

Modelling of solute transport in a mild heterogeneous porous medium using stochastic finite element method: Effects of random source conditions

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SUMMARY

Randomness in the source condition other than the heterogeneity in the system parameters can also be a major source of uncertainty in the concentration field. Hence, a more general form of the problem formulation is necessary to consider randomness in both source condition and system parameters. When the source varies with time, the unsteady problem, can be solved using the unit response function. In the case of random system parameters, the response function becomes a random function and depends on the randomness in the system parameters. In the present study, the source is modelled as a random discrete process with either a fixed interval or a random interval (the Poisson process). In this study, an attempt is made to assess the relative effects of various types of source uncertainties on the probabilistic behaviour of the concentration in a porous medium while the system parameters are also modelled as random fields. Analytical expressions of mean and covariance of concentration due to random discrete source are derived in terms of mean and covariance of unit response function. The probabilistic behaviour of the random response function is obtained by using a perturbation-based stochastic finite element method (SFEM), which performs well for mild heterogeneity. The proposed method is applied for analysing both the 1-D as well as the 3-D solute transport problems. The results obtained with SFEM are compared with the Monte Carlo simulation for 1-D problems. Copyright © 2007 John Wiley & Sons, Ltd.

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1. INTRODUCTION

Probabilistic studies of solute transport in porous media are mainly focused on predicting the concentration uncertainty due to the heterogeneity of the governing flow and transport parameters [1–10]. In addition to the effective parameters, the spatial variability of governing parameters which

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is modelled as random space functions, results in the prediction uncertainty of the concentration distribution. The prediction uncertainty is represented as the variance of concentration [11–15]. Using moment equation method Andricevic [11] showed that the local dispersion tensor plays a role in the dissipation of concentration later when favourable concentration gradient is created. Kapoor and Kitanidis [14] also showed a similar behaviour using moment equation method and compared it with Monte Carlo simulation. Using Lagrangian approach, Pannone and Kitanidis [15] determined the concentration variance and they incorporated additional tensor that described the kinetics of dilution of a plume. Fiori and Dagan [13] showed that the coefficient of variation (COV) of concentration decreases with the decrease in the Peclet number for 2-D and 3-D transport problems. The input/source conditions can also be spatially and/or temporally varying random processes in a natural hydrologic system. The randomness in the source or boundary conditions can also be a major source of uncertainty in the concentration field while analysing the solute transport problems. The contaminant source often can be continuous or discrete in time [16]. For continuous source, the temporal correlation is described by a correlation function. On the other hand, the discrete source is modelled as a sequence of instantaneous injections with random mass. For the discrete source the time interval of mass injection can be either uniform or random. Wang and Zheng [16] analysed the contaminant transport under random sources in a deterministic groundwater system. In their model, the random source was considered either as a continuous source with random fluctuations in time or as a discrete instantaneous source. In certain cases, it is required to assess the relative effects of system uncertainty and source/boundary condition uncertainty on the probabilistic behaviour of concentration. Li and Graham [17] considered the boundary condition due to recharge as a spatial/temporal random process and analysed its effect on the contaminant transport problem in a heterogeneous groundwater system. Using analytical methods, they found that the spreading of the mean concentration was enhanced when the random spatio-temporal variation of recharge was considered. Stochastic modelling of the concentration variables with random inputs has received attention in the surface water quality literature [18, 19] as well. Recently, Boano *et al.* [20] modelled stochastically the DO and BOD components in a stream with random inputs using a semi-analytical approach for the solution of the stochastic ordinary differential equations (ODE's), which was computationally more efficient than the common Monte Carlo Simulation method (MCSM).

However, for the solution of the stochastic partial differential equations (SPDE's) when analytical methods are not applicable due to the complicated initial and source conditions, non-uniform flow fields and non-stationary parameters, numerical methods are required. The popular and simple MCSM is computationally exhaustive when a few thousands of realizations are required especially for higher degrees of medium heterogeneities along with higher space–time grid resolutions. To avoid this difficulty, alternate perturbation-based methods such as moment equation method [21–24], stochastic finite element method (SFEM) [25–27] were proposed. In moment equation method, the PDEs for statistical moments which are derived analytically based on stochastic manipulation, are solved numerically [22, 23]. Lu and Zhang [24] proposed higher-order moment equation method using Karhunen–Loeve expansion of the random system parameters and polynomial chaos expansion of the hydraulic head. An iterative moment equation method was proposed by Morales-Casique *et al.* [28]. This method was found to provide good results for the non-reactive solute transport problem. In SFEM, a set of coupled algebraic equations are derived from the PDE and the output vector (such as discretized hydraulic head or concentration) are expressed in a Taylor series of the discretized random hydraulic conductivity, porosity, etc. [25]. The sensitivity of output variable is solved by matrix operation in a recursive way and ensemble statistics are estimated

using the Taylor series expansion. The higher-order SFEM is based on polynomial chaos expansion, as proposed by Ghanem [26]. This higher-order SFEM is more accurate for strong heterogeneity but less efficient compared to the SFEM using Taylor series expansion. The SFEM was used for analysing multi-dimensional solute transport problems for reactive solutes with hydraulic conductivity, porosity, sorption coefficient, etc. as random fields [25, 29, 30]. The method was shown to be efficient and accurate in simulating the mean and variance of concentration field for mild heterogeneous case. In these cases, the source/boundary conditions are assumed as deterministic.

These alternate methods such as SFEM or moment equation method can be developed for the probabilistic analysis of flow and solute transport in a heterogeneous porous medium when source conditions are also a random process. In such circumstances these are used to obtain the stochastic unit response function, which can be combined with the analytically derived expressions of the statistical moments of the concentration of the system. In a recent study, Chaudhuri and Sekhar [31] presented the results for a random source condition, which was modelled as a discrete process with uniform time interval. Using SFEM the unit response function was obtained, which was used in the analytical expressions of the statistical moments of the concentration due to random source and random system parameters. However, when the source is to be modelled as a process with random time interval (the Poisson process), the derivation of the expressions of mean and covariance of concentration is relatively complicated.

In the present study, the analytical expressions of mean and covariance of concentration are obtained when the random source is modelled as a Poisson process. SFEM is used to obtain the unit response function for a case with random system parameters (viz hydraulic conductivity, porosity, etc.). The results obtained by the proposed method are compared with Monte Carlo simulations for a 1-D problem. The effect of random source condition is analysed on cases with varying levels of uncertainty in the random system parameters. Further, the combined effect of randomness of system parameters and source is studied on a 3-D problem, which considers a realistic and complicated source condition [32]. It is demonstrated that the prediction uncertainty of the concentration is enhanced due to the randomness of the time interval of the mass injection events. Results are also presented to discuss the relative effects of source uncertainty and parametric uncertainty. Since the local-scale dispersion parameters have influence on the variance of concentration, the combined effects of system and source uncertainty are studied for different values of local-scale dispersion parameters.

2. PROBLEM DESCRIPTION

The governing equation for the transport of a linearly sorbing and decaying solute in a 3-D porous media is

$$\begin{aligned} (n(\mathbf{x}) + \rho_b k_d(\mathbf{x})) \frac{\partial c(\mathbf{x}, t)}{\partial t} + \frac{\partial}{\partial x_i} \left(n(\mathbf{x}) v_i(\mathbf{x}) c(\mathbf{x}, t) - n(\mathbf{x}) D_{ij}(\mathbf{x}) \frac{\partial c(\mathbf{x}, t)}{\partial x_j} \right) \\ + (n(\mathbf{x}) + \rho_b k_d(\mathbf{x})) \gamma_d(\mathbf{x}) c(\mathbf{x}, t) = 0 \end{aligned} \quad (1)$$

where $c(\mathbf{x}, t)$ is the concentration at location \mathbf{x} and time t . Here $n(\mathbf{x})$, $k_d(\mathbf{x})$ and $\gamma_d(\mathbf{x})$ are, respectively, spatially varying porosity, sorption and decay coefficient. In Equation (1), $\mathbf{v}(\mathbf{x})$ is the pore water velocity vector which is defined as $\mathbf{v}(\mathbf{x}) = \mathbf{q}(\mathbf{x})/n(\mathbf{x})$. The seepage flux vector $\mathbf{q}(\mathbf{x})$ is

obtained using the hydraulic conductivity tensor ($\mathbf{K}(\mathbf{x})$) and hydraulic head ($h(\mathbf{x})$), based on the Darcy equation,

$$q_i(\mathbf{x}) = -K_{ij}(\mathbf{x}) \frac{\partial h(\mathbf{x})}{\partial x_j} \quad (2)$$

$\mathbf{D}(\mathbf{x})$ is the hydrodynamic dispersion coefficient tensor, which is combined with the molecular diffusion coefficient ($D_m(\mathbf{x})$). The expression for it is given as

$$D_{ij}\mathbf{x} = \alpha(\mathbf{x}) \left((1 - \varepsilon) \frac{v_i(\mathbf{x})v_j(\mathbf{x})}{v(\mathbf{x})} + \varepsilon v(\mathbf{x})\delta_{ij} \right) + D_m(\mathbf{x})\delta_{ij} \quad (3)$$

where $\alpha(\mathbf{x})$ is the longitudinal local dispersivity and ε is the ratio of transverse to longitudinal local dispersivity. Equation (1) is solved for a set of initial and boundary conditions which, in general, are written as

$$c(\mathbf{x}, 0) = c_0(\mathbf{x}) \quad \text{for } \mathbf{x} \in \Omega$$

$$c(\mathbf{x}, t) = c_b(\mathbf{x}, t) \quad \text{for } \mathbf{x} \in \Gamma_1$$

and

$$\left(n(\mathbf{x})v_i(\mathbf{x})c(\mathbf{x}, t) - n(\mathbf{x})D_{ij}(\mathbf{x}) \frac{\partial c(\mathbf{x}, t)}{\partial x_j} \right) n_{x_i} = f_b(\mathbf{x}, t) \quad \text{for } \mathbf{x} \in \Gamma_2 \quad (4)$$

Here $c_0(\mathbf{x})$ is initial distribution of concentration in the domain Ω while $c_b(\mathbf{x}, t)$ and $f_b(\mathbf{x}, t)$ are, respectively, the time-dependent specified concentration at the boundary Γ_1 and flux at the at the boundary Γ_2 . Further n_{x_i} is the direction cosine of the normal to the boundary surface along x_i axis.

The equation for a steady-state flow in the domain with spatially varying hydraulic conductivity field is given by

$$\frac{\partial}{\partial x_i} \left(K_{ij}(\mathbf{x}) \frac{\partial h(\mathbf{x})}{\partial x_j} \right) = 0 \quad (5)$$

with specified boundary conditions governing the flow in the domain represented as,

$$h(\mathbf{x}) = h_b(\mathbf{x}) \quad \text{for } \mathbf{x} \in \Gamma_1^h \quad \text{and} \quad K_{ij}(\mathbf{x}) \frac{\partial h(\mathbf{x})}{\partial x_j} n_{x_i} = q_b(\mathbf{x}) \quad \text{for } \mathbf{x} \in \Gamma_2^h \quad (6)$$

Using finite element method for spatial discretization and finite difference method (Crank–Nicholson formulation) for temporal discretization the global equation for transport (Equation (1)) with specified boundary conditions (Equation (4)) is obtained as

$$[D_1]\{c^{t+1}\} = [D_2]\{c^t\} + \theta\{c_b^{t+1}\} + (1 - \theta)\{c_b^t\} \quad (7)$$

where the matrices $[D_1]$ and $[D_2]$ can be called as transport matrices. These matrices are functions of the vector $\{r\}$ which consists of the discretized vector of velocity components, local dispersivity,

molecular diffusion, porosity, sorption coefficient and decay rate of each element. In general, the vector can be written as $\{r\} = \{v_{1p}, v_{2p}, v_{3p}, \alpha_p, D_{mp}, n_p, k_{dp}, \gamma_{dp}; p = 1, 2, \dots, N^e\}^T$, where N^e is the number of elements used to discretize the domain. Thus, the nodal vectors of dependent variables ($\{c^{t+1}\}$ and $\{c^t\}$) become the function of the discretized random parameters. The global equation for the flow (Equation (5)) for a given head and flux boundary conditions (Equation (6)) is obtained as,

$$[K]\{h\} = \{h_0\} \tag{8}$$

Here $[K]$ is the global hydraulic conductivity matrix in the flow equation. The velocity is obtained for a specified hydraulic head gradient. The i th component of seepage flux of p th element is obtained by taking average of that at all of the Gauss points (\mathbf{x}_k , for $k = 1, \dots, N_G$, where N_G is the number of Gauss points) and is given as

$$q_{ip} = - \frac{1}{N_G} K_{ijp} \sum_{k=1}^{N_G} \frac{\partial N_l(\mathbf{x})}{\partial x_j} \bigg|_{\mathbf{x}_k} h_l = - \frac{1}{N_G} K_p \sum_{k=1}^{N_G} \frac{\partial N_l(\mathbf{x})}{\partial x_i} \bigg|_{\mathbf{x}_k} h_l \tag{9}$$

For isotropic cases the hydraulic conductivity tensor becomes a scalar quantity (K_p). Using the relation between seepage flux and flow velocity ($q_{ip} = n_p v_{ip}$) the product $n_p D_{ijp}$ can be written in terms of water flux as

$$n_p D_{ijp} = \alpha_p \left((1 - \varepsilon) \frac{q_{ip} q_{jp}}{q_p} + \varepsilon q_p \delta_{ij} \right) + n_p D_{mp} \delta_{ij} \tag{10}$$

where the resultant seepage flux ($q_p = (\sum_{i=1}^3 q_{ip}^2)^{1/2}$).

Since the seepage flux (Equation (9)) or flow velocity is used as a system parameter for the transport problem (Equation (1)), the transport matrices (D_1) and (D_2) in Equation (7) are functions of the velocity vector. Thus, the transport matrices are related to the random hydraulic conductivity.

3. SFEM FORMULATION

In SFEM, the matrices (D_1 and D_2) and nodal vectors of dependent variable ($\{c^{t+1}\}$ and $\{c^t\}$) are expanded using the Taylor series about the mean value of the random parameters ($r'_p, p=1, 2, \dots, N_r$).

$$\begin{aligned} [D_1] &= [\bar{D}_1] + [D_1]' = [\bar{D}_1] + \sum_{p=1}^{N_r} [D_1]_{r'_p}^{(I)} r'_p \quad \text{and} \\ [D_2] &= [\bar{D}_2] + [D_2]' = [\bar{D}_2] + \sum_{p=1}^{N_r} [D_2]_{r'_p}^{(I)} r'_p \end{aligned} \tag{11}$$

where $[D_1]_{r'_p}^{(I)} = \partial[D_1]/\partial r_p$ and $[D_2]_{r'_p}^{(I)} = \partial[D_2]/\partial r_p$. Similarly, the concentration can be expanded in Taylor series about the mean value of the random parameters and can be expressed as follows:

$$\{c^{t+1}\} = \{c^{t+1}\}^{(0)} + \sum_{p=1}^{N_r} \{c^{t+1}\}_{r'_p}^{(I)} r'_p + \frac{1}{2} \sum_{p=1}^{N_r} \sum_{q=1}^{N_r} \{c^{t+1}\}_{r'_p r'_q}^{(II)} r'_p r'_q \dots \tag{12}$$

After substituting Equations (12) and (11) into Equation (7), the various orders of derivatives of concentration are obtained as,

$$\{c^{t+1}\}^{(0)} = [\overline{D}_1]^{-1}([\overline{D}_2]\{c^t\}^{(0)} + \theta\{\overline{c}_b^{t+1}\} + (1 - \theta)\{\overline{c}_b^t\}) \quad (13)$$

$$\{c^{t+1}\}_{r_p}^{(I)} = [\overline{D}_1]^{-1}(-[D_1]_{r_p}^{(I)}\{c^{t+1}\}^{(0)} + [D_2]_{r_p}^{(I)}\{c^t\}^{(0)} + [\overline{D}_2]\{c^t\}_{r_p}^{(I)}) \quad (14)$$

$$\begin{aligned} \{c^{t+1}\}_{r_p r_q}^{(II)} &= [\overline{D}_1]^{-1}(-[D_1]_{r_p}^{(I)}\{c^{t+1}\}_{r_q}^{(I)} - [D_1]_{r_q}^{(I)}\{c^{t+1}\}_{r_p}^{(I)} \\ &\quad + [D_2]_{r_p}^{(I)}\{c^t\}_{r_q}^{(I)} + [D_2]_{r_q}^{(I)}\{c^t\}_{r_p}^{(I)} + [\overline{D}_2]\{c^t\}_{r_p r_q}^{(II)}) \end{aligned} \quad (15)$$

By taking the expectation of Equation (12) and neglecting the higher-order terms, the second-order accurate mean concentration is obtained as

$$\{\overline{c}^{t+1}\} = \{c^{t+1}\}^{(0)} + \frac{1}{2} \sum_{p=1}^{N_r} \sum_{q=1}^{N_r} \{c^{t+1}\}_{r_p r_q}^{(II)} \overline{r_p r_q} \quad (16)$$

Under the approximation that $\overline{r_p r_q} - r_p r_q \approx 0$, the first-order accurate covariance matrix of the concentration at any two different time instances (t_1 and t_2) is obtained as

$$[CV]_{cc} = \sum_{p=1}^{N_r} \sum_{q=1}^{N_r} \{c^{t_1}\}_{r_p}^{(I)} \{c^{t_2}\}_{r_q}^{(I)} \overline{r_p r_q} \quad (17)$$

This approximation is valid for the mild heterogeneity of the random system parameters.

The covariance matrix of the random parameters (which are piece-wise linear inside an element) is derived from the given variances and spatial correlation functions for the random fields. For solving the transport problem, the statistical moments of the flow velocity need to be derived from the statistical moments of the hydraulic conductivity. The derivation of this is presented in the Appendix. In this study all the flow and transport parameters, which are considered as random fields, are assumed to follow a log normal distribution since they take positive values and also vary considerably. However, this assumption is not a limitation for the proposed SFEM. The random fields are assumed as statistically homogeneous and are described by a Gaussian (squared exponential)-type correlation function. However, it may be noted that for specific applications, experimentally derived correlation functions if available can also be used in the SFEM. The correlation coefficient between the log parameters of any two points is given by $\rho(\mathbf{x}) = \exp(-(x_1/\lambda_1)^2 - (x_2/\lambda_2)^2 - (x_3/\lambda_3)^2)$, where λ_1 , λ_2 and λ_3 are the correlation lengths. The covariance matrix for a random element parameters is determined from the correlation function and the variances using the local averaging method [33].

4. RANDOM SOURCE CONDITION

In the present study a contaminant source, which is discrete in time is considered. The source is modelled as a sequence of instantaneous mass injections with random amount. For the discrete source the time interval of mass injection may be uniform (Figure 1(a)) or random (Figure 1(b)). In addition, the amount of input mass can also vary with the time. The total amount of cumulative

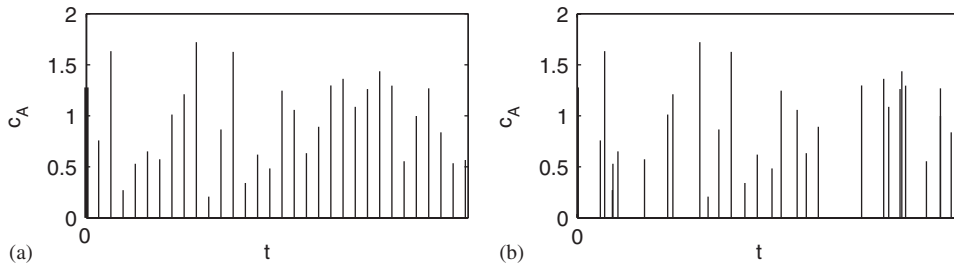


Figure 1. The random mass release at: (a) uniform time interval and (b) random time interval.

mass of contaminant injected into the porous medium is the sum of random number of incidents: $\sum_{k=1}^{M(t)} c_{A_k}$, where $M(t)$ is the number of mass injections that occurred during the interval $[0, t]$. Here c_{A_k} is the amount of random mass injected at time τ_k . In this study it is also assumed that the magnitude of the mass released at each time follows a normal distribution, however, the formulation can consider any distribution. In the present work, for the solute transport problem under the general assumptions, the concentration can be obtained as a random sum of the impulse/response function, which is expressed as

$$c(\mathbf{x}, t) = \sum_{k=1}^{M(t)} c_{A_k} G_c(\mathbf{x}, t, \tau_k) \tag{18}$$

In the above expression, $G_c(\mathbf{x}, t, \tau_k)$ is a response function of the concentration due to a unit magnitude of mass of solute which is applied at time $t=0$ following a given set of boundary conditions. For some cases with simple boundary conditions, the unit response function can be derived analytically. In the case of complicated system and boundary conditions, any numerical method is adopted to obtain the unit response function in the form of a discretized array instead of a closed-form function. Since the response function must satisfy the condition: $G_c(\mathbf{x}, t, \tau) = 0$ for $t < \tau$, in Equation (18), $M(t)$ can be replaced by $M(T)$ where T is the total time of simulation. Since the output concentration here is a nonlinear function of system parameters, a truncated Taylor series expansion is used for the stochastic analysis of output concentration while incorporating the uncertainty in the system parameters. When numerical methods are used for the solution with discretization in space and time, the response function is obtained as a vector of concentration at all nodes $\{G_c(t, \tau_k)\}$. The concentration at i th node due to the multiple sources can be expressed as

$$c_i(t) = \sum_{k=1}^{M(T)} c_{A_k} G_{c_i}(t, \tau_k) \tag{19}$$

When the governing transport parameters are also random fields, the unit response function in Equation (19) can be expressed as,

$$G_{c_i}(t, \tau) = (G_{c_i}(t, \tau))^{(0)} + \sum_{p=1}^{N_r} (G_{c_i}(t, \tau))_{r_p}^{(I)} r'_p + \frac{1}{2} \sum_{p=1}^{N_r} \sum_{q=1}^{N_r} (G_{c_i}(t, \tau))_{r_p r_q}^{(II)} r'_p r'_q \tag{20}$$

The mean of the response function is written as

$$\overline{G_{c_i}(t, \tau)} = (G_{c_i}(t, \tau))^{(0)} + \frac{1}{2} \sum_{p=1}^{N_r} \sum_{q=1}^{N_r} (G_{c_i}(t, \tau))_{r_p r_q}^{(II)} \overline{r'_p r'_q} \quad (21)$$

and the expectation of the product of the response function is written as

$$\begin{aligned} \overline{G_{c_i}(t_1, \tau_1) G_{c_j}(t_2, \tau_2)} &= (G_{c_i}(t_1, \tau_1))^{(0)} (G_{c_j}(t_2, \tau_2))^{(0)} + \frac{1}{2} (G_{c_i}(t_1, \tau_1))^{(0)} \\ &\quad \times \left(\sum_{p=1}^{N_r} \sum_{q=1}^{N_r} (G_{c_j}(t_2, \tau_2))_{r_p r_q}^{(II)} \overline{r'_p r'_q} \right) \\ &\quad + \frac{1}{2} \left(\sum_{p=1}^{N_r} \sum_{q=1}^{N_r} (G_{c_i}(t_1, \tau_1))_{r_p r_q}^{(II)} \overline{r'_p r'_q} \right) (G_{c_j}(t_2, \tau_2))^{(0)} \\ &\quad + \sum_{p=1}^{N_r} \sum_{q=1}^{N_r} ((G_{c_i}(t_1, \tau_1))_{r_p}^{(I)} (G_{c_j}(t_2, \tau_2))_{r_q}^{(I)}) \overline{r'_p r'_q} \quad (22) \end{aligned}$$

In this study, SFEM is used to compute $\overline{G_{c_i}(t, \tau)}$ and $\overline{G_{c_i}(t_1, \tau_1) G_{c_j}(t_2, \tau_2)}$ numerically considering the source as a unit pulse. The random transport parameters are usually uncorrelated with the source conditions, i.e. c_{A_k} and r'_p are uncorrelated. Since the random pulse inputs of concentration at two different times are independent, it may be noted that $\overline{c_{A_k} c_{A_l}} = (\overline{c_A})^2 + \sigma_{c_A}^2 \delta_{kl}$.

4.1. Uniform time interval case

In this case the mass releases are assumed to be at uniform time intervals which are fixed and known. Hence, c_{A_k} ($k = 1, 2, \dots, M(T)$) are random variables but τ_k ($k = 1, 2, \dots, M(T)$) are deterministic. Taking the expectation of Equation (20) the mean concentration is obtained as

$$\overline{c_i(t)} = E[c_i(t)] = E \left[\sum_{k=1}^{M(T)} c_{A_k} G_{c_i}(t, \tau_k) \right] = \overline{c_A} A \quad (23)$$

where

$$A = \sum_{k=1}^{M(T)} \overline{G_{c_i}(t, \tau_k)} = \sum_{k=1}^{M(T)} \left((G_{c_i}(t, \tau_k))^{(0)} + \frac{1}{2} \sum_{p=1}^{N_r} \sum_{q=1}^{N_r} (G_{c_i}(t, \tau_k))_{r_p r_q}^{(II)} \overline{r'_p r'_q} \right) \quad (24)$$

The expression for the expectation of the product of concentration at two different times and locations is written as

$$\overline{c_i(t_1) c_j(t_2)} = E[c_i(t_1) c_j(t_2)] = E \left[\sum_{k=1}^{M(T)} \sum_{l=1}^{M(T)} c_{A_k} c_{A_l} G_{c_i}(t_1, \tau_k) G_{c_j}(t_2, \tau_l) \right] \quad (25)$$

Due to the fact that $\overline{c_{A_k} c_{A_l}} = (\overline{c_A})^2 + \sigma_{c_A}^2 \delta_{kl}$, the above equation is decomposed as

$$\overline{c_i(t_1) c_j(t_2)} = \sigma_{c_A}^2 \sum_{k=1}^{M(T)} \overline{G_{c_i}(t_1, \tau_k) G_{c_j}(t_2, \tau_k)} + (\overline{c_A})^2 \sum_{k=1}^{M(T)} \sum_{l=1}^{M(T)} \overline{G_{c_i}(t_1, \tau_k) G_{c_j}(t_2, \tau_l)} \quad (26)$$

In a simplified manner Equation (26) can be written as

$$\Rightarrow \overline{c_i(t_1)c_j(t_2)} = \sigma_{c_A}^2 A_1 + (\bar{c}_A)^2 A_2 \tag{27}$$

Here the terms A_1 and A_2 are obtained by substituting the expression of $\overline{G_{c_i}(t_1, \tau_k)G_{c_j}(t_2, \tau_l)}$ from Equation (22) in the first and the second terms in the right-hand side of Equation (26), respectively. Considering $k = l$ the coefficient A_1 in the first term is obtained as

$$\begin{aligned} A_1 = & \sum_{k=1}^{M(T)} \left((G_{c_i}(t_1, \tau_k))^{(0)}(G_{c_j}(t_2, \tau_k))^{(0)} + \frac{1}{2}(G_{c_i}(t_1, \tau_k))^{(0)} \left(\sum_{p=1}^{N_r} \sum_{q=1}^{N_r} (G_{c_j}(t_2, \tau_k))_{r_p r_q}^{(II)} \overline{r'_p r'_q} \right) \right. \\ & + \left. \frac{1}{2} \left(\sum_{p=1}^{N_r} \sum_{q=1}^{N_r} (G_{c_i}(t_1, \tau_k))_{r_p r_q}^{(II)} \overline{r'_p r'_q} \right) (G_{c_j}(t_2, \tau_k))^{(0)} \right) \\ & + \sum_{p=1}^{N_r} \sum_{q=1}^{N_r} ((G_{c_i}(t_1, \tau_k))_{r_p}^{(I)}(G_{c_j}(t_2, \tau_k))_{r_q}^{(I)} \overline{r'_p r'_q}) \end{aligned} \tag{28}$$

Substituting the expression of $\overline{G_{c_i}(t_1, \tau_k)G_{c_j}(t_2, \tau_l)}$ in the second term of Equation (26) and then rearranging based on possible decoupling of the double summation, the coefficient A_2 is obtained as

$$\begin{aligned} A_2 = & \left(\sum_{k=1}^{M(T)} (G_{c_i}(t_1, \tau_k))^{(0)} \right) \left(\sum_{k=1}^{M(T)} (G_{c_j}(t_2, \tau_k))^{(0)} \right) + \frac{1}{2} \left(\sum_{k=1}^{M(T)} (G_{c_i}(t_1, \tau_k))^{(0)} \right) \\ & \times \left(\sum_{k=1}^{M(T)} \left(\sum_{p=1}^{N_r} \sum_{q=1}^{N_r} (G_{c_j}(t_2, \tau_k))_{r_p r_q}^{(II)} \overline{r'_p r'_q} \right) \right) + \frac{1}{2} \left(\sum_{k=1}^{M(T)} \left(\sum_{p=1}^{N_r} \sum_{q=1}^{N_r} (G_{c_i}(t_1, \tau_k))_{r_p r_q}^{(II)} \overline{r'_p r'_q} \right) \right) \\ & \times \left(\sum_{k=1}^{M(T)} (G_{c_j}(t_2, \tau_k))^{(0)} \right) + \sum_{k=1}^{M(T)} \sum_{l=1}^{M(T)} \left(\sum_{p=1}^{N_r} \sum_{q=1}^{N_r} (G_{c_i}(t_1, \tau_k))_{r_p}^{(I)}(G_{c_j}(t_2, \tau_l))_{r_q}^{(I)} \overline{r'_p r'_q} \right) \end{aligned} \tag{29}$$

The cross covariance for concentration at two different times and locations is obtained as

$$\begin{aligned} \overline{c'_i(t_1)c'_j(t_2)} &= \overline{c_i(t_1)c_j(t_2)} - \bar{c}_i(t_1)\bar{c}_j(t_2) \\ &= \sigma_{c_A}^2 A_1 + (\bar{c}_A)^2 A_2 - (\bar{c}_A A)^2 \\ &= \sigma_{c_A}^2 A_1 + (\bar{c}_A)^2 \sum_{k=1}^{M(T)} \sum_{l=1}^{M(T)} \left(\sum_{p=1}^{N_r} \sum_{q=1}^{N_r} (G_{c_i}(t_1, \tau_k))_{r_p}^{(I)}(G_{c_j}(t_2, \tau_l))_{r_q}^{(I)} \overline{r'_p r'_q} \right) \\ &= \sigma_{c_A}^2 A_1 + (\bar{c}_A)^2 \sum_{k=1}^{M(T)} \sum_{l=1}^{M(T)} CV_{G_{ij}}(t_1 - \tau_k, t_2 - \tau_l) \end{aligned} \tag{30}$$

where $CV_{G_{ij}}(t_1 - \tau_k, t_2 - \tau_l)$ is the covariance function of the concentration with time due to unit pulse. In deriving the random component of the concentration, the term $r'_p r'_q - \overline{r'_p r'_q}$ has

been ignored, which results in the first-order accurate covariance matrix of concentration. The standard deviation of concentration, $\sigma_{c_i}(t) = \sqrt{c_i(t)c_i(t)}$ increases due to additive effect of the source uncertainty. If the unit response function at any location in the domain is obtained using SFEM for the specified unit pulse source condition, the mean and covariance of concentration for a random source condition at that location can be obtained using Equations (23) and (30), respectively. These expressions of mean and covariance of concentration are also applicable when the source is a random continuous process. The covariance of source concentration at different times $\overline{c_{A_k}c_{A_l}}$ is obtained from the known mean and autocorrelation function of the random source condition.

4.2. Random time interval

In the case of random time interval between injection of source events, the statistical moments are obtained using the probability density function of the random time interval. When the time intervals follow exponential distribution the random process is known as Poisson process. The output variable, i.e. concentration at any specific location is called a filtered Poisson process. The probabilistic structure of the filtered Poisson process may be derived by using the characteristic functional [34, pp. 87–89], when the response function is deterministic. When the system parameters are assumed to vary randomly in space, the derivation of the statistics through the characteristic functional becomes very complicated. In the present study, the expressions of the mean and the cross covariance of the filtered Poisson process are derived directly using the probability density function of the random time interval.

In this section the source is modelled as a Poisson process, i.e. the arrival time of the random mass releases ($\tau_1, \tau_2, \tau_3, \dots$ where $0 < \tau_1 < \tau_2 < \tau_3 < \dots$), are random variables. The joint probability density function of the random vector ($\{\tau\}_m = [\tau_1, \tau_2, \tau_3, \dots, \tau_m]^T$) is given as

$$p(\{\tau\}_m) = \exp\left(\int_0^{\tau_m} \lambda_p(\tau) d\tau\right) \prod_{k=1}^m \lambda_p(\tau_k) \quad (31)$$

and the marginal probability density function of τ_k follows the Gamma distribution, which is given as

$$p(\tau_k) = \lambda_p(\tau_k) \exp\left(\int_0^{\tau_k} \lambda_p(\tau) d\tau\right) \frac{\left(\int_0^{\tau_k} \lambda_p(\tau) d\tau\right)^{k-1}}{(k-1)!} \quad (32)$$

Here $\lambda_p(\tau)$ is the time-varying intensity of the Poisson events, i.e. the expected number of events of mass injection per unit of time. In the case when the number of events in the interval $[0, T]$ is exactly the same as m , the conditional probability density function of the arrival times, ($0 < \tau_1 < \tau_2 < \tau_3 < \dots < \tau_m \leq T$), is given as

$$p(\{\tau\}_m | M(T) = m) = \frac{m!}{\left(\int_0^T \lambda_p(\tau) d\tau\right)^m} \prod_{k=1}^m \lambda_p(\tau_k) \quad (33)$$

and the conditional marginal probability density fluctuation of τ_k follows the Beta distribution, which is given as

$$p(\tau_k | M(T) = m) = \lambda_p(\tau_k) \frac{\left(\int_0^{\tau_k} \lambda_p(\tau) d\tau\right)^{k-1}}{(k-1)!} \frac{\left(\int_{\tau_k}^T \lambda_p(\tau) d\tau\right)^{m-k}}{(m-k)!} \frac{m!}{\left(\int_0^T \lambda_p(\tau) d\tau\right)^m} \quad (34)$$

It is assumed that the instantaneous amount of mass release follows the normal distribution. The mean concentration can be derived as

$$\bar{c}_i(t) = E[c_i(t)] = E \left[\sum_{k=1}^{M(T)} c_{A_k} G_{c_i}(t, \tau_k) \right] = \sum_{m=0}^{\infty} P_M(m, T) E \left[\sum_{k=1}^m c_{A_k} G_{c_i}(t, \tau_k) \mid M(T) = m \right] \quad (35)$$

In the above expression $P_M(m, T)$ is the probability density function of the number of releases of solute mass in the time interval T . The conditional expectation on the right-hand side of the above equation can be simplified and can be given as follows:

$$\begin{aligned} E \left[\sum_{k=1}^m c_{A_k} G_{c_i}(t, \tau_k) \mid M(T) = m \right] &= E \left[\sum_{k=1}^m E[c_{A_k} G_{c_i}(t, \tau_k) \mid M(T) = m] \right] \\ &= \bar{c}_A E \left[\sum_{k=1}^m \bar{G}_{c_i}(t, \tau_k) \mid M(T) = m \right] \end{aligned} \quad (36)$$

Using the joint conditional pdf of the arrival time, the conditional expectation is obtained as

$$\begin{aligned} &E \left[\sum_{k=1}^m \bar{G}_{c_i}(t, \tau_k) \mid M(T) = m \right] \\ &= \int_0^T d\tau_1 \int_{\tau_1}^T d\tau_2 \cdots \int_{\tau_{m-1}}^T \sum_{k=1}^m \bar{G}_{c_i}(t, \tau_k) \frac{m! \prod_{l=1}^m \lambda_p(\tau_l)^m}{(\int_0^T \lambda_p(\tau) d\tau)^m} d\tau_m \\ &= \frac{1}{m!} \int_0^T d\tau_1 \int_0^T d\tau_2 \cdots \int_0^T \sum_{k=1}^m \bar{G}_{c_i}(t, \tau_k) \frac{m! \prod_{l=1}^m \lambda_p(\tau_l)}{(\int_0^T \lambda_p(\tau) d\tau)^m} d\tau_m \\ &= \sum_{k=1}^m \int_0^T d\tau_1 \int_0^T d\tau_2 \cdots \int_0^T \bar{G}_{c_i}(t, \tau_k) \frac{\prod_{l=1}^m \lambda_p(\tau_l)}{(\int_0^T \lambda_p(\tau) d\tau)^m} d\tau_m \end{aligned} \quad (37)$$

Since the function $\bar{G}_{c_i}(t, \tau_k)$ is same for all k , the integral $\sum_{k=1}^m \bar{G}_{c_i}(t, \tau_k)$ is symmetric about its arguments, $(\tau_1, \tau_2, \tau_3, \dots, \tau_m)$. Due to this fact the domain of the multiple integration has been transformed to a m -dimensional hyper-cube. The integration with respect to each variable can be decoupled. It can be noted that for any k in the last expression of Equation (37), the integration with respect to $\tau_l; l \neq k$ boils down to $\int_0^T \lambda_p(\tau) d\tau$ and the integration with respect to τ_k is similar to $\int_0^T \lambda_p(\tau) \bar{G}_{c_i}(t, \tau) d\tau$. Thus, in the above expression the integration is invariant of k . In a simplified manner the conditional expectation can be expressed as

$$E \left[\sum_{k=1}^m \bar{G}_{c_i}(t, \tau_k) \mid M(T) = m \right] = \sum_{k=1}^m \frac{(\int_0^T \lambda_p(\tau) d\tau)^{m-1} \int_0^T \lambda_p(\tau) \bar{G}_{c_i}(t, \tau) d\tau}{(\int_0^T \lambda_p(\tau) d\tau)^m} = \frac{mA}{\int_0^T \lambda_p(\tau) d\tau} \quad (38)$$

where

$$A = \int_0^T \lambda_p(\tau) \bar{G}_{c_i}(t, \tau) d\tau = \int_0^T \lambda_p(\tau) \left((G_{c_i}(t, \tau))^{(0)} + \frac{1}{2} \sum_{p=1}^{N_p} \sum_{q=1}^{N_q} (G_{c_i}(t, \tau))_{r_p r_q}^{(II)} \overline{r'_p r'_q} \right) d\tau \quad (39)$$

Substituting the probability

$$P(m, T) = \frac{1}{m!} \exp\left(-\int_0^T \lambda_p(\tau) d\tau\right) \left(\int_0^T \lambda_p(\tau) d\tau\right)^m$$

and the conditional expectation (Equation (38)) in Equation (35), the mean concentration is obtained as

$$\begin{aligned} \bar{c}_i(t) &= \sum_{m=0}^{\infty} \frac{1}{m!} \exp\left(-\int_0^T \lambda_p(\tau) d\tau\right) \left(\int_0^T \lambda_p(\tau) d\tau\right)^m \frac{m\bar{c}_A A}{\int_0^T \lambda_p(\tau) d\tau} \\ &= (\bar{c}_A A) \exp\left(-\int_0^T \lambda_p(\tau) d\tau\right) \sum_{m=1}^{\infty} \frac{\left(\int_0^T \lambda_p(\tau) d\tau\right)^{m-1}}{(m-1)!} = \bar{c}_A A \end{aligned} \quad (40)$$

The expectation of the product of the concentration at two different times and locations can be written as

$$\begin{aligned} \overline{c_i(t_1)c_j(t_2)} &= E[c_i(t_1)c_j(t_2)] = E\left[\sum_{k=1}^{M(T)} c_{A_k} G_{c_i}(t_1, \tau_k) \sum_{l=1}^{M(T)} c_{A_l} G_{c_j}(t_2, \tau_l)\right] \\ &= \sum_{m=0}^{\infty} P_M(m, T) E\left[\sum_{k=1}^m \sum_{l=1}^m c_{A_k} c_{A_l} G_{c_i}(t_1, \tau_k) G_{c_j}(t_2, \tau_l) \middle| M(T) = m\right] \end{aligned} \quad (41)$$

In order to derive the expectation of the term in expression (Equation (41)) for $k=l$, both τ_k and τ_l have to be treated as a single random variable and similarly for c_{A_k} and c_{A_l} . The expectation of the summation of the terms where $k=l$ can be derived following the similar steps as discussed for mean concentration. The expectation is written as

$$\begin{aligned} &E\left[\sum_{k=1}^m c_{A_k}^2 G_{c_i}(t_1, \tau_k) G_{c_j}(t_2, \tau_k) \middle| M(T) = m\right] \\ &= E\left[\sum_{k=1}^m E[c_{A_k}^2 G_{c_i}(t_1, \tau_k) G_{c_j}(t_2, \tau_k)] \middle| M(T) = m\right] \\ &= ((\bar{c}_A)^2 + \sigma_{c_A}^2) E\left[\sum_{k=1}^m \overline{G_{c_i}(t_1, \tau_k) G_{c_j}(t_2, \tau_k)} \middle| M(T) = m\right] \\ &= ((\bar{c}_A)^2 + \sigma_{c_A}^2) \frac{mA_1}{\int_0^T \lambda_p(\tau) d\tau} \end{aligned} \quad (42)$$

Here it can be proved following the same approach as discussed for the mean concentration that the conditional expectation is also independent of k and the coefficient A_1 is written as,

$$A_1 = \int_0^T \lambda_p(\tau) \overline{G_{c_i}(t_1, \tau) G_{c_j}(t_2, \tau)} d\tau \quad (43)$$

After substituting the expression of $\overline{G_{c_i}(t_1, \tau)G_{c_j}(t_2, \tau)}$ from Equation (22), A_1 can be explicitly written as

$$\begin{aligned}
 A_1 = & \int_0^T \lambda_p(\tau) \left((G_{c_i}(t_1, \tau))^{(0)}(G_{c_j}(t_2, \tau))^{(0)} + \frac{1}{2}(G_{c_i}(t_1, \tau))^{(0)} \right. \\
 & \times \left. \left(\sum_{p=1}^{N_r} \sum_{q=1}^{N_r} (G_{c_j}(t_2, \tau))_{r_p r_q}^{(II)} \overline{r'_p r'_q} \right) \right. \\
 & + \frac{1}{2} \left. \left(\sum_{p=1}^{N_r} \sum_{q=1}^{N_r} (G_{c_i}(t_1, \tau))_{r_p r_q}^{(II)} \overline{r'_p r'_q} \right) (G_{c_j}(t_2, \tau))^{(0)} \right. \\
 & \left. + \sum_{p=1}^{N_r} \sum_{q=1}^{N_r} ((G_{c_i}(t_1, \tau))_{r_p}^{(I)}(G_{c_j}(t_2, \tau))_{r_q}^{(I)} \overline{r'_p r'_q}) \right) d\tau \tag{44}
 \end{aligned}$$

In Equation (41) the terms where $k \neq l$, τ_k and τ_l should be treated as two different random variables. The conditional expectation of these terms is derived as follows:

$$\begin{aligned}
 E \left[\sum_{k=1}^m \sum_{l=1; l \neq k}^m c_{A_k} c_{A_l} G_{c_i}(t_1, \tau_k) G_{c_j}(t_2, \tau_l) \middle| M(T) = m \right] \\
 = (\bar{c}_A)^2 E \left[\sum_{k=1}^m \sum_{l=1; l \neq k}^m \overline{G_{c_i}(t_1, \tau_k) G_{c_j}(t_2, \tau_l)} \middle| M(T) = m \right] \tag{45}
 \end{aligned}$$

It can be noted that the number of terms within the double summation in Equation (45) is $m(m - 1)$. Using a similar approach as shown in Equation (37) for the multiple integration, the conditional expectation in the above equation can be derived. It can also be shown that the multiple integration of each terms becomes invariant to both k and l . Hence, the expectation of the summation of $m(m - 1)$ terms can be expressed as

$$(\bar{c}_A)^2 E \left[\sum_{k=1}^m \sum_{l=1; l \neq k}^m \overline{G_{c_i}(t_1, \tau_k) G_{c_j}(t_2, \tau_l)} \middle| M(T) = m \right] = (\bar{c}_A)^2 \frac{m(m - 1)A_2}{\left(\int_0^T \lambda_p(\tau) d\tau\right)^2} \tag{46}$$

where A_2 is given as

$$A_2 = \int_0^T \int_0^T \lambda_p(\tau_1) \lambda_p(\tau_2) \overline{G_{c_i}(t_1, \tau_1) G_{c_j}(t_2, \tau_2)} d\tau_1 d\tau_2 \tag{47}$$

After substituting the expression of $\overline{G_{c_i}(t_1, \tau_1)G_{c_j}(t_2, \tau_2)}$ from Equation (22) and decoupling the double integration for the terms wherever possible, A_2 can be explicitly written as

$$\begin{aligned}
 A_2 = & \left(\int_0^T \lambda_p(\tau) (G_{c_i}(t_1, \tau))^{(0)} d\tau \right) \left(\int_0^T \lambda_p(\tau) (G_{c_j}(t_2, \tau))^{(0)} d\tau \right) \\
 & + \frac{1}{2} \left(\int_0^T \lambda_p(\tau) (G_{c_i}(t_1, \tau))^{(0)} d\tau \right)
 \end{aligned}$$

$$\begin{aligned}
& \times \left(\int_0^T \lambda_p(\tau) \left(\sum_{p=1}^{N_r} \sum_{q=1}^{N_r} (G_{c_j}(t_2, \tau))_{r_p r'_q}^{(II)} \overline{r'_p r'_q} \right) d\tau \right) \\
& + \frac{1}{2} \left(\int_0^T \lambda_p(\tau) \left(\sum_{p=1}^{N_r} \sum_{q=1}^{N_r} (G_{c_i}(t_1, \tau))_{r_p r'_q}^{(II)} \overline{r'_p r'_q} \right) d\tau \right) \\
& \times \left(\int_0^T \lambda_p(\tau) (G_{c_j}(t_2, \tau))^{(0)} d\tau \right) + \int_0^T \int_0^T \lambda_p(\tau_1) \lambda_p(\tau_2) \\
& \times \sum_{p=1}^{N_r} \sum_{q=1}^{N_r} ((G_{c_i}(t_1, \tau_1))_{r_p}^{(I)} (G_{c_j}(t_2, \tau_2))_{r'_q}^{(I)} \overline{r'_p r'_q}) d\tau_1 d\tau_2
\end{aligned} \tag{48}$$

Using Equations (42)–(48), the Equation (41) can be simplified as

$$\begin{aligned}
\overline{c_i(t_1)c_j(t_2)} &= \sum_{m=0}^{\infty} \frac{1}{m!} \exp\left(-\int_0^T \lambda_p(\tau) d\tau\right) \left(\int_0^T \lambda_p(\tau) d\tau\right)^m \\
& \times \left(\frac{m((\bar{c}_A)^2 + \sigma_{c_A}^2)A_1}{\int_0^T \lambda_p(\tau) d\tau} + \frac{m(m-1)(\bar{c}_A)^2 A_2}{(\int_0^T \lambda_p(\tau) d\tau)^2} \right) \\
& = ((\bar{c}_A)^2 + \sigma_{c_A}^2)A_1 + (\bar{c}_A)^2 A_2
\end{aligned} \tag{49}$$

The cross covariance of the concentration in this case is obtained as

$$\begin{aligned}
\overline{c'_i(t_1)c'_j(t_2)} &= \overline{c_i(t_1)c_j(t_2)} - \bar{c}_i(t_1)\bar{c}_j(t_2) = ((\bar{c}_A)^2 + \sigma_{c_A}^2)A_1 + (\bar{c}_A)^2 A_2 - (\bar{c}_A A)^2 \\
& = ((\bar{c}_A)^2 + \sigma_{c_A}^2)A_1 + (\bar{c}_A)^2 \int_0^T \int_0^T \lambda_p(\tau_1) \lambda_p(\tau_2) \\
& \quad \times \sum_{p=1}^{N_r} \sum_{q=1}^{N_r} ((G_{c_i}(t_1, \tau_1))_{r_p}^{(I)} (G_{c_j}(t_2, \tau_2))_{r'_q}^{(I)} \overline{r'_p r'_q}) d\tau_1 d\tau_2 \\
& = ((\bar{c}_A)^2 + \sigma_{c_A}^2)A_1 + (\bar{c}_A)^2 \int_0^T \int_0^T \lambda_p(\tau_1) \lambda_p(\tau_2) \text{CV}_{G_{ij}} \\
& \quad \times (t_1 - \tau_1, t_2 - \tau_2) d\tau_1 d\tau_2
\end{aligned} \tag{50}$$

The additional terms in the expressions of the mean of the concentration (Equations (23) and (40)) and the cross covariance of the concentration (Equations (30) and (50)) are derived using SFEM based on the covariance of random parameters and derivatives of concentration with respect to discretized random parameters. These terms include the effect of uncertainty in the governing parameters on the probabilistic structure of the concentration. The terms in the expressions of the statistical parameters of the concentration due to uniform and random time intervals are very similar. The summation in the case of uniform time intervals are replaced by integration with respect to time.

In the MCSM, for each realization the response function ($G_{c_i}(t, 0)$) due to a unit solute mass injection is obtained using a generated random vector of the discretized hydraulic conductivity field. A sequence of random solute mass and associated injection times are generated based on the specified statistical parameters of the source, to compute the concentration using Equation (18). The ensemble mean and standard deviation of the concentration are estimated from 10000 realizations.

5. RESULTS AND DISCUSSION

5.1. 1-D problem

The model formulation is applied to a problem of transport of solutes in a 1-D porous media column. Here, a solute of unit concentration is assumed to be injected instantaneously and the transport of solute is assumed to be due to advection and dispersion. For numerical simulation all variables are made dimensionless with respect to the height of the column (H) and pore water velocity (v_d). The dimensionless concentration of the pollutant at dimensionless depth ($x_3 = \tilde{x}_3/H$) and dimensionless time ($t = v_d \tilde{t}/H$) is defined as $c(x_3, t) = \tilde{c}(\tilde{x}_3, \tilde{t})/c_0$. Further, $\alpha = \tilde{\alpha}/H$, $D_m(x_3) = \tilde{D}_m(\tilde{x}_3)/(v_d H)$ are, respectively, the dimensionless local-scale dispersivity, diffusion coefficient. Here ‘ \sim ’ corresponds to the dimensional quantity. The solute is injected at $x = 0.1$ and the concentration is measured at $x = 0.2$. The velocity for the deterministic system (homogeneous hydraulic conductivity field) is taken as $v_d = 1.0$. The values of the governing parameters are chosen as $n = 0.4$, $\rho_b k_d = 0.2$, $\alpha = 0.0025$ and $D_m = 0.0025$. The correlation length is taken as $\lambda = 0.01$. The uncertainty in the random parameters is quantified in terms of COV. The study has been performed for a range of COV of random system parameters. A constant head boundary conditions at the two ends of the column have been used for this 1-D problem. The spatial distribution of the mean and standard deviation of concentrations due to a single pulse input in a heterogeneous column ($COV_K = 0.4$) are presented in Figure 2. Figure 2(a) shows that the solute movement is slow for the heterogeneous case in comparison to the homogeneous case, since the effective velocity is less for the modelled random hydraulic conductivity field. The break through for the mean concentration obtained by SFEM and MCSM match well. The plot of standard deviation of concentration with time (Figure 2(b)) shows two peaks associated with rapid changes in the mean concentration. This is expected since σ_{G_c} is computed from the derivative of \bar{c} .

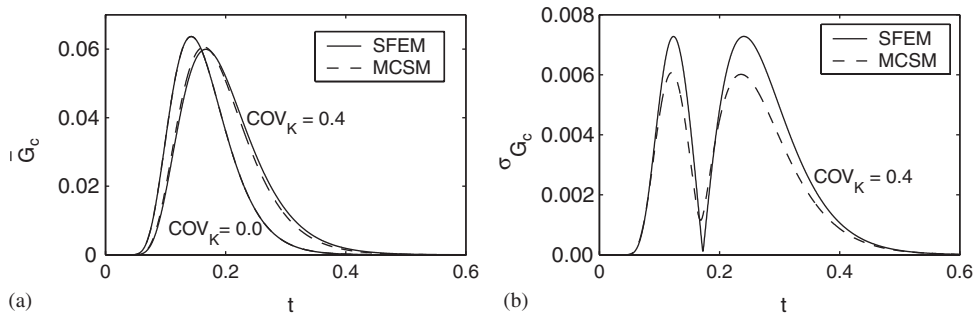


Figure 2. Comparison of mean and standard deviation of concentration obtained using SFEM and MCSM for unit mass injected.

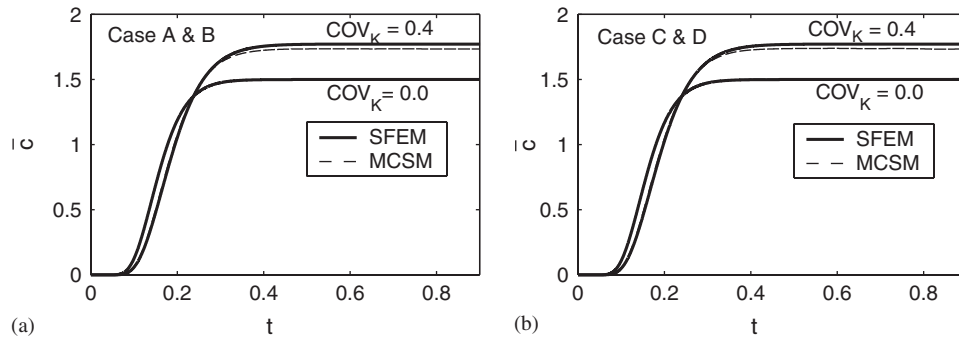


Figure 3. Comparison of mean of concentration obtained using SFEM and MCSM for various types of source conditions.

Based on the time interval and the amount of solute mass injected, the random source condition modelled is categorized into four cases as follows:

Case A—uniform time interval with fixed mass ($COV_{c_A} = 0.0$): deterministic source.

Case B—uniform time interval with random mass ($COV_{c_A} = 0.5$): random source.

Case C—random time interval with fixed mass ($COV_{c_A} = 0.0$): random Poisson-type source.

Case D—random time interval with random mass ($COV_{c_A} = 0.5$): random Poisson-type source.

The expected number of mass injections per unit time is taken as same for all the cases, $\lambda_p = 200$. The expressions in (Equations (23) and (40)) show that the mean concentration is independent of the randomness in the amount of solute mass injected. It is shown in Figures 3(a) and (b) that the mean concentration is same for both uniform and random time interval cases. If the time intervals between source injections are assumed to be large, then the mean break through of concentration shows oscillations. Since the theoretical analysis for the deterministic system with random source condition is exact, the results match with Monte Carlo simulation. As the area under the mean concentration break through (Figure 2(a)) is higher, the steady-state mean concentration happens to be higher for the heterogeneous system. The standard deviation of concentration for the deterministic source (Figure 4(a)) shows a dip similar to the single pulse input due to the random hydraulic conductivity field (Figure 2(b)). It increases monotonically with time as shown in Figure 4(b) when random amount of solute is injected at uniform time intervals into a homogeneous column. The error with standard deviation is found to be more for the case of random conductivity field. Figures 4(c) and (d) show that the standard deviation becomes significantly higher when the source is modelled as a Poisson process, i.e. the time interval follows an exponential distribution. Since the randomness in source injection dominates, the non-monotonic behaviour due to the random hydraulic conductivity, is masked. Figure 5 shows that the concentration variance increases with COV of hydraulic conductivity. The increase in variance due to randomness in the solute mass injected is contributed by the term $(\sigma_{c_A}^2 A_1)$ in the expressions of covariance (Equations (30) and (50)). Comparing Equations (30) and (50), it is interestingly observed that an additional term $(\langle \bar{c}_A \rangle^2 A_1)$ has appeared in Equation (50) due to the Poisson process. The impact of this term is quite high as seen in Figure 5. A similar effect due to various types of source conditions are observed as shown in Figure 5 for the simulations with MCSM for $COV_K = 0.4$. In addition to this, comparisons have been made for the case of multiple

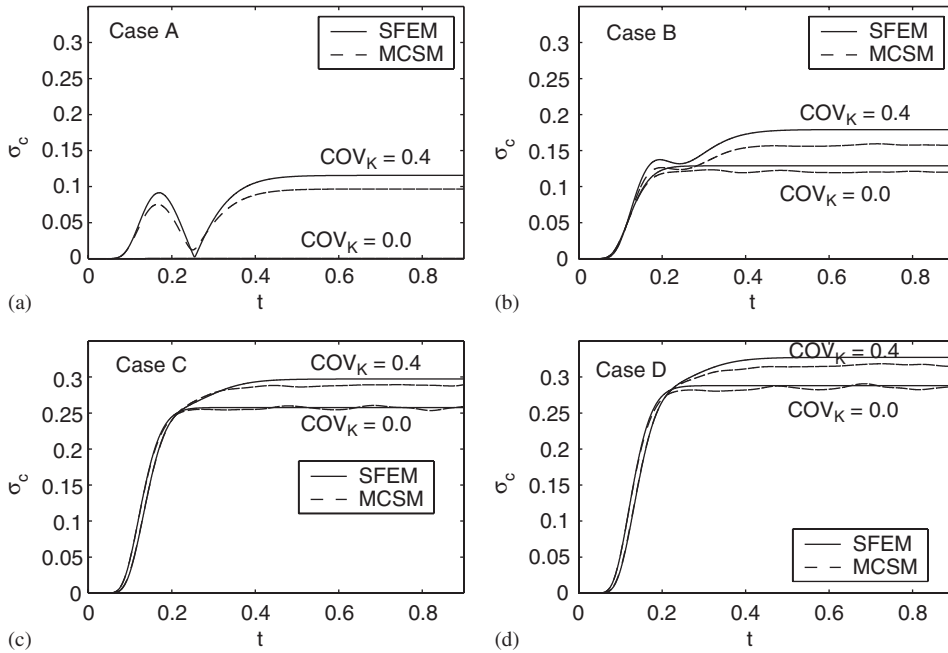


Figure 4. Comparison of standard deviation of concentration obtained using SFEM and MCSM for various types of source conditions.

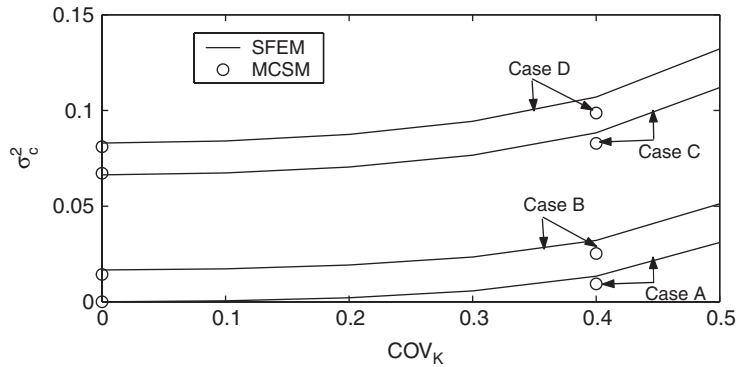


Figure 5. Variance of concentration with respect to the COV of hydraulic conductivity for Cases A–D.

random parameters (hydraulic conductivity, porosity, local dispersivity, molecular diffusion and sorption coefficient). In this case hydraulic conductivity, porosity, local dispersivity and diffusion coefficient are considered as positively correlated while sorption coefficient is negatively correlated. The reason for choosing a case with such correlation among the parameters is that it may generate higher prediction uncertainty. For the case of the perfect correlation (either positive or negative)

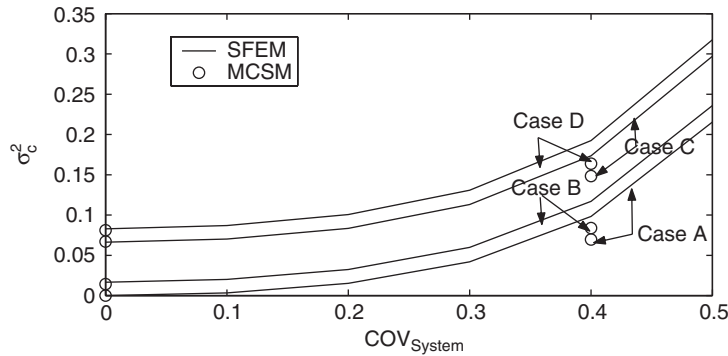


Figure 6. Variance of concentration with respect to the COV of random system parameters for Cases A–D.

case all random parameters follow the same correlation function and the correlation length. For the case of multiple random parameters (i.e. $\text{COV}_{K,n,x,D_m,k_d} = \text{COV}_{\text{system}} = 0.4$) the prediction uncertainty increases. However, for this case the comparison of results with MCSM becomes relatively inferior (Figure 6).

5.2. 3-D problem

The methodology developed in Section 4 combining SFEM with random source conditions is applied to the problem of solute transport in a 3-D heterogeneous medium with random solute source located at the top domain of the soil system in a landfill problem as illustrated in Figure 7. For the flow problem a constant head boundary condition is used along the direction of flow with no flow conditions in the other directions. It is also assumed that a vertical recharge (q) is taking place through the landfill. The solute from this source at the top of the domain is assumed to get transported through the underlying soil layers by both vertical recharge and horizontal subsurface flow. The governing parameters for flow and transport are assumed to vary randomly in space. A typical mixed flux boundary condition is used here, which is used for the 1-D vertical transport of leachate from a source [32]. It is assumed that the source has an initial specified mass of contaminant with a known concentration (c_0). The total mass of pollutant in the source decreases in time as it leaches down. Hence, a time-varying concentration boundary condition has to be applied at the source location. The top boundary condition for this problem may be given as

$$c(\mathbf{x}, t) = c_0 - \frac{1}{H_f} \int_0^t f(\mathbf{x}, \tau) d\tau \quad \text{for } -\frac{l_{x_1}}{2} \leq x_1 \leq \frac{l_{x_1}}{2}, \quad -\frac{l_{x_2}}{2} \leq x_2 \leq \frac{l_{x_2}}{2} \quad \text{and } x_3 = 0 \quad (51)$$

This boundary condition can also be rewritten as

$$f(\mathbf{x}, t) = -H_f \frac{\partial c(\mathbf{x}, t)}{\partial t} \quad \text{and } c(\mathbf{x}, 0) = c_0$$

$$\text{for } -\frac{l_{x_1}}{2} \leq x_1 \leq \frac{l_{x_1}}{2}, \quad -\frac{l_{x_2}}{2} \leq x_2 \leq \frac{l_{x_2}}{2} \quad \text{and } x_3 = 0 \quad (52)$$

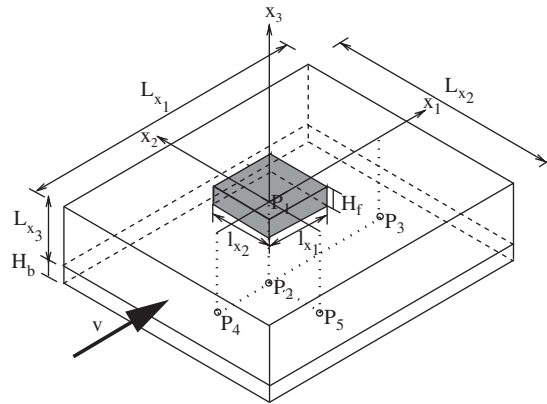


Figure 7. Schematic diagram of a 3-D domain in a landfill problem with a source at the top surface of the soil layer.

where the solute flux is expressed as, $f(\mathbf{x}, \tau) = (n(\mathbf{x})v_i(\mathbf{x})c(\mathbf{x}, t) - n(\mathbf{x})D_{ij}(\mathbf{x})\partial c(\mathbf{x}, t)/\partial x_j)n_{x_i}$. At the top surface $n_{x_1} = 0$ and $n_{x_2} = 0$. In Equation (52), H_f is the height of the leachate source. It is considered that soil is stratified below the source with upper layer close to the source having a lower permeability while the bottom layer having a higher permeability. The flow field in this problem becomes non-uniform due to the constant continuous recharge from the pollutant source combined with the lateral groundwater flow in the permeable layer. In this study, hydraulic conductivity, porosity, local dispersivity, molecular diffusion and decay coefficient are considered as positively correlated while sorption coefficient is negatively correlated. The mean and covariance of the random flow field are derived from the random hydraulic conductivity field. Along with covariance matrices of the other random fields, the covariance of velocity is also used for the probabilistic analysis of contaminant transport. A square contaminant source of dimension $l_{x_1} = l_{x_2} = l$ is assumed to be located in an aquifer. The governing equation (1), the boundary conditions and the parameters are made dimensionless with respect to the size of the source (l) and the horizontal velocity of flow (v_d). Here $c(\mathbf{x}, t) = \tilde{c}(\tilde{\mathbf{x}}, \tilde{t})/c_0$ is the dimensionless concentration of the pollutant at a dimensionless distance $\mathbf{x} = \tilde{\mathbf{x}}/l$, a dimensionless time ($t = v_d \tilde{t}/l$) and c_0 is the concentration at the top of the soil. Further, $v(\mathbf{x}) = \tilde{v}(\tilde{\mathbf{x}})/v_d$, $\alpha(\mathbf{x}) = \tilde{\alpha}(\tilde{\mathbf{x}})/l$, $D_m(\mathbf{x}) = \tilde{D}_m(\tilde{\mathbf{x}})/(v_d l)$, $\gamma_d(\mathbf{x}) = \tilde{\gamma}_d(\tilde{\mathbf{x}})/v_d$ and $q = \tilde{q}/v_d$ are, respectively, the dimensionless velocity of flow, dispersivity, molecular diffusion, decay coefficient and recharge at the top. Here v_d is the horizontal velocity of flow, for a deterministic case without any recharge. Following numerical values of parameters are chosen for solving the 3-D problem: $\bar{n} = 0.4$, $\bar{k}_d = 0.5$, $\bar{\gamma}_d = 0.4$, $q = 0.5$, $\varepsilon = 0.1$. The dimensionless thickness of the upper layer with low permeability is assumed as $L_{x_3} = 0.5$ while the thickness of the bottom layer with high permeability is taken as, $H_b = 0.01$. The horizontal dimensionless size of the domain is 11×6.5 . The height of the leachate source and ratio of conductivity are, respectively, taken as $H_f = 0.2$ and $R_K = 100$. In this study, the correlation scales along the horizontal plane are assumed to be the same (i.e. $\lambda_1 = \lambda_2 = \lambda_h$). The horizontal correlation scale (λ_h) is considered much larger in comparison to the vertical correlation scale (λ_3). The dimensionless correlation lengths with respect to the size of the source are chosen: $\lambda_h = 2.0$ and $\lambda_v = 0.5$. In this study the results are obtained for the following values of COVs of the system

parameters:

Case 1: $\text{COV}_K = 0.0$, $\text{COV}_\eta = 0.0$, $\text{COV}_\alpha = 0.0$, $\text{COV}_{D_m} = 0.0$, $\text{COV}_{K_d} = 0.0$ and $\text{COV}_{\gamma_d} = 0.0$, $\bar{\alpha} = 0.2$, $\bar{D}_m = 0.2$.

Case 2: $\text{COV}_K = 0.4$, $\text{COV}_\eta = 0.4$, $\text{COV}_\alpha = 0.4$, $\text{COV}_{D_m} = 0.4$, $\text{COV}_{K_d} = 0.4$ and $\text{COV}_{\gamma_d} = 0.4$, $\bar{\alpha} = 0.2$, $\bar{D}_m = 0.2$.

Case 3: $\text{COV}_K = 1.0$, $\text{COV}_\eta = 0.4$, $\text{COV}_\alpha = 0.4$, $\text{COV}_{D_m} = 0.4$, $\text{COV}_{K_d} = 0.4$ and $\text{COV}_{\gamma_d} = 0.4$, $\bar{\alpha} = 0.2$, $\bar{D}_m = 0.2$.

Case 4: $\text{COV}_K = 0.0$, $\text{COV}_\eta = 0.0$, $\text{COV}_\alpha = 0.0$, $\text{COV}_{D_m} = 0.0$, $\text{COV}_{K_d} = 0.0$ and $\text{COV}_{\gamma_d} = 0.0$, $\bar{\alpha} = 0.1$, $\bar{D}_m = 0.1$.

Case 5: $\text{COV}_K = 1.0$, $\text{COV}_\eta = 0.4$, $\text{COV}_\alpha = 0.4$, $\text{COV}_{D_m} = 0.4$, $\text{COV}_{K_d} = 0.4$ and $\text{COV}_{\gamma_d} = 0.4$, $\bar{\alpha} = 0.1$, $\bar{D}_m = 0.1$.

Since the dispersion parameters have large impact on the variance of concentration in heterogeneous porous media, two different sets of $\bar{\alpha}$ and \bar{D}_m have been chosen. The present FEM-based numerical methods cannot perform for very low dispersion parameters and hence moderate values are used. In addition to this, the solute transport from a landfill through the underneath porous media is sometimes dispersion dominated. The mean and the standard deviation of the concentration at various locations in the domain for a unit concentration pulse condition are the source are computed using SFEM. It may be noted that these means and standard deviations are functions of the random system parameters. Figure 8 shows a typical plot of the temporal variation of the mean and standard deviations of concentration due to unit pulse at locations: $P_1(0, 0, 0)$, $P_2(0, 0, -0.5)$, $P_3(2.5, 0, -0.5)$, $P_4(-1.0, 0, -0.5)$ and $P_5(0, -1.0, -0.5)$ (marked in Figure 7). For the locations away from the source (P_2 , P_3 , P_4 and P_5) both the mean and the standard deviation of concentration are initially zero. The behaviours of the mean and the standard deviation of concentration are observed to be quite similar to the 1-D problem. It is observed that the concentration at the location away from the source is largely influenced by the heterogeneity of the system parameters. Since the spatial variability of the hydraulic conductivity causes the reduction of the effective velocity of flow and spreading of the solutes, the concentration at the downstream locations is less and more at the upstream locations in heterogeneous case. At some locations where the mean concentration is higher for Case 2, in comparison to Case 3, the standard deviation of concentration is also found to be higher. In this case, the higher variance of conductivity mainly affects the mean velocity of flow rather than the variance of velocity. Due to this, higher variance of conductivity does not always cause the higher standard deviation of concentration. The unit response function is affected by the local dispersion parameters, as the spreading of the solute is also controlled by these parameters. The effect of heterogeneity on the distribution of mean and standard deviation of concentration is similar for different local dispersion parameters. Both the mean and the standard deviation of concentration are found to be more for Cases 4 and 5, in comparison to Cases 1 and 3. But the standard deviation increase is relatively higher for Case 5 compared to Case 3 since lower dispersion and diffusion coefficients are used. Though the degree of heterogeneity is kept the same for Cases 3 and 5, the effect of spatial variability of the parameters differs due to the values of the physical parameters. Hence, the behaviour of the stochastic unit response function is not very simple.

Analysis is extended for assessing the probabilistic behaviour of concentration distribution in the domain (Figure 7) due to various source conditions (Cases A – D) as mentioned in the section for 1-D problem as well as degrees of heterogeneity for Cases 1 – 5. A single realization of concentration break through curves at location P_1 due to multiple random pulses for Case 3B and Case 3D

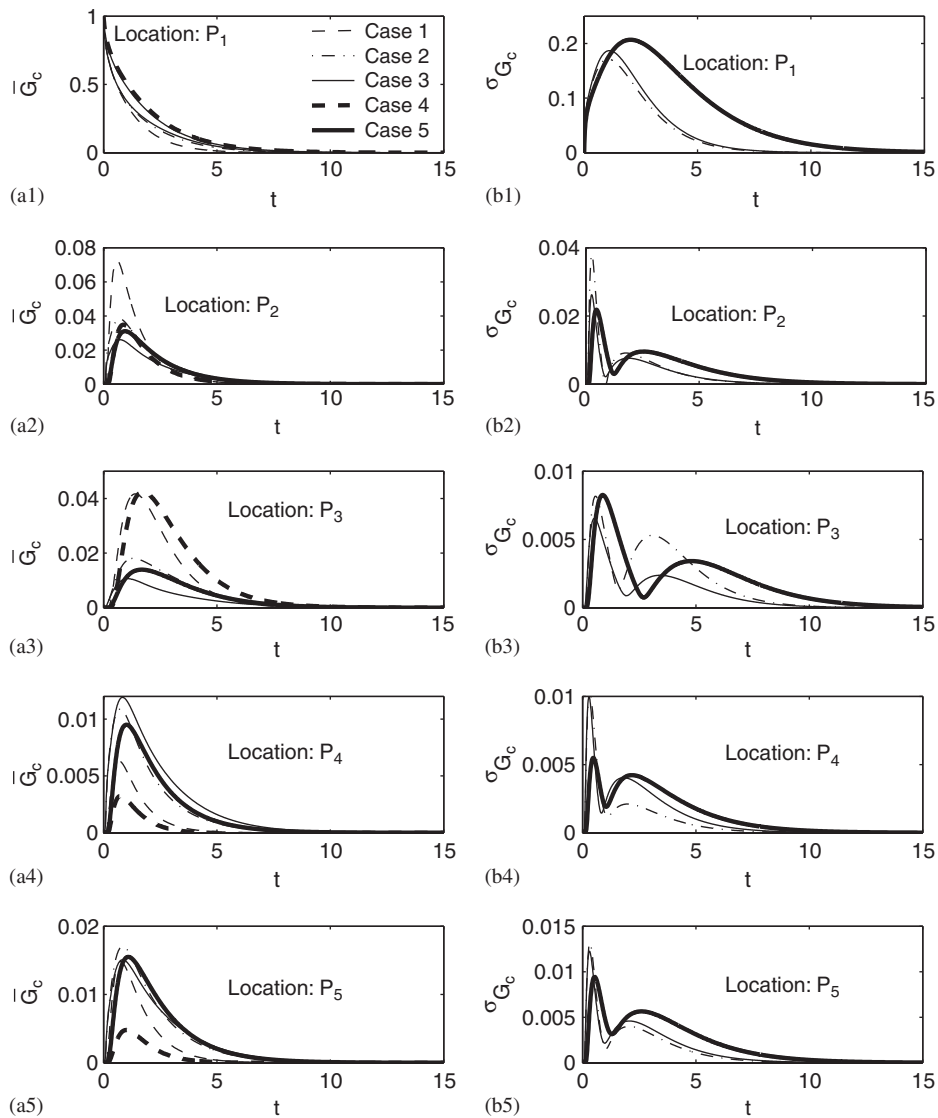


Figure 8. The temporal behaviour of mean and standard deviation of concentration at different locations for the 3-D problem (as shown in Figure 7) due to a unit concentration pulse in the leachate source.

are shown in Figures 9(a) and (b), respectively. Figures 10–14 present the behaviour of the mean and standard deviation of concentration at the upstream and the downstream locations of the source for homogeneous cases (Cases 1 and 4) and heterogeneous cases (Cases 2, 3 and 5). The mean and the standard deviation of the concentration behaviour show a pattern of reaching a steady-state condition, which is expected due to random multiple pulses as noted by Wang and Zheng [16]. For Cases A and B (i.e. when the arrival time of pulses is fixed) the mean and the standard deviation of

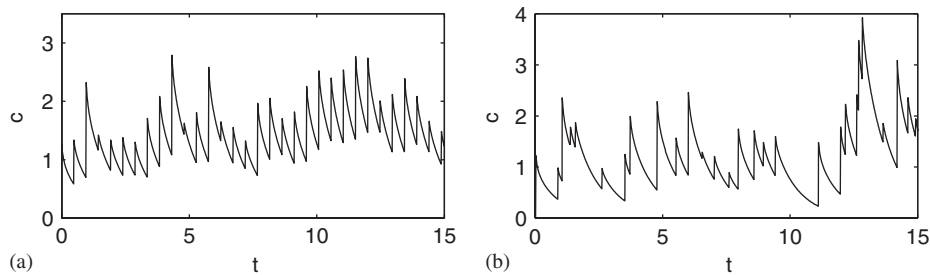


Figure 9. The concentration break through curve at the location P_1 of a single realization.

the concentration show a fluctuating response with the same frequency of the solute mass injection. The fluctuating pattern may not be distinguishable if the time interval of the mass injection is very small in comparison to the spread of the temporal break through curve of concentration for a single pulse. It has been observed that for the 1-D problem both the mean and the standard deviation in Cases A and B are quite smooth. For the random time interval cases (i.e. Cases C and D), the mean and the standard deviation of the concentration smoothly reach a steady state since the continuous integration has replaced the discrete summation for uniform time interval. When the random solute mass is injected at fixed time interval, the probability of mass injection at any instance is either 0 or 1. But for the case of random time interval the probability is a continuous function of time which is discussed in Section 4.2. From Figure 9 it can be inferred that the ensemble average of the break through curve for a uniform interval (Figure 9(a)) will have the same fluctuating pattern while for the random interval case due to the process of averaging the fluctuation will be wiped out.

For the homogeneous case (i.e. Cases 1 and 4), the second term in the expressions of covariance of concentration (Equations (30) and (50)) vanishes. In particular, Cases 1A and 4A correspond to the homogeneous system with a deterministic source and hence the standard deviation of the concentration is zero. During the comparison of the mean and the standard deviations of the concentration for Cases 1 – 3 with the random source conditions (Cases B – D), it is found that the standard deviation for the homogeneous case (Cases 1B – 1D) is higher than the heterogeneous cases at some locations (P_2 and P_3) where the mean concentration is also quite larger in homogeneous cases than in heterogeneous cases. This is more prominent for stronger random source conditions (Cases C and D). It should be noted that the second term (which is related to the covariance of the unit response function) is relatively higher for higher heterogeneity. The standard deviation of the concentration at most of the locations is higher in Case 3A than in Case 2A. As the relative effect of randomness of source condition is lesser with the increase in the degree of heterogeneity of system parameters, the gap among the various random source cases (Cases A – D) gets lesser from Case 1 to Case 3. It can be explained using the behaviours of the unit response function in Figure 8 and comparing the terms in Equations (30) and (50). From Figure 8, it can be found that for the parametric heterogeneity cases chosen, the area under the mean of concentration is larger than the area under the standard deviation of concentration. At location P_3 (far away in the downstream of the source), the standard deviation is quite smaller than the mean. In both Equations (30) and (50) the first terms are related to the mean of stochastic unit while the second terms are related to the covariance of the unit response function. For uniform time interval case (Equation (30)),

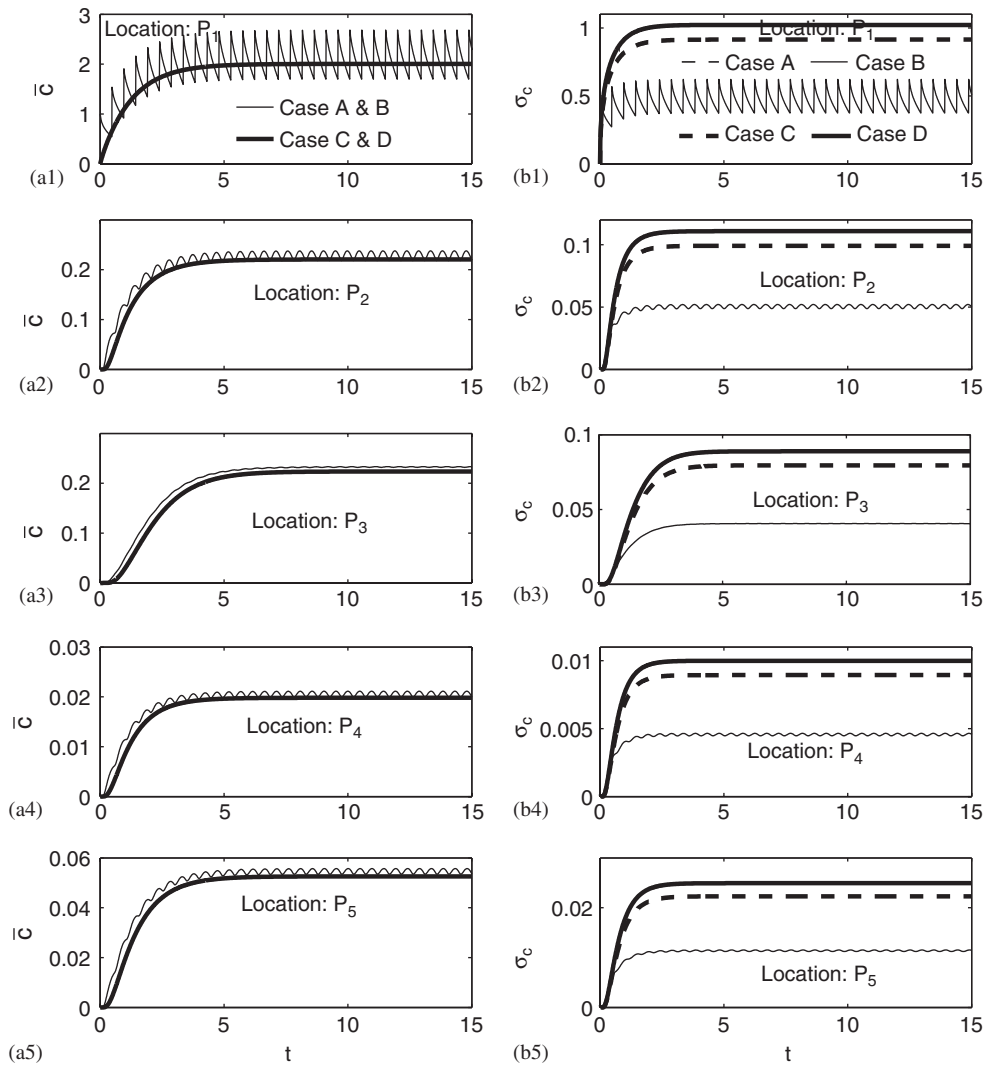


Figure 10. The temporal behaviour of mean and standard deviation of concentration at different locations for 3-D problems (as shown in Figure 7) of Case 1 due to various types of random source conditions (Cases A – D).

the first term contains $\sigma_{c_A}^2$ and the second term contains $(\bar{c}_A)^2$. Since in this study, COV_{c_A} is taken as 0.5, both the terms are comparable and prediction uncertainty of concentration increases with the degree of parametric heterogeneity. In the case of random time interval of the multiple source (Poisson process), the first term contains $((\bar{c}_A)^2 + \sigma_{c_A}^2)$ and largely dominates over the second term. Hence for Poisson process-type source case, the randomness in the source condition dominates over the heterogeneity of governing system parameters. At the locations where the unit response function is higher for homogeneous case, the standard deviation of concentration due to multiple

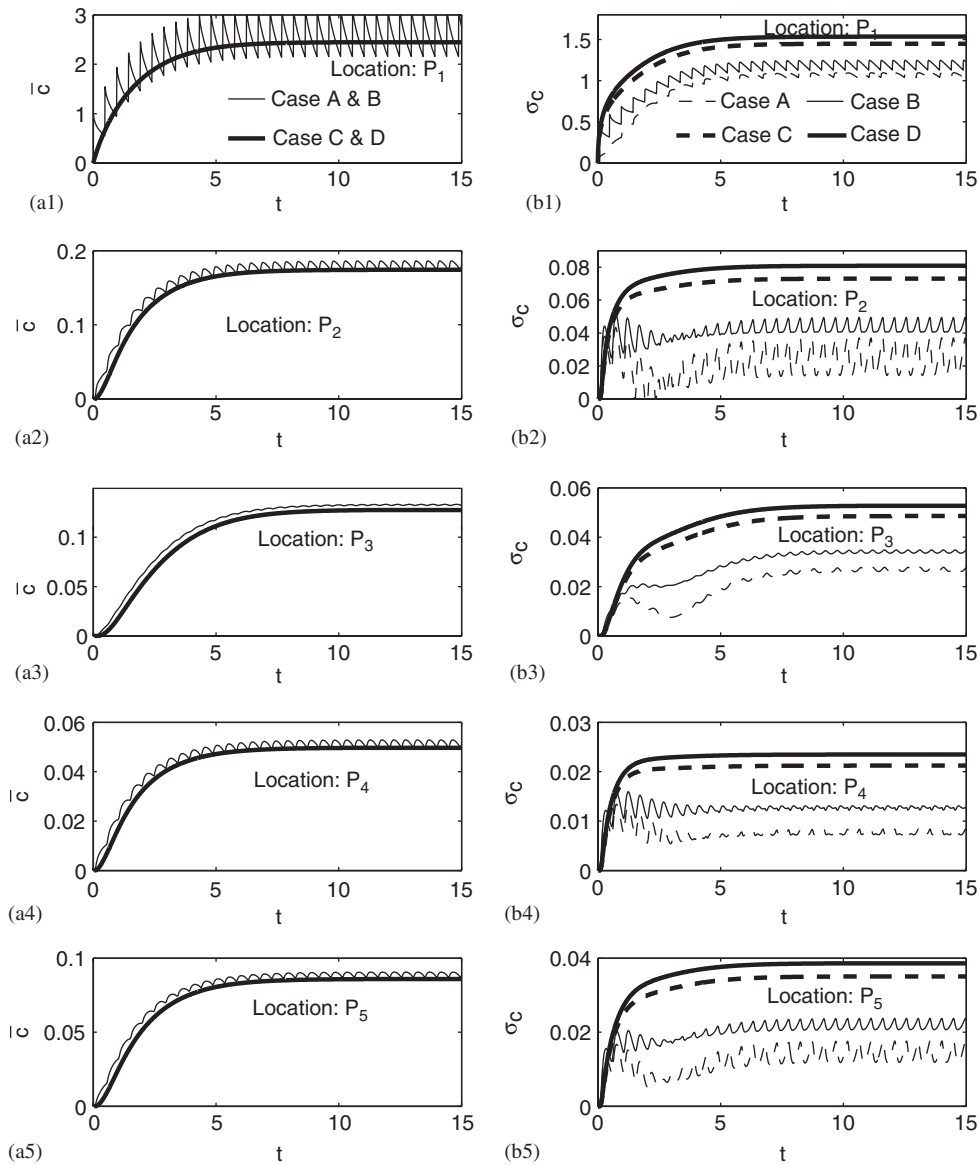


Figure 11. The temporal behaviour of mean and standard deviation of concentration at different locations for 3-D problems (as shown in Figure 7) of Case 2 due to various types of random source conditions (Cases A – D).

pulse is lesser even for heterogeneous case. Similar effects of heterogeneity are found in the cases with lower dispersion parameters (Cases 4 and 5) for various statistically different source conditions (Cases A – D).

In cases of homogeneous system parameters (Cases 1 and 4), only the mean of unit response function is affected by dispersion parameters and in these cases both mean and standard deviation

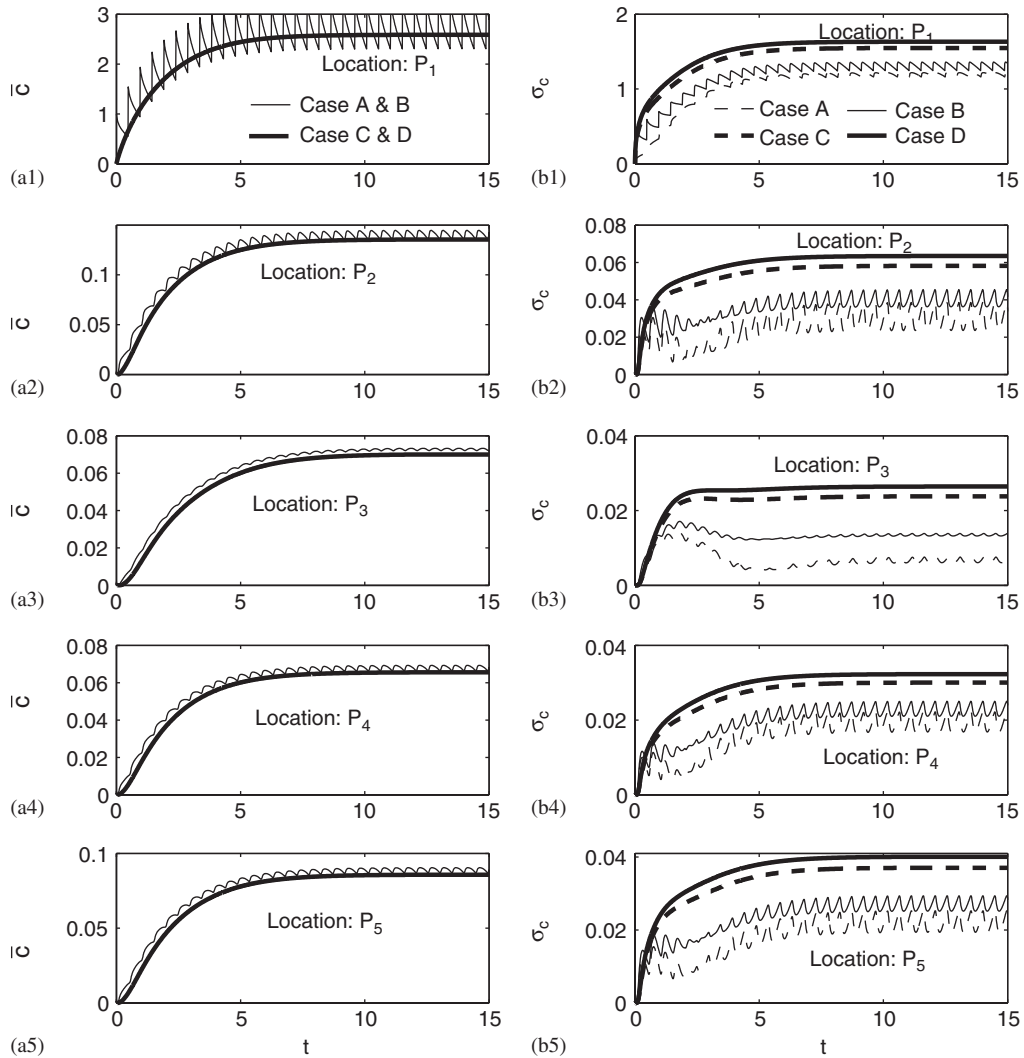


Figure 12. The temporal behaviour of mean and standard deviation of concentration at different locations for 3-D problems (as shown in Figure 7) of Case 3 due to various types of random source conditions (Cases A – D).

of concentration result from the mean of unit response function. As the unit response function is higher at locations P_1 and P_3 (Figures 8(a1) and (a3)), the mean as well the standard deviation of concentration due to random source conditions are also higher in Case 4 than in Case 1 (as shown in Figures 10 and 13). In heterogeneous case, for different random source conditions (Cases A – D) the mean concentrations in Case 5 are higher or lower at different locations compared to Case 3. This variation in mean is similar to the mean unit response functions in Cases 3 and 5. The standard deviation of concentration is affected by both mean and standard

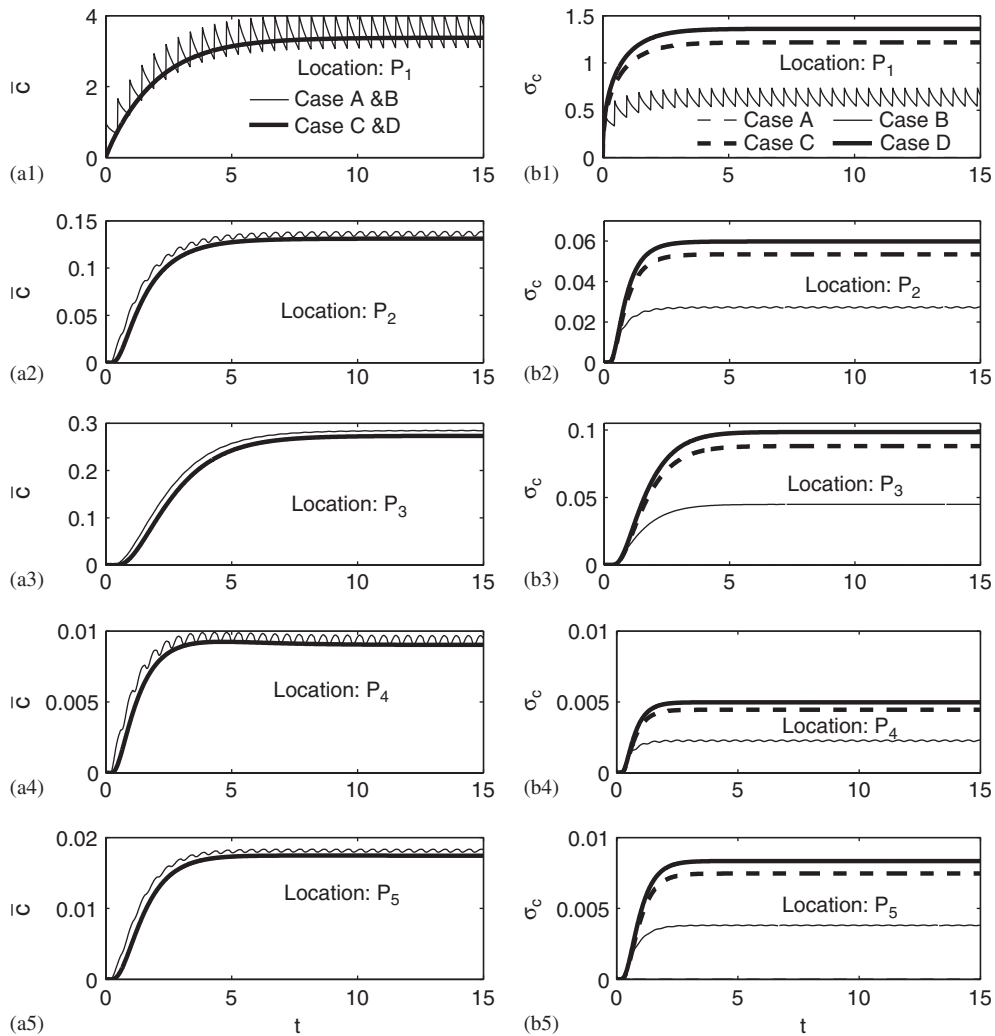


Figure 13. The temporal behaviour of mean and standard deviation of concentration at different locations for 3-D problems (as shown in Figure 7) of Case 4 due to various types of random source conditions (Cases A – D).

deviation of unit response function. Due to lower dispersion parameters, the standard deviation of concentration at all locations for all types of source conditions are higher in Case 5 compared to Case 3. For the deterministic source condition (Case A), the increase in the standard deviation from Case 3 to Case 5 is higher. In this case the first terms in the expressions of covariance (Equations (30) and (50)) are zero. For Case 5 (in comparison to Case 3), the increase in the standard deviation due to stronger uncertainty in the source condition is found to be smaller.

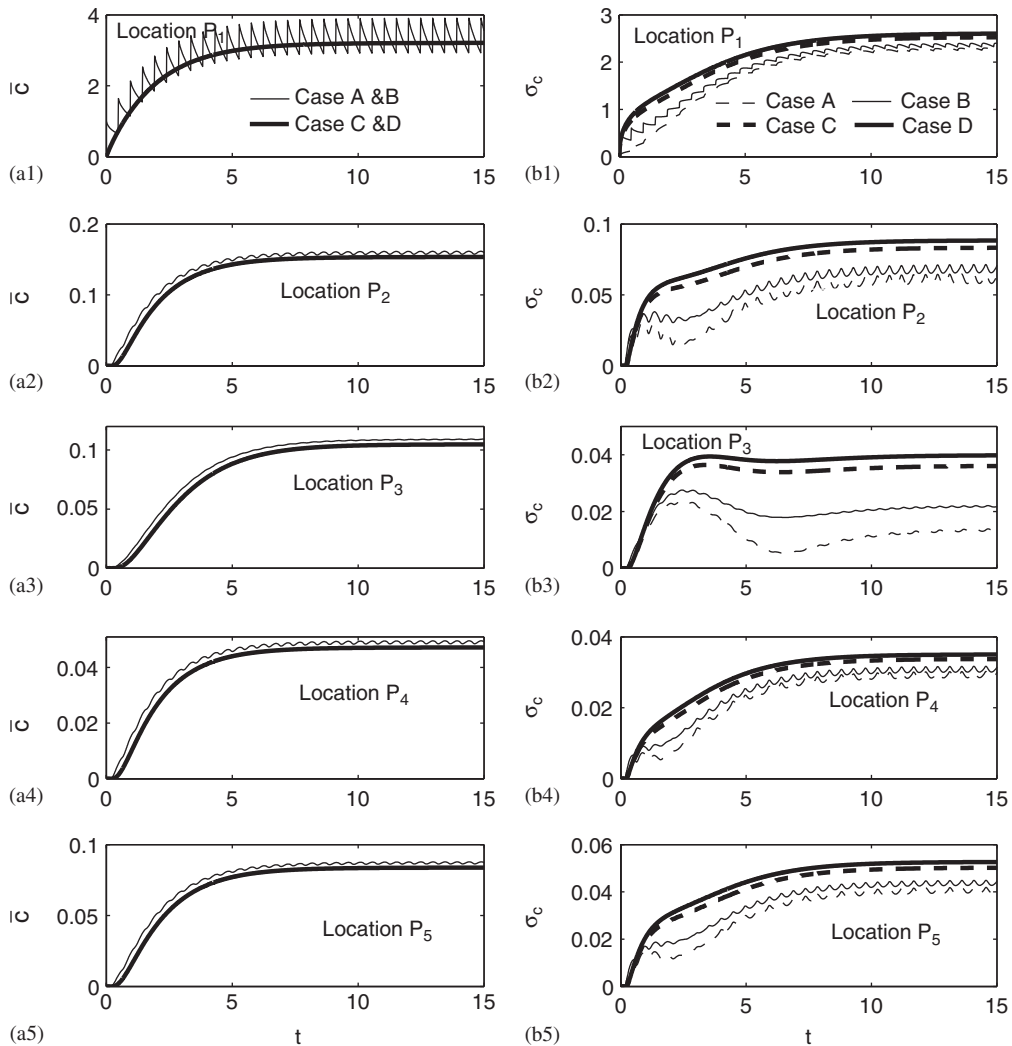


Figure 14. The temporal behaviour of mean and standard deviation of concentration at different locations for 3-D problems (as shown in Figure 7) of Case 5 due to various types of random source conditions (Cases A – D).

6. CONCLUSIONS

A semi-analytical approach is presented for the stochastic modelling of the concentration in a heterogeneous porous medium due to random inputs. This approach combines the analytical expressions for the random input conditions with the numerical methods to derive the stochastic behaviour of the unit response function. In this present study SFEM is used for the solution of SPDEs resulting from spatial heterogeneity. For mild heterogeneity (i.e. $COV \leq 1.0$) the performance of the method is found to be good.

The expressions derived for random input with a uniform and a random time interval indicate that an additional term due to randomness of time interval results in the later case, which has a significant effect on the prediction uncertainty of the concentration. This result has a practical implication while modelling the input conditions in groundwater contamination problems.

For the same degree of heterogeneity of the random system parameters, the standard deviation of the concentration increases with lower local dispersion parameters. Hence, the relative effect of source and system uncertainty depends on the values of the physical parameters.

The random spatial variability of the governing system parameters affects the distribution of the mean concentration in addition to the increase in the standard deviation. For a simple linear system, the effect of randomness in the source is additive but this is not the case with the randomness in the system parameters. Due to this fact the combined effects of randomness in the system parameters and the source condition are not straightforward.

The expressions of the mean and the covariance of the concentration have been derived analytically based on the stochastic unit response function for the case of a system with random parameters. Since no approximation has been made to derive these analytical expressions, they are exact. These are very useful for a random source condition. The errors, which have been observed during the comparison with MCSM, are due to the approximation made in the SFEM formulation to compute numerically the mean and the covariance of the stochastic unit response function. The statistical moments of the stochastic unit response function can be derived by any analytical method or any other numerical method such as the moment equation method. Any efficient and higher-order numerical method which is capable of obtaining accurately the statistical moments of concentration, in the heterogeneous porous media can be combined with the analytical expressions derived for the mean and the covariance of the concentration using a stochastic unit response function.

APPENDIX

Using a similar methodology presented for the transport problem (Section 3), the perturbation approach can also be applied on the flow problem (Equation (8)), to obtain the mean and the random perturbed components of the hydraulic head. In the case of the flow problem, the random properties are only the hydraulic conductivities of the elements K_p , ($p = 1, 2, \dots, N$) and hence the mean and the random components of the hydraulic head are expressed as,

$$\{\bar{h}\} = \left([\mathbf{I}] + \sum_{p=1}^{N_k} \sum_{q=1}^{N_k} [\bar{K}]^{-1} [K]_{K_p}^{(1)} [\bar{K}]^{-1} [K]_{K_q}^{(1)} \overline{K'_p K'_q} \right) [\bar{K}]^{-1} \{h_0\} \quad (\text{A1})$$

$$\{h\}' = \sum_{p=1}^{N_k} \{h\}'_{K_p} K'_p \quad \text{where } \{h\}'_{K_p} = -[\bar{K}]^{-1} [K]_{K_p}^{(1)} [\bar{K}]^{-1} \{h_0\} \quad (\text{A2})$$

Using (A1) and (A2) the mean and random components of the seepage flux (q_{i_p}) are written as,

$$\bar{q}_{i_p} = -\frac{1}{N_G} \sum_{k=1}^{N_G} \frac{\partial N_l(\mathbf{x})}{\partial x_i} \bigg|_{\mathbf{x}_k} \left(\bar{K}_p \bar{h}_l + \sum_{q=1}^{N_k} h_{l,K_q}^{(1)} \overline{K'_p K'_q} \right) \quad (\text{A3})$$

$$q'_{ip} = - \sum_{q=1}^{N_k} \frac{1}{N_G} \sum_{k=1}^{N_G} \frac{\partial N_l(\mathbf{x})}{\partial x_i} \bigg|_{\mathbf{x}_k} (\bar{h}_l \delta_{pq} + \bar{K}_p h_{l,K_q}^{(I)}) K'_q = \sum_{q=1}^{N_k} q_{ip,K_q}^{(I)} K'_q \tag{A4}$$

From the above expressions one can obtain the auto covariance of velocity and the cross covariance with any other random property (r'_j) using the auto covariance of the hydraulic conductivity and cross covariance of hydraulic conductivity with r'_j , which may be given as,

$$\overline{q'_{ip_1} q'_{jp_2}} = \sum_{q_1=1}^{N_k} \sum_{q_2=1}^{N_k} q_{ip_1,K_{q_1}}^{(I)} q_{jp_2,K_{q_2}}^{(I)} \overline{K'_{q_1} K'_{q_2}} \quad \text{and} \quad \overline{q'_{ip} r'_j} = \sum_{q=1}^{N_k} q_{ip,K_q}^{(I)} \overline{K'_q r'_j} \tag{A5}$$

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