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# Impact of spatial variability in hydraulic parameters on plume migration within unsaturated surficial formations

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## Abstract

Heterogeneities in textural properties of surficial formations at field scale result in spatial variations in hydraulic parameters governing unsaturated zone flow. This study aims at quantifying the influence of such variations on solute transport resulting from a localized source of radioactive contaminant at ground level.

The study focuses on three hydraulic parameters related to the Mualem-van Genuchten formalism, namely the saturated hydraulic conductivity  $K_s$ , the parameter  $\alpha$  inversely proportional to the air-entry value, and the parameter  $n$  related to the pore-size distribution. Sets of random fields accounting for spatial variability of these parameters are generated using lognormal distributions with different variances and correlation lengths. These random fields are used as inputs to an unsaturated flow and transport model to simulate radionuclide plume migration.

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Each simulated plume is characterized by its size (plume surface area), position (location of center of mass) and shape (elongation ratio) within the unsaturated zone. By comparison with the homogeneous medium,  $K_s$ -,  $\alpha$ - and  $n$ - random fields generated with the mean variances computed through the analysis of a global soil database respectively result in average in (i) 25 (variable  $K_s$ ), 20 (variable  $\alpha$ ) and 65% (variable  $n$ ) increase in plume size; (ii) 0.8, 1 and 1.8 m horizontal offsets of the plume center; and (iii) 20, 30 and 50% decrease in plume circularity. In addition, changes in the variance values within one order of magnitude appear to have critical consequences only for the  $n$  parameter.

The issue of spatial variability of hydraulic parameters is thus crucial for characterizing the evolution of pollutant plumes within an unsaturated zone and for developing better remediation strategies for industrial sites.

*Keywords:* Richards equation, Mualem-van Genuchten model, Random fields, Radionuclide migration, Sensitivity analysis.

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## 1. Introduction

Spatial heterogeneities found in porous formations at field scale impact groundwater flow and solute transport. The role of geometrical patterns, especially connected features, in concentrating flow and reducing travel time has been widely highlighted, in both saturated (Knudby and Carrera, 2005; Renard and Allard, 2013) and unsaturated (Appels et al., 2018) zones. Most of the related studies assume that such patterns mainly result from the arrangement of large-scale discrete geological structures with high contrast in hydraulic parameters (Feyen and Caers, 2006; Ye and Khaleel, 2008). The

10 spatial distribution of these parameters can be represented by either several  
11 unimodal random fields populating a discontinuous facies model (Matheron  
12 et al., 1987; Le Coz et al., 2011; Le Coz et al., 2013) or a single continuous  
13 random field whose properties are multimodal (Russo, 2012).

14 Smaller scale variability, *i.e.* within individual facies, is known to con-  
15 tribute significantly to the overall hydraulic parameters variability within  
16 the unsaturated zone (Botros et al., 2009). When considering such individ-  
17 ual facies, the corresponding parameters are generally represented by a single  
18 population whose distribution is unimodal, more particularly lognormal for  
19 the hydraulic conductivity (Fogg et al., 1998; Paleologos et al., 2015; Tan  
20 et al., 2017), and whose spatial variability can be characterized through a  
21 two-point spatial covariance (Botros et al., 2009; Russo, 2012). However,  
22 identifying a relevant covariance model requires a large amount of data at  
23 field scale (*e.g.*, Botros et al., 2009) and is a complex task because the mea-  
24 surements of hydraulic parameters that govern unsaturated zone flow are  
25 difficult and time-consuming (Shaap et al., 2004).

26 The spatial variability within individual facies is thus often neglected,  
27 for instance in most of the studies that focus on the spread of radioactive  
28 contaminants in the unsaturated zone (Skuratović et al., 2016; Testoni et  
29 al., 2017). In particular, Testoni et al. (2017) simulated the barrier effect  
30 of the unsaturated zone as a delay and capture system by considering var-  
31 ious medium configurations and initial conditions in case of an accidental  
32 release of Cesium 137. These simulations were performed by coupling of (i)  
33 a distributed one-dimensional unsaturated flow and transport model and (ii)  
34 a three-dimensional saturated flow and transport model. This coupling is

thus based on the assumption that the unsaturated water flow is mainly in the vertical direction. This commonly formulated assumption (Bugai et al., 2012; Jakimavičiūtė-Maseliienė et al., 2016) is questionable since anisotropy induced by lateral variability in hydraulic parameters can refract flow lines away from vertical (Gannon et al., 2017).

This study aims at quantifying the influence of the spatial variability in hydraulic parameters on plume migration in unsaturated surficial formations, within the framework of a punctual release of radionuclides to the subsurface. First, the degree of spatial variability in hydraulic parameters at field scale, for typical soils, is assessed based on the analysis of a global soil database. Then, numerical simulations are run using a full two-dimensional (2D) flow and transport model to reproduce lateral flow induced by such spatial variability.

## 2. Unsaturated flow in porous media

This section recalls the main parameters governing flow in unsaturated surficial formations and quantifies their spatial variability in natural environment. The variance of three of these parameters is computed at field scale based on soil texture information recorded in a global soil database.

### 2.1. Governing equation and parameterization

Variably saturated flow processes in porous media are typically described by the Richards equation:

$$\frac{\partial \theta(\psi)}{\partial t} = \nabla \cdot (K(\psi) \nabla (\psi + z)) + q_{ss} \quad (1)$$

56 where  $\theta$  is the volumetric moisture content [ $L^3.L^{-3}$ ],  $t$  is the time [T],  $K$  is  
 57 the hydraulic conductivity tensor [ $L.T^{-1}$ ],  $\psi$  is the water pressure head [L],  $z$   
 58 is the vertical coordinate directed upward [L] and  $q_{ss}$  represents distributed  
 59 source (positive) or sink (negative) terms [ $L^3.L^{-3}.T^{-1}$ ]. Solving the Richards  
 60 equation requires the moisture retention curve and the relative hydraulic  
 61 conductivity function as inputs describing the links between pressure head,  
 62 water content and relative hydraulic conductivity. These relationships are  
 63 based on a model, *e.g.*, the Mualem-van Genuchten model (Mualem, 1976;  
 64 van Genuchten, 1980):

$$\theta(\psi) = \theta_r + \frac{\theta_s - \theta_r}{(1 + |\alpha\psi|^n)^m} \quad \text{with} \quad m = 1 - \frac{1}{n} \quad (2)$$

65 and

$$K(\psi) = K_s S_e^{\frac{1}{2}} [1 - (1 - S_e^{\frac{1}{m}})^2] \quad \text{with} \quad S_e = \frac{\theta(\psi) - \theta_r}{\theta_s - \theta_r} \quad (3)$$

66 where  $\theta_r$  and  $\theta_s$  are respectively the residual and saturated volumetric wa-  
 67 ter contents [ $L^3.L^{-3}$ ],  $\alpha$  is inversely proportional to the of air-entry value [ $L^{-1}$ ],  
 68  $n$  is a pore-size distribution index [-] and  $K_s$  is the saturated hydraulic con-  
 69 ductivity tensor [ $L.T^{-1}$ ].

70 Although the hydraulic parameters are linked to physical soil proper-  
 71 ties, the direct measurement of some of them from samples is subject to  
 72 experimental limitations (Schaap et al., 2004). Many investigations are thus  
 73 undertaken to estimate hydraulic parameters using empirical relationships  
 74 deduced from more readily available data, such as soil texture and bulk den-  
 75 sity (*e.g.*, Wösten et al., 1999; Schaap et al., 2001; Saxton and Rawls, 2006;  
 76 Tóth et al., 2015; Zhang and Schaap, 2017). These relationships, commonly

referred as pedotransfer functions (PTFs), are mostly based on regression analysis of existing soil databases. The ROSETTA PTF (Schaap et al., 2001) is based on artificial neural network analysis coupled with bootstrap re-sampling which allows to estimate hydraulic parameters of the Mualem-van Genuchten model.

The parameter  $\theta_r$  can be quite precisely derived from soil texture using PTFs (Vereecken et al., 1989); both  $\theta_r$  and  $\theta_s$  can be directly obtained from measurements under extremely dry or saturated conditions at the subsurface respectively (Jadoon et al., 2012). In addition, preliminary sensitivity analyses performed in temperate climate conditions show that the spatial variability of these two parameters does not significantly influence the solute transport. The sensitivity analysis conducted in this study thus only focuses on the  $K_s$ ,  $\alpha$  and  $n$  parameters.

## 2.2. Variability of hydraulic parameters at field scale

### 2.2.1. Database analysis

The World Soil Information Service (WoSIS) aims at providing consistent harmonized (*i.e.*, georeferenced, quality-assessed and standardized) soil data on a global scale based on soil profiles compiled by the International Soil Reference and Information Center (ISRIC). The WoSIS database currently contains some 96,000 georeferenced soil profiles, described in terms of analytical and physical soil properties, among which 20% were so far quality-assessed and standardized (Batjes et al., 2017). The number of recorded data for each property varies between profiles; yet a majority of samples is described at least in term of soil texture, *i.e.* sand, silt and clay contents. These textural properties are thus used to estimate the Mualem-van Genuchten hydraulic

102 parameters by mean of the ROSETTA PTF. The soil profiles from the WoSIS  
 103 database are then grouped by locations. A location is defined as a circular  
 104 area of radius 500 m that contains at least 10 soil profiles. For each of the 49  
 105 identified locations, the mean ( $\mu$ ) and variance ( $\sigma^2$ ) of  $\log K_s$ ,  $\log \alpha$  and  $\log n$   
 106 (the parameters  $K_s$ ,  $\alpha$  and  $n$  are assumed to follow lognormal distributions)  
 107 are computed based on the whole corresponding soil samples. The Pearson  
 108 correlation coefficients ( $r$ ) are also computed between the three parameter  
 109 pairs, *i.e.*,  $\log K_s$  and  $\log \alpha$ ;  $\log K_s$  and  $\log n$ ; and  $\log \alpha$  and  $\log n$ .

### 110 2.2.2. Variances and correlations

111 On the 49 identified locations, the log-variances (which enables to work  
 112 with a dimensionless parameter  $\sigma^2$ ) of the hydraulic parameters generally  
 113 vary within about one order of magnitude (Figure 1) in relation with the  
 114 mean parameter values. Indeed,  $\sigma^2(\log K_s)$  increases from  $2.5 \cdot 10^{-2}$  (1st decile)  
 115 to  $2.5 \cdot 10^{-1}$  (9th decile) when  $\mu(K_s)$  varies from 0.1 to 0.6 m.d<sup>-1</sup>;  $\sigma^2(\log \alpha)$   
 116 decreases from  $8 \cdot 10^{-2}$  to  $8 \cdot 10^{-3}$  when  $\mu(\alpha)$  varies from 0.9 to 2.7 m<sup>-1</sup>; and  
 117  $\sigma^2(\log n)$  increases from  $8 \cdot 10^{-4}$  to  $1 \cdot 10^{-2}$  when  $\mu(n)$  varies from 1.25 to 1.65.

118 The Pearson correlation coefficients between the parameters also appear  
 119 to depend on the mean parameter values (Figure 2). For  $\mu(K_s)$  lower than  
 120 0.2 m.d<sup>-1</sup>,  $r$  values are highly scattered: from -0.2 to 0.8 between  $\log K_s$  and  
 121  $\log \alpha$ ; from -0.7 to 0.9 between  $\log K_s$  and  $\log n$ ; and from -0.9 to 0.5 between  
 122  $\log \alpha$  and  $\log n$ . For  $\mu(K_s)$  higher than 0.2 m.d<sup>-1</sup>,  $r$  values are less scattered  
 123 and tend to stabilize: from 0.3 to 0.7 between  $\log K_s$  and  $\log \alpha$ ; from 0.7 to 1  
 124 between  $\log K_s$  and  $\log n$ ; and from -0.1 to 0.6 between  $\log \alpha$  and  $\log n$ .



### 125 3. Methods

126 This section describes the modeling tools and approaches carried out for  
 127 assessing the influence of spatial variability in  $K_s$ ,  $\alpha$  and  $n$  on the migration  
 128 of a radionuclide plume in unsaturated surficial formations. A 2D hydroge-  
 129 ological flow and transport numerical model is run in simulation mode with  
 130 various input parameter fields representing distinct levels of spatial variabil-  
 131 ity.

#### 132 3.1. Flow and transport numerical model

133 The MELODIE numerical code developed by the French Institute for  
 134 Radiation Protection and Nuclear Safety (IRSN) aims at making available a  
 135 tool for evaluating the long term safety of a radioactive waste disposal facil-  
 136 ity (IRSN, 2009; Amor et al., 2014; Amor et al., 2015; Bouzid et al., 2018).  
 137 This code simulates underground flow and solute transport in saturated or  
 138 variably saturated porous media based on a mixed finite volume - finite el-  
 139 ement scheme, namely, Godunov development for the convective term and  
 140 Galerkin development for the diffusion - dispersion term (Amaziane et al.,  
 141 2008). In this study, MELODIE is set for solving in 2D (i) the Richards equa-  
 142 tion describing flow in variably saturated porous media (Eq. 1); and (ii) the  
 143 following advection-dispersion-reaction equation representing the migration  
 144 of radionuclides:

$$\nabla \cdot [(D|\vec{u}| + \omega d)\nabla C - \vec{u}C] = \omega' R \frac{\partial C}{\partial t} + \omega' \lambda RC \quad (4)$$

145 where  $C$  is the volumetric radionuclide concentration [M.L<sup>-3</sup>],  $D$  is the  
 146 dispersivity tensor [L],  $d$  is the molecular diffusion coefficient [L<sup>2</sup>.T<sup>-1</sup>],  $\vec{u}$  is

the pore water velocity [ $L.T^{-1}$ ],  $\omega$  and  $\omega'$  are respectively total and effective porosities [-],  $\lambda$  is the decay constant [ $T^{-1}$ ] and  $R$  is the retardation factor [-].

The modeling domain (Figure 3a) consists of a 2D vertical section of surficial deposits of 100 m large (x axis) by 15 m deep (z axis), discretized in triangles of base 0.5 m and height 0.25 m. The boundary conditions are defined as follows:

1. a fixed head corresponding to the mean water table elevation (7.5 m above the bottom boundary with a  $0.004 \text{ m.m}^{-1}$  lateral gradient) is set on both sides of the domain;
2. no-flow conditions are set on the bottom boundary;
3. a time variable flow corresponding to the daily percolation rate, typical from center of France, and estimated from the water balance method (Thorntwaite and Mather, 1955) is imposed on the top boundary (Figure 3b).

A point source of tritium is simulated by setting an activity of 1,000  $\text{Bq.d}^{-1}$  during one month on the top surface node located at the center of the modeling domain. The evolution of the activity within the domain is then simulated during six years with an adaptative time stepping (from  $10^{-20}$  to 1 day) by considering a retardation factor ( $R$ ) of 1 and a decay constant ( $\lambda$ ) of  $1.54.10^{-4} \text{ d}^{-1}$  (Table 1). The tritium plume is delimited by the nodes for which the simulated activity is higher than  $1 \text{ Bq.m}^{-3} \text{H}_2\text{O}$ .

### 3.2. Parameter fields

First, the internal model parameters are fixed homogeneously for the whole domain as representative of sandy loam material (Table 2). Then, sets

171 of 100 simulations are run by considering (i) one hydraulic parameter (either  
 172  $K_s$  or  $\alpha$  or  $n$ ) variable in space through sets of 100 random fields with similar  
 173 properties (*i.e.* same parameters used for the geostatistical simulations of  
 174 random fields) and (ii) three hydraulic parameters ( $K_s$  and  $\alpha$  and  $n$ ) varying  
 175 simultaneously through sets of 100 correlated random fields (Table 3).

176 Hereafter,  $Z$  is a random field referring indifferently to one of the hy-  
 177 draulic parameters under consideration. Since  $K_s$ ,  $\alpha$  and  $n$  are assumed to  
 178 follow lognormal distributions, the logarithm of  $Z$  is a gaussian random field  
 179 such that :

$$\log Z \sim \mathcal{N}(\mu, \sigma^2) \quad (5)$$

180 where  $\mu$  and  $\sigma^2$  are the mean and variance of  $\log Z$ , respectively. The var-  
 181 iogram chosen to account for the spatial variability of  $\log Z$  is the spherical  
 182 model, which was shown to fit experimental data of soil hydraulic parameters  
 183 (Herbst et al., 2006; Botros et al., 2009; Bevington et al., 2016):

$$\gamma(h) = \begin{cases} \frac{3}{2} \frac{|h|}{\delta} - \frac{1}{2} \left( \frac{|h|}{\delta} \right)^3, & \text{if } |h| \leq \delta \\ 1, & \text{if } |h| > \delta \end{cases} \quad (6)$$

184 where  $\gamma(h)$  is the variogram value for pairs of points separated by a dis-  
 185 tance  $|h|$  and  $\delta$  is the range (correlation length). In order to take into account  
 186 the anisotropy of geologic structures, a geometric anisotropy with different  
 187 ranges along the horizontal and vertical directions (namely  $\delta_X$  and  $\delta_Z$ ) is  
 188 introduced. The simulations of  $\log Z$  are performed by keeping a constant  
 189 hazard as far as possible. Thus, the following sensitivity analysis takes into  
 190 account the effect of changes in simulation parameters, rather than the effect  
 191 of random sampling (Nguyen et al., 2018).

Each set of 100 random fields is built with specific  $\sigma^2$ ,  $\delta_X$  and  $\delta_Z$  values (Table 3) defined consistently to studies performed at field scale showing that a significant spatial correlation typically exists within distances ranging from a few meters to 20 m (Taskinen et al., 2008). Yet, the  $\mu$  value is fixed so that the geometric mean of  $Z$  corresponds to the mean sandy loam properties (Table 2).

### 3.3. Sensitivity analyses

For each of the three hydraulic parameters under study, the sensitivity of the simulated tritium plume to the properties ( $\sigma^2$ ,  $\delta_X$  and  $\delta_Z$ ) of the 100 corresponding random fields is analyzed with regards to the following features (only the unsaturated zone is considered):

1. the surface area of the plume, defined as the number of grid cells with a volumic activity higher than 1 Bq.m<sup>-3</sup><sub>H<sub>2</sub>O</sub>;
2. the distance between the center of mass of each plume and the center of mass of the plume simulated in the homogeneous medium (where  $\sigma^2(\log K_s) = \sigma^2(\log \alpha) = \sigma^2(\log n) = 0$ );
3. the elongation ratio ( $a/b$ ) of the equivalent ellipse (Figure 4), defined as the ellipse whose center coincides with the center of mass of the plume and whose semi-minor and semi-major axes lengths (namely  $a$  and  $b$ ) are proportional to the eigen values of the inertia matrix.

The number of simulations is set to 100 to ensure that the averages of the previous features reach a stabilization. The modeling conditions (initial and boundary conditions, transient fluxes, source term) are similar for all the

simulations (see section 3.1). Only the soil properties ( $\sigma^2$ ,  $\delta_X$  and  $\delta_Z$ ) differ from one simulation to another.

#### 3.4. Additional tests

In order to assess the effect of both radionuclide type and soil texture, two additional tests are conducted by considering different model parameterizations:

1. the tritium (ideal groundwater tracer) is replaced with a reactive radionuclide (e.g.,  $^{90}\text{Sr}$ ) with a retardation factor ( $R$ ) of 3 and a decay constant ( $\lambda$ ) of  $6.59 \cdot 10^{-4} \text{ d}^{-1}$  (Table 1);
2. the sandy loam mean properties are replaced with silty clay loam mean properties, i.e. related to significantly finer texture (Table 2).

## 4. Results

This section presents the results averaged over each set of simulations for the last modeling time step, i.e. six years after the radionuclide injection. In order to assess the influence of the variance of hydraulic parameters on the plume features, the focus is first put on simulations with one hydraulic parameter varying (Table 3, sets 1., 2., 3.) and then on simulations with the three parameters varying simultaneously (Table 3, sets 4.). Then, the influence of the range values is described.

#### 4.1. Sensitivity to the variance of one hydraulic parameter

Whatever the hydraulic parameter under consideration ( $K_s$  or  $\alpha$  or  $n$ ), the increase in the variance of the corresponding random field results both

237 in the increase in the average plume surface areas (from 220, corresponding  
 238 to the homogeneous plume, to 350 grid cells) and in the variability of this  
 239 surface areas (standard deviation ranging from 0, corresponding to the ho-  
 240 mogeneous plume, to 110 grid cells) among the simulated plumes (Figure 5).  
 241 The relationships between  $\sigma^2$  and the surface areas of the plumes appear to  
 242 be similar for the three hydraulic parameters under study when considering  
 243  $\sigma^2(\log K_s)/\sigma^2(\log n) = 200$  and  $\sigma^2(\log K_s)/\sigma^2(\log \alpha) = 5$ .

244 In addition, the increase in variance for any hydraulic parameter results  
 245 in the increase of the offset in both directions of the mean center of mass  
 246 regarding the center of the homogeneous plume (Figure 6). The trend is yet  
 247 stronger in the horizontal direction (mean offsets ranging from 0.5 to 2 m)  
 248 than in the vertical direction (offsets ranging from 0.5 to 0.7 m). Besides,  
 249 the relationships between  $\sigma^2$  and the offsets are similar when considering  
 250  $\sigma^2(\log K_s)/\sigma^2(\log n) = 200$  and  $\sigma^2(\log K_s)/\sigma^2(\log \alpha) = 5$ .

251 Finally, the increase in variance results in the decrease of the elongation  
 252 ratio (from 0.88, corresponding to the homogeneous plume, to 0.45), re-  
 253 gardless of which hydraulic parameter is considered (Figure 7). Thus, when  
 254 the variability of hydraulic parameters is high, pollutant plumes are more  
 255 likely to have an elongated shape. Besides, the relationships between  $\sigma^2$   
 256 and the offsets are similar when considering  $\sigma^2(\log K_s)/\sigma^2(\log n) = 200$  and  
 257  $\sigma^2(\log K_s)/\sigma^2(\log \alpha) = 5$ .

#### 258 4.2. Sensitivity to the variance of the three hydraulic parameters

259 The mean and standard deviation of the plume surface areas, the mean  
 260 offsets between each centers and the homogeneous center and the mean elon-  
 261 gation ratios are computed for sets of 100 simulations with the three param-

ters varying simultaneously and in a correlated manner, with  $\sigma^2(\log K_s)/\sigma^2(\log n) =$   
 200 and  $\sigma^2(\log K_s)/\sigma^2(\log \alpha) = 5$ .

These features are close to the values obtained for the sets with only  
 one parameter varying, considering the corresponding variances (Figures 5, 6  
 and 7). Here, the results are given for a coefficient  $r$  of 0.8. Other values of  
 $r$  coefficient have been tested and lead to similar results.

#### 4.3. Sensitivity to the ranges

Given the values of  $\sigma^2$ ,  $\delta_X$  and  $\delta_Z$  chosen for this sensitivity analysis, the  
 range in the horizontal direction ( $\delta_X$ ) does not seem to have a significant  
 influence on the simulated plume features (size, center of mass and shape),  
 regardless of which hydraulic parameter is considered (Figure 8). The influ-  
 ence of the range in the vertical direction ( $\delta_Z$ ) appears to be slightly more  
 visible (Figure 9), especially regarding the mean plume surface areas and the  
 location of the mean centers of mass: when the vertical range increases from  
 1 to 5 m, the mean plume surface areas decrease, especially for  $K_s$  and  $n$   
 (diminution from 300 to 250 and from 290 to 260 grid cells respectively) and  
 the horizontal offsets regarding the homogeneous plume center increase of  
 around 0.3 m for the three hydraulic parameters.

#### 4.4. Additional tests

When considering a reactive radionuclide ( $R = 3$ ), the simulated plumes  
 are logically less developed than for the ideal groundwater tracer ( $R = 1$ ).  
 However, both the surface area and the related standard deviation increase  
 significantly when the variance of each of the three hydraulic parameters

285 considered increase. These features tend to stabilize when the average sur-  
286 face reaches about 130% of the surface simulated in a homogenous medium,  
287 consistently with the results for tritium (Figure 10).

288 The results obtained with mean hydraulic parameters related to a finer  
289 texture (silty clay loam) are similar to those above presented for sandy loam  
290 (Figure 10a). This could be explained by the fact that silty clay loam mean  
291 properties remain sufficiently favorable for leading the infiltration of the wa-  
292 ter height imposed at the top surface of the model (Figure 3b). The standard  
293 deviation computed for highest variances of the hydraulic parameters is yet  
294 lower (Figure 10b). This could be explained by the fact that a large distri-  
295 bution of the hydraulic parameters centered on sandy loam material could  
296 result in very high hydraulic conductivity values more favorable to the de-  
297 velopment of preferential flow paths and therefore to the lateral migration of  
298 the plume.

## 299 5. Discussion

300 This study allows to quantify the impact of the variance of the random  
301 fields representing the logarithms of the hydraulic parameters  $K_s$ ,  $\alpha$  and  $n$   
302 on solute transport within unsaturated surficial formations. By focusing on  
303 variances estimated for porous media in natural environment at field scale, it  
304 appears that the  $n$  and, in a lesser extent,  $K_s$  parameters are the most critical  
305 (Figures 5, 6 and 7). Thus, by comparison with homogeneous parameter  
306 fields, random fields generated with  $\sigma^2(\log K_s) = 0.14$ ,  $\sigma^2(\log \alpha) = 0.036$   
307 and  $\sigma^2(\log n) = 0.005$ , *i.e.* mean variances calculated based on the WoSIS  
308 database, respectively result in (i) 25, 20 and 65% increase in plume size;



309 (ii) 0.8, 1 and 1.8 m horizontal offsets of the plume center; and (iii) 20, 30  
 310 and 50% decrease in plume circularity. Besides, within the range of variation  
 311 of  $\sigma^2(\log n)$ , the average surface areas range from 260 to 350 grid cells. In  
 312 comparison, within the range of variation of  $\sigma^2(\log K_s)$  and  $\sigma^2(\log \alpha)$ , the  
 313 mean surface areas only range from 235 to 290, which highlights that  $n$  is  
 314 the most critical parameter. Such results are consistent with previous studies  
 315 showing the high influence of the spatial variability of the parameter  $n$  on  
 316 flow (Lu and Zhang, 2002 and Tan et al., 2017) in the unsaturated zone.

317 This study also shows that the simulated plumes are far less sensitive to  
 318 changes in ranges than changes in variances of the random fields representing  
 319 the hydraulic parameters, at least for the intervals of values chosen. Yet, the  
 320 impact of the vertical range appears to be more pronounced than the hori-  
 321 zontal range: when the zones of high or low values of hydraulic parameters  
 322 are more expanded in depth (higher value of vertical ranges), the preferential  
 323 flow paths are potentially more continuous and the plume surface areas tend  
 324 to decrease and get closer to the homogeneous plume surface area. The val-  
 325 ues of horizontal ranges considered here are of the same order of magnitude  
 326 as the pollutant plumes extensions (a few meters): it could explain the low  
 327 influence of the changes in horizontal range on the simulated plumes. Never-  
 328 theless, these ranges are consistent with analyses of soil parameter variability  
 329 at field scale (Russo et al., 1997 and Botros et al., 2009). However, for higher  
 330 values of ranges (from hundreds of meters to a thousand meter), the influ-  
 331 ence of horizontal range on the flow rate in unsaturated zone was shown to  
 332 be more significant (Tan et al., 2017).

333 The sensitivity analysis performed in this study mainly focuses on an

334 ideal groundwater tracer (tritium) in sandy texture. However, additional  
 335 tests show that the results obtained for reactive radionuclides and a finer  
 336 texture are comparable (at least for similar infiltration rate). From a general  
 337 standpoint, this study thus indicates that taking into account the weak intra-  
 338 facies (small scale) spatial variability is critical for characterizing plume mi-  
 339 gration within unsaturated zone. This requires to develop in-situ approaches  
 340 for estimating more efficiently soil hydraulic parameters and their variabil-  
 341 ity at field scale (*e.g.*, Léger et al., 2014; Léger et al., 2016). When these  
 342 parameters remain poorly recognized, a set of simulations based on typical  
 343 variability deduced from local or global database can be used for identifying  
 344 the plume properties and the related uncertainties at a given time.

## 345 6. Conclusion

346 This work has highlighted the critical influence of the spatial variability  
 347 in hydraulic parameters, in particular pore-size distribution index ( $n$ ), on  
 348 radionuclide migration in unsaturated surficial formations. In practice, this  
 349 work demonstrates the need for well characterizing this spatial variability  
 350 in order to quantify and locate soil volumes that need to be removed or  
 351 depolluted in the context of remediation of nuclear or industrial sites. It also  
 352 quantifies the corresponding uncertainties.

353 In addition, although the quantitative results presented in this paper are  
 354 related to a theoretical example and are dependent on the model settings  
 355 (environmental parameters, radionuclide, etc.), the proposed method (ran-  
 356 dom field generation, physically-based groundwater transport simulations,  
 357 plume properties characterization) can be transposed to other contexts, such

as chemical pollutions or design of devices to contain pollutant spread.

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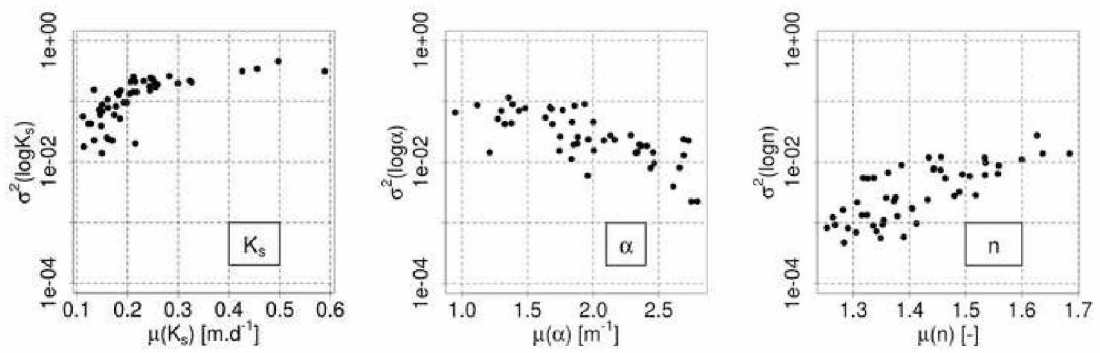
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Figure1  
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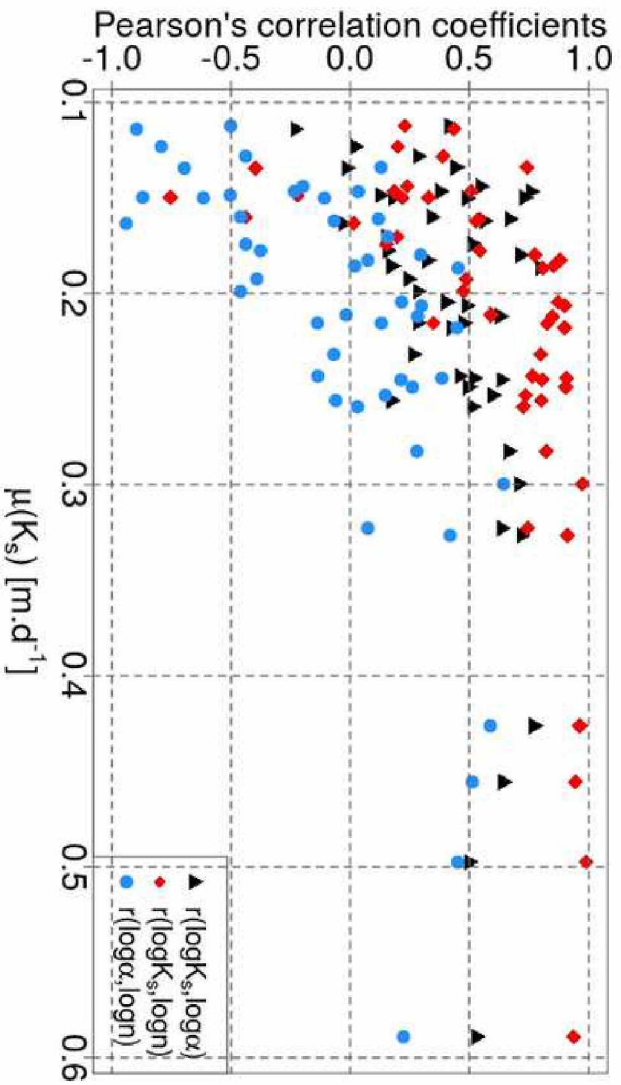
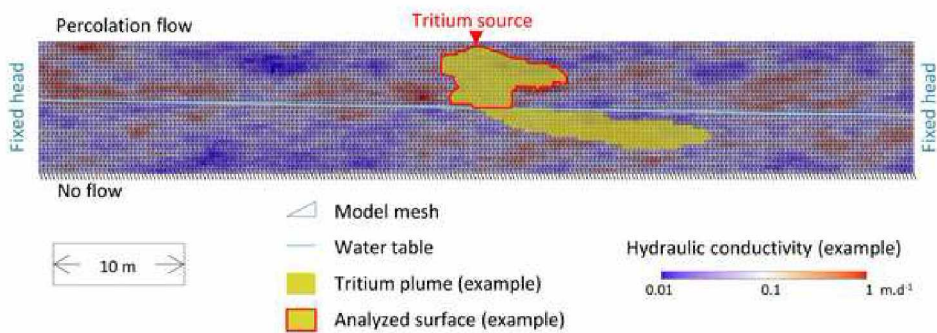


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(a)



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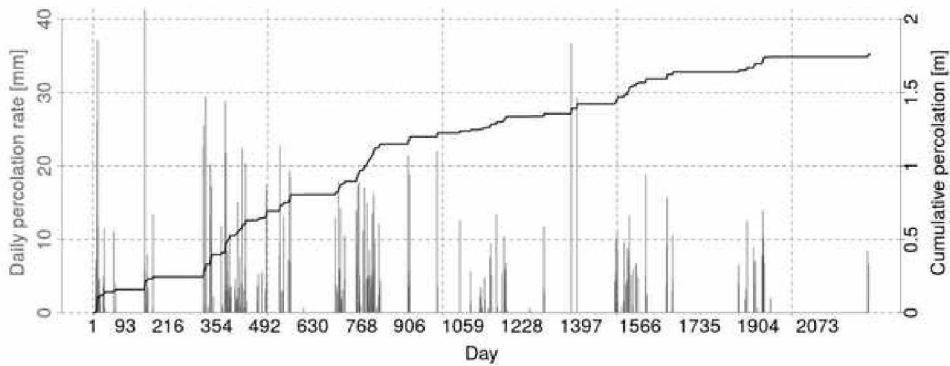
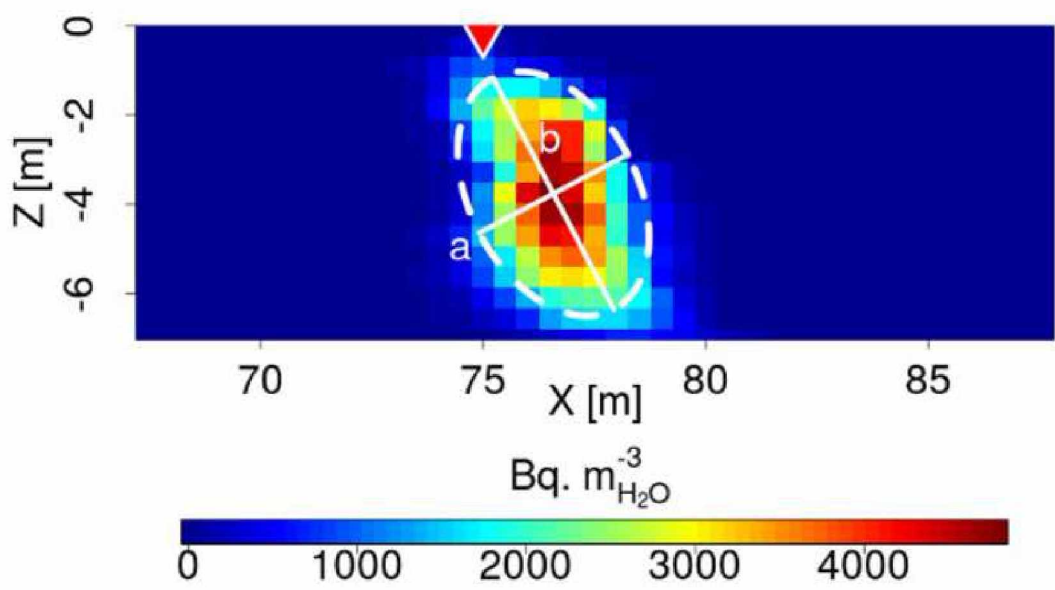


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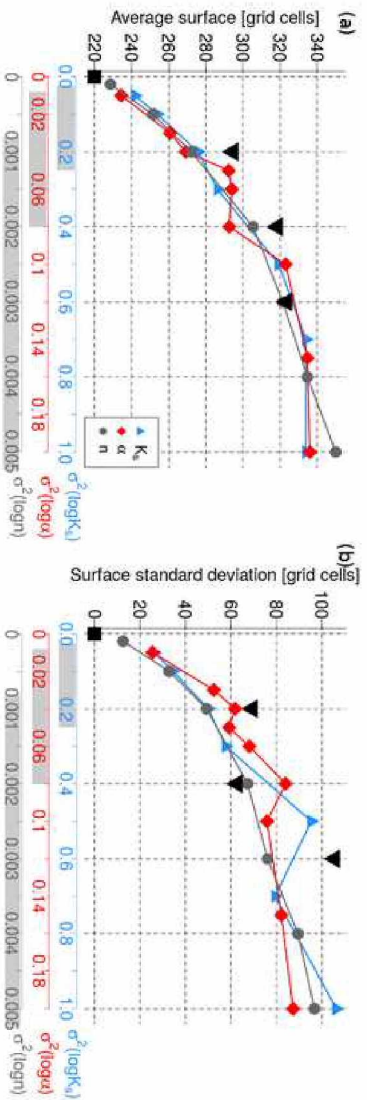


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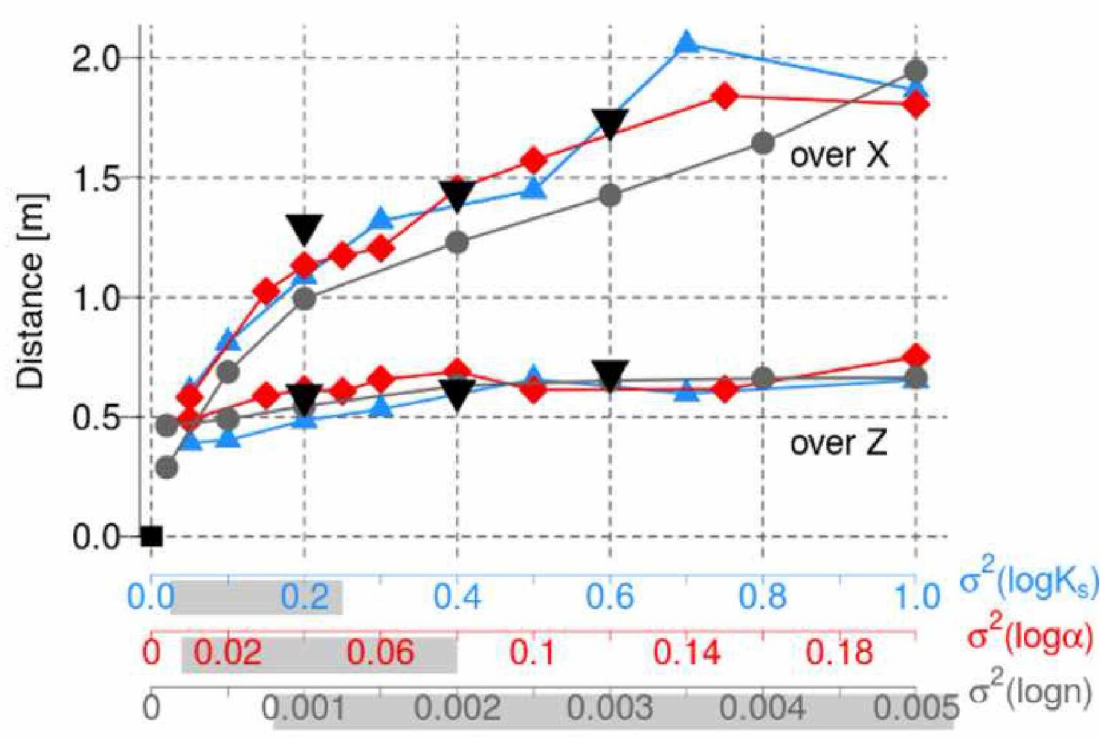


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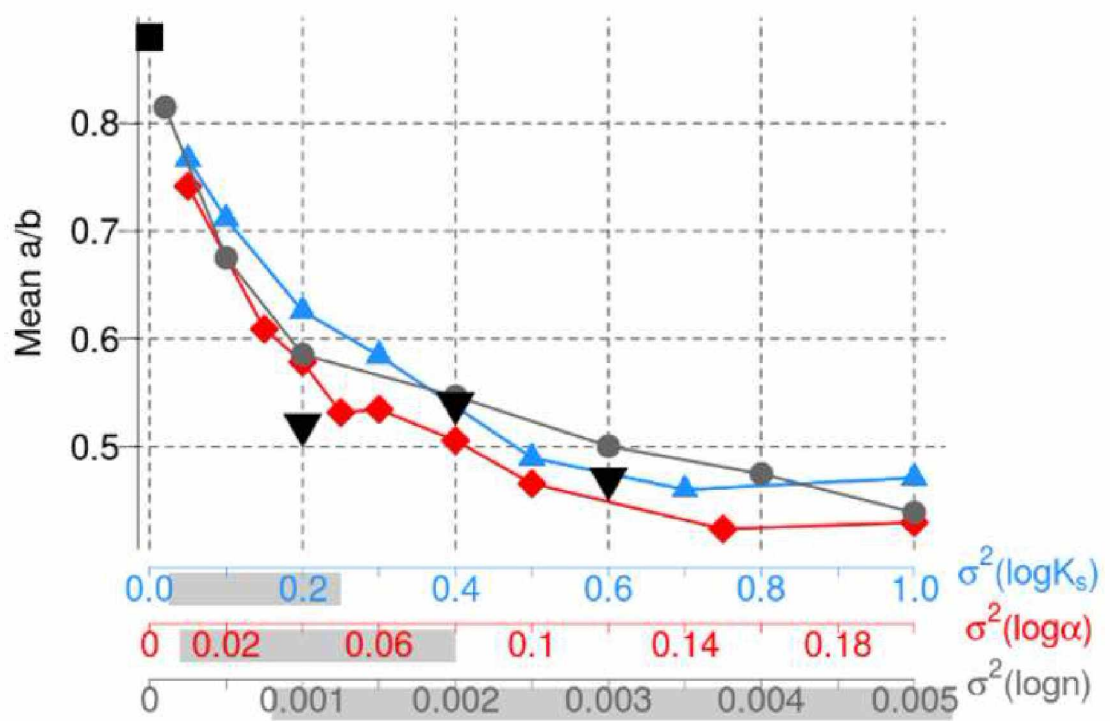




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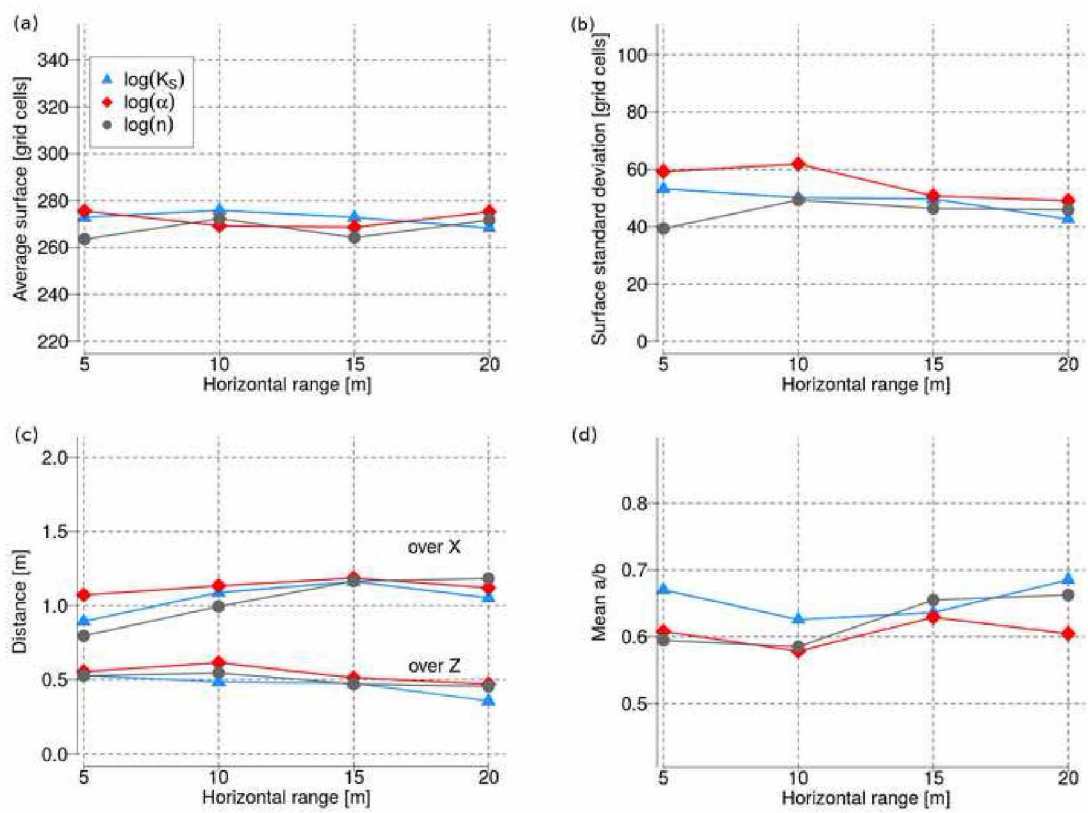


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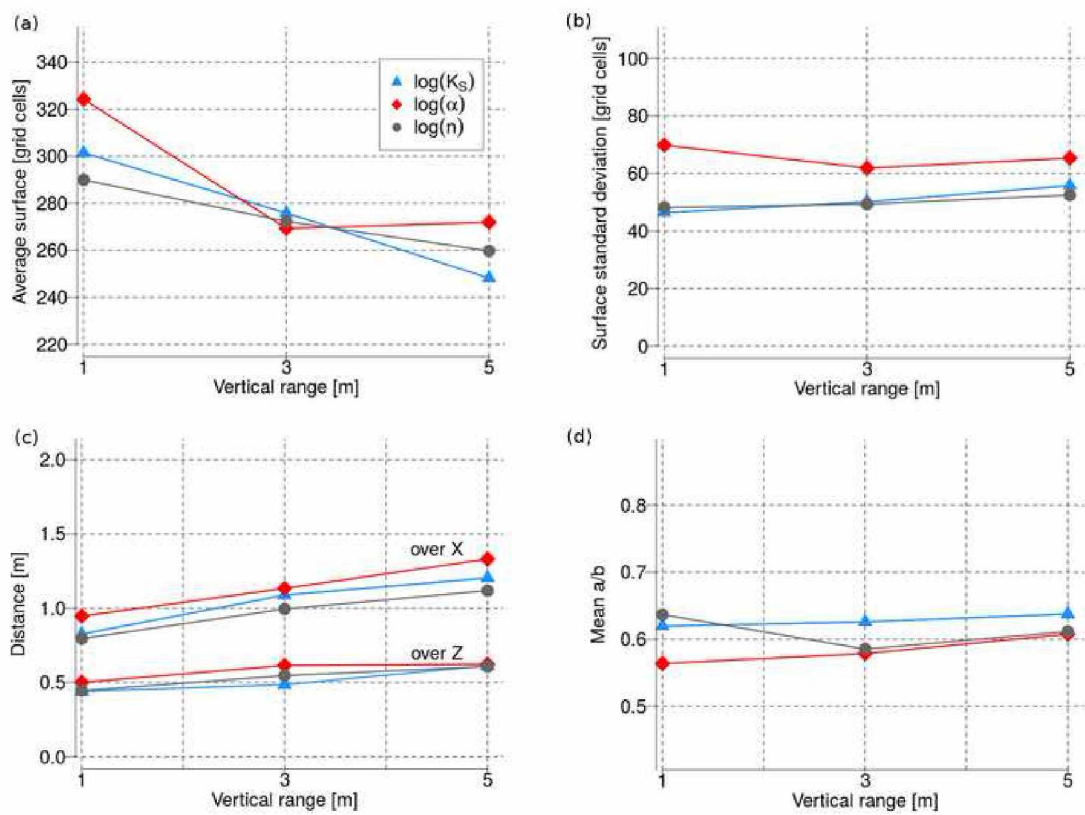


Figure 10  
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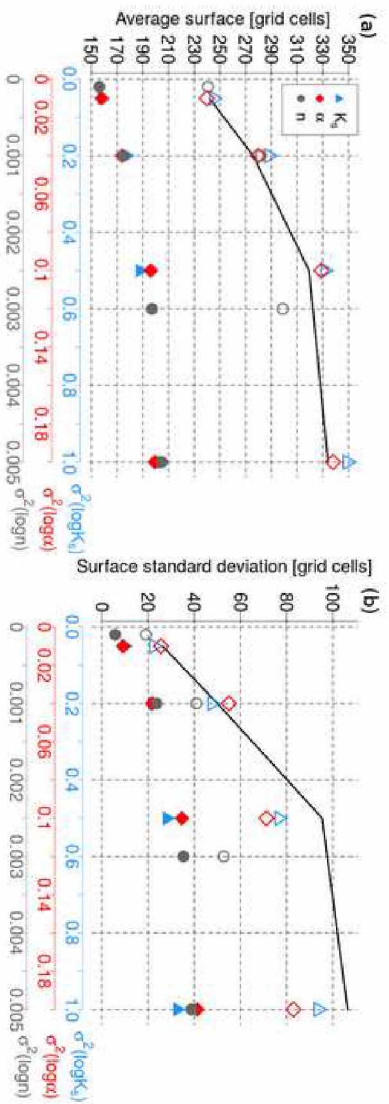


Figure captions

**Figure 1:** Log-variances in function of means for  $K_s$ ,  $\alpha$  and  $n$  for each location built via WoSIS database analysis.

**Figure 2:** Pearson's correlation coefficients between the log-parameters in function of  $\mu(K_s)$  for each location built via WoSIS database analysis.

**Figure 3:** (a) Modeling domain with an example of hydraulic conductivity random field and resulting tritium plume. (b) Daily percolation rate and cumulative percolation rate used for the simulations.

**Figure 4:** Example of pollutant plume within the unsaturated zone with its equivalent ellipse in dotted line. The  $a/b$  ratio gives a hint about the elongation of the plume: the more  $a/b$  is close to 0, the more the plume has an elongated shape. The triangle highlights the tritium source.

**Figure 5:** Average plume surface areas (a) and standard deviation of surface areas (b) depending on  $\sigma^2$  for the three hydraulic parameters. The squares correspond to the homogeneous medium and the three upside-down triangles on each graph correspond to media where the three hydraulic parameters are varying in a correlated manner. The values of  $\delta_x$  and  $\delta_z$  are set to 10m and 3m. The shaded rectangles on the axes correspond to the range of variances found at field scale via WoSIS database.

**Figure 6:** Mean distances with the center of the plume simulated in the homogeneous medium in function of  $\sigma^2$  for the three hydraulic parameters. The square corresponds to the homogeneous medium and the six upside-down triangles correspond to the media where the three hydraulic parameters are varying in a correlated manner. The values of  $\delta_x$  and  $\delta_z$  are set to 10m and 3m. The shaded rectangles on the axes correspond to the range of variances found at field scale via WoSIS database.

**Figure 7:** Mean  $a/b$  ratio in function of  $\sigma^2$  for the three hydraulic parameters. The square corresponds to the homogeneous medium and the three upside-down triangles correspond to the media where the hydraulic parameters are varying in a correlated manner. The values of  $\delta_x$  and  $\delta_z$  are set to 10m and 3m. The shaded rectangles on the axes correspond to the range of variances found at field scale via WoSIS database.

**Figure 8:** Average (a) and standard deviation (b) of the plume surface areas depending on the horizontal range for the three hydraulic parameters. Average distances (c) with the homogeneous center and mean elongation ratio (d) in function of the horizontal range. The values of  $\sigma^2$  are set to 0.2 for  $\log K_s$ , 0.04 for  $\log \alpha$  and 0.001 for  $\log n$  and  $\delta_z$  is set to 3m.

**Figure 9:** Average (a) and standard deviation (b) of the plume surface areas depending on the vertical range for the three hydraulic parameters. Average distances (c) with the homogeneous center and mean elongation ratio (d) in function of the vertical range. The values of  $\sigma^2$  are set to 0.2 for  $\log K_s$ , 0.04 for  $\log \alpha$  and 0.001 for  $\log n$  and  $\delta_x$  is set to 10m.

**Figure 10:** Average (a) and standard deviation (b) of the plume surface areas depending on  $\sigma^2$  for the three hydraulic parameters. The black line corresponds to the results obtained for tritium and sandy loam texture (for the parameter  $K_s$ ). The filled symbols correspond to the results for the reactive radionuclide and sandy loam texture. The empty symbols correspond to the results for tritium and silty clay loam texture.

Table1 (revised)  
[Click here to download Table: table1.pdf](#)

	$\omega$ [-]	$D_z$ [m]	$D_z$ [m]	$d$ [m <sup>2</sup> .d <sup>-1</sup> ]	$\lambda$ [d <sup>-1</sup> ]	$R$ [-]
Tritium	0.38	0.05	0.005	0.00001	$1.54.10^{-4}$	1
Reactive radionuclide	0.38	0.05	0.005	0.00001	$6.59.10^{-4}$	3

	$\theta_r$ [L <sup>3</sup> L <sup>-3</sup> ]	$\theta_s$ [L <sup>3</sup> L <sup>-3</sup> ]	$K_s$ [m.d <sup>-1</sup> ]	$\alpha$ [m <sup>-1</sup> ]	$n$ [-]	$K_x/K_z$ [-]
Sandy loam	0.039	0.38	0.38	2.67	1.45	10
Silty clay loam	0.090	0.48	0.11	0.84	1.52	10

Table3  
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	$\sigma^2(\log K_s)$	$\sigma^2(\log \alpha)$	$\sigma^2(\log \eta)$	$\delta_X$ [m]	$\delta_Z$ [m]	$r(\log K_s, \log \alpha)$ and $r(\log K_s, \log \eta)$
Set 1.1	0.05	0	0	10	3	-
Set 1.2	0.1	0	0	10	3	-
Set 1.3	0.2	0	0	5	3	-
Set 1.4	0.2	0	0	10	3	-
Set 1.5	0.2	0	0	15	3	-
Set 1.6	0.2	0	0	20	3	-
Set 1.7	0.2	0	0	10	1	-
Set 1.8	0.2	0	0	10	5	-
Set 1.9	0.3	0	0	10	3	-
Set 1.10	0.5	0	0	10	3	-
Set 1.11	0.7	0	0	10	3	-
Set 1.12	1	0	0	10	3	-
Set 2.1	0	0.01	0	10	3	-
Set 2.2	0	0.03	0	10	3	-
Set 2.3	0	0.04	0	5	3	-
Set 2.4	0	0.04	0	10	3	-
Set 2.5	0	0.04	0	15	3	-
Set 2.6	0	0.04	0	20	3	-
Set 2.7	0	0.04	0	10	1	-
Set 2.8	0	0.04	0	10	5	-
Set 2.9	0	0.05	0	10	3	-
Set 2.10	0	0.06	0	10	3	-
Set 2.11	0	0.08	0	10	3	-
Set 2.12	0	0.1	0	10	3	-
Set 2.13	0	0.15	0	10	3	-
Set 2.14	0	0.20	0	10	3	-
Set 3.1	0	0	0.0001	10	3	-
Set 3.2	0	0	0.0005	10	3	-
Set 3.3	0	0	0.001	5	3	-
Set 3.4	0	0	0.001	10	3	-
Set 3.5	0	0	0.001	15	3	-
Set 3.6	0	0	0.001	20	3	-
Set 3.7	0	0	0.001	10	1	-
Set 3.8	0	0	0.001	10	5	-
Set 3.9	0	0	0.002	10	3	-
Set 3.10	0	0	0.003	10	3	-
Set 3.11	0	0	0.004	10	3	-
Set 3.12	0	0	0.005	10	3	-
Set 4.1	0.2	0.04	0.001	10	3	0.8
Set 4.2	0.4	0.08	0.002	10	3	0.8
Set 4.3	0.6	0.12	0.003	10	3	0.8



Table captions (revised)

**Table 1:** Transport parameters used in (Eq. 4) for tritium and for a reactive radionuclide.

**Table 2:** Soil hydraulic (flow) parameters derived from ROSETTA PTF for sandy loam and silty clay loam materials, used in (Eq. 1), (Eq. 2) and (Eq. 3).

**Table 3:** Values of variances, ranges and Pearson's correlation coefficients for each set of 100 simulations.