ABSTRACT

In this paper we present the application of a non-intrusive spectral techniques we recently developed for the evaluation of the uncertainties associated with a radionuclide migration problem. Spectral techniques can be used to reconstruct stochastic quantities of interest by means of a Fourier-like expansion. Their application to uncertainty propagation problems can be performed by evaluating a set of realizations which are chosen adaptively, in this work the main details about how this is done are presented. The uncertainty quantification problem we are going to deal with was first solved in a recent work where the authors used a spectral technique based on an intrusive approach. In this paper we are going to reproduce the results of this reference work, compare them and discuss the main numerical aspects. Key Words: compartment model, uncertainty quantification, spectral techniques

1. INTRODUCTION

The design of a nuclear waste repository requires robust simulation tools in order to accurately assess the risk associated with such a system. Because of the inherent complexity of this type of problem approximate mathematical models must be introduced. These are used in order to tackle the large temporal and spatial scales involved with the transport of radionuclides within the biosphere. The International Atomic Energy Agency presented a set of recommendations [1] which can be used as a reference for this type of problems. Based on these recommendations, a simplified model has been derived in [2] by treating different components of the biosphere, i.e. soil, plants, animals and humans, as separate compartments whose interconnections are modeled through transfer coefficients.

The use of such a simplified model requires the assessment of the influence of the uncertainty present within its input parameters on the numerical predictions of interest. In [3] such a task has been dealt with by using a spectral technique based on the representation of stochastic inputs and outputs by means of a Polynomial Chaos Expansion [4]. This was done in an intrusive way, by defining a new mathematical model and by developing a dedicated numerical solver.

In this paper we present the application of an alternative method which can be used to perform the same task, known as the non-intrusive spectral projection. This method is also based on the definition of
Polynomial Chaos, however its application does not require the development of a new mathematical model but it can use, similarly to sampling techniques, the solver of the original problem as a “black box”.

In the first section of this paper an introduction to the compartment model is presented. Then, the uncertainty quantification problem is presented and a new non-intrusive spectral approach developed by us is described. Finally, the results of its application to the compartment model are discussed from a numerical point of view and compared to the outcomes presented in [3]

2. THE RADIONUCLIDE MIGRATION MODEL

Given a decay chain containing \( N_r \) radionuclides, their migration across a medium (representing the biosphere) can be modeled by treating the water and the rock phases separately. Let us introduce the concentration of the radionuclide \( k \) in water as \( C^k \) and the contentation of the same radionuclide adsorbed on the solid phase as \( F^k \). Assuming the transport of each radionuclide \( k \) can be accurately modeled using advection-dispersion theory, the corresponding governing equations are [5]

\[
\frac{\partial}{\partial t} C^k(r, t) = -\nabla \cdot J^k(r, t) - \lambda^k \epsilon C^k(r, t) + \lambda^{-1} \epsilon C^{k-1}(r, t) - f(F^k, C^k) + S^k(r, t) \tag{1}
\]

\[
\frac{\partial}{\partial t} (1 - \epsilon) F^k(r, t) = -\lambda^k (1 - \epsilon) F^k(r, t) + \lambda^{-1} (1 - \epsilon) F^{k-1}(r, t) + f(F^k, C^k) \tag{2}
\]

where each concentration is expressed in \( [\text{Mol} \cdot \text{m}^{-3}] \), \( \epsilon \) is the porosity of the rock, \( \lambda^k \) the decay constant of radionuclide \( k \), \( S^k \) an external source term (representing, for example, the radionuclide release in the repository), and \( f(F, C) \) the adsorption isotherm \( [\text{Mol} \cdot \text{m}^{-3} \cdot \text{y}^{-1}] \). This term is used to define the transfer between the two phases occurring because of adsorption-desorption mechanisms.

The current term \( J^k \) in Eq. 1 is finally used to model the radionuclide transport within the water phase

\[
J^k(r, t) = -D(r, t) \nabla C^k(r, t) + U(r, t) C^k(r, t) \tag{3}
\]

where \( D \) and \( U \) are the dispersion coefficient and the advective flow velocity of water in the medium. This system of partial differential equation is fairly complex and challenging to solve, for this reason Williams [2] simplified these equations showing that, under the condition that \( C^k \) and \( F^k \) are proportional, these can be combined into

\[
\frac{\partial}{\partial t} (R^k \epsilon C^k) = -\nabla \cdot J^k - \lambda^k R^k \epsilon C^k + \lambda^{-1} R^{k-1} \epsilon C^{k-1} + S^k \tag{4}
\]

Where \( R^k \) is the retardation factor of radionuclide species \( k \) defined as

\[
R^k = 1 + \frac{1 - \epsilon}{\epsilon} K_{d,k} \rho_s. \tag{5}
\]

This retardation factor is a function of the porosity of the rock \( \epsilon \), its density \( \rho_s \), and the distribution coefficient \( K_{d,k} \). This distribution coefficient is equal to the ratio between the equilibrium concentration of radionuclide \( k \) in water over the concentration of the same radionuclide adsorbed on the solid phase. This parameter, which depends specifically on the chemical properties of a radionuclide, is usually associated with large uncertainties.

In general, Eq. 4 can be solved numerically, however the lack of knowledge about the transport terms \( D \) and \( U \) makes the task difficult. In [2] a further approximation was introduced in order to tackle the spatial
component of the differential problem. This consists in splitting up the domain in a convenient number of volumes $N_c$ (or compartments) and integrating Eq. 4 over each of these volumes.

In this way it is possible to deal with the total concentration on a certain compartment $V_i$ rather than the local concentration defined on $r$. Furthermore it is also possible, by using this formulation, to substitute the current term $J^k_i$ with a transfer term defining the transport between compartments. After integrating Eq. 4 over the spatial domain and performing few manipulations (described in details in [2]) we can express the transfer rate between compartments $i$ to $j$ as

$$
\int_{S_i} dS \cdot J^k_i(\mathbf{r}_s, t) \approx - \sum_j a_{i,j}C^k_j(t)
$$

(6)

Where $a_{ij}$ defines the transfer rates between compartments. Using this formulation it is possible to rewrite Eq. 4 using the following compartment model

$$
\frac{d}{dt} R^k_i C^k_i = \sum_{j=1}^{N_c} a_{i,j} C^k_j - \lambda^k R^k_i C^k_i + \lambda^{k-1} R_i^{k-1} C_i^{k-1} + S^k_i
$$

(7)

where the concentration $C^k_i$ now defines the number of moles of radionuclide $k$ present within compartment $i$.

Equation 7 describes a set of $N_c \cdot N_r$ coupled first order, Ordinary Differential Equations. In [2] a fictitious system, containing 8 (made-up) radionuclides and 31 compartments, was introduced and solved for illustrative purposes, the same model was then used again in [3]. We adopted the same compartment problem for the present work since our scope was to compare the outcome of our adaptive spectral techniques with the results presented in [3]. We refer to one of the two papers just discussed for a detailed description of the data (retardation factors, transfer rates, source terms, etc) used in the following sections.

In general, this mathematical model is relatively easy to solve. A code for the evaluation of the 248 unknowns characterizing the problem (31 compartments $\times$ 8 nuclides) has been written by using one of MATLAB’s built-in ODE solvers.

In the next section the uncertainty quantification problem is presented, together with the non-intrusive spectral technique used to solve it.

3. UNCERTAINTIES IN THE COMPARTMENT MODEL AND APPLICATION OF AN ADAPTIVE SPECTRAL TECHNIQUE

In [3] an uncertainty quantification problem was introduced by considering each of the distribution coefficients $K^k_d$ within Eq. 5 to be a random quantity. The reason behind this choice is that the physical and chemical processes behind absorption-desorption phenomena are not fully understood yet. The distribution coefficients depend furthermore on a variety of parameters such as temperature, soil texture and pH.

For these reasons a log-uniform distribution was chosen to better represent the large uncertainty associated with each of these coefficients. This corresponds to assuming that each of them can be represented in the following way

$$
K^k_d(\xi_k) = K^k_d,min \left( \frac{K^k_d,max}{K^k_d,min} \right)^{\xi_k}
$$

(8)
Where $K_{d,\text{min}}^k$ and $K_{d,\text{max}}^k$ are respectively, the lower and the upper bound of the distribution and $\xi_k$ is a uniformly distributed random variable. We assume that each of the distribution coefficients is associated with an independent random variable $\xi_k$, we can therefore represent our input uncertainties with an eight-dimensional vector $\xi = [\xi_1, ..., \xi_8]$.

Using this notation, the task of uncertainty quantification is to determine the influence of the vector $\xi$ on the outputs of our compartment model. The most straightforward way to deal with this task is to sample $\xi$ for a sufficiently large amount of times and to evaluate the solutions corresponding to the different realizations of the distribution coefficients. Provided this sampling set is large enough, we will have enough information to reconstruct the statistical moments of interest (i.e. mean, standard deviation and skewness) of our output quantities.

An alternative way to this approach is represented by the application of spectral techniques. These are based on the spectral representation of the stochastic inputs/outputs of our problem by means of a Fourier-like expansion, usually known as the Polynomial Chaos Expansion (PCE). The application of this concept within the uncertainty quantification framework was first proposed by Ghanem [4]. Since then, several applications and techniques have been presented, a complete overview about these can be found in [6].

Using this approach we can, for example, represent the dependency of a generic unknown of the model presented in Eq. 7 with respect to $\xi$ as

$$C_j^k(t) = \sum_{i=0}^{P} c_{ki}^j(t) \Psi_i(\xi)$$

(9)

Where $c_{ki}^j$ is the $i$th coefficient of the expansion and $\Psi_i$ the corresponding multi-dimensional (since it depends on $\xi$) polynomial of the PCE. These polynomials can be chosen from different families according to the particular type of stochastic processes one is dealing with. Legendre polynomials, for example, are the best choice when dealing with uniformly distributed random quantities. The most important characteristic associated with the PCE is that its polynomials have to be orthogonal according to the following definition of inner product

$$<\Psi_n, \Psi_m> = \int_\Theta \Psi_m(\xi)\Psi_n(\xi)w(\xi)d\xi = h_n^2 \delta_{mn}$$

(10)

where $w$ is the probability measure defining the inner product and $h_n^2$ is a constant. The only approximation introduced when representing a stochastic quantity with a PCE is given by the number of terms $P$ employed in the expansion of Eq. 9. This number depends on the dimension of $\xi$ and on the maximum polynomial order $p$ used within the PCE

$$P + 1 = \frac{(N + p)!}{N!p!}$$

According to the notation just introduced, performing uncertainty analysis by means of spectral techniques corresponds to evaluating the $P + 1$ coefficients $c_{ki}^j$ associated with the spectral representation of the output. Several techniques can be used to perform this operation, the main distinction is whether their application is intrusive or non-intrusive with respect to the numerical code used to solve the problem.

Intrusive techniques can be used to define a new mathematical problem whose unknowns are the coupled set of spectral coefficients of the expansion for each of the original unknowns. This corresponds to the
definition of a new system which is $P + 1$ larger than the original one whose solution needs the
development a completely new solver. In [3] this kind of approach have been used in order to evaluate the
coefficients of Eq. 9.

The same information can also be obtained in a non-intrusive fashion. This can be done by reconstructing
the spectral outputs through the collection of a set of their realizations, which can be evaluated by using
exclusively the original solver. We can, for example, perform the following projection using the
orthogonality property of the PCE

$$ c_{ki}^j = \frac{< C_{ki}^j, \Psi_i >}{< \Psi_i, \Psi_i >} \quad (11) $$

The evaluation of this integral can be performed numerically by using a quadrature formula, in this case the
only requirement is the collection of a set of realizations of the stochastic outputs. This procedure is
usually known as the Non-Intrusive Spectral Projection. The main issue associated with this kind of
approach is that performing multi-dimensional quadratures can become exceptionally expensive in terms of
number of quadrature points needed to evaluate the integral. This issue becomes more important when
increasing the dimension (or, in other words, the number of random inputs) of the integral.

For this reason sparse grids [7] are usually implemented for the evaluation of the projection integrals.
Furthermore, their particular definition makes it relatively easy to perform the quadrature in an adaptive
fashion. This can be done by excluding the set of the quadrature points which do not have influence on the
integrals. An algorithm to evaluate sparse grids adaptively was, for example, presented in [8]. Based on this
definition we derived and developed a new adaptive algorithm which can be used to solve the projection
integrals when applying spectral techniques. Its details are introduced in the following section.

3.1. An adaptive non-intrusive spectral method

We recently developed an algorithm which can be used to adaptively evaluate sparse grids when applying
spectral techniques. This algorithm has been successfully applied to criticality calculations in [9] and to a
time-dependent multi-physics problem in [10]. We discuss in this section the main details about the
algorithm, however for a more detailed description we refer to the two works mentioned above.

In order to present the algorithm, we first need to introduce the following quadrature formula

$$ \Delta_{lev}^{(1)} f \equiv \left( Q_{lev}^{(1)} - Q_{lev-1}^{(1)} \right) f \quad (12) $$

where $Q_{lev}^{(1)}$ is a one-dimensional quadrature formula (i.e. Trapezoidal, Gauss-Legendre, etc.) and $lev$ its
desired accuracy. According to this definition $\Delta_{lev}^{(1)}$ is also a quadrature formula resulting from the
subtraction of two consecutive quadrature levels.

Using this definition, we can mathematically represent a sparse grid by using the following summation [6]

$$ Q_{lev}^{(N)} f \equiv \sum_{|l| \leq lev+N-1} \left( \Delta_{l_1}^{(1)} \otimes \cdots \otimes \Delta_{l_N}^{(1)} \right) f \quad (13) $$

Where $Q_{lev}^{N}$ is the final sparse quadrature formula (characterized by the accuracy $lev$) and $\Delta_{l_1}^{(1)} \otimes \cdots \otimes \Delta_{l_N}^{(1)}$
is a set of multi-dimensional quadrature rules obtained as the tensorization of Eq. 12. This tensorization is...
performed by using different quadrature levels in each direction. Each of these rules can be characterized by the multi-index \( \mathbf{l} = (l_1, \ldots, l_N) \in \mathbb{N}^N \) which is a vector as large as the number of stochastic directions, whose indexes \( l_i \) are used to associate an accuracy level with each direction.

In Eq. 13 the norm of this multi-index is defined as

\[
|\mathbf{l}| \equiv \sum_{i=1}^{N} l_i
\]

If we analyze Eq. 13 based on this definition we can see that the final sparse grid is built by adding a number of “sub-grids” (each of them defined by the multi-index \( \mathbf{l} \)) according to the constraint \( |\mathbf{l}| \leq \text{lev} + N - 1 \). Each sub-grid has an associated size according to the corresponding multi-index: this size is equal to the number of entries whose value is larger than one.

In practice not all the sub-grids included in the final sparse quadrature rule will give the same contribution to the integral, in theory we can optimize the sparse quadrature process by excluding a subset of sub-grids. We developed an algorithm in order to perform this task. This algorithm is applied by adding the sub-grids according to their size and their resolution. In this way, the information associated with the subgrids already evaluated is analyzed in order to adaptively select the subgrids that still need to be included within the quadrature rule. The following indicator is used to quantify the contribution of each sub-grid

\[
\epsilon_i = \max \left[ \frac{\left| \delta E_i(R) \right|}{E(R)}, \frac{\left| \delta \sigma_i(R) \right|}{\sigma(R)} \right]
\]

(14)

where \( R \) is a generic stochastic output whose spectral expansion needs to be evaluated. The changes in mean and standard deviation used in Eq. 14 are calculated by using the spectral expansion of \( R \).

In general, the following process is repeated, starting from the set of one-dimensional sub-grids associated with the lowest norm, until the largest dimension is reached

1. Evaluate the indicator \( \epsilon_i \) associated with each \( d \)-dimensional grid \( \mathbf{l} \) characterized by the same norm
2. If \( \epsilon_i \) is smaller than a specified tolerance do not include the grids obtained as a tensorization of \( \mathbf{l} \) or by increasing its resolution
3. Repeat the previous step for all the sub-grids with the same size but associated with a higher norm \( |\mathbf{l}| = |\mathbf{l}| + 1 \). Increase the norm until all the sub-grids having dimension \( d \) are either included or excluded.
4. Increase the size \( d \) of the sub-grids and repeat step 1 starting from the sub-grid associated with the smallest norm.

The algorithm will stop when every grid defined by the constraint \( |\mathbf{l}| \leq \text{lev} + N - 1 \) in Eq. 13 has either been evaluated or discarded.

The algorithm includes another part. After the one-dimensional sub-grids have been evaluated, a convergence check is performed in order to determine the polynomial order needed to represent the
stochastic outputs along each direction. This is done by considering each stochastic direction as a separate one-dimensional random problem. The main assumption is that the spectral convergence along each of the directions of the multi-dimensional random problem is the same as the corresponding one-dimensional problem. This procedure allows to determine a truncated PCE which can be used for the evaluation of the rest of the sub-grids.

In the next section the algorithm just introduced will be applied to the radionuclide migration problem discussed in the first part of the paper.

4. RESULTS

In this section we present some of the results we obtained by applying the adaptive algorithm to the radionuclide compartment problem. Although in [3] this application is presented for several compartments and two points in time, all the results included in this section refer only to the radionuclide concentrations in compartment 31 after 100000 years.

First, the probability density functions for each of the radionuclides in compartment 31 have been obtained by applying a standard unbiased sampling technique. 100000 realizations were collected for this purpose. Then, the adaptive algorithm introduced in the previous section was applied to the problem by using 5 different tolerances. Figure 1 presents the comparison of the probability density functions obtained by using the sampling technique with the ones built by using the adaptive spectral technique with a 0.001% tolerance. The figure clearly verifies the convergence and the robustness of the spectral algorithm. Fifth order PCEs were required to reach convergence, this is caused by the pronounced skewness of the probability distributions. In [3], the same PCE order was also used to build the distributions using the intrusive method. Altough The computational cost of the technique was also studied by collecting the outcomes obtained using different tolerances. Table I shows the corresponding error calculated by averaging the errors present within the standard deviation of each radionuclide concentration. Each relative error was evaluated by considering as a reference value the one obtained by implementing a complete sparse grid (corresponding to 6069 points). The number of realizations needed to reach convergence in each case is also included. It is important to notice two things from this table: first, the error reduction achieved by using smaller tolerances is relatively small, and secondly the prediction obtained by using the largest tolerance (1%) is fairly accurate.

The comparison between the number of realizations needed to reach convergence clearly show that there is
Figure 1. Radionuclides concentrations in compartment 31 at $t = 100000$ y. Comparison between the values obtained by using the adaptive algorithm with 0.001\% tolerance (blue lines) and the values obtained by using a standard random sampling (red lines, 100000 samples).

an optimal number of points after which the cost-benefit ratio becomes too large. In this case we can state
that by collecting 165 realizations we have a fairly accurate estimate of our output's standard deviation.

From Figure 2 we can see that this is also true for the other statistical moments of interest. Here the difference between the functions obtained by using the largest and the smallest tolerances is almost negligible. These results suggest that by using relatively large tolerances we can accurately reconstruct our stochastic outputs by using a fraction of realizations compared to standard sampling techniques. Finally,

Table II. Probability for the concentrations of each nuclide to exceed their mean value. Comparison between the values obtained in [3] and the values obtained by using the adaptive algorithm.

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Hagues et al.</th>
<th>Adaptive spectral algorithm (tol=1%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0.4985</td>
<td>0.4988</td>
</tr>
<tr>
<td>2</td>
<td>0.3615</td>
<td>0.3591</td>
</tr>
<tr>
<td>3</td>
<td>0.3367</td>
<td>0.3352</td>
</tr>
<tr>
<td>4</td>
<td>0.3461</td>
<td>0.3451</td>
</tr>
<tr>
<td>5</td>
<td>0.3355</td>
<td>0.3340</td>
</tr>
<tr>
<td>6</td>
<td>0.3438</td>
<td>0.3435</td>
</tr>
<tr>
<td>7</td>
<td>0.3383</td>
<td>0.3362</td>
</tr>
<tr>
<td>8</td>
<td>0.3354</td>
<td>0.3342</td>
</tr>
</tbody>
</table>

the probability of each nuclide's concentration to be larger than its mean value was also calculated as in [3]. This parameter is relevant from a safety point of view because it can be used as an estimator of the risk associated with high radionuclide concentrations. Table II includes a comparison between the values obtained by using the adaptive algorithm and 1% tolerance with the ones presented in [3].

All the values are in good agreement, especially if we take into account the statistical errors and the fact that the two methods are inherently different from the numerical point of view. For the intrusive case the application of the spectral technique was performed by solving a mathematical problem whose size is 1287 times larger than the original problem. For this particular example, the advantage of using an adaptive non-intrusive technique is quite considerable.

5. CONCLUSIONS

In this paper we solved an uncertainty propagation problem associated with a radionuclide release model by means of a non-intrusive spectral technique. This spectral technique is based on the definition of a sparse grid and it is applied by adaptively collecting a set of realizations of the stochastic outputs. Once the adaptive algorithm is defined, the application to a particular problem is, unlike intrusive approaches, straightforward.

We presented a set of probability density functions characterizing the stochastic outputs of the problem, and compared them to the outcome of a traditional sampling approach. The results obtained with the
Figure 2. Radionuclides concentrations in compartment 31 at $t = 100000y$. Comparison between the values obtained by using the adaptive algorithm with 1% tolerance (blue lines) and the values obtained by using the same algorithm with 0.001% tolerance (red lines).
adaptive technique proved to be fairly accurate even when using a large tolerance within the algorithm. This corresponded to a relatively small set of samples required to converge, leading to considerable computational savings. In general, the number of realizations needed to reconstruct the spectral expansion increases when increasing the number of stochastic outputs. Nevertheless for the present example we demonstrated that the application of the adaptive non-intrusive technique is considerably cheaper than standard sampling approaches.

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REFERENCES


