Stochastic analysis of the effect of spatial variability of diffusion parameters on radionuclide transport in a low permeability clay layer

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Abstract Most studies that incorporate subsurface heterogeneity in groundwater flow and transport models only analyze and simulate the spatial variability of hydraulic conductivity. Heterogeneity of the other flow and transport parameters is usually neglected. This approach is often justified, but there are however cases in which disregarding the heterogeneity of the other flow and transport parameters can be questionable. In low permeability media, for instance, diffusion is often the dominant transport mechanism. It therefore seems logical to incorporate the spatial variability of the diffusion parameters in the transport model. This study therefore analyzes and simulates the spatial variability of the effective diffusion coefficient and the diffusion accessible porosity with geostatistical techniques and incorporates their heterogeneity in the transport model of a low permeability formation. The calculated output radionuclide fluxes of this model are compared with the fluxes calculated with a homogeneous model and a model with a heterogeneous hydraulic conductivity distribution. This analysis shows that the heterogeneity of the diffusion parameters has a much larger effect on the calculated output radionuclide fluxes than the heterogeneity of hydraulic conductivity.

Keywords geostatistics, heterogeneous media, diffusion, waste disposal, radionuclide transport

INTRODUCTION

It is generally recognized that subsurface heterogeneity may have a large influence on groundwater flow and transport of contaminants. Therefore, a large number of recent studies incorporate the underground heterogeneity in hydrogeological flow and transport models using geostatistical techniques. These studies usually only analyze and simulate the spatial variability of hydraulic conductivity, thereby neglecting the heterogeneity of the other flow and transport parameters. This approach is often justified, since advection is usually the dominant transport process. There are however cases in which disregarding the heterogeneity of the other flow and transport parameters is questionable. In low permeability media, for instance, diffusion is often the dominant transport mechanism. The effect of transport by advection in such media can be relatively small compared to the effect of transport by diffusion. It therefore seems logical to incorporate the spatial variability of the diffusion parameters in the hydrogeological transport model. The main diffusion parameters are the effective diffusion coefficient and the diffusion accessible porosity as the diffusive mass flux in porous media is given by:

$$F = -\eta D_e \mathrm{grad} C \qquad (1)$$

where F is the diffusive mass flux [kg m^{-2} s^{-1}], η is the diffusion accessible porosity [-], D_e is the effective diffusion coefficient [m^2 s^{-1}] and C is the solute concentration [kg m^{-3}]. The diffusion accessible porosity is not always equal to the total porosity but may be smaller. Only a fraction of the total water-filled porosity is available for diffusive transport. This is caused by size-exclusion effects, i.e. some pores are narrower than the ion size, and by the permanent structural negative charge on the clay surface, which can cause negatively charged ions to be excluded from the narrower interparticle spaces of the clay (Horseman et al. 1996). This study analyzes and simulates the spatial variability of the two diffusion parameters and incorporates their heterogeneity in the transport model of a low permeability formation.

The studied formation is the Boom Clay in Belgium. This low permeability clay layer is a candidate host rock for the deep geological disposal of high-level radioactive waste. In previous studies, the fate of radionuclides released from a potential repository in the Boom Clay was calculated under different assumptions. Mallants et al. (2001) examined radionuclide migration from the vitrified waste through the Boom Clay into the surrounding aquifers, assuming that the clay layer was homogeneous. These calculations showed that the magnitude of the fluxes released into the surrounding aquifers was strongly limited by the Boom Clay, so that the dose rates were hundreds times lower than the internationally recommended dose limit. In a later study (Huysmans and Dassargues, submitted), the effect of fractures and the spatial variability of hydraulic conductivity was investigated. The output fluxes of this heterogeneous model differed at most 8% from the fluxes of the homogeneous model. In the present study, the effect of the spatial variability of the diffusion parameters of the Boom Clay is examined. A large number of equally probable random realizations of the clay layer are generated with stochastic simulation and co-simulation procedures using all available hard and soft data. Each of these equiprobable fields is used as input for a transport model that calculates radionuclide transport by advection, diffusion, dispersion, adsorption and decay through the heterogeneous medium. Radionuclide fluxes at the clay-aquifer interfaces are calculated, taking the heterogeneity of the effective diffusion coefficient and the diffusion accessible porosity into account. Radionuclide fluxes computed with this model are compared with fluxes obtained from the previous models.

METHODOLOGY

The study site is the nuclear zone of Mol/Dessel (province of Antwerp). The research activities of the Belgian nuclear repository program, conducted by ONDRAF/NIRAS (Belgian agency for radioactive waste and enriched fissile materials) are concentrated at SCK●CEN (Belgian Nuclear Research Centre) located in this zone. An underground experimental facility (HADES-URF) was built in the Boom Clay at 223 m depth. In this area, the Boom Clay has a thickness of about 100 m and is overlain by 180 m of water bearing sand formations.

On the Mol/Dessel site, a 570 m deep borehole (Mol-1 borehole) was drilled. Several transport and geological parameters have been intensively measured in the laboratory on cores taken at the Mol-1 borehole. Geophysical logging was also performed in the same borehole. The resulting data set comprises 41 diffusion coefficient and diffusion accessible porosity measurements, 52 hydraulic conductivity values, a gamma ray log, an electrical resistivity log, 71 grain size measurements and a porosity log estimated from the nuclear magnetic resonance log. The diffusion coefficient, hydraulic conductivity, electrical resistivity and grain size show higher values in the lower part of the Boom Clay while gamma ray shows lower values in that part. Diffusion accessible porosity and nuclear magnetic resonance porosity do not show this trend.

The diffusion accessible porosity has a symmetric distribution while the diffusion coefficient shows a skewed distribution. The diffusion accessible porosity has an average value of 0.16 and a standard deviation of 0.02. The diffusion coefficient D_e has an average value of 1.62×10^{-10} m² s^{-1} and a standard deviation of 9.03×10^{-11} m² s^{-1}.

Correlation coefficients of η and D_e with the secondary variables are calculated. Diffusion accessible porosity shows very little correlation with the other variables. The diffusion coefficient, on the contrary, shows a good correlation with hydraulic conductivity, grain size, resistivity and gamma ray.

Since diffusion accessible porosity shows no correlation with the other variables, only a direct variogram is calculated and fitted. It is fitted with the sum of a nugget model of 0.00015 and a spherical model with a range of 5.8 m and a sill of 0.00018. The small range and the relatively large nugget effect suggest that this variable shows little spatial correlation.

The diffusion coefficient D_e shows a strong correlation with all secondary variables. These secondary variables are therefore all incorporated in the simulation procedure. Experimental variograms and cross-variograms of all variables are calculated in 25 lags with a lag distance of 3 m. The shape of most variograms suggests fitting by a spherical model. The average range is approximately 35 m. Variograms and cross-variograms of D_e and all secondary variables are modeled as the sum of a nugget model and a spherical model with a range of 35 m. The sills are fitted by the optimization program LCMFIT2 (Pardo-Iguzquiza and Dowd, 2002).

A large number of equally probable random realizations of the clay layer are generated, using the modeled variograms and cross-variograms. Since the Boom Clay shows a lateral continuity that largely exceeds the extent of the local scale model (Wouters and Vandenberghe, 1994), it is assumed that the properties of the Boom Clay do not vary in the horizontal direction and one-dimensional vertical realizations of the diffusion accessible porosity and the diffusion coefficient were generated. The diffusion coefficient and the diffusion accessible porosity for iodide are simulated with direct sequential simulation with histogram reproduction. This approach creates realizations that reproduce the (1) local point and block data in the original data units, (2) the mean, variance and variogram of the variable and (3) the histogram of the variable (Oz et al., 2003). The simulation of the diffusion accessible porosity reflects the symmetric distribution of η, while the simulation of the diffusion coefficient shows a skewed distribution. The simulated diffusion accessible porosity shows no trend, whereas the simulated diffusion coefficient has higher values in the lower part of the Boom Clay.

A local 3D hydrogeological model of the Boom Clay is constructed. The model width in the *x*-direction is 20m, i.e. half the distance between the disposal galleries. The model length in the *y*-direction is 15m. The model dimension in the *z*-direction is 102 m, i.e. the total thickness of the Boom Clay in the nuclear zone of Mol-Dessel. The grid spacing is 1m in the *x*-direction and in the *y*-direction and varies between 0.2m and 1m in the *z*-

direction. The horizontal and vertical hydraulic conductivity are 7×10^{-12} m s^{-1} and 2.8×10^{-12} m s^{-1} respectively. The vertical boundary conditions for groundwater flow are zero flux boundary conditions since the hydraulic gradient is vertical. The specified head at the upper boundary is 2 m higher than the specified head at the lower boundary since the vertical hydraulic gradient is approximately 0.02 in the 100 m thick Boom Clay (Wemaere and Marivoet, 1995).

Transport by advection, dispersion, molecular diffusion and radioactive decay is calculated for 3 radionuclides: Se-79, I-129 and Tc-99. Previous calculations revealed that they were the most important in terms of dose rates from a potential high-level waste repository for vitrified waste (Mallants et al., 1999). The boundary conditions for transport at the upper and lower boundaries are zero concentration boundary conditions (Mallants et al., 1999) since the hydraulic conductivity contrast between the clay and the aquifer is so large that solutes reaching the boundaries are assumed to be flushed away by advection in the aquifer. The source term models for the 3 radionuclides are as described by Mallants et al., 1999. The radionuclides are contained in borosilicate glass and as the glass corrodes, the radionuclides become available for dissolution into the groundwater. A constant glass dissolution rate of 3 μm per year is assumed. The glass matrix would be completely dissolved after approximately 70000 years. The source term model is therefore a constant flux over a period of 70000 years equal to the total radionuclide inventory divided by 70000 years. If, however, this source term model results in calculated concentrations higher than the solubility limit, the source term model is replaced by a constant concentration model. A constant concentration equal to the solubility limit is then prescribed until exhaustion of the source.

For the radionuclide I-129, the different equiprobable realizations of the diffusion coefficient and diffusion accessible porosity of iodide are directly imported in the model. For Se-79 and Tc-99, previous studies indicate that the diffusion coefficient is approximately equal to the diffusion coefficient of iodide. Therefore the realizations of the diffusion coefficient were also used to model transport of Se-79 and Tc-99. The diffusion accessible porosity of these radionuclides is however different. While the average value of the diffusion accessible porosity of iodide is 0.16, Se-79 and Tc-99 are reported to have diffusion accessible porosities of 0.13 and 0.30 respectively. Therefore the simulations of the diffusion accessible porosity of iodide were rescaled for Se-79 and Tc-99 so that the average values of the simulated porosities were equal to 0.13 and 0.30.

This local 3D hydrogeological model was run with FRAC3DVS, a simulator for three-dimensional groundwater flow and solute transport in porous, discretely-fractured porous or dual-porosity formations (Therrien et al., 1996, Therrien et al., 2003). This model was run for 10 different random combinations of simulations of the diffusion coefficient and the diffusion accessible porosity. The results of this model were compared with the results of a homogeneous model.

RESULTS

A comparison is made between the radionuclide activities calculated with the heterogeneous simulations and a homogeneous model with a homogeneous diffusion coefficient and diffusion accessible porosity equal to the average values. Compared to the homogeneous model, the radionuclide activity flowing through the lower clay-aquifer is between 21 % smaller and 3 % larger in the heterogeneous model. The radionuclide activity flowing through the upper clay-aquifer is between 25 % smaller and 2.5 % larger.

DISCUSSION

A model with a heterogeneous diffusion coefficient and diffusion accessible porosity distribution results in fluxes that are up to 25 % different from the fluxes calculated with a homogeneous model with a homogeneous diffusion coefficient and diffusion accessible porosity equal to the average values. The output fluxes of a previous model with a heterogeneous hydraulic conductivity distribution differed at most 8% from the fluxes of the homogeneous model (Huysmans and Dassargues, submitted). The coefficient of variation, i.e. standard deviation divided by mean value, of hydraulic conductivity is 2.75. This value is much larger than the coefficients of variation of the diffusion coefficient and diffusion accessible porosity, 0.125 and 0.557 respectively. Although hydraulic conductivity shows a much larger relative spatial variability than the diffusion coefficient and the diffusion accessible porosity, the heterogeneity of the diffusion parameters has a much larger effect on the output fluxes than the heterogeneity of hydraulic conductivity. Only incorporating the spatial variability of hydraulic conductivity would thus results in a serious underestimation of the effect of heterogeneity on the output fluxes.

This can be explained by the large importance of transport by diffusion in low permeability media. The effect of transport by advection in such media is usually small compared to the effect of transport by diffusion. The solute concentrations and fluxes are thus much more sensitive to changes in diffusion parameters than to changes in hydraulic conductivity.

CONCLUSION

Most studies that incorporate subsurface heterogeneity in groundwater flow and transport models only analyze and simulate the spatial variability of hydraulic conductivity. The heterogeneity of the other flow and transport parameters is usually neglected. This study has shown that this approach is not always justified. Radionuclide transport in a low permeability clay was simulated, taking the spatial variability of the diffusion coefficient and the diffusion accessible porosity into account. The output fluxes of this model were compared with a homogeneous model and with a model with a heterogeneous hydraulic conductivity distribution. Although hydraulic conductivity has a much larger relative spatial variability than the diffusion coefficient and the diffusion accessible porosity, the heterogeneity of the diffusion parameters proved to have a much larger effect on the output fluxes than the heterogeneity of hydraulic conductivity. This can be explained by the large importance of transport by diffusion in low permeability media. The solute concentrations and fluxes are much more sensitive to changes in diffusion parameters than to changes in hydraulic conductivity.

A hydrogeological study incorporating subsurface heterogeneity should therefore start with a sensitivity analysis of the different flow and transport parameters. The effect of the expected spatial variation in hydraulic conductivity K, effective porosity n_e, diffusion coefficient D_e, diffusion accessible porosity n, distribution coefficient K_d, etc. on the results should be examined. Based on this analysis, a selection should be made of the parameters for which the spatial variability should be incorporated in the model.

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