemented as Dirichlet type flow boundary conditions. These flow BCs are imposed on nodes located on top of the numerical model as well as on both lateral limits of the two limestone sequences (Figure 2).

Regional piezometric maps based on field measures (Linard et al., 2011) were used to constrain the hydraulic gradients in both carbonate sequences. The flow BCs imposed on the lateral boundaries of the two limestone sequences derive from a 25 km transect starting from the Gondrecourt trough and extending in a North-West direction, the main regional flow direction. The hydraulic gradient set on top of the model corresponds to the average topographic gradient of the region covered by the transect.

Under these conditions, the general groundwater flow direction is oriented from right to left. The proportions of the total outflowing rates are approximately 2%, 60% and 38% for the top of the model, the Oxfordian and the Dogger discharge boundaries, respectively. In layer C2, the groundwater flows downward in the very right part of the model and then upward in the remainder, with a hydraulic gradient inversion in the vicinity of the TZ (see Figure 2). As a summary, the flow BCs are gathered in Table 1.

To account for uncertainties in the flow BCs, the hydraulic gradients in the two limestone sequences and on the top of the model are considered as uncertain input factors included in the following SA (Section 4). A change

Table 1: Flow boundary conditions.

| Boundary | Position | Hydraulic head |
|------------------|-----------------------------|-----------------------|
| right Oxfordian | $x = 25000, z = [500, 760]$ | $H = 305$ m |
| left Oxfordian | $x = 0, z = [500, 760]$ | $H = 230$ m |
| right Dogger | $x = 25000, z = [110, 360]$ | $H = 295$ m |
| left Dogger | $x = 0, z = [110, 360]$ | $H = 275$ m |
| top of the model | $x = [0, 25000], z = 1040$ | $H = 225 + 85x/25000$ |
| elsewhere | | <i>no flow</i> |

in the hydraulic gradients may shift the position of the vertical groundwater flux inversion in layer C2, and thus the MLE calculated at the TZ.

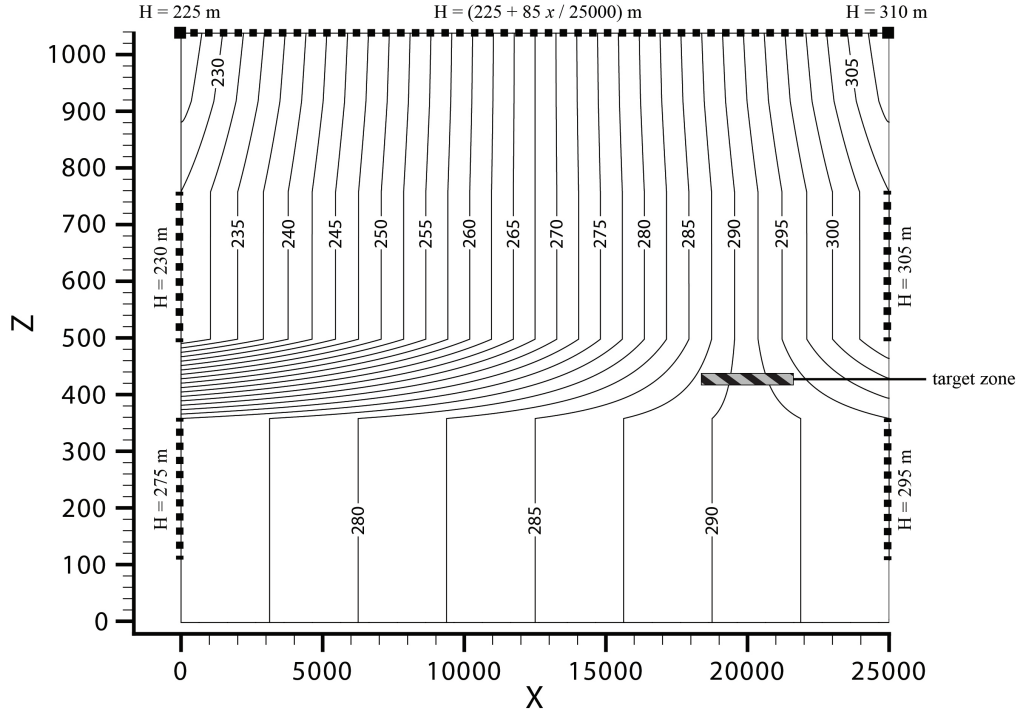


Figure 2: Flow boundary conditions and head contours (vertical exaggeration: 20).

2.4 Hydraulic conductivity and porosity values

Many studies have undertaken the inventory of hydraulic conductivity (K) and porosity (ϕ) values in the various geological formations of the Paris Basin.

For a large number of wells and boreholes within a wide area around the experimental site of Bure, laboratory and field measurements were conducted to provide $\{K, \phi\}$ datasets for the two limestone sequences (Brigaud et al., 2010; Linard et al., 2011; Fourre et al., 2011; Delay and Distinguin, 2004; Delay et al., 2007a).

However, very few $\{K, \phi\}$ datasets are available for the four low-permeability formations implemented in the present model (*i.e.* K3, K1-K2, C2 and T). Hence, data extracted from the literature (Cosenza et al., 2002; Delay et al., 2007b, 2006; Enssle et al., 2011; Mazurek et al., 2011; Vinsot et al., 2011), and employed in previous studies (Contoux et al., 2013; de Hoyos et al., 2012; Goncalves et al., 2004a,b), were used to define the uncertainty ranges for the $\{K, \phi\}$ sets in these layers.

In the geological formations of the Oxfordian and Dogger sequences, large variabilities of the $\{K, \phi\}$ couples are noticed with the presence of dependencies (*e.g.* low K and low ϕ values are correlated). However, to simplify the conceptual approach in a first stage, a perfect dependence between $\log_{10}(K)$ and ϕ is defined in each layer by making use of mathematical functions approximating the relationship between these two. In the sequel, both parameters are referred to as a whole under the name *petrofacies*. This approach reduces the computational burden of the subsequent SA by avoiding the use of correlation functions between the two uncertain factors. For each layer, the estimated value of hydraulic conductivity \hat{K} is retrieved through a relationship: $\log_{10}(\hat{K}) = f(\phi)$.

Although no explicit information is available on the following, the geological formations are believed to feature anisotropic hydraulic conductivity tensors \mathbf{K} , *i.e.* anisotropy in the two principal components of the tensor (K_x and K_z) defined as the ratio $A_K = K_z/K_x$. The hydraulic conductivity values deriving from the $\{K, \phi\}$ distributions in each layer are attributed to the longitudinal component of the hydraulic conductivity tensor, K_x . In the nominal case, $A_K = 0.1$ is assumed for every layer of the model.

Preferential flow directions are supposedly taking place within each individual layer. For each layer, the Euler angle θ defines the orientation of the hydraulic conductivity tensor \mathbf{K}_p in the Cartesian space (see Eq. (2)-(3)). In the nominal case, $\theta = 0$ degree is assumed in every layer, which corresponds to the two principal components of the hydraulic conductivity tensors \mathbf{K}_p being oriented along the x and z axes. The orientation of the groundwater flux \mathbf{q} in the model is principally due to the static hydraulic gradients ∇H resulting from flow BCs implemented on the edges of the model. Note however that the Euler angle θ may locally change the orientation of \mathbf{q} in a given layer and thus drive the groundwater into adjacent layers where magnitudes might be different. This phenomenon may have a significant effect on the

MLE calculated from the target zone, which is explored in Section 4.

Table 2 summarizes the nominal values for ϕ and the corresponding \widehat{K}_x in each of the 15 hydrogeological layers comprised in the model. The values for ϕ correspond to the mean value (or the median value of the CDF) of the distribution in each layer, whereas the values for \widehat{K}_x derive from approximation functions. As a reminder, the present study assumes homogeneous parameters in every layer. Although this feature is unrealistic, it is recalled that the purpose of this preliminary study is to bring insights into the global effect of equivalent advective-dispersive parameters of the multi-layered hydrogeological model and to provide recommendations for a similar application on a high-resolution integrated hydrogeological model of the Paris Basin.

Table 2: Nominal values for the porosity (ϕ) and the estimated longitudinal hydraulic conductivity (\widehat{K}_x) in the 15 hydrogeological layers.

| Layer | \widehat{K}_x [m/s] | ϕ [-] |
|-------|-----------------------|------------|
| K3 | 9.01E-09 | 0.0100 |
| K1-K2 | 4.53E-09 | 0.1150 |
| L2c | 1.10E-06 | 0.1389 |
| L2b | 3.46E-07 | 0.1110 |
| L2a | 1.62E-07 | 0.1139 |
| L1b | 1.49E-05 | 0.1604 |
| L1a | 1.17E-06 | 0.1549 |
| C3ab | 4.59E-08 | 0.0984 |
| C2 | 1.99E-13 | 0.1580 |
| C1 | 1.89E-06 | 0.0470 |
| D4 | 1.65E-05 | 0.0905 |
| D3 | 1.76E-06 | 0.1016 |
| D2 | 2.62E-07 | 0.0623 |
| D1 | 3.23E-06 | 0.0688 |
| T | 1.95E-12 | 0.0810 |

2.5 Dispersion parameters

The mean lifetime expectancy formulation (Eq. (6)) is an advective-dispersive solute transport equation, where the longitudinal and transverse components of the macro-dispersion tensor (α_L and α_T respectively) control the particles dispersion. These two uncertain factors depend particularly on the rock type, on the tortuosity of the porous media and also on the scale considered. Homogeneous values of α_L and α_T are set within each layer, with the values

$\alpha_L = 15$ m and $A_\alpha = \alpha_T/\alpha_L = 0.1$ considered in the entire numerical model in the nominal case.

As mentioned previously, no decay or adsorption effects are accounted in the computation of the MLE. The coefficient of molecular diffusion corresponds to the theoretical self-diffusion coefficient for the water molecule, $D_m = 2.3 \cdot 10^{-9}$ m²/s.

3 Polynomial chaos expansions for sensitivity analysis

Let us denote by \mathcal{M} the computational model describing the behavior of the considered physical system. Let $\mathbf{X} = \{X_1, \dots, X_M\}$ denote the M -dimensional random input vector with joint PDF $f_{\mathbf{X}}(\mathbf{x})$ and marginal PDFs $f_{X_i}(x_i)$, $i = 1, \dots, M$. Due to the input uncertainties represented by \mathbf{X} , the quantity of interest becomes random. The computational model is thus considered as the map

$$\mathbf{X} \in \mathcal{D}_{\mathbf{X}} \subset \mathbb{R}^M \mapsto Y = \mathcal{M}(\mathbf{X}) \in \mathbb{R}, \quad (7)$$

where $\mathcal{D}_{\mathbf{X}}$ is the support of \mathbf{X} . In the description of the theoretical framework hereafter, we assume that the components of \mathbf{X} are *independent*, which is the case for the model in the present study.

As explained in the Introduction, the aim of global sensitivity analysis is to identify random input variables and combinations thereof with significant contributions to the variability of Y , as described by its variance. A concise description of the employed method of PCE-based Sobol' sensitivity indices is given in the following; for further details on the topic, the reader is referred to Sudret (2008) and Blatman and Sudret (2010a). The extension to the case of mutually dependent random variables is presented in Kucherenko et al. (2012); Li et al. (2010).

3.1 Sobol' indices

Assuming that the function $\mathcal{M}(\mathbf{X})$ is square-integrable with respect to the probability measure associated with $f_{\mathbf{X}}(\mathbf{x})$, the Sobol' decomposition of $Y = \mathcal{M}(\mathbf{X})$ in summands of increasing dimension is given by Sobol' (1993)

$$\mathcal{M}(\mathbf{X}) = \mathcal{M}_0 + \sum_{i=1}^M \mathcal{M}_i(X_i) + \sum_{1 \leq i < j \leq M} \mathcal{M}_{ij}(X_i, X_j) + \dots + \mathcal{M}_{12\dots M}(\mathbf{X}) \quad (8)$$

or equivalently, by

$$\mathcal{M}(\mathbf{X}) = \mathcal{M}_0 + \sum_{\mathbf{u} \neq \emptyset} \mathcal{M}_{\mathbf{u}}(\mathbf{X}_{\mathbf{u}}), \quad (9)$$

where \mathcal{M}_0 is the mean value of Y , $\mathbf{u} = \{i_1, \dots, i_s\} \subset \{1, \dots, M\}$ are index sets and $\mathbf{X}_{\mathbf{u}}$ denotes a subvector of \mathbf{X} containing only those components of which the indices belong to \mathbf{u} . The number of summands in the above equations is $2^M - 1$.

The Sobol' decomposition is unique under the condition

$$\int_{\mathcal{D}_{X_k}} \mathcal{M}_{\mathbf{u}}(\mathbf{x}_{\mathbf{u}}) f_{X_k}(x_k) dx_k = 0, \quad \text{if } k \in \mathbf{u}, \quad (10)$$

where \mathcal{D}_{X_k} and $f_{X_k}(x_k)$ respectively denote the support and marginal PDF of X_k . Eq. (10) leads to the orthogonality property

$$\mathbb{E} [\mathcal{M}_{\mathbf{u}}(\mathbf{X}_{\mathbf{u}}) \mathcal{M}_{\mathbf{v}}(\mathbf{X}_{\mathbf{v}})] = 0, \quad \text{if } \mathbf{u} \neq \mathbf{v}. \quad (11)$$

The uniqueness and orthogonality properties allow decomposition of the variance D of Y as

$$D = \text{Var} [\mathcal{M}(\mathbf{X})] = \sum_{\mathbf{u} \neq \emptyset} D_{\mathbf{u}}, \quad (12)$$

where $D_{\mathbf{u}}$ denotes the partial variance

$$D_{\mathbf{u}} = \text{Var} [\mathcal{M}_{\mathbf{u}}(\mathbf{X}_{\mathbf{u}})] = \mathbb{E} [\mathcal{M}_{\mathbf{u}}^2(\mathbf{X}_{\mathbf{u}})]. \quad (13)$$

The Sobol' index $S_{\mathbf{u}}$ is defined as

$$S_{\mathbf{u}} = D_{\mathbf{u}}/D, \quad (14)$$

and describes the amount of the total variance that is due to the uncertainties in the set of input parameters $\mathbf{X}_{\mathbf{u}}$. By definition, $\sum_{\mathbf{u} \neq \emptyset} S_{\mathbf{u}} = 1$. First-order indices, $S_i^{(1)}$, describe the influence of each parameter X_i considered separately, also called *main effects*. Second-order indices, $S_{ij}^{(2)}$, describe influences from pairs of parameters $\{X_i, X_j\}$. Higher-order indices describe combined influences from larger sets of parameters.

The total sensitivity indices, S_i^{Tot} , represent the *total effect* of an input parameter X_i , accounting for its main effect and all interactions with other parameters. They are derived from the sum of all partial sensitivity indices $S_{\mathbf{u}}$ that involve parameter X_i , *i.e.*

$$S_i^{Tot} = \sum_{\mathcal{I}_i} D_{\mathbf{u}}/D, \quad \mathcal{I}_i = \{\mathbf{u} \supset i\}. \quad (15)$$

It follows that $S_i^{Tot} = 1 - S_{\sim i}$, where $S_{\sim i}$ is the sum of all $S_{\mathbf{u}}$ with \mathbf{u} not including i .

Evaluation of the Sobol' indices by Monte Carlo simulation is based on a recursive relationship which requires computing 2^M Monte Carlo integrals involving $\mathcal{M}(\mathbf{X})$. Clearly, this is not affordable when the computational model is a time-consuming algorithmic sequence. On the other hand, when PCE of the quantity of interest are available, Sobol' indices can be obtained analytically at almost no additional computational cost.

3.2 Polynomial chaos expansions

3.2.1 Computation of polynomial chaos expansions

A PCE approximation of $Y = \mathcal{M}(\mathbf{X})$ in Eq. (8) has the form Xiu and Karniadakis (2002)

$$\hat{Y} = \mathcal{M}^{\text{PCE}}(\mathbf{X}) = \sum_{\alpha \in \mathcal{A}} y_{\alpha} \Psi_{\alpha}(\mathbf{X}), \quad (16)$$

where $\{\Psi_{\alpha}, \alpha \in \mathcal{A}\}$ is a set of multivariate polynomials that are orthonormal with respect to $f_{\mathbf{X}}$, with multi-indices $\alpha = (\alpha_1, \dots, \alpha_M)$, and y_{α} denotes the corresponding polynomial coefficients.

The multivariate polynomials that comprise the PCE basis are obtained by tensorization of appropriate univariate polynomials, *i.e.*

$$\Psi_{\alpha}(\mathbf{X}) = \prod_{i=1}^M \psi_{\alpha_i}^{(i)}(X_i), \quad (17)$$

where $\psi_{\alpha_i}^{(i)}(X_i)$ is a polynomial of degree α_i in the i -th input variable belonging to a family of polynomials that are orthonormal with respect to f_{X_i} . For standard distributions, the associated family of orthonormal polynomials is well-known (*e.g.* a uniform variable with support $[-1, 1]$ is associated with the family of Legendre polynomials), whereas a general case can be treated through an isoprobabilistic transform of \mathbf{X} to a basic random vector. The set of multi-indices \mathcal{A} in Eq. (16) is determined by an appropriate truncation scheme. In the present study, a hyperbolic truncation scheme is employed, which corresponds to selecting all multi-indices satisfying

$$\|\alpha\|_q = \left(\sum_{i=1}^M \alpha_i^q \right)^{1/q} \leq p. \quad (18)$$

for appropriate $0 < q \leq 1$ and $p \in \mathbb{N}$ Blatman and Sudret (2010b).

Once the basis has been specified, the set of coefficients $\mathbf{y} = \{y_\alpha, \alpha \in \mathcal{A}\}$ may be computed by minimizing the mean-square error of the approximation over a set of realizations of the input vector, $\mathcal{E} = \{\mathbf{x}^{(1)}, \dots, \mathbf{x}^{(N)}\}$, called *experimental design*. Efficient solution schemes are obtained by considering the regularized problem

$$\mathbf{y} = \arg \min_{\mathbf{v} \in \mathbb{R}^{\text{card} \mathcal{A}}} \sum_{i=1}^N \left(\mathcal{M}(\mathbf{x}^{(i)}) - \sum_{\alpha \in \mathcal{A}} v_\alpha \Psi_\alpha(\mathbf{x}^{(i)}) \right)^2 + C \|\mathbf{v}\|_1^2, \quad (19)$$

where $\|\mathbf{v}\|_1 = \sum_{j=1}^{\text{card} \mathcal{A}} |v_j|$ and C is a non-negative constant. A nice feature of the above regularized problem is that it provides a *sparse* meta-model by disregarding insignificant terms from the set of predictors. In the present application, we solve Eq. (19) using the *hybrid Least Angle Regression* (LAR) method as originally proposed in Blatman and Sudret (2011). Hybrid LAR employs the LAR algorithm Efron et al. (2004) to select the best set of predictors and subsequently, estimates the coefficients with standard least-squares minimization.

3.2.2 Error estimates

A good measure of the accuracy of PCE is the mean-square error of the residual, $Err_G = \mathbb{E} \left[\left(Y - \widehat{Y} \right)^2 \right]$, called *generalization error*. In practice, this could be estimated by Monte Carlo simulation using a sufficiently large set of realizations of the input vector, $\mathcal{X}_{\text{val}} = \{\mathbf{x}_1, \dots, \mathbf{x}_{n_{\text{val}}}\}$, called *validation set*. The estimate of the generalization error is given by

$$\widehat{Err}_G = \frac{1}{n_{\text{val}}} \sum_{i=1}^{n_{\text{val}}} \left(\mathcal{M}(\mathbf{x}_i) - \sum_{\alpha \in \mathcal{A}} y_\alpha \Psi_\alpha(\mathbf{x}_i) \right)^2. \quad (20)$$

The relative generalization error, \widehat{err}_G , is estimated by normalizing \widehat{Err}_G with the empirical variance of $\mathcal{Y}_{\text{val}} = \{\mathcal{M}(\mathbf{x}_1), \dots, \mathcal{M}(\mathbf{x}_{n_{\text{val}}})\}$.

However, PCE are typically used as surrogate models in cases when evaluating a large number of model responses is not affordable. It is thus desirable to get an error estimate of Err_G using only the information obtained from the experimental design. One such error measure is the *Leave-One-Out* (LOO) *error* Allen (1971). The idea of the LOO cross-validation is to set apart one point of the experimental design, say $\mathbf{x}^{(i)}$, and use the remaining points to build the PCE, denoted $\mathcal{M}^{\text{PCE} \setminus i}$. The LOO error is obtained after alternating over all points of the experimental design, *i.e.*

$$\widehat{Err}_{LOO} = \frac{1}{N} \sum_{i=1}^N (\mathcal{M}(\mathbf{x}^{(i)}) - \mathcal{M}^{\text{PCE} \setminus i}(\mathbf{x}^{(i)}))^2. \quad (21)$$

Although the above definition outlines a computationally demanding procedure, algebraic manipulations allow evaluation of the LOO error from a *single* PCE based on the full experimental design. Let us denote by h_i the i -th diagonal term of matrix $\Psi(\Psi^T\Psi)^{-1}\Psi^T$, where $\Psi = \{\Psi_{ij} = \Psi_j(\mathbf{x}^{(i)}), i = 1, \dots, N; j = 1, \dots, \text{card } \mathcal{A}\}$. Then, the LOO error can be computed as

$$\widehat{Err}_{LOO} = \frac{1}{N} \sum_{i=1}^N \left(\frac{\mathcal{M}(\mathbf{x}^{(i)}) - \mathcal{M}^{\text{PCE}}(\mathbf{x}^{(i)})}{1 - h_i} \right). \quad (22)$$

The relative LOO error, \widehat{err}_{LOO} , is obtained by normalizing \widehat{Err}_{LOO} with the empirical variance of $\mathcal{Y} = \{\mathcal{M}(\mathbf{x}^{(i)}), \dots, \mathcal{M}(\mathbf{x}^{(N)})\}$. Because this error estimate may be too optimistic, we subsequently employ a corrected estimate, given by Chapelle et al. (2002)

$$\widehat{err}_{LOO}^* = \widehat{err}_{LOO} \left(1 - \frac{\text{card } \mathcal{A}}{N} \right)^{-1} (1 + \text{tr}((\Psi^T\Psi)^{-1})). \quad (23)$$

This corrected LOO error is a good compromise between fair error estimation and affordable computational cost.

3.3 Sobol' indices from polynomial chaos expansions

Let us consider the PCE $\widehat{Y} = \mathcal{M}^{\text{PCE}}(\mathbf{X})$ of the quantity of interest $Y = \mathcal{M}(\mathbf{X})$. It is straightforward to obtain the Sobol' decomposition of \widehat{Y} in an analytical form by observing that the summands $\mathcal{M}_{\mathbf{u}}^{\text{PCE}}(\mathbf{X}_{\mathbf{u}})$ in Eq. (9) can be written as

$$\mathcal{M}_{\mathbf{u}}^{\text{PCE}}(\mathbf{X}_{\mathbf{u}}) = \sum_{\alpha \in \mathcal{A}_{\mathbf{u}}} y_{\alpha} \Psi_{\alpha}, \quad (24)$$

where $\mathcal{A}_{\mathbf{u}}$ denotes the set of multi-indices that depend *only on* \mathbf{u} , *i.e.*

$$\mathcal{A}_{\mathbf{u}} = \{\alpha \in \mathcal{A} : \alpha_k \neq 0 \text{ if and only if } k \in \mathbf{u}\}. \quad (25)$$

Clearly, $\bigcup \mathcal{A}_{\mathbf{u}} = \mathcal{A}$. Consequently, due to the orthogonality of the PCE basis, the partial variance $D_{\mathbf{u}}$ reduces to

$$D_{\mathbf{u}} = \text{Var} [\mathcal{M}_{\mathbf{u}}^{\text{PCE}}(\mathbf{X}_{\mathbf{u}})] = \sum_{\alpha \in \mathcal{A}_{\mathbf{u}}} y_{\alpha}^2, \quad (26)$$

whereas the total variance reads

$$D = \text{Var} [\mathcal{M}^{\text{PCE}}(\mathbf{X})] = \sum_{\alpha \in \mathcal{A}} y_{\alpha}^2. \quad (27)$$

Accordingly, the Sobol' indices of any order can be obtained by a mere combination of the squares of the PCE coefficients. For instance, the first-order Sobol' indices are given by

$$S_i^{(1)} = \sum_{\alpha \in \mathcal{A}_i} y_{\alpha}^2 / D, \quad \mathcal{A}_i = \{\alpha \in \mathcal{A} : \alpha_i > 0, \alpha_{i \neq j} = 0\}, \quad (28)$$

whereas the total Sobol' indices are given by

$$S_i^{Tot} = \sum_{\alpha \in \mathcal{A}_i^{Tot}} y_{\alpha}^2 / D, \quad \mathcal{A}_i^{Tot} = \{\alpha \in \mathcal{A} : \alpha_i > 0\}. \quad (29)$$

It is evident that once a PCE representation of $Y = \mathcal{M}(\mathbf{X})$ is available, the complete list of Sobol' indices can be obtained at a nearly costless post-processing of the PCE coefficients requiring only elementary mathematical operations.

4 Results and discussion

Figure 3 provides an overview of the distribution of the mean lifetime expectancy (MLE) throughout the entire model in the nominal case. In this case, the parameters are distributed homogeneously in each of the 15 layers; K_x and ϕ take on the values given in Table 2 for each layer, whereas for all layers the anisotropy ratio is $A_K = 0.1$, the Euler angle is $\theta = 0$ degree, the longitudinal component of the macro-dispersion tensor is $\alpha_L = 15\text{m}$ and the anisotropy ratio is $A_{\alpha} = 0.1$. The hydraulic gradients follow the boundary conditions settings described in Section 2.3.

Because of its highly confining properties, the middle layer (C2) presents values of MLE $> 40,000$ years. On average, it takes approximately 75,000 years for a solute departing from the target zone (TZ) to reach any outlet of the model. Much lower MLE values are found in the two aquifer sequences, with the Oxfordian displaying slightly smaller values. The effect of conductive layers is clearly distinguishable as fringes of low MLE values stretch in layers D4, L1a and L1b in particular. As a result of the low permeability in the top two layers (K3 and K1-K2) and the bottom layer (T), water molecules can take more than a 100,000 years to flow through the model.

In the following, we compute the PCE-based Sobol' indices for the MLE at the TZ by implementing the theory presented in Section 3. We conduct the analysis in two stages: we begin with a simplified description of the input by accounting for the uncertainty in the petrofacies only (case 1); in the sequel, we consider a higher-dimensional random input encompassing the entirety

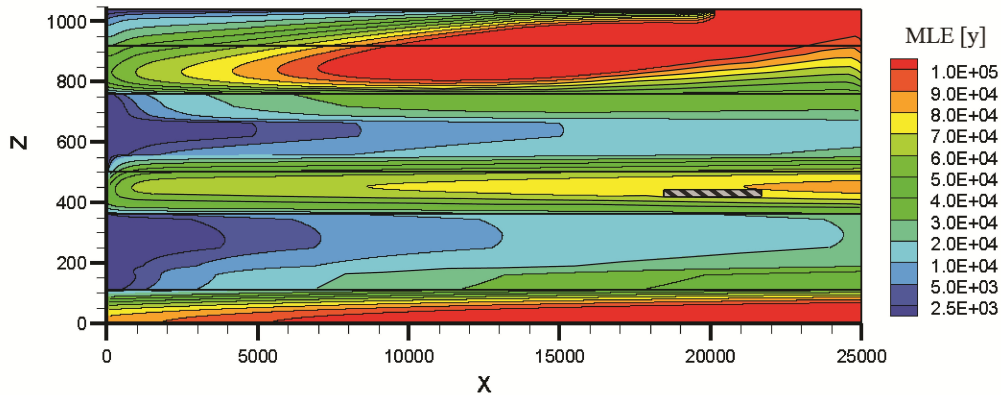


Figure 3: Spatial distribution of mean lifetime expectancy in the reference case (vertical exaggeration: 10).

of hydro-dispersive parameters described in Section 2 as well as the flow BC (case 2).

Note that the term *porosity*, ϕ , is construed in the discussion of Sobol' indices. Since the values for the hydraulic conductivities are retrieved through approximation functions, the estimation of the sensitivity for the ϕ variables is implicitly associated to that of the \hat{K}_x variables in the respective layers. This is singularly important when interpreting the Sobol' indices for aquifer formations where the hydraulic conductivity governs the ageing process (see Section 4.3).

Computations of the PCE and Sobol' indices are performed with the uncertainty quantification software UQLab (Marelli and Sudret (2014)).

4.1 Case 1: 15 input random variables

In the first case, the uncertain input comprises the petrofacies P in each of the 15 layers of the hydrogeological model. As stated before, a deterministic relationship $\log_{10}(\hat{K}_x) = f(\phi)$ is assumed for each layer, *i.e.* the uncertainty regarding the petrofacies P of a layer is treated through the porosity ϕ , resulting in a random input vector of dimension $M = 15$. The uncertain porosities are modeled as *independent* uniform random variables, each bounded by the values $\phi^{(min)}$ and $\phi^{(max)}$ listed in Table 3 together with the respective coefficients of variation (CoV); the porosity ranges along the model cross-section are shown graphically in Figure 4. The bounds $\phi^{(min)}$ and $\phi^{(max)}$ represent the 1st and 9th deciles of the corresponding CDF derived from porosity values measured in each geological layer. This approach is justified by the presence

of local extreme measures that cannot be representative for the whole layer. Bounds for the \widehat{K}_x parameters are also provided in Table 3, consistently with the $\log_{10}(\widehat{K}_x) = f(\phi)$ approximation functions, and presented graphically in Figure 5.

Table 3: Ranges of porosity ϕ with the respective CoV and the estimated permeability values \widehat{K} in the 15 geological layers.

| Layer | $\phi^{(min)}$ [-] | $\phi^{(max)}$ [-] | CoV | $\widehat{K}_x^{(min)}$ [m/s] | $\widehat{K}_x^{(max)}$ [m/s] |
|-------|--------------------|--------------------|--------|-------------------------------|-------------------------------|
| K3 | 0.0840 | 0.1160 | 0.0924 | 3.3734e-10 | 2.4078e-07 |
| K1-K2 | 0.0870 | 0.1430 | 0.1406 | 9.8116e-11 | 2.0928e-07 |
| L2c | 0.1019 | 0.1759 | 0.1538 | 3.6186e-08 | 2.6212e-06 |
| L2b | 0.0645 | 0.1574 | 0.2417 | 8.7318e-10 | 6.3950e-06 |
| L2a | 0.0651 | 0.1627 | 0.2474 | 4.7005e-10 | 9.9336e-06 |
| L1b | 0.1375 | 0.1833 | 0.0824 | 3.4324e-09 | 2.8913e-04 |
| L1a | 0.0991 | 0.2107 | 0.2080 | 3.1165e-08 | 2.1523e-06 |
| C3ab | 0.0747 | 0.1221 | 0.1391 | 7.8488e-09 | 1.2945e-06 |
| C2 | 0.1284 | 0.1876 | 0.1082 | 5.0349e-14 | 6.2570e-13 |
| C1 | 0.0142 | 0.0799 | 0.4031 | 1.8184e-07 | 1.6195e-05 |
| D4 | 0.0237 | 0.1573 | 0.4262 | 1.6408e-07 | 3.1521e-03 |
| D3 | 0.0237 | 0.1795 | 0.4427 | 1.7470e-07 | 4.3539e-06 |
| D2 | 0.0185 | 0.1061 | 0.4059 | 6.6071e-08 | 1.7049e-06 |
| D1 | 0.0191 | 0.1186 | 0.4172 | 6.2552e-08 | 1.8425e-05 |
| T | 0.0696 | 0.0925 | 0.0816 | 1.2325e-13 | 8.1328e-12 |

4.1.1 PCE

To build PCE of the MLE in terms of the 15 input random variables, we use an experimental design comprising $N = 1,000$ points drawn with Latin hypercube sampling (LHS) McKay et al. (1979). LHS is a popular technique for obtaining random experimental designs ensuring uniformity of each sample on the margin input variables $\{X_1, \dots, X_M\}$. A histogram of the model response at the considered LHS design is shown in Figure 6, indicating a positively skewed distribution with the mode situated at $\text{MLE} \approx 75,000$ years.

We develop two PCE meta-models, respectively denoted 1A and 1B, by applying the procedure in Section 3.2.1, first, on the original scale and then, on the logarithmic transform of the MLE. For both PCE, the candidate basis is determined using a hyperbolic truncation scheme (see Eq. (18)) with $q = 0.5$. The maximum degree p is varied from 1 to 15 and the optimal sparse PCE is selected by means of the corrected relative LOO error (see Eq. (23)), simply called LOO error and denoted err_{LOO} hereafter.

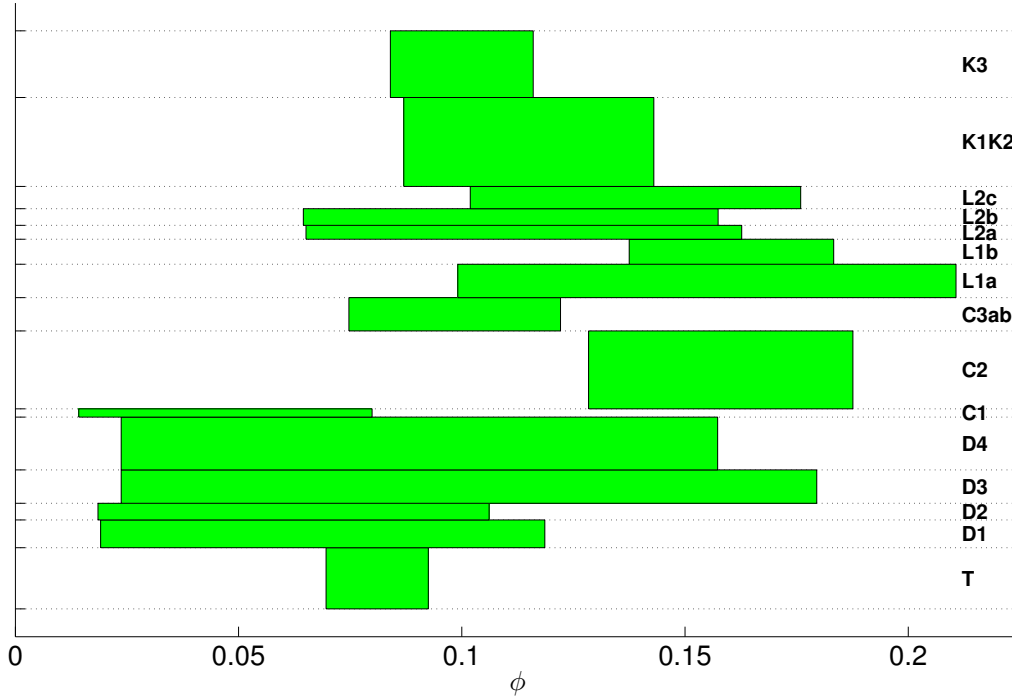


Figure 4: Porosity ranges along the model cross-section.

When the non-transformed response is considered, the optimal PCE 1A is obtained for $p = 10$ and the corresponding LOO error is $err_{LOO} = 0.0321$. The sparse meta-model includes 166 basis elements, whereas the total number of basis elements for $p = 10$ and $q = 0.5$ (resp. $q = 1$) is 1,656 (resp. 3.2×10^6). When the logarithmic response is considered, the optimal PCE 1B is obtained for $p = 13$ and the corresponding LOO error is $err_{LOO} = 0.0120$. In this case, the sparse meta-model includes 245 basis elements, whereas the total number of basis elements for $p = 13$ and $q = 0.5$ (resp. $q = 1$) is 3,801 (resp. 3.7×10^7). For both PCE, the sparse bases involve polynomials in all 15 input variables. The index of sparsity, defined as the number of basis elements in the sparse expansion divided by the size of a full basis ($q = 1$) with the same maximum degree, is $166/3.2 \times 10^6 \approx 5.2 \times 10^{-5}$ for case 1A and $245/3.7 \times 10^7 \approx 6.6 \times 10^{-6}$ for case 1B. These numbers indicate the interest in developing sparse PCE for such analyses.

The left graph of Figure 7 compares the values of PCE 1A, \hat{Y} , with the respective values of the exact model, Y , at the input samples of the experimental design; the right graph shows a similar comparison for the exponential transform of PCE 1B. Assessing the relative accuracy of the two meta-models, first, we note that 1B yields a smaller LOO error; furthermore, it results in

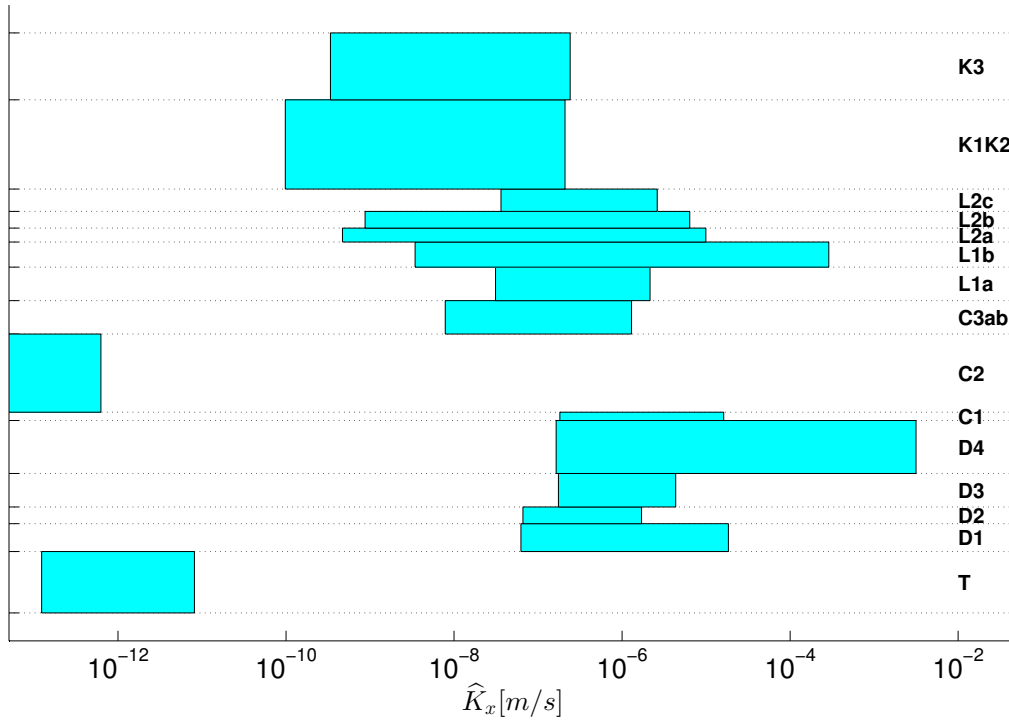


Figure 5: Hydraulic conductivity ranges along the model cross-section.

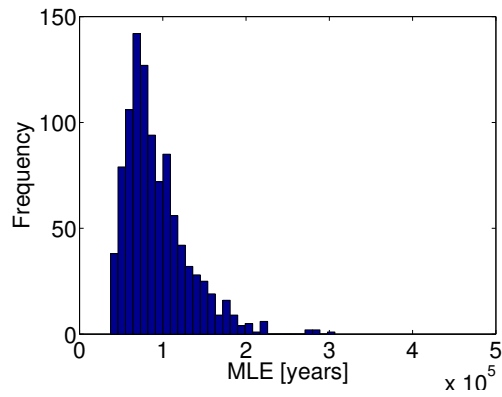


Figure 6: Histogram of mean lifetime expectancy values.

a smaller dispersion of $\hat{Y} - Y$ around zero and a better approximation of the exact response at the upper tail of its distribution. However, we should bear in mind that in the subsequent SA, the Sobol' indices obtained from the coefficients of PCE 1B represent contributions to the variance of the *logarithmic* MLE.

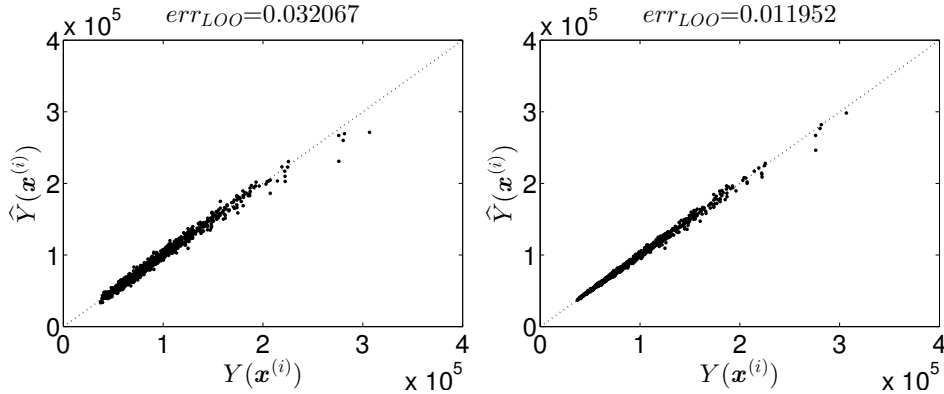


Figure 7: Comparison of PCE 1A (left) and the exponential transform of PCE 1B (right) with the actual model response at the experimental design.

4.1.2 Sobol' indices

Figures 8 and 9 show bar-plots of the first-order and total Sobol' indices of the porosities at the 15 layers using PCE 1A and 1B, respectively. To identify unimportant effects, the threshold of 0.01 is marked with a horizontal dashed line.

Both figures indicate that the variability in the MLE is mainly due to main effects and interactions associated with layers D4, L1b, C3ab and L1a in order of importance in terms of total effects, with layer D4 being clearly dominant. All aforementioned layers are located close to the host layer C2; D4 is the thickest among those and has the highest hydraulic conductivity. The condition $S_i^{Tot} > 0.01$ additionally classifies as important layers C1 and D1 for both PCE 1A and 1B, and marginally layers D3 and L2b for PCE 1A only. Note that although C1 is adjacent to the host layer C2, it is associated with smaller total and first-order indices than other neighboring layers, which may be attributed to its small thickness.

To gain further insight into the effects of the important variables on the model response, we examine the behavior of first-order summands comprising univariate polynomials only, *i.e.*

$$\mathcal{M}_i^{(1)}(x_i) = \mathbb{E} [\mathcal{M}(\mathbf{X} | X_i = x_i)] \quad (30)$$

or equivalently

$$\mathcal{M}_i^{(1)}(x_i) = \sum_{\alpha \in \mathcal{A}_i} y_\alpha \Psi_\alpha(x_i), \quad \mathcal{A}_i = \{\alpha \in \mathcal{A} : \alpha_i > 0, \alpha_{i \neq j} = 0\}. \quad (31)$$

Figures 10 and 11 depict such univariate effects considering PCE 1A and 1B, respectively, for the porosities at the six layers classified as important

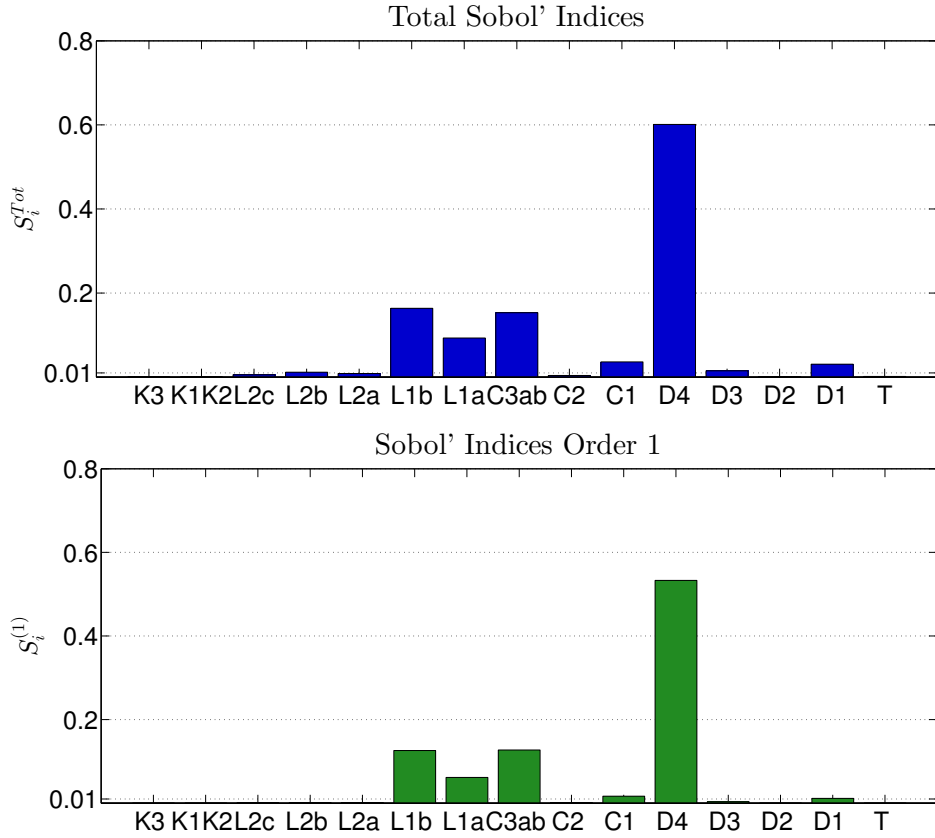


Figure 8: Sobol' indices using PCE 1A.

with both meta-models. The scales in the vertical axes in the two figures are different due to consideration of the non-transformed response in one and the logarithmic response in the other. The figures demonstrate that an increasing porosity, and thus an increasing permeability, is associated with a decreasing algebraic contribution to the MLE value, though this relationship is not strictly monotonic for all layers. Despite the different scales, the shapes of respective curves in the two figures demonstrate similar trends. However, we note the presence of higher-order terms for PCE 1B, which is consistent with its higher degree p and lower sparsity as compared to 1A. For both PCE, the sum in Eq. (31) tends to include more higher-order terms for the important variables.

For model 1A, main and second-order effects respectively account for 88.5% and 10.3% of the response variance, indicating that the contribution of higher-order effects is merely 1.2%. For model 1B, main and second-order effects respectively account for 94.7% and 4.8% of the response variance, indicating that the contribution of higher-order effects is smaller than 1.0%.

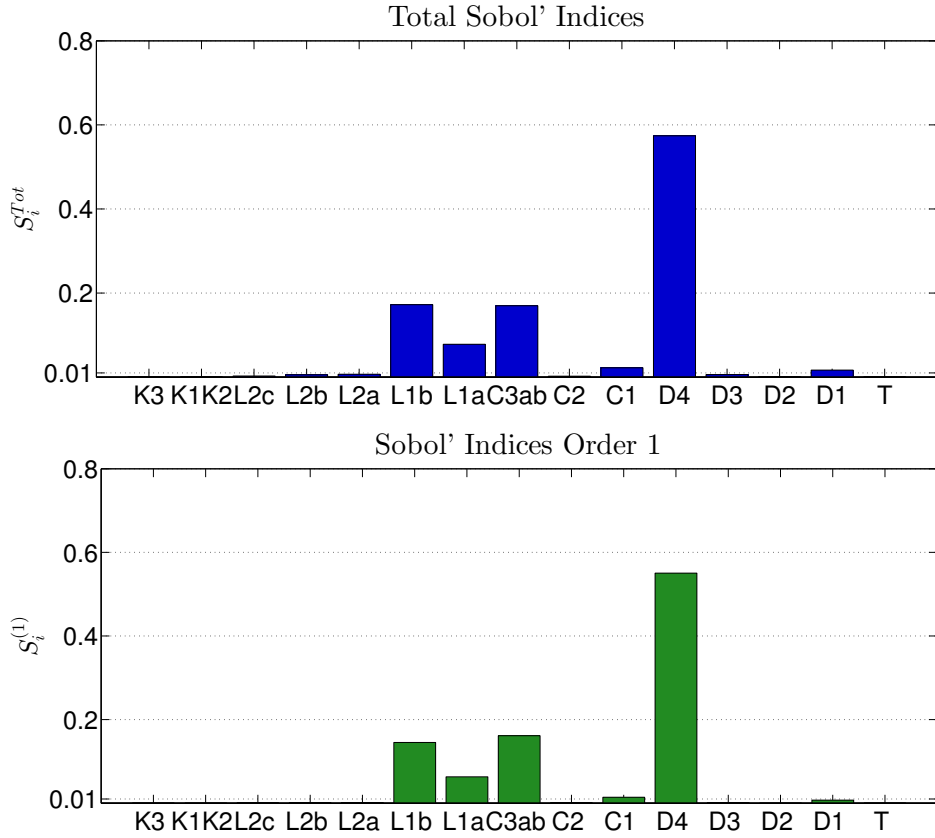


Figure 9: Sobol' indices using PCE 1B.

For both models, the highest second-order indices involve one of the layers D4 or L1b. Overall, effects from interactions are slightly more significant for the variance of MLE in the original scale.

4.2 Case 2: 78 input random variables

In case 2, we employ a more detailed description of the input uncertainty. In addition to the petrofacies, the uncertain input in each layer comprises the following four parameters governing the advective-dispersive processes: the anisotropy in the components of the hydraulic conductivity tensor, A_K ; the Euler angle of the hydraulic conductivity tensor, θ ; the longitudinal component of the dispersivity tensor, α_L ; and the anisotropy in the longitudinal and vertical components of the dispersivity tensor, A_α . As in case 1, a deterministic relationship $\log_{10}(\tilde{K}_x) = f(\phi)$ is assumed for each layer. Furthermore, the uncertain input includes the hydraulic gradients, ∇H , at three zones: the Dogger sequence, the Oxfordian sequence and the top of the model,

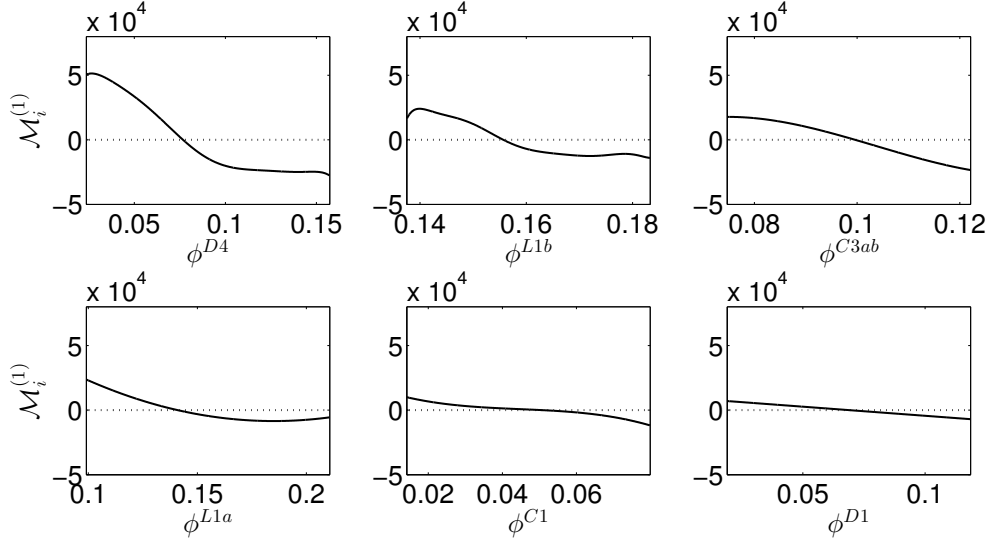


Figure 10: Univariate effects of important variables using PCE 1A.

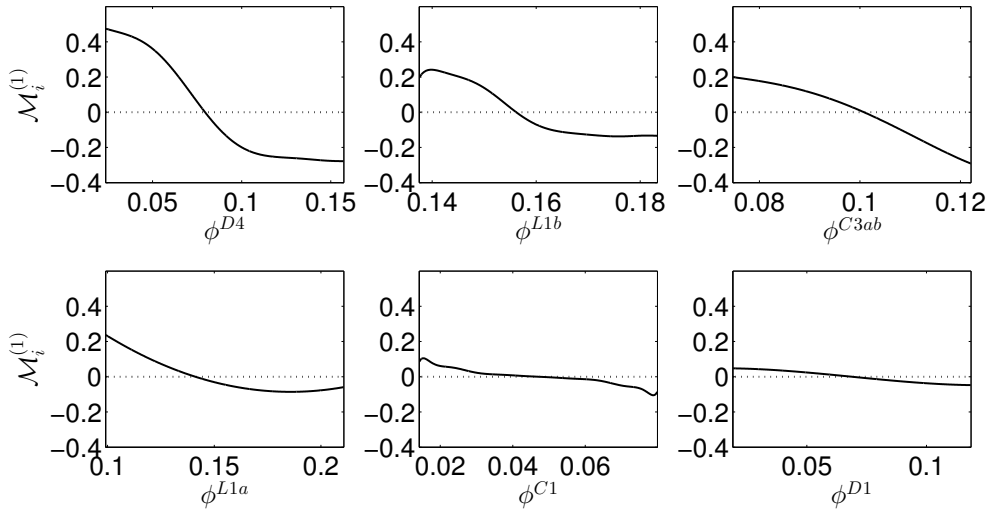


Figure 11: Univariate effects of important variables using PCE 1B.

respectively denoted zone 1, zone 2 and zone 3. In total, the model subsumes $M = 78$ independent input random variables upon the MLE of water molecules outflowing from the TZ in the middle of layer C2.

The uncertain porosities, and the associated hydraulic conductivities, are modeled as in case 1. Due to the lack of explicit information, each of the parameters A_K , θ , α_L and A_α is identically distributed in the different layers. In particular, the anisotropy ratios A_K and A_α both follow a uniform distri-

bution in $[0.01, 1]$, the Euler angle θ is uniformly distributed in $[-30, 30]$ (in degrees) and the parameter α_L is uniformly distributed in $[5, 25]$ (in meters). The static hydraulic gradients are also uniformly distributed in the ranges given in Table 4. These were obtained by applying a perturbation of 20% of the nominal hydraulic gradient within each of the three zones. Although hypothetical, these conservative uncertainty ranges were purposely chosen to provide insights into the behavior of the multi-layered model.

Table 4: Ranges of hydraulic gradient at the three zones of interest.

| Zone number | $\nabla H^{(min)}$ | $\nabla H^{(max)}$ |
|-------------|--------------------|--------------------|
| 1 | 0.00064 | 0.00096 |
| 2 | 0.00240 | 0.00360 |
| 3 | 0.00272 | 0.00408 |

4.2.1 PCE

In this case, the available data for building PCE consist of the MLE values for (i) an experimental design of size $N = 2,000$, drawn with LHS and denoted \mathcal{E} , and (ii) an enrichment of \mathcal{E} of equal size $N' = 2,000$, denoted \mathcal{E}' . The enrichment is built so that the joint set $\{\mathcal{E}, \mathcal{E}'\}$ forms a LHS experimental design as well. Histograms of the model response for the two sets of input vectors are shown in Figure 12. Positively skewed distributions are observed for both output sets, while modes are situated at $\text{MLE} \approx 85,000$ years.

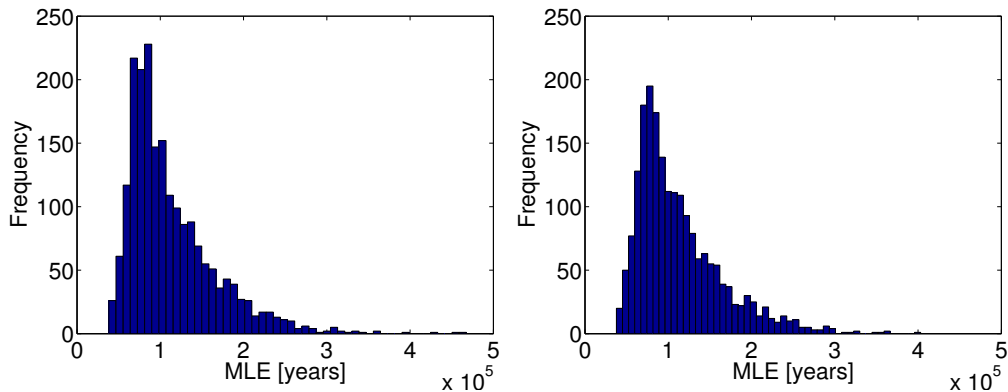


Figure 12: Histograms of mean lifetime expectancy values calculated with sets \mathcal{E} (left) and \mathcal{E}' (right).

In the following, we develop PCE based on \mathcal{E} and the joint set $\{\mathcal{E}, \mathcal{E}'\}$. For the set \mathcal{E} , we consider the MLE response in both the original and the

logarithmic scales; in this case, the enrichment \mathcal{E}' serves as a validation set for computing the generalization error (see Section 3.2.2). For all PCE, the candidate basis is determined using a hyperbolic truncation scheme with $q = 0.5$. The maximum degree p is varied from 1 to 15 and the optimal sparse meta-model is selected by means of the LOO error.

The first PCE, denoted 2A, is built using \mathcal{E} as the experimental design and considering the response in the original scale. The optimal degree is $p = 8$ and the corresponding LOO error is $err_{LOO} = 0.0565$. The sparse PCE includes 185 basis elements, whereas the total number of basis elements for $p = 8$ and $q = 0.5$ (resp. $q = 1$) is 18,643 (resp. 5.3×10^{10}). Thus, the index of sparsity is $185/5.3 \times 10^{10} \approx 3.5 \times 10^{-9}$. The sparse basis consists of polynomials in 68 out of the 78 total input parameters, meaning that the output does not depend at all on the values of the 10 excluded parameters. Note that 3 out of the 10 excluded parameters are properties of layer T. The generalization error (evaluated with \mathcal{E}') is $err_G = 0.0759$. The left graph of Figure 13 compares the values of the meta-model, \hat{Y} , with the respective values of the exact model, Y , at the input samples of the experimental design, \mathcal{E} . A similar comparison but for the validation set, \mathcal{E}' , is shown in the right graph of the same figure.

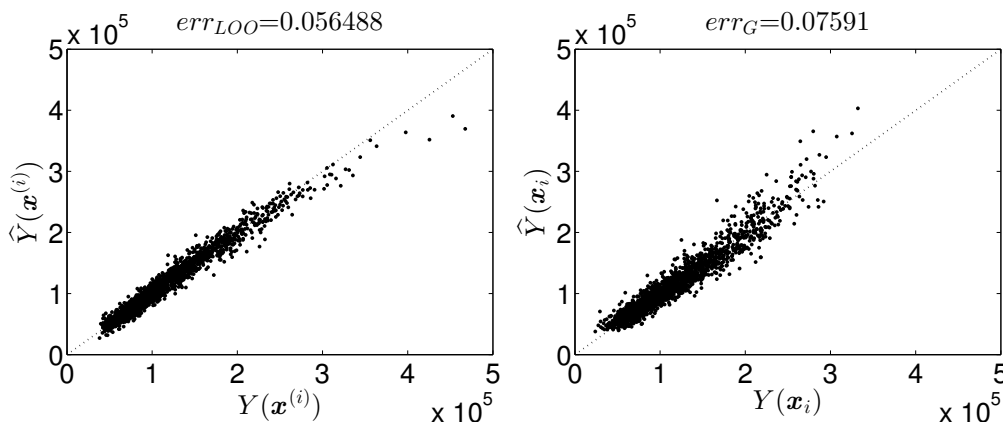


Figure 13: Comparison of PCE 2A with the actual model response at the experimental design, \mathcal{E} , (left) and at the validation set, \mathcal{E}' (right).

A second PCE, denoted 2B, is built by using again \mathcal{E} as the experimental design, but employing a logarithmic transform of the MLE. The optimal sparse PCE is obtained for $p = 8$ (same degree as for 2A) and comprises 163 basis elements; the corresponding LOO error is $err_{LOO} = 0.0287$. The sparse basis consists of polynomials in 65 out of the 78 total input parameters; note that 6 out of the 13 excluded parameters are dispersivity anisotropy ratios (A_α). The resulting generalization error for the MLE response in the

original scale (considering the exponential transform of the obtained PCE) is $err_G = 0.0452$, which is slightly lower than that of PCE 2A. The left and right graphs of Figure 14 compare the exponential transform of PCE 2B with the exact model response at \mathcal{E} and \mathcal{E}' , respectively.

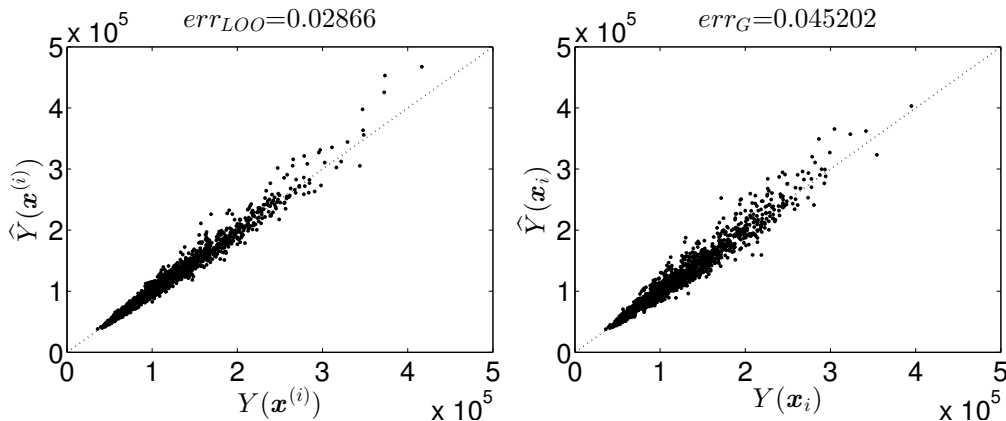


Figure 14: Comparison of exponential transform of PCE 2B with the actual model response at the experimental design, \mathcal{E} , (left) and at the validation set, \mathcal{E}' (right).

Finally, we use as experimental design the joint set $\{\mathcal{E}, \mathcal{E}'\}$ consisting of $N + N' = 4,000$ points. The optimal PCE is obtained for $p = 10$ and the corresponding LOO error is $err_{LOO} = 0.0384$. The sparse PCE comprises 312 basis elements and has an index of sparsity $312/4.5 \times 10^{12} \approx 6.9 \times 10^{-11}$. The only two parameters excluded from the sparse basis are A_K^{L2b} and α_L^T . The comparison between the meta-model, denoted 2C, and the exact model response at the input samples of the experimental design, $\{\mathcal{E}, \mathcal{E}'\}$, is shown in Figure 15.

Assessing the relative accuracies of the three meta-models, we note that all have LOO errors of the same order of magnitude, with the smallest error corresponding to PCE 1B. Because it is of interest to limit the number of costly evaluations of the exact hydrogeological model, an experimental design comprising 2,000 points is deemed most appropriate. We therefore conduct SA for the MLE response by post-processing the coefficients of PCE 2A and 2B and compare the results. We should bear in mind that the Sobol' indices obtained from the coefficients of 2B represent contributions to the variance of the *logarithmic* MLE.

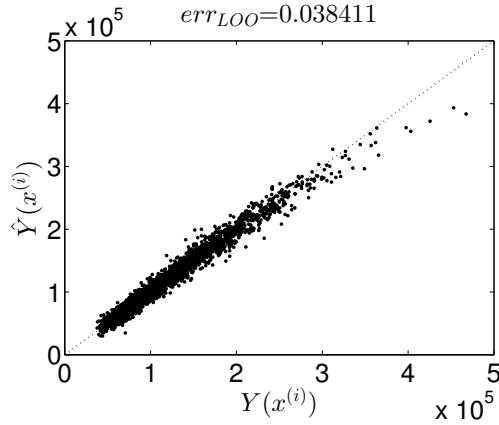


Figure 15: Comparison of PCE 2C with the actual model response at the experimental design, $\{\mathcal{E}, \mathcal{E}'\}$.

4.2.2 Sobol' indices

Figures 16 and 17 show bar-plots of the total and first-order Sobol' indices for PCE 2A and 2B, respectively. In each graph, bar-plots of the ten largest indices are presented in descending order. The superscripts on the parameter symbols on the horizontal axes denote layer names or zone numbers. To identify unimportant effects, the threshold of 0.01 is marked with a horizontal dashed line.

A first observation is that both PCE lead to the same ranking of the five variables with the highest total indices. These are the porosities of layers D4, C3ab, L1b, L1a and C1 in order of importance, with the porosities of layer D4 being the by-far dominant. For both PCE, these are also the variables with the highest first-order indices following the same ranking. Note that PCE 1A and 1B identified the same five layers as most significant in terms of both total and main effects, which indicates the consistency of the analyses in cases 1 and 2. However, the ranking of layers C3ab and L1b in terms of the total indices is interchanged in the two cases; also, layers D4 and C1 have slightly higher contributions in case 2. Employing the criterion $S_i^{Tot} < 0.01$ to sort out unimportant variables, the porosities of the aforementioned five layers comprise the only important parameters for both 2A and 2B. Univariate effects for these variables follow similar trends as in case 1 and are thus not shown. The screening allows one to consider 73 out of 78 parameters as unimportant, meaning that they could be given a deterministic value without affecting essentially the predicted MLE.

Let us now examine higher-order effects. For both PCE, the largest second-order effects comprise interactions between the five porosities clas-

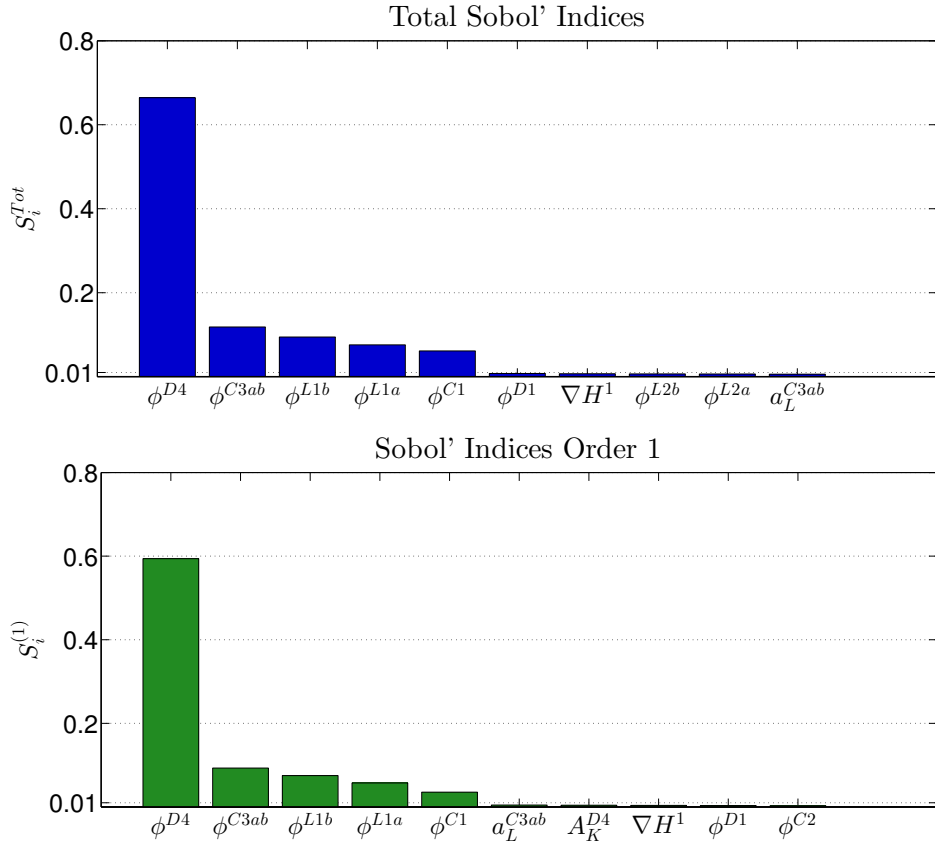


Figure 16: Sobol' indices using PCE 2A.

sified above as important and involve one of the layers D4 or L1b; their sum explains 10.9% and 4.2% of the total variance for PCE 2A and 2B, respectively. Overall, second-order effects are slightly less important for the logarithmic response than for the response in the original scale, which is consistent with the results in case 1. Contributions of higher than second-order effects are practically zero.

The above analysis indicated that among the set of random variables, only the porosities of certain layers are important for explaining the response variance. As highlighted earlier, because of the assumed relationship between porosity and hydraulic conductivity in each layer, the Sobol' indices for the ϕ variables are also indicative of the importance of \widehat{K}_x in the respective layers. Accordingly, in the subsequent discussion of the SA results (Section 4.3), we will interpret the variability of MLE in terms of joint effects of the petrofacies P .

For a more in-depth investigation of the contributions of the different types of hydro-dispersive parameters, Table 5 lists the sums of the first-order

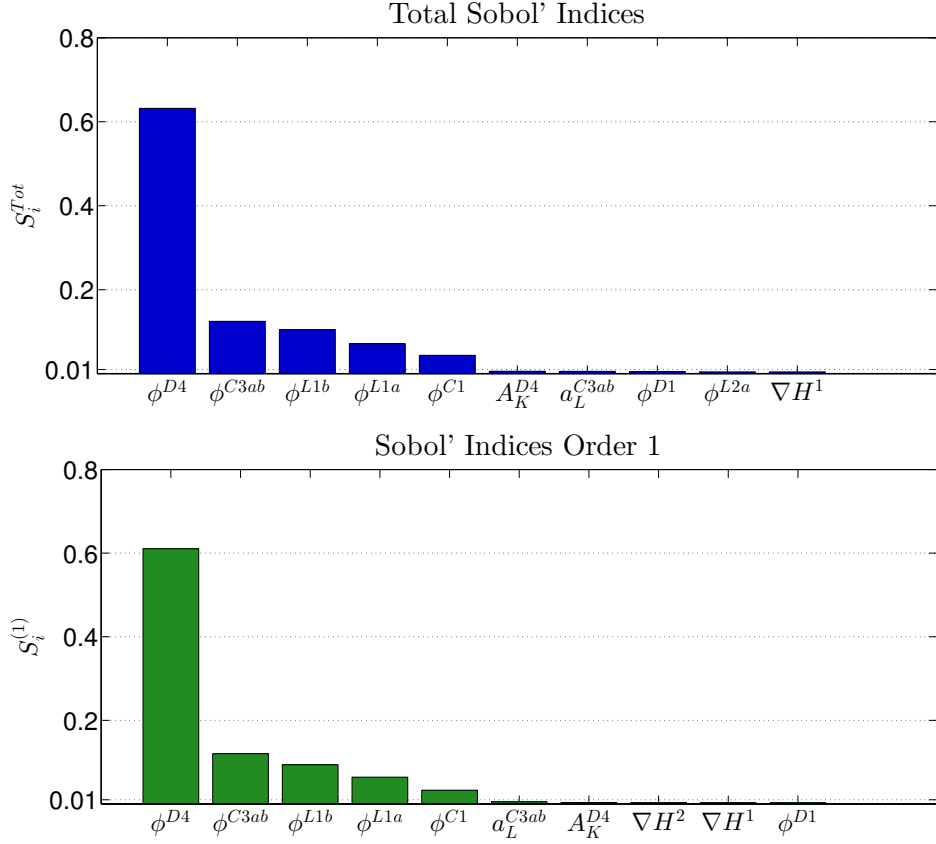


Figure 17: Sobol' indices using PCE 2B.

indices per type of property over all layers or zones of the considered cross-section. According to this table, the added main effects of the porosities account for approximately 87% – 93% of the response variance for the two PCE, whereas the added main effects of all remaining input random variables account for a mere 2.5% – 2.8%. We note the slightly higher contributions of main effects for 2B and the zero main effects of A_α for both PCE.

Table 5: Sums of first-order Sobol' indices over all layers per type of property.

| meta-model | ϕ | A_K | θ | α_L | A_α | ∇H |
|------------|--------|--------|----------|------------|------------|------------|
| 2A | 0.8664 | 0.0088 | 0.0029 | 0.0076 | 0 | 0.0057 |
| 2B | 0.9302 | 0.0096 | 0.0032 | 0.0088 | 0 | 0.0061 |

4.3 Discussion of results

In both cases 1 and 2, the petrofacies of aquifer formations have the largest effects on the variability of the MLE at the TZ. For layers D4 and L1b in particular, for which the upper bounds of the hydraulic conductivity ranges are the highest (see Table 3, Figure 5), strong advective processes within their own volume may reduce the overall groundwater residence time in the model. Besides, the wider the ranges of \widehat{K}_x values in these permeable formations, the higher are the contributions of the respective petrofacies to the variability of the MLE. The position of the layers relatively to layer C2 is also relevant: the further the layer is, the lower is its effect on the MLE of water molecules departing from the TZ. Layer L1a, which is the first aquifer encountered in the Oxfordian sequence (at a distance of 60 meters from layer C2), has a significant effect on the output variance, whereas layers D2 and D3 that have similar \widehat{K}_x ranges have much smaller contributions.

The three most significant aquifer formations, namely D4, L1b and L1a, have rather non-linear univariate effects on the output response (see Figures 10 and 11). Substantial changes within the response are observed with small shifts in the ranges of low porosity-permeability values, revealing the effect of the balance between advective and dispersive-diffusive transport processes. But for higher porosity-permeability values, advective fluxes prevail, thus yielding more linear and moderate changes in the response.

The relatively large uncertainty range and high upper-bound value of permeability for layer D1 results in a marginal effect on the response variability in case 1 (see Figures 8 and 9). The uncertainty in the petrofacies of this layer influences linearly or nearly linearly and rather moderately the transit time of water molecules from the TZ (see Figures 10 and 11). However, accounting for uncertain dispersion processes and groundwater fluxes in case 2 may reduce the quantity of water molecules departing from the TZ and reaching layer D1, thus lessening the contribution of P^{D1} to the variance of the output quantity, which becomes unimportant with respect to the threshold $S_i^{Tot} < 0.01$.

The petrofacies of semi-permeable formations, P^{C3ab} and P^{C1} , are also significantly influencing the variability of the MLE at the TZ. By isolating layer C2 from major aquifer formations, they buffer solute intrusions into the Oxfordian and Dogger aquifer sequences, thus acting like a geological barrier. Figures 10 and 11 show that their univariate effects on the output quantity are relatively non-linear despite their limited amplitude.

We underline that the petrofacies of the host layer attributed to the Callovo-Oxfordian claystone (P^{C2}) are insignificant with regard to the MLE variability. Slow diffusive processes take place into highly impermeable rocks,

which induces large values of the MLE response. Modifying the magnitudes of advective-dispersive transport processes in layer C2 does not add a significant variability to the time required for solutes to leave the layer's volume.

The magnitude of the transverse advective fluxes in each layer is related to the respective value of K_z , in which the uncertainty is accounted for through the anisotropy ratio A_K . Although A_K represents the second most influential group of factors, considering added effects from all layers (see Table 5), the uncertainty in this property adds a small amount of variability to the MLE ($< 1\%$) compared to the petrofacies ($\approx 90\%$). We note however that factor A_K^{D4} is only marginally excluded from the important factors when PCE 2B is considered (see Figure 17). Indeed, for the highest \hat{K}_x values, strong advective fluxes take place within the layer's volume. Under the assumption of strong transverse fluxes ($A_K^{D4} \rightarrow 1$), solutes can be oriented toward neighbouring layers where slower fluxes occur, thus raising the MLE.

For each layer, the Euler angle θ could deviate groundwater fluxes from an orientation parallel to the x -axis and toward the main discharge boundaries, thus raising the variability of the response. Although it could be assumed as especially influential in the most advective layers, the total contribution of this group of uncertain factors to the variance of the MLE is negligible in comparison to that of the petrofacies P (see Table 5).

In aquifer formations, the effects of the uncertainty regarding the macro-dispersion tensors upon the response quantity, *i.e.* the magnitude (α_L) and anisotropy (A_α) of the tensors, are concealed by the strong effect of petrofacies on the advective part of the transport processes (see Table 5). We note however that the longitudinal component of the macro-dispersion tensor in layer C3ab (α_L^{C3ab}) appears among the ten factors with the highest total Sobol' indices for both PCE 2A and 2B. The anisotropy ratios, A_α , have no contribution at all to the response variance when considered independently; the uncertainty in these factors contributes to the variability of MLE only through interaction terms.

The sensitivity of the MLE with respect to flow BCs considered in the model is directly related to the magnitude and orientation of the advective fluxes in the entire model. In the case of high gradients in both limestone sequences, the advective solute transport processes would raise within their volume, and thus reduce the MLE. Table 5 indicates that the three random hydraulic gradients have a small added effect on the output variance. Note however that the total Sobol' index for the hydraulic gradient in the Dogger sequence (∇H^1) belongs to the ten highest indices for both PCE 2A and 2B. The uncertainty regarding this factor can alter the advective processes occurring notably in layer D4, which has the far highest contribution to the

variance of the MLE calculated at the TZ.

5 Conclusions

The model introduced in this paper stands as a two-dimensional vertical cross-section of the subsurface of Paris Basin in the vicinity of Bure (Haute-Marne). While encompassing most of the hydrogeological features of the underground media, it was simplified with regard to geometries, discontinuities, fractures and heterogeneities. This numerical model is intended to explore the behavior of a complex multi-layered hydrogeological system at low computational cost and provide insights into the effect of uncertain parameters upon various types of model output.

Sensitivity analysis (SA) was carried out for two cases of modeling the input uncertainty. For the sake of simplicity, homogeneous parameters were assumed within each of the 15 hydrogeological layers comprising the model in both cases. In case 1, only the petrofacies, P , regarded as the couple permeability-porosity, $\{K, \phi\}$, were considered uncertain. In case 2, the uncertain factors at each layer included four additional hydro-dispersive parameters: the anisotropy ratio and the orientation of the hydraulic conductivity tensor, A_K and θ respectively, the magnitude and anisotropy ratio in the macro-dispersion tensor, α_L and A_α respectively; additionally, the hydraulic gradients, ∇H , in three zones of the numerical model were considered random, summing up to 78 uncertain input factors.

In the present study, a target zone (TZ) located within the middle layer (C2) of the model provides the output response of interest. Latin hypercube sampling was employed to address the propagation of the uncertainty from the input factors upon the mean lifetime expectancy (MLE) of water molecules departing from the TZ. Polynomial chaos expansion (PCE) meta-models were used to compute the Sobol' sensitivity indices for each input factor at low computational costs. Sparse PCE proved particularly efficient in providing fairly accurate representations of the MLE in terms of the considered high-dimensional input. The accuracy was enhanced when the PCE were fitted to the logarithmic MLE.

Focusing on the effects of petrofacies solely, case 1 demonstrated the large contributions of aquifer formations to the variance of the model output. In particular, (i) the closer the aquifer formation to layer C2, (ii) the thicker the layer, (iii) the wider the ranges of the petrofacies and (iv) the higher the upper bound of the hydraulic conductivity range, the larger were the effects of the petrofacies on the variability of the response. Investigation of the univariate effects of petrofacies highlighted that for these permeable

formations, the response is more sensitive to changes occurring within low porosity-permeability ranges. Hence, within a certain range of $\{K, \phi\}$ values, the dispersive-diffusive processes counterbalance with the strong advective fluxes in the ageing process.

The SA results in case 2 showed that the variability of the MLE is almost entirely due to the uncertainty regarding the petrofacies of the hydrogeological layers. The other hydro-dispersive parameters are insignificant for explaining the response variance and may be considered as deterministic factors in future works. SA in cases 1 and 2 were consistent in identifying the layers of which the petrofacies are important.

In formations characterized by highly advective processes, the longitudinal hydraulic conductivities applying in the main groundwater direction have large contributions to the MLE variability. The two semi-confining formations encompassing the C2 layer buffer the transfer of solute from the latter toward the further aquifer sequences. Besides, it is acknowledged that longitudinal dispersion processes occurring within their own volume also retard the solute transfer toward the adjacent aquifers. Because of the diffusion-dominated transport processes occurring within its volume, the petrofacies of the highly-confining C2 layer have a negligible effect on the variance of the output response.

It is important to remind that the use of a 2D model tends to overestimate the output response of interest by omitting the advection and dispersion along the third dimension. Recognizing this limitation, we underline that the purpose of the synthetic model introduced in this study is to shed light on the relative effects of various uncertain factors governing the advective and dispersive processes in a complex multi-layered hydrogeological system. The methods employed in this study can be applied to a real-case study employing a realistic 3D numerical model.

The sensitivity analysis performed in this work is deemed very informative for future applications with the high-resolution integrated Meuse/Haute-Marne hydrogeological model. In the frame of a real-case uncertainty analysis with concern to a solute transport in the subsurface of the Paris Basin, the authors recommend defining as thoroughly as possible the spatial distributions of hydraulic conductivity values, with a main focus on the large aquifer sequences of Oxfordian and Dogger ages.

Acknowledgement

The authors are very grateful to Mr Benjamin Brigaud (Université Paris-Sud) and Mrs Agnès Vinsot (ANDRA) for having provided the hydro-dispersive datasets used in this analysis, and to Mr Fabien Cornaton (DHI-WASY GmbH) for providing the code *GroundWater*.

References

- Renard, P.. Stochastic hydrogeology: What professionals really need? *Ground Water* 2007;45(5):531–541.
- Saltelli, A., Ratto, M., Andres, T., Campolongo, F., Cariboni, J., Gatelli, D., et al. *Global Sensitivity Analysis – The Primer*. Wiley; 2008.
- Sobol', I.M.. Sensitivity estimates for nonlinear mathematical models. *Math Modeling & Comp Exp* 1993;1:407–414.
- Archer, G., Saltelli, A., Sobol', I.. Sensitivity measures, ANOVA-like techniques and the use of bootstrap. *J Stat Comput Simul* 1997;58:99–120.
- Sobol', I.. Global sensitivity indices for nonlinear mathematical models and their Monte Carlo estimates. *Mathematics and Computers in Simulation* 2001;55(1-3):271–280.
- Saltelli, A.. Making best use of model evaluations to compute sensitivity indices. *Comput Phys Comm* 2002;145:280–297.
- Sobol', I., Kucherenko, S.. Global sensitivity indices for nonlinear mathematical models. *Review. Wilmott magazine* 2005;1:56–61.
- Saltelli, A., Annoni, P., Azzini, V., Campolongo, F., Ratto, M., Tarantola, S.. Variance based sensitivity analysis of model output. Design and estimator for the total sensitivity index. *Comput Phys Comm* 2010;181:259–270.
- Sobol, I.M., Tarantola, S., Gatelli, D., Kucherenko, S.S., Mauntz, W.. Estimating the approximation error when fixing unessential factors in global sensitivity analysis. *Reliability Engineering & System Safety* 2007;92(7):957–960.
- Janon, A., Klein, T., Lagnoux, A., Nodet, M., Prieur, C.. Asymptotic normality and efficiency of two Sobol index estimators. *ESAIM: Probability and Statistics* 2013;:1–20.
- Storlie, C.B., Swiler, L.P., Helton, J.C., Sallaberry, C.J.. Implementation and evaluation of nonparametric regression procedures for sensitivity analysis of computationally demanding models. *Reliab Eng Sys Safety* 2009;94:1735–1763.

- Zuniga, M.M., Kucherenko, S., Shah, N.. Metamodelling with independent and dependent inputs. *Computer Physics Communications* 2013;184(6):1570–1580.
- Sudret, B.. Global sensitivity analysis using polynomial chaos expansions. *Reliab Eng Sys Safety* 2008;93:964–979.
- Blatman, G., Sudret, B.. Efficient computation of global sensitivity indices using sparse polynomial chaos expansions. *Reliab Eng Sys Safety* 2010a;95:1216–1229.
- Fajraoui, N., Ramasomanana, F., Younes, A., Mara, T., Ackerer, P., Guadagnini, A.. Use of global sensitivity analysis and polynomial chaos expansion for interpretation of nonreactive transport experiments in laboratory-scale porous media. *Water Resources Research* 2011;47.
- Younes, A., Mara, T., Fajraoui, N., Lehmann, F., Belfort, B., Beydoun, H.. Use of global sensitivity analysis to help assess unsaturated soil hydraulic parameters. *Vadose Zone Journal* 2013;12(1).
- Sochala, P., Le Maître, O.. Polynomial chaos expansion for subsurface flows with uncertain soil parameters. *Advances in Water Resources* 2013;62:139–154.
- Ciriello, V., Federico, V., Riva, M., Cadini, F., Sanctis, J., Zio, E., et al. Polynomial chaos expansion for global sensitivity analysis applied to a model of radionuclide migration in a randomly heterogeneous aquifer. *Stochastic Environmental Research and Risk Assessment* 2013;27(4):945–954.
- Formaggia, L., Guadagnini, A., Imperiali, I., Lever, V., Porta, G., Riva, M., et al. Global sensitivity analysis through polynomial chaos expansion of a basin-scale geochemical compaction model. *Computational Geosciences* 2013;17(1):25–42.
- Delay, J., Trouiller, A., Lavanchy, J.. Hydrodynamic properties of the Callovo-Oxfordian formation in the East of the Paris Basin: comparison of results obtained through different approaches. *Comptes Rendus Geoscience* 2006;338(12-13):892–907.
- Distinguin, M., Lavanchy, J.. Determination of hydraulic properties of the Callovo-Oxfordian argillite at the Bure site: Synthesis of the results obtained in deep boreholes using several in situ investigation techniques. *Physics and Chemistry of the Earth* 2007;32(1-7):379–392.

- Enssle, C., Cruchaudet, M., Croise, J., Brommundt, J.. Determination of the permeability of the Callovo-Oxfordian clay at the metre to decametre scale. *Physics and Chemistry of the Earth* 2011;36(17-18):1669–1678.
- Brigaud, B., Vincent, B., Durllet, C., Deconinck, J., Blanc, P., Trouiller, A.. Acoustic properties of ancient shallow-marine carbonates: Effects of depositional environments and diagenetic processes (middle jurassic, Paris Basin, France). *Journal of Sedimentary Research* 2010;80(9-10):791–807.
- Linard, Y., Vinsot, A., Vincent, B., Delay, J., Wechner, S., De La Vaissiere, R., et al. Water flow in the Oxfordian and Dogger limestone around the Meuse/Haute-Marne underground research laboratory. *Physics and Chemistry of the Earth* 2011;36(17-18):1450–1468.
- Landrein, P., Vigneron, G., Delay, J., Lebon, P., Pagel, M.. Lithology, hydrodynamism and thermicity in the multi-layer sedimentary system intersected by the Andra deep borehole of Montiers-sur-Saulx (Meuse, France). *Bulletin de la Société Géologique de France* 2013;184(6):519–543.
- Demam, G., Kerrou, J., Benabderrahmane, H., Perrochet, P.. Sensitivity analysis of groundwater lifetime expectancy to hydro-dispersive parameters: the case of ANDRA Meuse/Haute-Marne site. *Reliability Engineering & System Safety* 2015;134(0):276–286.
- ANDRA. Modèle hydrogéologique intégré région-secteur à l’actuel. Phase II 2010-2012 - tâche 3 : Sensibilité du modèle de référence aux incertitudes sur les paramètres hydro-dispersifs et analyse de risque. Report C.RP.0CHYN.12.0001. Tech. Rep.; 2012.
- Castellini, A., Chawathe, A., Larue, D., Landa, J., Jian, F., Toldi, J., et al. What is relevant to flow? a comprehensive study using a shallow marine reservoir. 2003.
- Blatman, G., Sudret, B.. Adaptive sparse polynomial chaos expansion based on Least Angle Regression. *J Comput Phys* 2011;230:2345–2367.
- Sudret, B.. Uncertainty propagation and sensitivity analysis in mechanical models – Contributions to structural reliability and stochastic spectral methods. Université Blaise Pascal, Clermont-Ferrand, France; 2007. Habilitation à diriger des recherches, 173 pages.
- Bourgeat, A., Kern, M., Schumacher, S., Talandier, J.. The COUPLEX test cases: Nuclear waste disposal simulation. *Computational Geosciences* 2004;8(2):83–98.

- Uffink, G.. Application of the Kolmogorov's backward equation in random walk simulation of groundwater contaminant transport. *Contaminant Transport in Groundwater* 1989;:283–289.
- Cornaton, F., Perrochet, P.. Groundwater age, life expectancy and transit time distributions in advective-dispersive systems; 2. reservoir theory for sub-drainage basins. *Advances in Water Resources* 2006a;29(9):1292–1305.
- Cornaton, F., Perrochet, P.. Groundwater age, life expectancy and transit time distributions in advective-dispersive systems: 1. generalized reservoir theory. *Advances in Water Resources* 2006b;29(9):1267–1291.
- Kazemi, G., Lehr, J., Perrochet, P.. *Groundwater Age*. Wiley; 2006.
- Cornaton, F.. *GroundWater: a 3-D ground water and surface water flow, mass transport and heat transfer finite element simulator*. reference manual. 2007.
- Kerrou, J., Renard, P.. A numerical analysis of dimensionality and heterogeneity effects on advective dispersive seawater intrusion processes. *Hydrogeology Journal* 2010;18(1):55–72.
- Fourey, E., Jean-Baptiste, P., Dapoigny, A., Lavielle, B., Smith, T., Thomas, B., et al. Dissolved helium distribution in the Oxfordian and Dogger deep aquifers of the Meuse/Haute-Marne area. *Physics and Chemistry of the Earth* 2011;36(17-18):1511–1520.
- Delay, J., Distinguin, M.. Hydrogeological investigations in deep wells at the Meuse/Haute Marne underground research laboratory; vol. 104 of *Lecture Notes in Earth Sciences*; chap. 26. Springer Berlin Heidelberg; 2004, p. 219–225.
- Delay, J., Distinguin, M., Dewonck, S.. Characterization of a clay-rich rock through development and installation of specific hydrogeological and diffusion test equipment in deep boreholes. *Physics and Chemistry of the Earth* 2007a;32(1-7):393–407.
- Cosenza, P., Ghoreychi, M., de Marsily, G., Vasseur, G., Violette, S.. Theoretical prediction of poroelastic properties of argillaceous rocks from in situ specific storage coefficient. *Water Resources Research* 2002;38(10).
- Delay, J., Rebours, H., Vinsot, A., Robin, P.. Scientific investigation in deep wells for nuclear waste disposal studies at the Meuse/Haute Marne underground research laboratory, Northeastern France. *Physics and Chemistry of the Earth* 2007b;32(1-7):42–57.

- Mazurek, M., Alt-Epping, P., Bath, A., Gimmi, T., W.H., N., Buschaert, S., et al. Natural tracer profiles across argillaceous formations. *Applied Geochemistry* 2011;26(7):1035–1064.
- Vinsot, A., Delay, J., de La Vaissière, R., Cruchaudet, M.. Pumping tests in a low permeability rock: Results and interpretation of a four-year long monitoring of water production flow rates in the Callovo-Oxfordian argillaceous rock. *Physics and Chemistry of the Earth, Parts A/B/C* 2011;36(17-18):1679–1687.
- Contoux, C., Violette, S., Vivona, R., Goblet, P., Patriarche, D.. How basin model results enable the study of multi-layer aquifer response to pumping: the Paris Basin, France. *Hydrogeology Journal* 2013;21(3):545–557.
- de Hoyos, A., Viennot, P., Ledoux, E., Matray, J., Rocher, M., Certes, C.. Influence of thermohaline effects on groundwater modelling - application to the Paris sedimentary Basin. *Journal of Hydrology* 2012;464:12–26.
- Goncalves, J., Violette, S., Guillocheau, F., Robin, C., Pagel, M., Bruel, D., et al. Contribution of a three-dimensional regional scale basin model to the study of the past fluid flow evolution and the present hydrology of the Paris Basin, France. *Basin Research* 2004a;16(4):569–586.
- Goncalves, J., Violette, S., Robin, C., Bruel, D., Guillocheau, F., Ledoux, E.. Combining a compaction model with a facies model to reproduce permeability fields at the regional scale. *Physics and Chemistry of the Earth* 2004b;29(1):17–24.
- Kucherenko, S., Tarantola, A., Annoni, P.. Estimation of global sensitivity indices for models with dependent variables. *Comput Phys Comm* 2012;183:937–946.
- Li, G., Rabitz, H., Yelvington, P., Oluwole, O., Bacon, F., Kolb, C., et al. Global sensitivity analysis for systems with independent and/or correlated inputs. *J PhysChem* 2010;114:6022–6032.
- Xiu, D., Karniadakis, G.E.. The Wiener-Askey polynomial chaos for stochastic differential equations. *SIAM J Sci Comput* 2002;24(2):619–644.
- Blatman, G., Sudret, B.. An adaptive algorithm to build up sparse polynomial chaos expansions for stochastic finite element analysis. *Prob Eng Mech* 2010b;25:183–197.

- Efron, B., Hastie, T., Johnstone, I., Tibshirani, R.. Least angle regression. *Annals of Statistics* 2004;32:407–499.
- Allen, D.. The prediction sum of squares as a criterion for selecting predictor variables. Technical report, Dept of Statistics, University of Kentucky 1971;(23).
- Chapelle, O., Vapnik, V., Bengio, Y.. Model selection for small sample regression. *Machine Learning* 2002;48(1):9–23.
- Marelli, S., Sudret, B.. UQLab: a framework for uncertainty quantification in MATLAB. In: *Proc. 2nd Int. Conf. on Vulnerability, Risk Analysis and Management (ICVRAM2014)*, Liverpool, United Kingdom. 2014,.
- McKay, M., Beckman, R., Conover, W.. A comparison of three methods for selecting values of input variables in the analysis of output from a computer code. *Technometrics* 1979;21(2):239–245.