

AN ANALYSIS OF THE EFFECT OF HYDRAULIC PARAMETERS ON RADIONUCLIDE MIGRATION IN AN UNSATURATED ZONE

GYE-NAM KIM*, JEI-KWON MOON and KUNE-WOO LEE

Korea Atomic Energy Research Institute
1045 Daedeokdaero, Yusong-gu, Daejeon

*Corresponding author. E-mail : kimsum@kaeri.re.kr

Received March 23, 2010

Accepted for Publication July 24, 2010

A One-Dimensional Water Flow and Contaminant Transport in Unsaturated Zone (FTUNS) code has been developed in order to interpret radionuclide migration in an unsaturated zone. The pore-size distribution index (n) and the inverse of the air-entry value (α) for an unsaturated zone were measured by KS M ISO 11275 method. The hydraulic parameters of the unsaturated soil are investigated by using soil from around a nuclear facility in Korea. The effect of hydraulic parameters on radionuclide migration in an unsaturated zone has been analyzed. The higher the value of the n -factor, the more the cobalt concentration was condensed. The larger the value of α -factor, the faster the migration of cobalt was and the more aggregative the cobalt concentration was. Also, it was found that an effect on contaminant migration due to the pore-size distribution index (n) and the inverse of the air-entry value (α) was minute. Meanwhile, migrations of cobalt and cesium are in inverse proportion to the Freundlich isotherm coefficient. That is to say, the migration velocity of cobalt was about 8.35 times that of cesium. It was conclusively demonstrated that the Freundlich isotherm coefficient was the most important factor for contaminant migration.

KEYWORDS : Hydraulic Parameter, Cobalt, Cesium, Migration, Unsaturated Zone

1. INTRODUCTION

Decommissioning of a retired Training, Research, Isotope production, General Atomics (TRIGA) research reactor facility has been performed for the reuse of the decommissioned site. The residents adjacent to the decommissioned site should be safe from radiation exposure. Radionuclides around the decommissioned site were first distributed on the site surface, and then later moved from the surface to the groundwater table. Finally, radionuclides around the decommissioned site have been found mostly to be located in the unsaturated zone. Therefore, it is necessary to develop a technology for interpreting radionuclide-migration in an unsaturated zone to secure the radiation safety of the decommissioned site.

The Multimedia Environmental Pollutant Assessment System (MEPAS) [1] is a physics-based environmental analysis code that integrates source-term, transport, and exposure models for endpoints such as concentration, dose, or risk. Developed by the Pacific Northwest National Laboratory, MEPAS was designed for site-specific assessments using readily available information. Endpoints are computed for chemical and radioactive pollutants. The

Multimedia Contaminant Fate, Transport, and Exposure Model (MMSOILS) [2] was used to estimate the human exposure and health risk associated with releases of contamination from hazardous waste sites. The methodology consists of a multimedia model that addresses the transport of a chemical in groundwater and surface water, the soil erosion, the atmosphere, and the accumulation in the food chain. The RESidual RADioactive (RESRAD) family of computer codes [3] was developed by Environmental Science Division (EVS) to provide useful tools for evaluating human health risk at sites contaminated with radioactive residues. The RESRAD methodology was cited in a Department of Energy (DOE) order for dose assessment and determination of guidelines for clean-up of radiologically contaminated sites. The potential effect of rainfall and evapotranspiration on contaminant migration in the unsaturated soil zone was studied using a deterministic dynamic modeling approach at the University of California [4].

Recently, researchers at the University of Tubingen, Germany, investigated the transport of fate of reactive compounds in the unsaturated vadose zone using numerical simulation of steady-state and transient flow scenarios [5]. Also, researchers at the University of Liege, Belgium,

attempted to model the migration of contaminants through variably saturated dual-porosity, dual-permeability chalk [6]. The Korea Institute of Nuclear Safety has developed a simplified approximation method of the multi-compartments model [7] of contaminant migration in an unsaturated zone. The Pacific Northwest National Laboratory has estimated soil hydraulic parameters of a field drainage experiment using inverse techniques [8]. Researchers at the Lawrence Berkeley National Laboratory have studied modeling of mountain-scale radionuclide transport in an unsaturated zone at Yucca Mountain [9].

In this study, a numerical cord has been developed in order to interpret the radionuclide migration in an unsaturated zone. The pore-size distribution index (n) and the inverse of the air-entry value (α) for an unsaturated zone were measured with the KS M ISO 11275 method. The hydraulic parameters of the unsaturated soil are investigated by using soil from around a nuclear facility in Korea. Also, the effect of hydraulic parameters on radionuclide migration in an unsaturated zone has been analyzed.

2. DEVELOPMENT OF CONTAMINATANT MIGRATION MODEL FOR AN UNSATURATED ZONE

2.1 Water Flow Model in an Unsaturated Zone

The relationship between θ and h is described by the commonly used van Genuchten function [10], where α is fitting parameter that is inversely proportional to the air-entry pressure value, n is a parameter related to the width of pore-size distribution of the medium, and m is a constant that is commonly approximated by $m=1-1/n$. The Mualem Function [11] yields the hydraulic conductivity function. The unsaturated soil water migration model can be described by Richards' Function (1), and Van Genuchten Function and Mualem Function (2)-(3) as

$$\frac{\partial \theta}{\partial t} = \frac{\partial}{\partial x} \left[K(h, x) \frac{\partial h}{\partial x} \right] \quad (1)$$

$$\begin{aligned} K(h, x) &= K_s(x) K_r(h, x) \\ F(\theta) &= \left[1 - \left(\frac{\theta - \theta_r}{\theta_s - \theta_r} \right)^{1/m} \right]^m \\ \theta(h) &= \theta_r + \frac{\theta_s - \theta_r}{(1 + |\alpha h|^n)^m} \\ m &= 1 - 1/n \end{aligned} \quad (2)$$

$$K_r(\theta) = \sqrt{\frac{\theta - \theta_r}{\theta_s - \theta_r}} [1 - F(\theta)]^2 \quad (3)$$

where θ is water content, θ_r is residual water content, θ_s is saturated water content h is pressure head, K is hydraulic conductivity

2.2 Contaminant Migration Model in An Unsaturated Zone

The contaminant migration model [12] can be described by the following convection-diffusion function (4).

$$\frac{\partial \theta C}{\partial t} + \frac{\partial \rho s}{\partial t} = \frac{\partial}{\partial x} \left(\theta D \frac{\partial c}{\partial x} \right) - \frac{\partial qc}{\partial x} - \mu_w \theta c - \mu_s \rho s \quad (4)$$

$S = kc^n$ (nonlinear Freundlich isotherm)

D : dispersion coefficient q : water velocity

μ_w, μ_s : first-order decay

3. MODELING OF CONTAMINANT MIGRATION IN AN UNSATURATED ZONE AROUND A NUCLEAR FACILITY

3.1 Drawing Up of Contaminant Migration Program

Fig. 1 provides the main flow chart of the contaminant migration program in an unsaturated zone. The numerical programs used the Galerkin finite element technique in order to calculate Richards' equation and the convection-dispersion equation. One-Dimensional Water Flow and Contaminant Transport in Unsaturated Zone (FTUNS) code were developed, which codes can analyze water flow and contaminant transport by precipitation. The FTUNS code is written in FORTRAN 77, and can be compiled and run on any PC compatible computer.

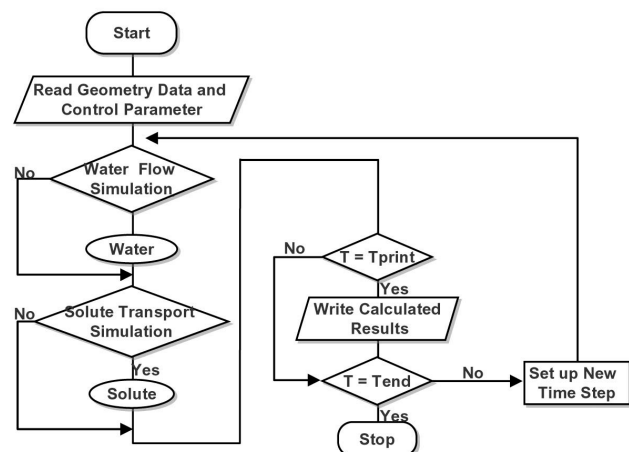


Fig. 1. Main Flow Chart for Contaminant Migration

3.2 Outline on Contaminant Migration Modeling in Unsaturated Zone

A soil column with dimensions of 30×30×58 cm was manufactured to verify the contaminant migration model, as shown in Fig. 2; this column was filled with soil from around a nuclear facility. A soil surface of 5 cm thickness was artificially contaminated with cobalt and cesium. Rainfall was occasionally injected by nozzle for 55 days, as shown in Table 1. The measured initial condition for modeling is shown in Fig. 3.



Fig. 2. Soil Column for Contaminant Migration Test

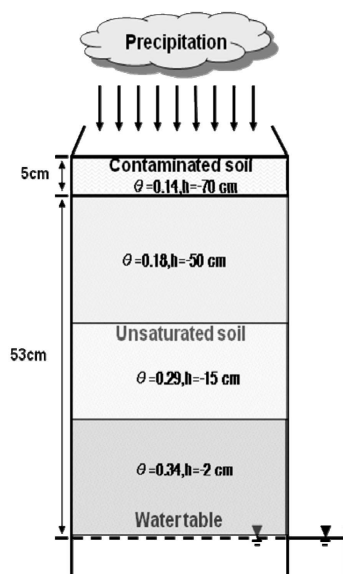


Fig. 3. Initial Condition for Modeling

Table 1. Daily Rainfall Rate for Migration Test

Period (55 days)		Rainfall (Total 19.05 L)
6/11	15:00	1.00
6/15	10:00	0.95
6/17	16:00	1.30
6/23	10:00	1.40
6/26	16:30	1.20
6/30	17:00	1.20
7/3	10:00	1.50
7/8	10:00	1.50
7/10	8:00	1.20
7/14	14:00	1.00
7/21	10:00	2.00
7/24	10:00	1.30
7/29	10:00	2.00
8/4	11:00	1.50
8/4	14:00	Completion

Table 2. The Hydraulic Parameters of the Soil Around a Nuclear Facility

Para. Time	Bulk density	Moisture content	Particle density	Porosity
1 st	1.479	0.145	2.430	0.365
2 nd	1.477	0.125	2.370	0.395
Aver.	1.478	0.135	2.400	0.380

3.3 Hydraulic Parameter Measurement

The soil around a nuclear facility in Korea was used for contaminant migration test; measured hydraulic parameters of the soil are shown in Table 2. Hydraulic conductivity and contaminant dispersion coefficient were measured through the soil column test, as shown in Fig. 4. The saturated hydraulic conductivity of the soil was 7.98×10^{-4} cm/sec and the contaminant dispersion coefficient of the soil, obtained using the Ogata function [12-13], was 3.0 m. The Freundlich isotherm coefficients of cobalt and cesium were 0.88 and 7.35, respectively, as shown in Fig. 5 and Fig. 6.

3.4 Measurement of the Pore-Size Distribution

Index (n) and the Inverse of the Air-Entry Value (α)

The pore-size distribution index (n) and the inverse of the air-entry value (α) for an unsaturated zone were

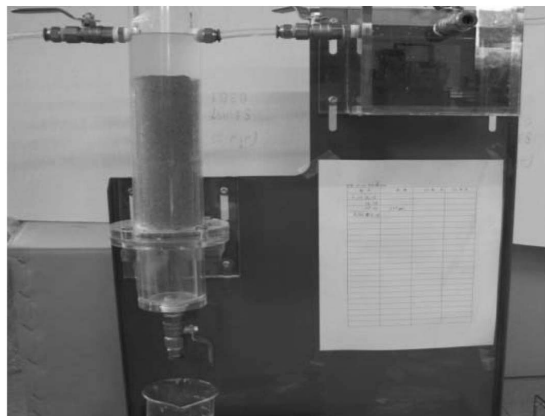


Fig. 4. Soil Column Test

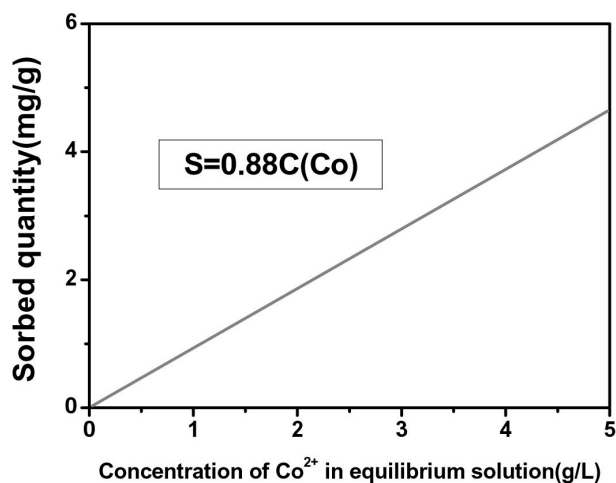


Fig. 5. Freundlich Isotherm Coefficient of Cobalt

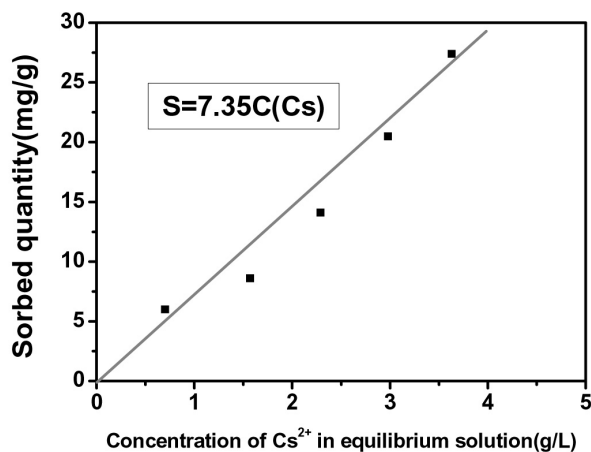


Fig. 6. Freundlich Isotherm Coefficient of Cesium

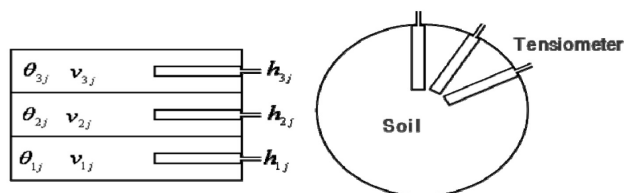


Fig. 7. Location of Tensiometer



Fig. 8. Measurement of Pressure Head and Weight of Soil Along Time by Tensiometer and Balance

measured with the KS M ISO 11275 method. The KS M ISO 11275 method is as follows. The soil column was divided into three layers and a tensiometer was injected into each soil layer, as shown in Fig. 7. The pressure head and the weight of the soil were reduced with time passage, because the water on the soil column surface evaporated. The pressure head and the weight of the soil were measured in hours by tensiometer and balance, as shown in Fig. 8. θ and h were calculated at the same time using functions (5)-(6) and a relation equation between θ and h was induced. Also, θ and K were calculated at the same time using functions (7)-(8) and a relation equation between θ and K was induced. The inverse of the air-entry value (α) was induced using function (2) and a relation equation between θ and h . The pore-size distribution index (n) was induced using function (3) and a relation equation between θ and K . That is, the measured pore-size distribution index (n) and the inverse of the air-entry value (α) were about 1.5 and 0.05, respectively.

$$\theta_j = \frac{m_j - m_e}{\rho_w V} + \theta_e \quad (5)$$

$$h_j = \frac{1}{n} \sum_{i=1}^n h_{ij} \quad (6)$$

m_e : soil weight after test completion
 θ_e : water content after test completion

$$\frac{\Delta h_p}{\Delta z} = \frac{-\sqrt{h_{i+1,j+1} \bullet h_{i+1,j}} + \sqrt{h_{i,j+1} \bullet h_{i,j}}}{z_{i+1} - z_i}$$

$$v_{ij} = \frac{1}{t_{j+1} - t_j} \sum_{k=1}^i a_k (\theta_{kj} - \theta_{k,j+1}) \quad (7)$$

$$K_{ij} = \frac{v_{ij}}{\frac{\Delta h_p}{\Delta z} + 1} \quad (8)$$

4. RESULTS AND DISCUSSION

Modeling for contaminant migration in the soil column that was filled with soil from around a nuclear facility in Korea was performed using the developed FTUNS code. The modeling results are as follows. Fig. 9 shows the effect of the pore-size distribution index (n) on cobalt migration. The higher the value of the n-factor is, the more the cobalt concentration along the depth was condensed. Also, the cobalt concentration at a depth of 12m was increased. Fig. 10 shows the effect of the inverse of the air-entry value (α) on cobalt migration. The larger the value of the α -factor is, the faster the migration of cobalt is and the more the cobalt concentration along the depth was condensed. Also, the cobalt concentration at a depth of 12m was increased. Fig. 11

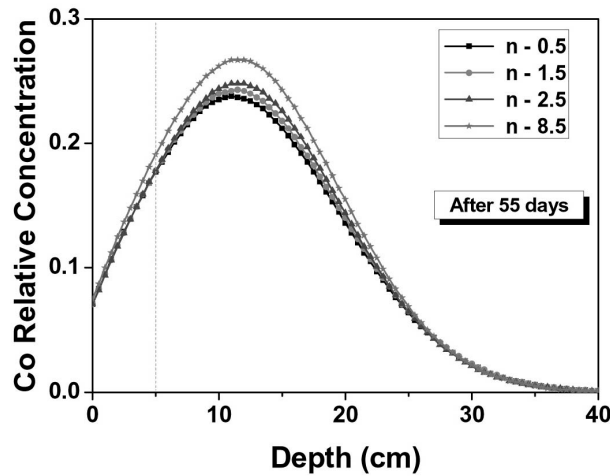


Fig. 9. The Effect of the Pore-size Distribution Index (n) for Cobalt Migration

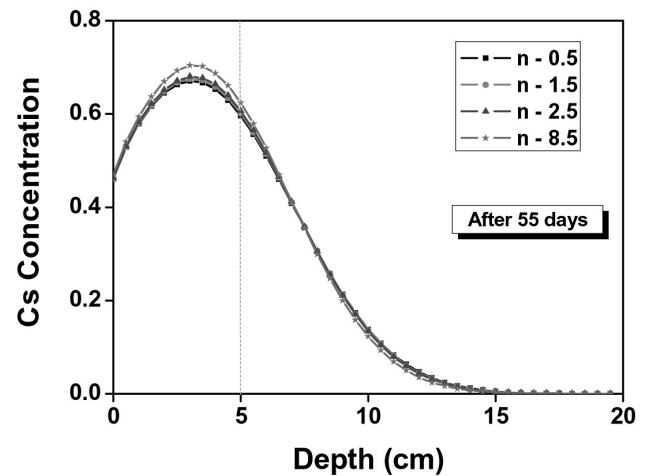


Fig. 11. The Effect of the Pore-size Distribution Index (n) for Cesium Migration

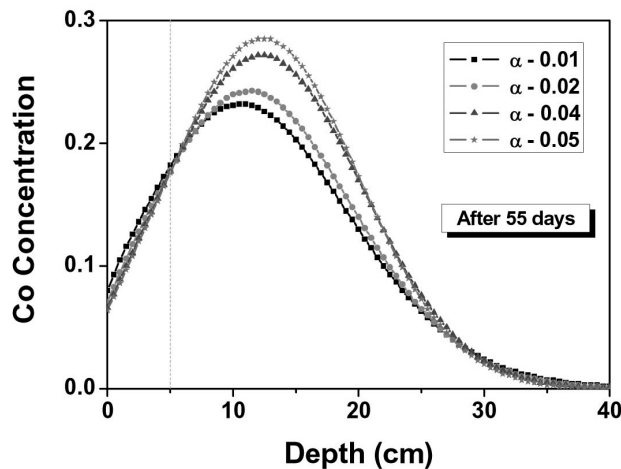


Fig. 10. The Effect of the Inverse of the Air-entry Value (α) for Cobalt Migration

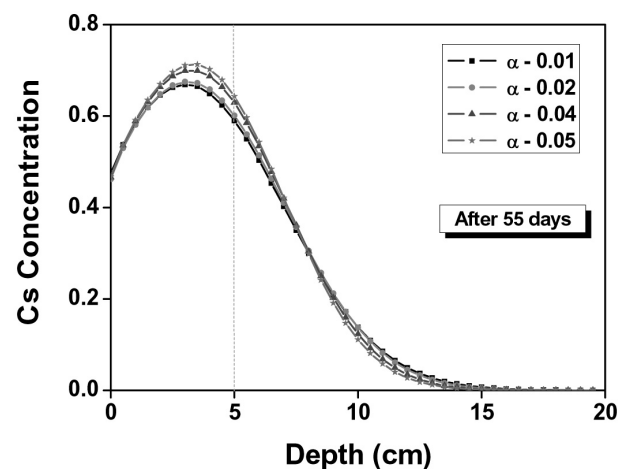


Fig. 12. The Effect of the Inverse of the Air-entry Value (α) for Cesium Migration

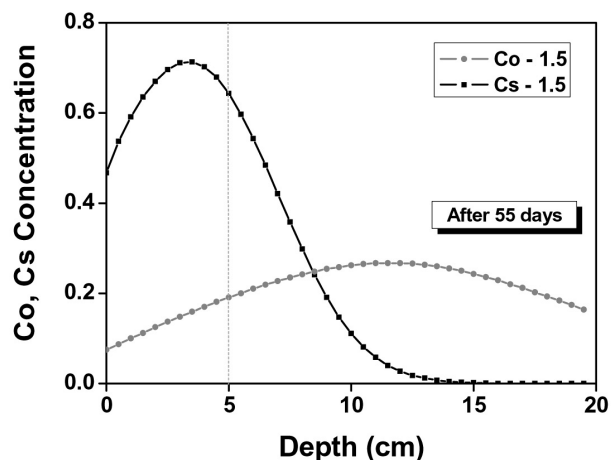


Fig.13. The Effect of Freundlich Isotherm Coefficient for Contaminant Migration

shows the effect of the pore-size distribution index (n) on cesium migration. The higher the value of the n -factor is, the more the cobalt concentration was condensed. Also, the cesium concentration at a depth of 3m was increased. Fig. 12 shows the effect of the inverse of the air-entry value (α) on cesium migration. The larger the value of the α -factor is, the faster the migration of cesium is and the more the cesium concentration was condensed. Also, the cesium concentration at a depth of 3m was increased. Finally, it was found that the effects of the pore-size distribution index (n) and the inverse of the air-entry value (α) on contaminant migration were both minute. Meanwhile, Fig. 13 shows concentration distributions of cobalt and cesium after rainfall for 55 days. The Freundlich isotherm coefficients of cobalt and cesium were 0.88 and 7.35, respectively, and migrations of cobalt and cesium was in inverse proportion to the Freundlich isotherm coefficient. That is, the migration velocity of cobalt was about 8.35 times that of cesium. It was conclusively demonstrated that the Freundlich isotherm coefficient was a more important factor than the pore-size distribution index (n) and the inverse of the air-entry value (α) for contaminant migration.

5. CONCLUSION

Modeling for contaminant migration in a soil column that was filled with soil from around a nuclear facility in Korea was performed using the developed FTUNS code. The pore-size distribution index (n) and the inverse of the air-entry value (α) for an unsaturated zone were measured with the KS M ISO 11275 method. The modeling results derived with the FTUNS code are as follows. The higher the value of the n -factor is, the more the contaminant concentration along the depth was condensed. Also, the contaminant concentration at a depth of 12m was increased.

The larger the value of the α -factor is, the faster the contaminant migration was and the more the contaminant concentration along the depth was condensed. It was found that the effect of the pore-size distribution index (n) and the inverse of the air-entry value (α) on contaminant migration were both minute. Meanwhile, migrations of cobalt and cesium were in inverse proportion to the Freundlich isotherm coefficient. That is, the migration velocity of cobalt was about 8.35 times that of cesium. It can be concluded that the Freundlich isotherm coefficient was a more important factor than the pore-size distribution index (n) and the inverse of the air-entry value (α) for contaminant migration.

ACKNOWLEDGMENT

This work was supported by a Nuclear Research & Development Program of the Korea Science and Engineering Foundation (KOSEF) grant funded by the South Korean government (MEST).

REFERENCES

- [1] D. L. Streng, M. A. Smith, "Multimedia Environmental Pollutant Assessment System (MEPAS)", PNNL-16164, Pacific Northwest National Laboratory (2006).
- [2] "The Multimedia Contaminant Fate, Transport, and Exposure Model," Environmental Protection Agency, USA, (2002).
- [3] C. Yu, A. J. Zielen, J. J. Cheng, D. J. LePore, E. Gnanapragasam, S. Kamboj, J. Arnish, A. Wallo III, "User's Manual for RESRAD Version 6," Argonne National Laboratory, (2001).
- [4] J. Grifoll and Y. Cohen, "Contaminant migration in saturated soil zone: an effect of rainfall and evapotranspiration," *Contaminant Hydrology*, **23**, 185 (1996).
- [5] D. Kuntz, P. Grathwohl, "Comparison of steady-state and transient flow conditions on reactive transport of contaminants in the vadose soil zone," *J. of Hydrology*, **369**, 225 (2009).
- [6] S. Brouyere, "Modelling the migration of contaminants through variably saturated dual-porosity, dual-permeability chalk," *J. of Contaminant Hydrology*, **82**, 195 (2006).
- [7] Jae Hah Cheong, "Simplified approximation method of the multi-compartments model on the migration of contaminant through unsaturated zone," *J. of Korean Radioactive Waste Society*, **5**(1), 29 (2007).
- [8] Z. F. Zhang, A. L. Ward, and G. W. Gee, "Estimating soil hydraulic parameters of a field drainage experiment using inverse techniques," *Vadose Zone Journal*, **2**, 201 (2003).
- [9] G. J. Moridis, Y. Seol, and Y. Wu, "Modeling studies of mountain-scale radionuclide transport in an unsaturated zone at Yucca mountain, Nevada," p. 12-14, Proc. TOUGH Symposium, Berkeley, California (2003).
- [10] M. Th. van Genuchten, "A closed-form equation for predicting the hydraulic conductivity of unsaturated soils," *Soil Sci. Soc. Am. J.*, **44**, 892 (1980).
- [11] Y. Mualem, "A new model for predicting the hydraulic conductivity of unsaturated porous media," *Water Resour. Res.*, **12**, 513 (1976).
- [12] R. A. Freeze and J. A. Cherry, "Groundwater," p. 604, Prentice Hall Inc., Upper Saddle River (1979).
- [13] A. Ogata, "Theory of dispersion in a granular medium," Prof. Paper 411, U. S. Geol. Surv. (1970).