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Polynomial Chaos Expansion for Global Sensitivity Analysis applied to a model of radionuclide migration in randomly heterogeneous aquifers

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Abstract

We perform Global Sensitivity Analysis (GSA) through Polynomial Chaos Expansion (PCE) on a contaminant transport model for the assessment of radionuclide concentration at a given control location in a heterogeneous aquifer, following a release from a near surface repository of radioactive waste. The aquifer hydraulic conductivity is modeled as a stationary stochastic process in space. We examine the uncertainty in the first two (ensemble) moments of the peak concentration, as a consequence of incomplete knowledge of (a) the parameters characterizing the variogram of hydraulic conductivity, (b) the partition coefficient associated with the migrating radionuclide, (c) the effective dispersivity at the scale of interest. These quantities are treated as random variables and a variance-based GSA is performed in a numerical Monte Carlo framework. This entails solving groundwater flow and transport processes within an ensemble of hydraulic conductivity realizations generated upon sampling the space of the considered random variables. The Sobol indices are adopted as sensitivity measures to provide an estimate of the role of uncertain parameters on the (ensemble) target moments of the variable of interest. The calculation of the indices is performed by employing PCE as a surrogate model of the migration process to reduce the computational burden. We show that the proposed methodology (a) allows identifying the influence of uncertain parameters on key statistical moments of the peak concentration (b) enables extending the number of Monte Carlo iterations to attain convergence of the (ensemble) target moments and (c) leads to considerable saving of computational time while keeping acceptable accuracy.

Keywords: Performance assessment, radionuclide migration, heterogeneous aquifers, Global Sensitivity Analysis, Sobol indices, Polynomial Chaos Expansion.

45 **1. Introduction**

46 Performance assessment of radioactive waste repositories aims at evaluating the risk
47 of groundwater contamination due to potential release of radionuclides. Modeling the
48 whole chain of processes involved in this analysis is extremely challenging and requires
49 employing highly complex theoretical and numerical models to couple radionuclide
50 migration within the repository and in the groundwater environment. Uncertainty
51 associated with, e.g., incomplete knowledge of initial and boundary conditions, nature
52 and structure of the groundwater system and related key parameters must be added to the
53 list of difficulties (e.g., Tartakovsky 2007; Winter 2010; Volkova et al. 2008 and
54 references therein).

55 We consider the analysis of the uncertainty associated with the first two (statistical)
56 moments of the peak solute concentration detected at a given location and time. The
57 source of uncertainty is incomplete/imprecise knowledge of the values of the
58 hydrogeological parameters characterizing the system (Rubin 2003; Zhang 2002). For a
59 rational management of the uncertainty analysis, we use Global Sensitivity Analysis
60 (GSA) to obtain information on the relative effects of the uncertain input parameters on
61 the model outputs (Saltelli et al. 2000). In particular, we resort to variance-based
62 methods, which can provide a comprehensive view on the uncertainty and allow
63 identifying the relative and joint contributions of the uncertain input parameters to the
64 uncertainty (variance) of the model outputs (Archer et al. 1997).

65 Within variance-based GSA, the Sobol indices are widely used as sensitivity metrics
66 (Sobol 1993), because they do not require any assumption of linearity in the interpretive
67 model adopted. Their estimation is traditionally performed by Monte Carlo (MC)
68 sampling (Sobol 2001). The sample size needed to attain statistical convergence of the
69 Monte Carlo estimates can be rather large, depending on the complexity and dimension
70 (number of uncertain input parameters) of the problem (e.g., Ballio and Guadagnini 2004,
71 Zhang et al. 2010, and references therein). This might result in a serious and sometimes
72 unsustainable computational burden in cases where repeated high-resolution simulations
73 of the model are required (Sudret 2008).

74 Techniques based on advanced sampling strategies can be introduced to reduce the
75 computational cost associated with Monte Carlo simulations. Among these, the Stochastic
76 Finite Element Method (SFEM) (Ghanem and Spanos 1991) is based on a spectral
77 analysis that allows the expansion of the model output into the probabilistic space, called
78 *Polynomial Chaos* (PC) (Wiener 1938). The Polynomial Chaos Expansion (PCE) of the
79 model can be used to build a surrogate model such that the variability of the output is
80 represented in the ensemble of the expansion coefficients (Sudret 2008). Once the

81 surrogate model has been derived, the calculation of the Sobol indices does not add
82 significant extra computational costs. The formulation of a surrogate model in a
83 polynomial form has the additional advantage of allowing performing Monte Carlo
84 simulations with negligible computational effort, as compared to the original, high-
85 complexity model.

86 In this work, we rely on PCE to analyze the uncertainty affecting the outputs of a
87 numerical model of radionuclide migration in an aquifer, following a release from a near
88 surface repository. The outflow from the repository is modeled within the Monte Carlo
89 framework proposed by Cadini et al. (2012). Radionuclide migration in the aquifer is
90 modeled through an Advection-Dispersion-Reaction-Equation (ADRE). The aquifer
91 hydraulic conductivity constitutes a (second-order stationary) randomly heterogeneous
92 field. In this context, the model outputs of interest are the first two (statistical) moments
93 (i.e., mean and variance) of the peak concentration at a given control location in the
94 aquifer. We study how the incomplete/imprecise knowledge of (a) the correlation scale,
95 λ , of the variogram of the log-conductivity field, (b) the partition coefficient associated
96 with the migrating radionuclide, k_d , and (c) the effective longitudinal dispersivity at the
97 scale of interest, α_L , propagates to the selected (ensemble) moments of the output
98 distribution.

99 GSA is performed jointly with PCE to compute the Sobol indices associated with the
100 three uncertain parameters (λ , k_d , α_L), which are treated as random variables. The PCE –
101 based surrogate model is then employed to perform an exhaustive set of Monte Carlo
102 (MC) simulations to attain convergence for the target moments of interest. Given the
103 prohibitive computational costs involved in performing a large number of MC
104 simulations on the original flow and transport model, the goodness of PCE-based results
105 is then assessed on the basis of a limited number of simulations, obtained upon sampling
106 the selected random parameter space.

107 **2. Theoretical Background and Methodology**

108 **2.1 Variance-based approaches for GSA**

109 In this context, the ANOVA (ANalysis Of VAriance) representation of a model
110 output (Archer et al. 1997) is a useful tool for the definition of the Sobol indices (Sobol
111 1993; Archer et al. 1997).

112 Consider a model function $y = f(\mathbf{x})$, y being a target random response of the
113 model at a prescribed space-time location. This response depends on the vector \mathbf{x} of n
114 independent random model parameters defined in the n -dimensional unit hypercube, I^n .
115 If $f(\mathbf{x})$ is integrable, the following representation holds:

116
$$f(\mathbf{x}) = f_0 + \sum_i f_i(x_i) + \sum_{i < j} f_{ij}(x_i, x_j) + \dots + f_{1,2,\dots,n}(x_1, x_2, \dots, x_n) \quad (1)$$

117
$$\int_0^1 f_{i_1, \dots, i_s}(x_{i_1}, \dots, x_{i_s}) dx_k = 0, \quad k = i_1, \dots, i_s \quad (2)$$

118 where $1 \leq i_1 < \dots < i_s \leq n$ ($s = 1, \dots, n$) are the indices specifying the parameters upon which
 119 each term depends and the 2^n summands in (1) are orthogonal functions that can be
 120 expressed as integrals of $f(\mathbf{x})$, e.g. $f_0 = \int f(\mathbf{x}) d\mathbf{x}$ is the mean of the model,

121
$$f_i(x_i) = \int f(\mathbf{x}) \prod_{k \neq i} dx_k - f_0$$
 and so on. Therefore condition (2) renders representation (1),

122 which is typically termed ANOVA decomposition, unique.

123 When $f(\mathbf{x})$ belongs to the space of square-integrable functions, then the total
 124 variance, V , of the model due to the uncertainty of its parameters is:

125
$$V = \int f^2(\mathbf{x}) d\mathbf{x} - f_0^2 = \sum_{s=1}^n \sum_{i_1 < \dots < i_s} V_{i_1, \dots, i_s}, \quad V_{i_1, \dots, i_s} = \int f_{i_1, \dots, i_s}^2 dx_{i_1} \dots dx_{i_s} \quad (3)$$

126 V_{i_1, \dots, i_s} being the partial variance, expressing the contribution to V due to the interaction of
 127 the set of model parameters $\{x_{i_1}, \dots, x_{i_s}\}$. The generic s -order Sobol index S_{i_1, \dots, i_s} is defined
 128 as (Sobol 1993):

129
$$S_{i_1, \dots, i_s} = V_{i_1, \dots, i_s} / V \quad (4)$$

130 The sum of the indices defined in (4) is unity. The first-order or principal
 131 sensitivity indices, S_i , describe the significance of each parameter individually
 132 considered. Higher-order indices describe the effects of interactions among parameters.
 133 The overall effect of a given parameter x_i is described by the total sensitivity index S_{T_i} ,
 134 defined as:

135
$$S_{T_i} = \sum_{\eta_i} S_{i_1, \dots, i_s}, \quad \eta_i = \{(i_1, \dots, i_s) : \exists k, 1 \leq k \leq s, i_k = i\}. \quad (5)$$

136 A complete GSA requires the estimation of 2^n integrals of the kind in (3). This is
 137 usually done by Monte Carlo simulation (Sobol 2001), but the computational cost
 138 becomes prohibitive when the model is complex and the number of uncertain parameters
 139 is large (Sudret 2008).

140 **2.2 Polynomial Chaos Expansion representation of a stochastic** 141 **model**

142 We focus on the identification of a surrogate model (or metamodel) of a high
 143 complexity model (which is hereafter termed full system model) by the Polynomial
 144 Chaos Expansion (PCE) technique. This involves the projection of the model equation

145 into a probabilistic space, termed Polynomial Chaos, to construct an approximation of the
 146 model response surface. Wiener (1938) showed that the expansion performed by adopting
 147 Hermite Polynomials as a basis converges, in L_2 -sense, for any random process
 148 characterized by finite second-order moments. While the Hermite basis is suitable for
 149 Gaussian processes, different types of orthogonal polynomials are required for optimum
 150 convergence rate in the case of non-Gaussian processes (Xiu and Karniadakis 2002).

151 In this framework, one starts by noting that any square-integrable random model
 152 response, S , admits the following expansion, or chaos representation (Soize and Ghanem
 153 2004):

$$154 \quad S = \sum_{j=0}^{\infty} s_j \Psi_j(\{\zeta_n\}_{n=1}^{\infty}) \quad (6)$$

155 Here, Ψ_j denotes the j -order multivariate orthogonal polynomial, $\{\zeta_n\}_{n=1}^{\infty}$ is the set of
 156 independent random variables whose distribution is linked to the choice of the
 157 polynomial basis (Xiu and Karniadakis, 2002), and s_j are the polynomial coefficients.

158 In various engineering fields one typically considers stochastic models associated
 159 with a finite number M of input random variables. The PCE of the random model output
 160 can be derived by approximating (6) to polynomials of degree not exceeding p as

$$161 \quad S(x_1, \dots, x_M) \cong \sum_{j=0}^{P-1} s_j \Psi_j(\zeta_1, \dots, \zeta_M), \quad P = \frac{(M+p)!}{M!p!} \quad (7)$$

162 where P is the number of (unknown) polynomial coefficients.

163 The distribution of the input random variables of the model, included in vector \mathbf{x} ,
 164 does not affect the applicability of the method. Note that in cases where this distribution
 165 is not interpreted by the one required by the chosen polynomial basis, an isoprobabilistic
 166 transformation is required to relate \mathbf{x} and $\zeta = (\zeta_1, \dots, \zeta_M)$. Correlation amongst random
 167 input model parameters can be accommodated in the methodology by applying the Nataf
 168 transformation (Nataf 1962), for which the knowledge of the marginal probability density
 169 functions of the parameters and the associated correlation matrix is required.

170 Assessment of the coefficients s_j in (7) can be performed by regression, upon
 171 minimization of the variance of a residual defined as the difference between the surrogate
 172 model response, \tilde{S} , and the exact solution given by the original model (Sudret 2008)

$$173 \quad \varepsilon = S(\mathbf{x}) - \tilde{S}(\zeta) = S(\mathbf{x}) - \sum_{j=0}^{P-1} s_j \Psi_j(\zeta) \quad (8)$$

174 Minimization with respect to the vector of the unknown coefficients ζ renders

$$175 \quad \zeta = \text{Min} \left\{ E \left[\left(S(\mathbf{x}) - \tilde{S}(\zeta) \right)^2 \right] \right\} \quad (9)$$

176 with $E[\cdot]$ denoting expected value. It is useful to rewrite (9) as

$$177 \quad \zeta = (\Psi^T \Psi)^{-1} \Psi^T \mathbf{S}', \quad \Psi_{ij} = \Psi_j(\zeta^i), \quad i = 1, \dots, N; j = 0, \dots, P-1 \quad (10)$$

178 where N is the number of regression points, \mathbf{S}' is the vector denoting the model response
179 at these points, while the product $\Psi^T \Psi$ defines the so-called information matrix.

180 The choice of the optimum set of regression points is performed following the
181 same criterion adopted in the context of integral estimation by Gaussian quadrature
182 (Huang et al. 2007). Solving (10) requires a minimum of $N = P$ regression points. One
183 typically selects $N > P$ to avoid singularity in the information matrix.

184 **2.3 Polynomial Chaos Expansion and Global Sensitivity Analysis**

185 Polynomial Chaos Expansion can be considered as a powerful tool for Global
186 Sensitivity Analysis because the entire variability of the original model is conserved in
187 the set of PCE coefficients (Ghanem and Spanos 1991). The Sobol indices can be
188 analytically calculated from these coefficients without additional computational cost
189 (Sudret 2008). Manipulating \tilde{S} by appropriate grouping of terms allows isolating the
190 contributions of the different (random) parameters to the system response:

$$191 \quad \tilde{S}(\zeta) = s_0 + \sum_{i=1}^n \sum_{\alpha \in \varphi_i} s_\alpha \Psi_\alpha(\zeta_i) + \sum_{1 \leq i_1 < \dots < i_s \leq n} \sum_{\alpha \in \varphi_{i_1 \dots i_s}} s_\alpha \Psi_\alpha(\zeta_{i_1}, \dots, \zeta_{i_s}) + \dots \quad (11)$$

$$+ \sum_{\alpha \in \varphi_{1,2,\dots,n}} s_\alpha \Psi_\alpha(\zeta_1, \dots, \zeta_n)$$

192 where φ denotes a general term depending only on the variables specified by the
193 subscript.

194 In this sense, a PCE is similar to the ANOVA representation of the model.
195 Orthogonality of the polynomial basis allows recognizing that the mean of the model
196 response coincides with the coefficient of the zero-order term, s_0 , in (11) while the total
197 variance of the response is

$$198 \quad V_{\tilde{S}} = \text{Var} \left[\sum_{j=0}^{P-1} s_j \Psi_j(\zeta) \right] = \sum_{j=1}^{P-1} s_j^2 E[\Psi_j^2(\zeta)] \quad (12)$$

199 The Sobol indices can then be derived as

$$200 \quad S_{i_1, \dots, i_s} = \frac{\sum_{\alpha \in \varphi_{i_1 \dots i_s}} s_\alpha^2 E[\Psi_\alpha^2]}{V_{\tilde{S}}} \quad (13)$$

201 calculation of $E[\Psi_\alpha^2]$ can be performed, e.g., according to Abramowitz and Stegun (1970).

202 **3. Application to a model of radionuclide**
203 **migration in a randomly heterogeneous aquifer**

204 We exemplify our approach by considering an environmental problem related to
205 the performance assessment of a radioactive waste repository. We use a Monte Carlo
206 simulation model to describe radionuclide release at the repository scale. This model of
207 release of radionuclides, i.e., ^{239}Pu , from the repository is linked to a groundwater flow
208 and transport numerical model to simulate radionuclide migration within a heterogeneous
209 aquifer.

210 The aquifer hydraulic conductivity is modeled as a second-order stationary
211 stochastic process in space. We take the first two (statistical) moments (i.e., mean and
212 variance) of the peak concentration detected at a given control location in the aquifer, as
213 the target model responses. Uncertainty in these variables is considered to be a
214 consequence of incomplete knowledge of (a) the correlation scale of the variogram of the
215 log-conductivity field (b) the partition coefficient associated with the migrating
216 radionuclide, and (c) the effective dispersivity at the scale of interest.

217 **3.1 Repository representation and modeling of radionuclide**
218 **release history**

219 The conceptual repository design considered in the performance assessment
220 illustrated in this study has been proposed by ENEA (Marseguerra et al. 2001a, b) and has
221 similarities with the currently operative disposal facility of El Cabril in Spain (Zuolaga
222 2006).

223 We model the repository as a one-dimensional (along the vertical direction)
224 system (Cadini et al. 2012). The major containment structures of the disposal facility are
225 the waste packages, the modules or containers, the cells and the disposal units. These
226 constitute a multiple-barrier system designed to limit water infiltration and subsequent
227 radionuclide migration. Figure 1a depicts a typical waste package consisting in a steel
228 drum containing the radioactive waste and immobilized in a concrete matrix. The
229 diameter and the height of the waste package have been set respectively to 0.791 m and
230 1.1 m, for a total volumetric capacity of around 400 l. Figure 1b shows a cross-section of
231 the containment module adopted in this study, i.e., a concrete box-shaped structure which
232 contains 6 waste packages and is sealed with a concrete top cover. The empty spaces
233 between the packages are filled by bentonite. The external length of the module is 3.05 m,
234 with a width and height of 2.09 m and 1.7 m, respectively. The corresponding internal
235 dimensions are 2.75 m, 1.79 m and 1.37 m. The modules are arranged in $5 \times 6 \times 8$ arrays
236 within concrete structure cells built below the natural ground level.

237 Figure 2 depicts the modules arrangement and the typical repository placement at
238 a given site. The disposal unit is a concrete structure embedding a row of 6 to 10 cells.
239 The disposal facility comprises several units, which are typically arranged into parallel
240 rows. Each unit can be modeled as an independent system which can be built and
241 operated without interfering with the remaining units.

242 In agreement with typical engineering scenarios we consider that (Marseguerra et
243 al. 2001a, b): (i) the modules are identical; (ii) the mass transport occurs chiefly along the
244 vertical direction; and (iii) lateral diffusive spreading is symmetric. Under these
245 assumptions, estimating the probability of radionuclide release into the groundwater
246 system below the repository can be reduced to the one-dimensional problem of estimating
247 the release from a column of five identical vertically stacked modules, i.e., the repository
248 column may be envisioned as a one-dimensional array of compartments, each
249 corresponding to a module.

250 The radionuclides transition across the compartments is described stochastically.
251 Under the assumption that solute displacement can be modeled as a Markovian process,
252 the transition rates can be identified from the classical advection/dispersion equation.
253 Non-Fickian transport can be modeled according to existing conceptual schemes
254 (Berkowitz et al. 2006 and references therein) where the relevant transport parameters
255 could be estimated by detailed data analysis at the temporal and spatial scales at which
256 the processes of interest occur.

257 For the purpose of our example we adopt the following criteria, which can be
258 considered as conservative in a performance assessment protocol: (i) the protection
259 offered by the concrete cell roof and ceiling and the backfill layers fails; (ii) the whole
260 column, which is formed by 5 modules, is saturated and a constant water head of 0.15 m
261 is applied at the top of the highest module, i.e., the water head at the top of the column is
262 $h(z = 5 \times 1.7 \text{ m}) = 8.65 \text{ m}$; (iii) the water head at the bottom of the column is zero; (iv)
263 each module is subject to constant head gradient $\Delta h/\Delta z = 1.018$, where $\Delta h = 8.65 \text{ m}$ and
264 $\Delta z = 5 \times 1.7 \text{ m} = 8.5 \text{ m}$ is the column height; (v) the ^{239}Pu radioactive decay and the
265 subsequent generation of other radionuclides from the decay chains are neglected within
266 the repository; (vi) the migration of ^{239}Pu occurs at linear isothermal equilibrium.

267 The numerical code MASCOT (Marseguerra and Zio 2001; Marseguerra et al.
268 2003; Cadini et al. 2012) has been adopted to compute the probability density function of
269 the release of ^{239}Pu from the modules. Details of the computations and the resulting
270 temporal dynamics of the radionuclide release history are presented in Cadini et al.
271 (2012).

272 **3.2 Radionuclide migration in the groundwater system**

273 For simplicity and for the purpose of our illustration we disregard the
274 radionuclide transfer time within the partially saturated zone and analyze only
275 contaminant residence time within the fully saturated medium. This assumption may be
276 regarded as conservative because it leads to overestimating the radionuclide concentration
277 detected downstream of the repository. This can also be considered as a viable working
278 assumption in the presence of shallow reservoirs. The effect of processes occurring within
279 the partially saturated region may require an additional analysis, which is outside the
280 scope of this work.

281 Groundwater flow and contaminant transport are modeled within a two-
282 dimensional system. The (natural) log-transformed hydraulic conductivity, $Y(\mathbf{x})$ (\mathbf{x}
283 denoting the space coordinates vector), is modeled as a second-order stationary spatial
284 random function. For our example, the parameters of the variogram of Y have been
285 selected as representative of a field case study, which we do not specifically report for
286 confidentiality reasons. We note, however, that the particular choice of these values does
287 not affect the generality of the methodology. Log-conductivity is characterized by an
288 isotropic variogram of the exponential type, with sill $\sigma^2 = 1.21$. For the purpose of our
289 illustrative example, we set the variogram sill and consider its correlation scale as an
290 uncertain parameter (see Section 4) because of its poor identifiability due to typical
291 horizontal spacing of available field-scale measuring locations. Monte Carlo realizations
292 of $Y(\mathbf{x})$ have been performed by employing the sequential Gaussian scheme implemented
293 in the code GCOSIM3D (Gómez-Hernández and Journel 1993).

294 We consider a two-dimensional domain of uniform lateral side equal to 2000 m.
295 As an example, a selected realization of the log-conductivity distribution is depicted in
296 Figure 3 together with the repository projection (R), with sides equal to 50 m and 80 m,
297 and the target control point (W), located 960 m downstream of the repository fence line.

298 The domain is discretized into square cells with uniform side of 10 m, ensuring
299 that there are at least five log-conductivity generation points per correlation scale (see
300 Section 4 for additional details). Each of the 8×5 cells located under the repository
301 projection area receives the release of a cluster of 4×3 columns of 5 modules. These
302 cells are modeled through a recharge boundary condition so that a time-dependent influx
303 solute mass is injected in the porous medium according to a suitable discretization in time
304 of the Monte Carlo-based outflow from the repository. As in Cadini et al. (2012), we set
305 the incoming water flow [m^3/y] from the repository at a constant value equal to
306 $\Phi_{in} = q_d S$, $q_d = 21.2$ [m/y] being the water Darcy flux at the bottom of the 5 modules
307 column and S [m^2] being the area of the source cells. The associated radionuclide
308 concentration [Bq/m^3] released to the aquifer is then:

309
$$C_{in}(t) = A_0 \frac{pdf_{out}(t)}{\Phi_{in}} \quad (14)$$

310 where $A_0 = 1.6 \times 10^6$ [Bq] is the total activity of ^{239}Pu (which we assumed to be
 311 uniformly distributed) in the repository at a reference time $t = 0$ and $pdf_{out}(t)$ [y^{-1}] is the
 312 release probability density function from the four compartment domain (i.e., the five
 313 module column). The adopted ^{239}Pu activity level corresponds to the Italian inventory
 314 (Enea 2000) and justifies the assumption of disregarding solubility-limited release. In our
 315 example, the concentration of ^{239}Pu within the repository is
 316 $C_{rep}^{Pu239} \cong \frac{\lambda_r A_0}{N_A V_{rep}} = 2.96 \cdot 10^{-14} < C_{sl}^{Pu239} = 2.30 \cdot 10^{-4}$ [mol/m^3], where $\lambda_r = 0.28761 \cdot 10^{-4}$ [y^{-1}] is
 317 the ^{239}Pu constant decay, N_A is the Avogadro constant, V_{rep} is the total volume of the
 318 repository and C_{sl}^{Pu239} is the solubility limit of ^{239}Pu . Additional details are presented in
 319 Cadini et al. (2012).

320 Base groundwater flow in the aquifer is driven by a constant hydraulic head drop
 321 between the East and West boundaries, resulting in a unit average head gradient. No-flow
 322 conditions are assigned to the North and South boundaries.

323 Simulations of the steady state flow problem for each conductivity realization are
 324 performed with the widely used and thoroughly tested finite difference code
 325 MODFLOW2000 (McDonald and Harbaugh 1988). Radionuclide migration in the
 326 groundwater system is then modeled by means of the classical Advection-Dispersion
 327 Equation (ADE), where the partition coefficient, k_d , governing sorption of the
 328 contaminant onto the host solid matrix and the effective longitudinal dispersivity, α_L (for
 329 simplicity, transverse dispersivity is assumed to be equal to $0.1 \alpha_L$), are considered to be
 330 random variables, as described in Section 4. A uniform effective porosity of 0.15 is
 331 considered.

332 4. Global Sensitivity Analysis of the (ensemble) 333 moments of radionuclide peak concentration

334 The three random parameters selected for our demonstration are assumed to be
 335 uniformly distributed within the intervals reported in Table 1. The ranges of variability of
 336 λ and α_L are compatible with the selected domain size, and consistent with the lack of a
 337 sufficiently large number of closely spaced Y measuring points. The degree of variability
 338 of k_d has been chosen according to ENEA (1997) and Nair and Krishnamoorthy (1999).

339 The model response, i.e., the radionuclide peak concentration, c_p , at the control
 340 point is then, in turn, a random variable. As introduced in Section 3, we perform our
 341 analysis in a numerical Monte Carlo framework according to the following steps: (a) a set

342 of $N_f = 100$ Y fields are generated by GCOSIM for given values of the random
 343 parameters sampled within the intervals presented in Table 1; (b) groundwater flow and
 344 transport are solved and (ensemble) mean, $\langle c_p \rangle$, and standard deviation, σ_{c_p} , of the peak
 345 concentration are computed; (c) steps (a) and (b) are repeated for different sampling
 346 points in the random parameters space; and (d) GSA is performed to discriminate the
 347 relative contribution of the random parameters to uncertainty of $\langle c_p \rangle$ and σ_{c_p} . Note that
 348 due to the random nature of $Y(x)$, we propose to perform GSA on the (ensemble)
 349 moments of c_p rather than on its actual value calculated at the selected control location
 350 for each random realization. Conceptually, this is equivalent to performing a GSA of the
 351 results stemming from the solution of transport equations satisfied by the ensemble
 352 moments of the evolving concentrations (e.g., Guadagnini and Neuman (2001) and
 353 Morales-Casique et al. (2006 a,b) for conservative solutes).

354 The procedure illustrated is rather cumbersome when considering the solution of
 355 the full system model, because of the large number of simulations required, so that a GSA
 356 might become impractical. Therefore, we adopt the PCE technique presented in Section 2
 357 and derive expansions of order $p = 2, 3$ and 4, for both $\langle c_p \rangle$ and σ_{c_p} . We resort to the
 358 Legendre Chaos space, because the uncertain input parameters are associated with
 359 uniform distributions.

360 The calibration of the coefficients of the surrogate models requires $N_R = 10, 38$
 361 and 78 (respectively for $p = 2, 3, 4$) sampling points in the space of the three selected
 362 uncertain parameters. In our example, this corresponds to $N_{MC} = 1000, 3800, 7800$ runs
 363 of the full model of groundwater flow and transport. Calculation of the Sobol indices is
 364 then performed with negligible additional computational requirements.

365 Figure 4 reports the Total Sensitivity Indices, S_T (left), and variances, V (right),
 366 of $\langle c_p \rangle$ versus the degree of polynomial expansion, p . Figure 5 reports the corresponding
 367 results for σ_{c_p} .

368 We start by noting that S_T and V are not dramatically influenced by the degree
 369 of polynomial expansion selected for both moments. The good agreement obtained
 370 between Total and Principal Sensitivity Indices (not shown) implies that the effects of
 371 parameters interactions can be neglected in this example.

372 Figure 4 reveals that k_d and α_L are the parameters which are most influential to
 373 $\langle c_p \rangle$, regardless of the degree of expansion adopted. On the other hand, the log-
 374 conductivity correlation scale, λ , and (to a lesser degree) the dispersivity, α_L , strongly

375 influence σ_{c_p} , while k_d does not have a significant impact for the specific values adopted
 376 in the case study. The uncertainty associated with the mean peak concentration is thus
 377 related mostly to the spatial structure of heterogeneity and to the strength of the
 378 dispersion phenomena, and less to the considered geochemical scenario.

379 The calibrated surrogate models allow extending with negligible computational
 380 cost the number of Monte Carlo simulation runs required for computing mean and
 381 standard deviation of $\langle c_p \rangle$ and σ_{c_p} , as illustrated in Section 2.2. Figures 6 and 7
 382 respectively depict the dependence of the mean and the standard deviation of $\langle c_p \rangle$ and
 383 σ_{c_p} on the number of Monte Carlo runs performed with the calibrated surrogate models.
 384 The high number ($\approx 10^4$) of simulations required to attain convergence denotes the
 385 complexity of the case study and supports the adoption of a surrogate model to assess the
 386 uncertainty associated with the model response at reasonable computational costs.

387 The reliability of the results obtained through the PCE-based surrogate model has
 388 been analyzed by comparison against a number of full model runs performed by uniform
 389 sampling of $N_s = 100$ points in the random parameters space, corresponding to a total of
 390 10^4 random realizations of $Y(\mathbf{x})$. The limited amount of sampling points selected is due to
 391 the excessive computational cost associated with the full model run (about 4 min for each
 392 simulation on a standard computer with a 3.16 GHz processor).

393 Figure 8 reports the relative fraction, $\mathcal{F}(\%)$, of the mean concentration values,
 394 $\langle c_p \rangle_l^{SM}$ ($l = 1, 2, \dots, N_s$), calculated with the PCE at different orders ($p = 2, 3, 4$) and
 395 comprised within intervals of width $w = \pm \left(\sigma_{c_p}^{FM} \right)_l$, $\pm 2 \left(\sigma_{c_p}^{FM} \right)_l$, and $\pm 3 \left(\sigma_{c_p}^{FM} \right)_l$ centered
 396 around $\langle c_p \rangle_l^{FM}$, $\langle c_p \rangle_l^{FM}$ and $\left(\sigma_{c_p}^{FM} \right)_l$ respectively being the mean and standard deviation
 397 of the peak concentration computed by means of the full system model. The latter is
 398 based on a standard Monte Carlo solution of radionuclide migration within $NMC = 100$
 399 log-conductivity realizations for each $1 \leq l \leq N_s$. It can be seen that at least 40% of the
 400 values calculated with the surrogate models of different orders are comprised within the
 401 intervals of width $\pm \sigma_{c_p}^{FM}$, while about 75% of the results are included within intervals
 402 not exceeding $\pm 2 \sigma_{c_p}^{FM}$. According to this criterion, Figure 8 suggests that the best
 403 results for our example appear to be provided by the PCE of order $p = 2$.

404 To complement these results, Table 2 reports the mean and standard deviation of
 405 $\langle c_p \rangle$ calculated on the basis of the $N_s = 100$ sampling points in the random parameters
 406 space for each model (standard Monte Carlo and surrogate models of different order).

407 Table 3 reports the corresponding results for σ_{c_p} . The limited number of simulations does
408 not allow to attain convergence of the target moments. However, it is possible to observe
409 that the PCE of order $p = 4$ provides the best approximation of both the mean and
410 standard deviation of $\langle c_p \rangle$ calculated with the full model. In other words, the Total
411 Sensitivity Indices for $\langle c_p \rangle$ calculated with the PCE of order $p = 4$ are candidates to
412 provide the best indications for a GSA, as one might expect. Finally, it can be noted that
413 the PCE of order $p = 3$ best approximates the mean and standard deviation of σ_{c_p}
414 calculated with the full model on the basis of the simulations performed.

415 **5. Conclusions**

416 In this work we proposed an approach for performing a Global Sensitivity
417 Analysis (GSA) of a high-complexity theoretical and numerical model descriptive of the
418 potential release of radionuclides from a near surface radioactive waste repository and
419 their subsequent migration in the groundwater system. We considered uncertainty
420 stemming from incomplete knowledge of the variogram and transport parameters (i.e., the
421 correlation length of the variogram of log-conductivity, the partition coefficient
422 associated with the migrating radionuclide and the effective dispersivity at the scale of
423 interest) and, due to the random nature of the hydraulic conductivity field. We identified
424 as target system responses the first two (ensemble) moments of the peak concentration at
425 a given control point. GSA has been performed through the Polynomial Chaos Expansion
426 (PCE) technique, leading to the following key results: (a) the analysis of the Sobol indices
427 has revealed that the (ensemble) mean of the peak concentration is strongly influenced by
428 the uncertainty in the partition coefficient and the longitudinal dispersivity, and the
429 effects of these parameters shadow the impact of the spatial coherence of the log-
430 conductivity field at the scale analyzed; (b) on the other hand, the log-conductivity
431 correlation scale is the most influential factor affecting the uncertainty of the standard
432 deviation of the peak concentration in our example; and (c) the PCE surrogate models
433 allow extending, with negligible computational cost and acceptable accuracy, the number
434 of Monte Carlo iterations to attain convergence of the selected target moments.

435 Our results support the relevance of adopting the proposed model reduction
436 technique for complex numerical models. This methodology allows performing in-depth
437 analyses which would be otherwise unfeasible, thus severely limiting our capability to
438 represent the relevant processes involved in a target environmental scenario.

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558 **Figure Captions**

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560 **Fig. 1** Conceptual design of: (a) a waste package, (b) a containment module (ENEA 1987).

561 **Fig. 2** Sketch of the $5 \times 6 \times 8$ array of modules considered in a repository cell (ENEA 1987;
562 Marseguerra et al. 2001a, b).

563 **Fig. 3** Sketch of the adopted two-dimensional groundwater flow domain, including the
564 repository projection (R) and the selected control point (W), for a selected realization of the log-
565 conductivity field.

566 **Fig. 4** Total Sensitivity Indices ($S_T(\Omega)$, $\Omega = \lambda, \alpha_L, k_d$), Total Variance (V) and Partial
567 Variances ($V(\Omega)$, $\Omega = \lambda, \alpha_L, k_d$) calculated for $\langle c_p \rangle$ and $p=2, 3, 4$.

568 **Fig. 5** Total Sensitivity Indices ($S_T(\Omega)$, $\Omega = \lambda, \alpha_L, k_d$), Total Variance (V) and Partial
569 Variances ($V(\Omega)$, $\Omega = \lambda, \alpha_L, k_d$) calculated for σ_{c_p} and $p=2, 3, 4$.

570 **Fig. 6** Dependence of the (a) mean and (b) standard deviation of $\langle c_p \rangle$ on the number of Monte
571 Carlo iterations performed with the calibrated surrogate models.

572 **Fig. 7** Dependence of the (a) mean and (b) standard deviation of σ_{c_p} on the number of Monte
573 Carlo iterations performed with the calibrated surrogate models.

574 **Fig. 8** Relative fraction, $\mathcal{F}(\%)$, of the mean concentration values, $\langle c_p \rangle_l^{SM}$ ($l = 1, 2, \dots, N_s$)
575 calculated with the PCE at different orders ($p = 2, 3, 4$) which are comprised within intervals of
576 width $w = \pm \left(\sigma_{c_p}^{FM} \right)_l, \pm 2 \left(\sigma_{c_p}^{FM} \right)_l$, and $\pm 3 \left(\sigma_{c_p}^{FM} \right)_l$ centered around $\langle c_p \rangle_l^{FM}$; $\langle c_p \rangle_l^{FM}$ and
577 $\left(\sigma_{c_p}^{FM} \right)_l$ respectively are the mean and standard deviation of the peak concentration computed
578 through the full system model on the basis of a standard Monte Carlo analysis of radionuclide
579 migration within $NMC = 100$ log-conductivity realizations for each l .
580

581 **Table 1** Intervals of variability of the selected uniformly distributed random model parameters.
 582

Random Variable	Distribution
Partition Coefficient, k_d	$U\left(1\frac{l}{g}; 3\frac{l}{g}\right)$
Longitudinal Dispersivity, α_L	$U(50m; 70m)$
Correlation length of log-conductivity, λ	$U(40m; 100m)$

583

584 **Table 2** Values of the mean and standard deviation of $\langle c_p \rangle$ calculated with the full model and the
 585 surrogate models on the basis of 100 sampling points in the random parameter space.
 586

Model	Mean of $\langle c_p \rangle$	Standard Deviation of $\langle c_p \rangle$
Full system model	2.738E-06	3.241E-07
Surrogate model $p = 2$	2.407E-06	7.175E-08
Surrogate model $p = 3$	3.190E-06	1.887E-07
Surrogate model $p = 4$	2.538E-06	3.462E-07

587

588 **Table 3** Values of the mean and standard deviation of σ_{c_p} calculated with the full system model
 589 and the surrogate models on the basis of 100 sampling points in the random parameter space.
 590

Model	Mean of σ_{c_p}	Standard Deviation of σ_{c_p}
Full system model	4.061E-07	8.169E-08
Surrogate model $p = 2$	4.708E-07	3.310E-08
Surrogate model $p = 3$	4.278E-07	5.719E-08
Surrogate model $p = 4$	4.530E-07	1.321E-07

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