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Hydrogeological modeling of radionuclide transport in low permeability media: a comparison between Boom Clay and Ypresian Clay

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Abstract Deep low-permeability clay layers are considered as suitable environments for disposal of high-level radioactive waste. In Belgium, the Boom Clay is the reference host formation and the Ypresian Clay an alternative host formation for research and safety and feasibility assessment of deep disposal of nuclear waste. In this study, two hydrogeological models are built to calculate the radionuclide fluxes that would migrate from a potential repository through these two clay formations. Transport parameter heterogeneity is incorporated in the models using geostatistical co-simulations of hydraulic conductivity, diffusion coefficient and diffusion accessible porosity. The calculated radionuclide fluxes in the two clay formations are compared. The results show that in the Ypresian Clay larger differences between the fluxes through the lower and the upper clay boundary occur, larger total output radionuclide amounts are calculated and a larger effect of parameter heterogeneity on the calculated fluxes is observed, compared to the Boom Clay.

Keywords Geostatistics · Waste disposal · Radionuclide transport · Heterogeneous media

Introduction

Safe disposal of nuclear waste is an important environmental challenge. Several countries are investigating deep geological disposal as a long-term solution for their high-level waste. In Belgium, the Oligocene Boom Clay is the reference host formation for research purposes and for the safety and feasibility assessment of the deep disposal of high-level and/or long-lived radioactive waste. The Mol-Dessel zone (province of Antwerp) is the reference site for research, development and demonstration studies on the Boom Clay (Fig. 1). The clay layers of the Eocene Ieper Group (the Kortrijk Formation and Kortemark Member) are an alternative host formation for the research and assessment of a deep disposal solution for high-level and/or long-lived radioactive waste in Belgium. The Doel nuclear zone (province of Antwerp) is an alternative reference site for methodological studies regarding the Ypresian Clays (ONDRAF/NIRAS 2002).

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. In two later studies (Huysmans and Dassargues 2005a, b), the effect of fractures and spatial variability of hydraulic conductivity and the effect of spatial variability of the diffusion parameters, respectively, was

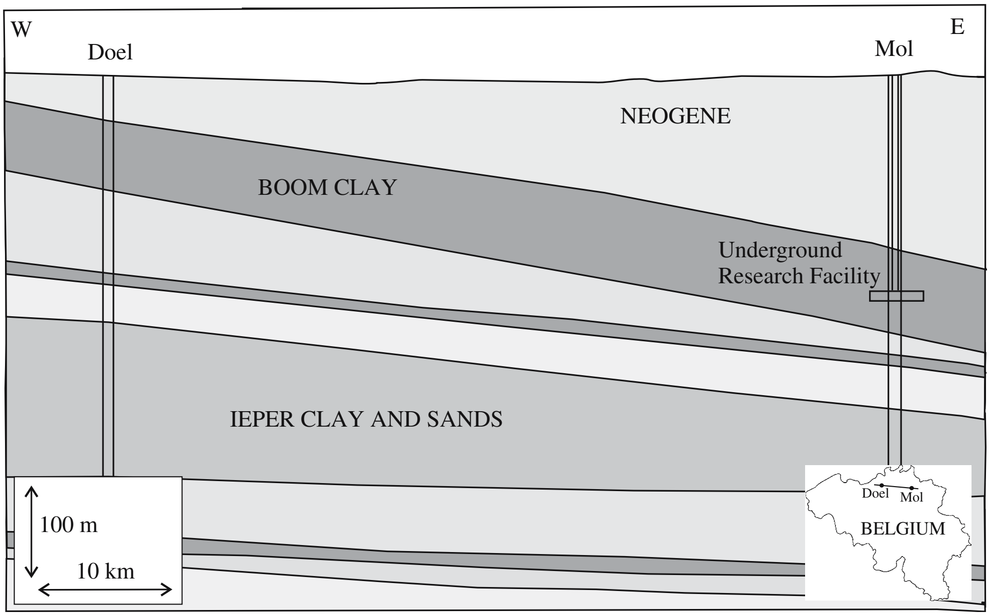

Fig. 1 E–W geological profile of north Belgium showing the location of the Doel borehole, the Mol-1 borehole and the underground research facility

investigated. Radionuclide transport through the Ypresian Clays has not yet been simulated.

Comparative studies of both formations improve the understanding of the two systems being studied, in particular by an analysis of the transferability of knowledge, methods, concepts, etc. from one formation or site to another. A comparison and assessment of the respective strengths and weaknesses of the disposal systems in these different host formations should also make it possible to optimize the design and development of a deep repository (ONDRAF/NIRAS 2002).

The aim of this study is therefore to calculate and compare the radionuclide fluxes that would migrate from a potential repository through the clays into the surrounding aquifers. Radionuclide transport through the clays into the surrounding aquifers is calculated by means of a hydrogeological model of both clay formations. The model results for both potential host formations are analyzed and compared. Since the previous studies of the Boom Clay (Huysmans and Dassargues 2005a, b) showed that spatial variability of the transport parameters may have an effect on the calculated radionuclide fluxes, the hydrogeological models in this study incorporate parameter heterogeneity. Hydraulic conductivity, diffusion coefficient and diffusion accessible porosity heterogeneity was included in the hydrogeological models using geostatistical simulation.

Method

Study sites

The Mol–Dessel zone (province of Antwerp) is the reference site for research, development and demonstration studies on the Oligocene Boom Clay. 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 (Fig. 1).

The Doel nuclear zone (province of Antwerp) is an alternative reference site for methodological studies regarding the Eocene Ypresian Clays. In this zone, the clay layers of the Ieper Group (the Kortrijk Formation and Kortemark Member) are situated at a depth of approximately 340 m and have a thickness of about 100 m (Fig. 1).

Data analysis

Two deep boreholes on the Mol/Dessel site and the Doel nuclear zone respectively provide the data for this study. On the Mol/Dessel site, a 570 m deep borehole (Mol-1 borehole) was drilled. Several transport and geological parameters (hydraulic conductivity K, diffusion coefficient D_e, diffusion accessible porosity η and grain size) have been intensively measured in the laboratory on cores taken at the Mol-1 borehole. To complement the knowledge about the primary variables of interest, i.e. the transport parameters hydraulic conductivity, diffusion coefficient and diffusion accessible porosity, measurements of secondary variables were also collected. Secondary variables, e.g. geophysical data, are usually spatially cross-correlated with the primary variables and contain useful information about the primary variables. This can be exploited to improve the estimates of the primary variables (Isaaks and Srivastava 1989). Geophysical

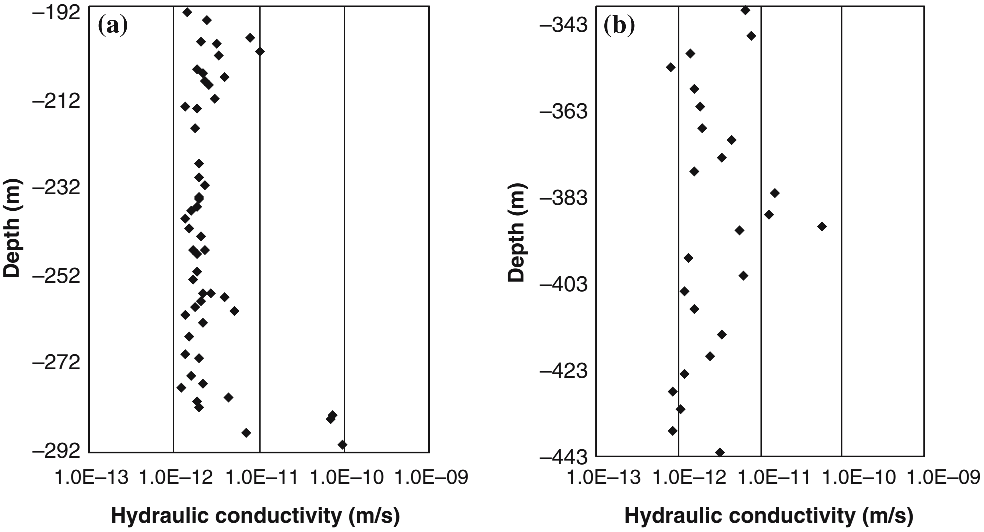

Fig. 2 Measured vertical hydraulic conductivity (m/s) of **a** the Boom Clay in the Mol-1 borehole and **b** the Ypresian Clay in the Doel borehole

logging was performed in the Mol-1 borehole to obtain logs of gamma ray, resistivity and nuclear magnetic resonance. The resulting data set for the Boom Clay comprises 52 hydraulic conductivity values (Fig. 2), 41 diffusion coefficient (Fig. 3) and diffusion accessible porosity measurements (Fig. 4), a gamma ray log, an electrical resistivity log, 71 grain size measurements and a porosity log estimated from the nuclear magnetic resonance log. On the Doel nuclear zone, a series of boreholes was drilled near the Doel nuclear power station (Van Marcke and Laenen 2005). The deepest borehole reaches a depth of 688 m. Laboratory experiments on cores from the Doel boreholes provided 25 hydraulic conductivity values (Fig. 2), 25 diffusion coefficient (Fig. 3) and diffusion accessible porosity (Fig. 4) measurements and 49 grain size measurements of the Ypresian Clay. Geophysical logging provided logs of gamma ray and resistivity.

Comparison of the statistics of the parameters of the Boom Clay and Ypresian Clay (Table 1) shows that the transport parameters have similar values for both clays. Comparison of the correlation coefficients between the parameters (Table 2) shows that hydraulic conductivity and diffusion coefficient are better correlated with the secondary variables in the Boom Clay than in the Ypresian Clay.

Geostatistical estimators, i.e. variograms and cross-variograms, were calculated and modeled for all primary and secondary measurements. Variograms and cross-variograms of variables are modeled as the sum of a nugget model and a spherical model with a range of 35m for the Boom Clay and 24m for the Ypresian Clay. The

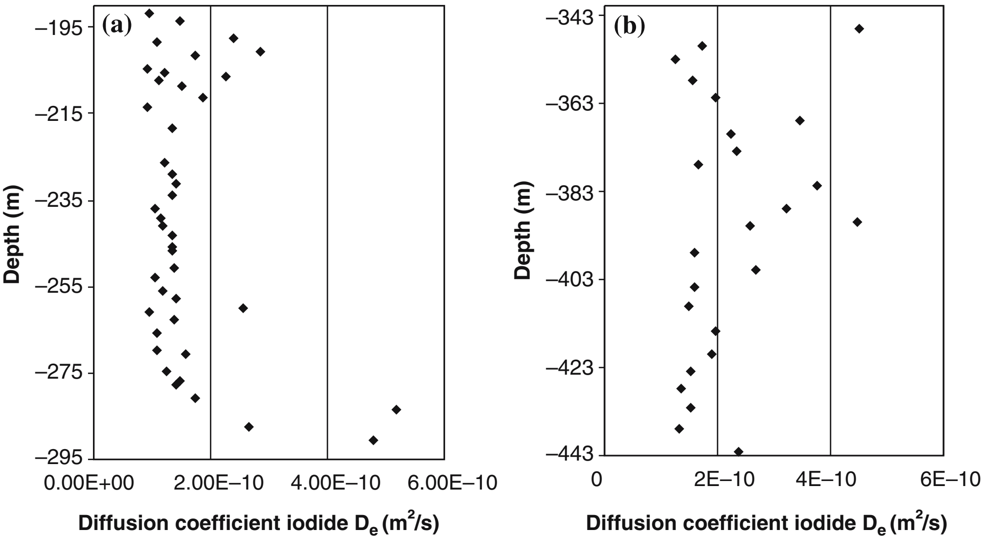

Fig. 3 Measured diffusion coefficient of iodide (m^2/s) of **a** the Boom Clay in the Mol-1 borehole and **b** the Ypresian Clay in the Doel borehole

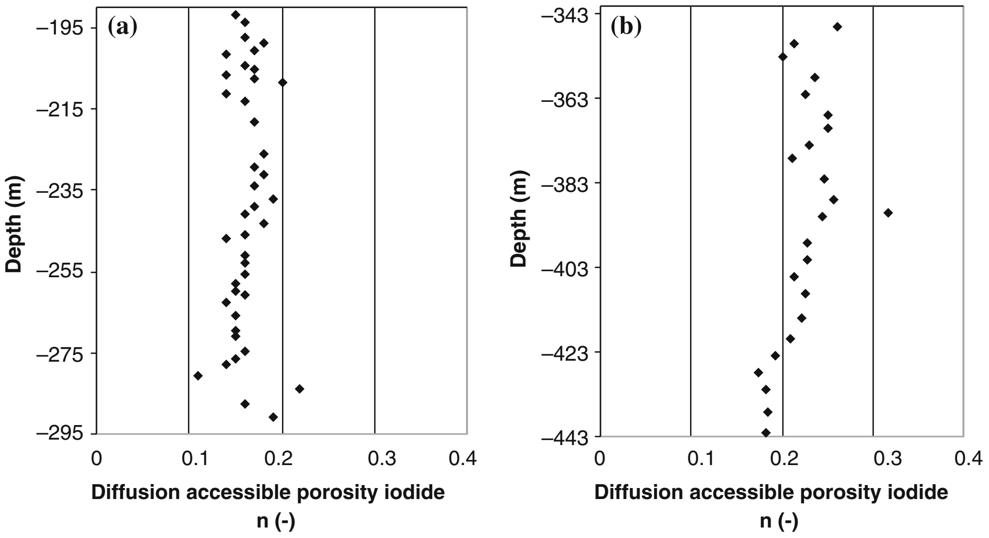

Fig. 4 Measured diffusion accessible porosity of iodide (m^2/s) of **a** the Boom Clay in the Mol-1 borehole and **b** the Ypresian Clay in the Doel borehole

sills are fitted by the optimization program LCMFIT2 (Pardo-Iguzquiza and Dowd 2002).

Stochastic sequential simulation of the transport parameters

The real spatial distributions of hydraulic conductivity, diffusion coefficient and diffusion accessible porosity of the Boom Clay and Ypresian Clay are not completely known. Therefore, a large number of equally probable random realizations of the clay layer are generated, using the modeled variograms and cross-variograms. The realizations honor the measured data and the mean, variance and variogram of the parameters. The Boom Clay and the Ypresian Clay shows a lateral continuity that largely exceeds the extent of the local scale model. Therefore it is assumed that the properties of the clays do not vary in the horizontal direction and one-dimensional vertical realizations of hydraulic conductivity, diffusion accessible porosity and diffusion coefficient are generated. The simulations are performed using direct sequential simulation with histogram reproduction (Oz et al. 2003). Figures 5 and 6 show examples of simulated fields of hydraulic conductivity, diffusion coefficient and diffusion accessible porosity in the Boom Clay and the Ypresian Clay.

Table 1 Average and variance of Boom Clay and Ypresian Clay parameters

	Boom Clay	Ypresian Clays
Vertical hydraulic conductivity average (m/s)	7.03e-12	5.84e-12
Vertical hydraulic conductivity variance (m^2/s^2)	3.42e-22	1.29e-22
Iodide diffusion coefficient average (m^2/s)	1.62e-10	2.30e-10
Iodide diffusion coefficient variance (m^4/s^2)	8.16e-21	9.65e-21
Iodide diffusion accessible porosity average	0.16	0.23
Iodide diffusion accessible porosity variance	0.00037	0.0012
Grain size (d_{40}) average[a] (μm)	3.79	7.43
Grain size (d_{40}) variance[a] (μm^2)	33.93	20.83
Gamma ray average (gAPI)	84.55	78.40
Gamma ray variance ($gAPI^2$)	104.41	116.65
Resistivity average (ohm m)	7.01	1.76
Resistivity variance ($ohm^2\ m^2$)	5.83	0.05

[a] Grain size is expressed by the parameter d_{40}, the grain size for which 40% of the total sample has a smaller grain size

Hydrogeological models

A local 3D hydrogeological model of the Boom Clay and a model of the Ypresian Clay are constructed. Both models have the same size and grid spacing. The model width in the x-direction is 20 m, i.e. half the assumed distance between the disposal galleries. The model length in the y-direction is 15 m. The model dimension of the Boom Clay model in the z-direction is 102 m, i.e. the total thickness of the Boom Clay in the nuclear zone of Mol-Dessel. The model dimension of the Ypresian Clay model in the z-direction is 104 m, i.e. the total thickness of the Ypresian Clay in the Doel nuclear zone. The grid spacing is 1m in the x-direction and in the y-direction and varies between 0.2 m and 1 m in the z-direction. The vertical boundary conditions for groundwater flow are zero flux boundary conditions since the hydraulic gradient is vertical. The horizontal boundary conditions for groundwater flow are Dirichlet conditions. The specified head at the upper boundary of

Table 2 Correlation coefficients between the parameters of the Boom Clay and the Ypresian Clays

	Boom clay	Ypresian clays
$\log_{10}K_{\mathrm{v}}-D$	0.97	0.88
$\log_{10} K_{\mathrm{v}}-\eta$	0.44	0.81
$\log_{10} K_{\mathrm{v}}$–GR	−0.65	−0.53
$\log_{10} K_{\mathrm{v}}$–RES	0.73	0.41
$\log_{10} K_{\mathrm{v}}$–grain size	0.95	0.78
$D-\eta$	0.36	0.80
D–GR	−0.63	−0.38
D–RES	0.66	0.53
D–grain size	0.93	0.92
η–GR	−0.20	−0.49
η–RES	0.20	0.36
η–grain size	0.28	0.03

the Boom Clay 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). The specified head at the lower boundary of the Ypresian Clay is 26.5 m higher than the specified head at the upper boundary (ONDRAF/NIRAS 2002). The vertical hydraulic gradient in the Ypresian Clay is more than 12 times larger than in the Boom Clay and oriented in the opposite direction. Although it is likely that these gradients vary over the long time periods considered, they are assumed to be constant in this study.

Transport by advection, dispersion, molecular diffusion and radioactive decay is calculated for three radionuclides: ^{79}Se, ^{129}I and ^{99}Tc. 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 properties of these radionuclides are given in Table 3. 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. In both models, the same source term is inserted. The source term models for the three 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. Since the initial radius of the cylindrical glass matrix would be 0.215 m, the glass matrix would be completely dissolved after approximately 70,000 years. The source term model is therefore a constant flux over a period of 70,000 years equal to the total radionuclide inventory divided by 70,000 years. If, however, this source term model resulted in calculated concentrations higher than the solubility limit, the source-term model was replaced by a constant concentration model. A constant concentration equal to the solubility limit was then prescribed until exhaustion of the source.

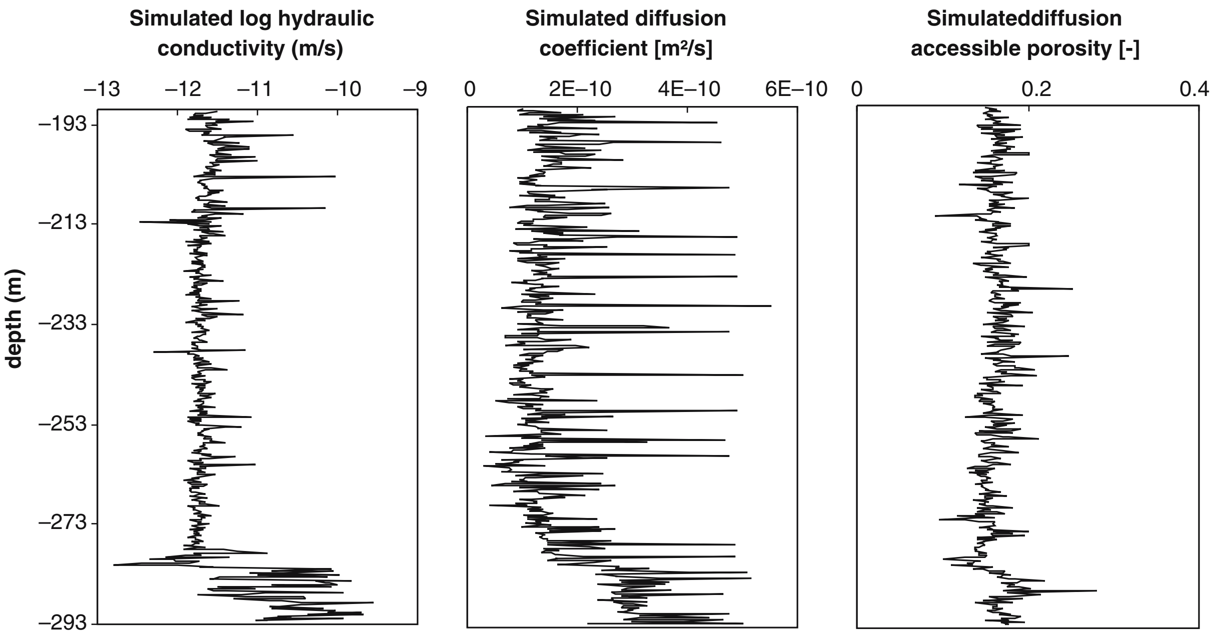

Fig. 5 Simulated hydraulic conductivity (m/s), diffusion coefficient (m^2/s) and diffusion accessible porosity of the Boom Clay in the Mol-1 borehole

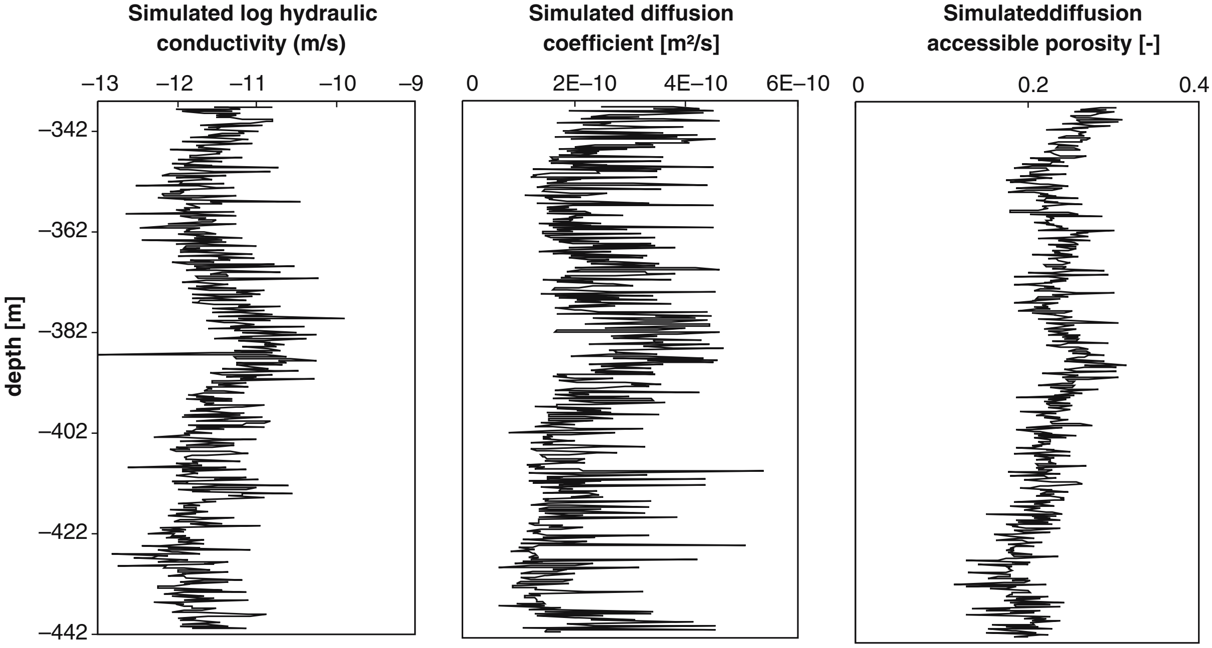

Fig. 6 Simulated hydraulic conductivity (m/s), diffusion coefficient (m^2/s) and diffusion accessible porosity of the Ypresian Clay in the Doel borehole

The random realizations of hydraulic conductivity are used as input in the model. For the radionuclide ^{129}I, the different equiprobable realizations of the diffusion coefficient and diffusion accessible porosity of iodide are directly imported in the model. For ^{79}Se and ^{99}Tc, 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 ^{79}Se and ^{99}Tc. The diffusion accessible porosity of these radionuclides is however different. While the average value of the diffusion accessible porosity of iodide is 0.16, ^{79}Se and ^{99}Tc 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 ^{79}Se and ^{99}Tc so that the average values of the simulated porosities were equal to 0.13 and 0.30.

The two local 3D hydrogeological models are run with FRAC3DVS, a simulator for 3D groundwater flow and solute transport in porous, discretely-fractured porous or dual-porosity formations (Therrien et al. 1996, 2003). The models are run for ten different random combinations of simulations of hydraulic conductivity, diffusion coefficient and diffusion accessible porosity.

Table 3 Properties of selected radionuclides (Values are taken from Mallants et al., 1999)

	^{79}Se	^{129}I	^{99}Tc
Half-life (year)	6.50×10^{4}	1.57×10^{7}	2.13×10^{5}
Decay constant (year^{-1})	1.07×10^{-5}	4.41×10^{-8}	3.25×10^{-6}
Solubility limit (mol l^{-1})	5.5×10^{-8}	-	3×10^{-8}
Retardation factor	1	1	1

Results

Figure 7 shows the computed total radionuclide fluxes through the lower and upper clay boundaries of the Boom Clay and the Ypresian Clay for ten different equally probably simulations. The total amount of radionuclides leaving the clay M (Bq) was calculated as flux integrated over time for each simulation and is also indicated on Fig. 7. For the Boom Clay, the calculated flux through the lower clay boundary is for all radionuclides larger than the calculated flux through the upper clay boundary. The difference between the total radionuclide amount leaving through the lower and upper clay boundary is between 6% (^{99}Tc) and 23% (^{79}Se). For the Ypresian Clay, the calculated flux through the upper clay boundary is for all radionuclides much larger than de calculated flux through the lower clay boundary. The total radionuclide amount leaving through the upper clay boundary is between 2.6 (^{99}Tc) and 4.8 (^{79}Se) times larger than the total radionuclide amount leaving through the lower clay boundary. Comparison of the total radionuclide amounts leaving the Boom Clay and the Ypresian Clay also shows that approximately twice as much radionuclides leave the Ypresian Clay compared to the Boom Clay.

In Fig. 8, a comparison is made between the radionuclide amounts leaving the clays calculated with heterogeneous simulations and homogeneous models with a homogeneous hydraulic conductivity, diffusion coefficient and diffusion accessible porosity equal to the arithmetic averages of the measurements. The figures show boxplots of the difference in percentage of total radionuclide amounts leaving the clays between the heterogeneous and the homogeneous model. A difference of 0% means that the calculated output radionuclide amount of the considered heterogeneous model is equal to the calculated output radionuclide amount of the homogeneous model. A negative difference means that the calculated output radionuclide amount of that heterogeneous model is smaller than the amount calculated with the homogeneous model. A positive difference means that the calculated output radionuclide amount of that heterogeneous model is larger than the amount calculated with the homogeneous model. Compared to the homogeneous model, the calculated radionuclide amount flowing through the lower Boom Clay boundary is between 27 and 2% smaller in the heterogeneous models. The calculated radionuclide activity flowing through the upper Boom Clay boundary is between 22 smaller and 19% larger. For the Ypresian Clay, the calculated radionuclide activity flowing through the lower clay boundary is between 5 smaller and 59% larger in the heterogeneous models. The calculated radionuclide activity flowing through the upper Ypresian Clay boundary is between 40 smaller and 8% smaller in the heterogeneous models. These values show that incorporating parameter heterogeneity has a larger effect in the Ypresian Clay than in the Boom Clay.

Discussion

In the Ypresian Clay, larger differences between the fluxes through the lower and the upper clay boundary and larger total output radionuclide amounts are calculated. Differences between the fluxes through the lower and the upper clay boundaries can only be attributed to transport by advection since in a pure diffusion model with a source in the middle of the clay the output fluxes through the lower and the upper clay boundary would be identical. These results show that the effect of upward advective transport in the Ypresian Clay is much larger than the effect of downward advective transport in the Boom Clay. Since all flow and transport parameters have similar values in both formations, this difference in results is probably due to the difference in hydraulic gradient. The gradient in the Ypresian Clay is more than twelve times larger than in the Boom Clay and oriented in the opposite direction. This results in a larger contribution of transport by advection in the Ypresian Clays and in larger differences between the fluxes through the lower and the upper clay boundary and larger total output radionuclide amounts in the Ypresian Clay.

The larger effect of parameter heterogeneity in the Ypresian Clay can also not be completely explained by differences in parameter variability. All transport parameters have mean values and variances in the same order of magnitude, as demonstrated by the statistics in Table 1. Detailed examination of the effect of heterogeneity in the Ypresian Clay shows that the heterogeneity of hydraulic conductivity has a larger effect than the heterogeneity of the diffusion parameters in this clay. The larger effect of parameter heterogeneity in the Ypresian Clay is therefore mainly a larger effect of hydraulic conductivity heterogeneity in the Ypresian Clay compared to the Boom Clay. Since the hydraulic conductivity variation is not significantly larger in the Ypresian Clay compared to the Boom Clay, the higher effect of K heterogeneity is probably also caused by the larger gradient. Since the gradient is larger, tranport by advection is a more important process in the Ypresian Clay. Therefore, the results are more sensitive to K heterogeneity.

Since both the radionuclides fluxes and the effect of heterogeneity on these fluxes seems to be largely affected by the direction and magnitude of the hydraulic gradient, modeling radionuclide transport with an incorrect gradient would result in erroneous predictions. The direction and magnitude of the gradient in nuclear waste disposal studies are however subject to large uncertainty due to the large time periods considered. This means that it is almost impossible to predict the evolution of the hydraulic gradient over periods of several hundred thousand years. Since the model results are sensitive to the magnitude and direction of the gradient, which are often uncertain over the considered time periods, it is advisable to calculate radionuclide fluxes with different gradient evolution scenarios. This study thus illustrates the importance of using a range of possible hydraulic gradients as input for safety studies.

Conclusion

In this study, the radionuclide fluxes that would migrate from a potential nuclear waste repository through the

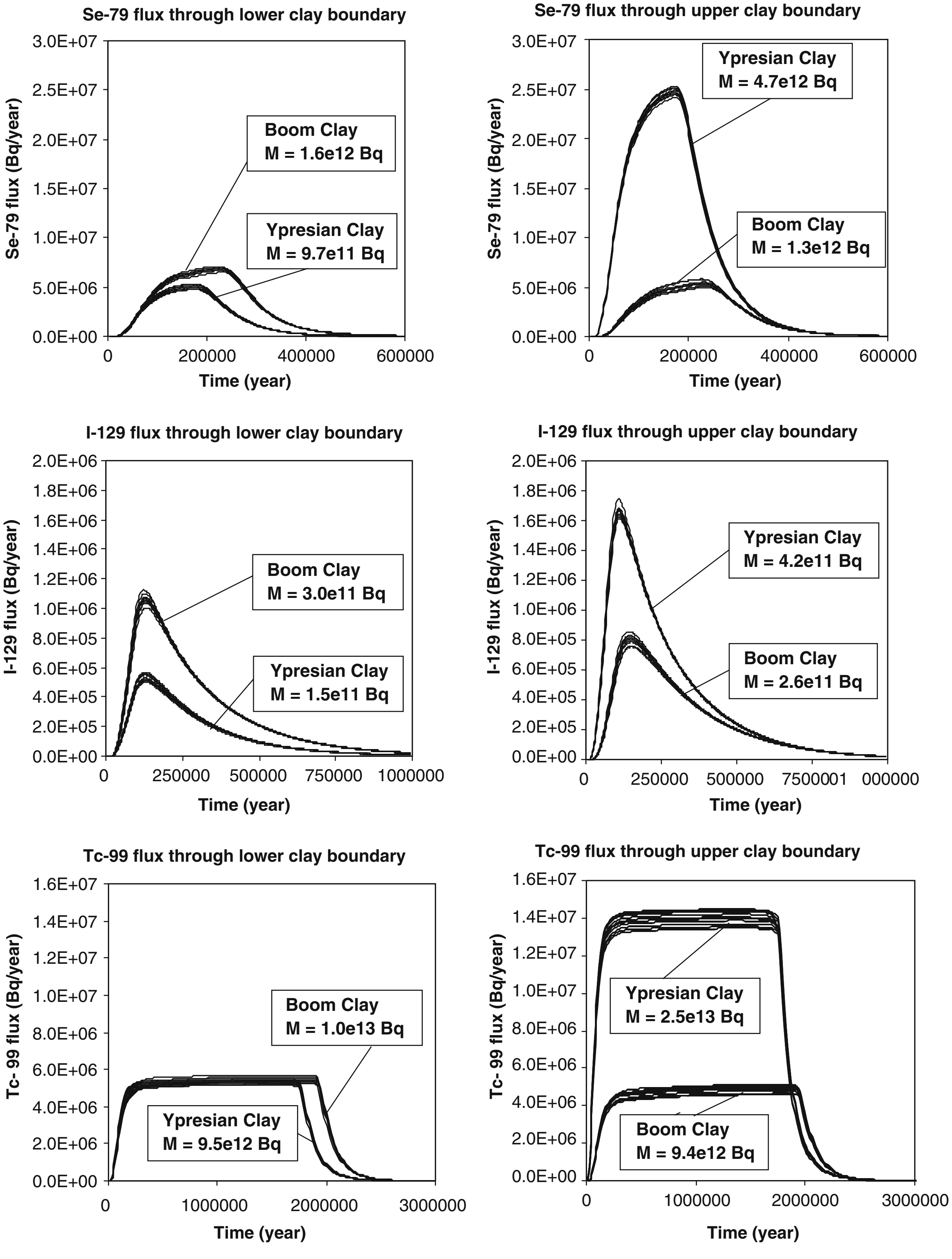

Fig. 7 Computed total radionuclide fluxes (Bq/year) versus time (year) through the lower and upper clay boundaries of the Boom Clay and the Ypresian Clay for ten different simulations

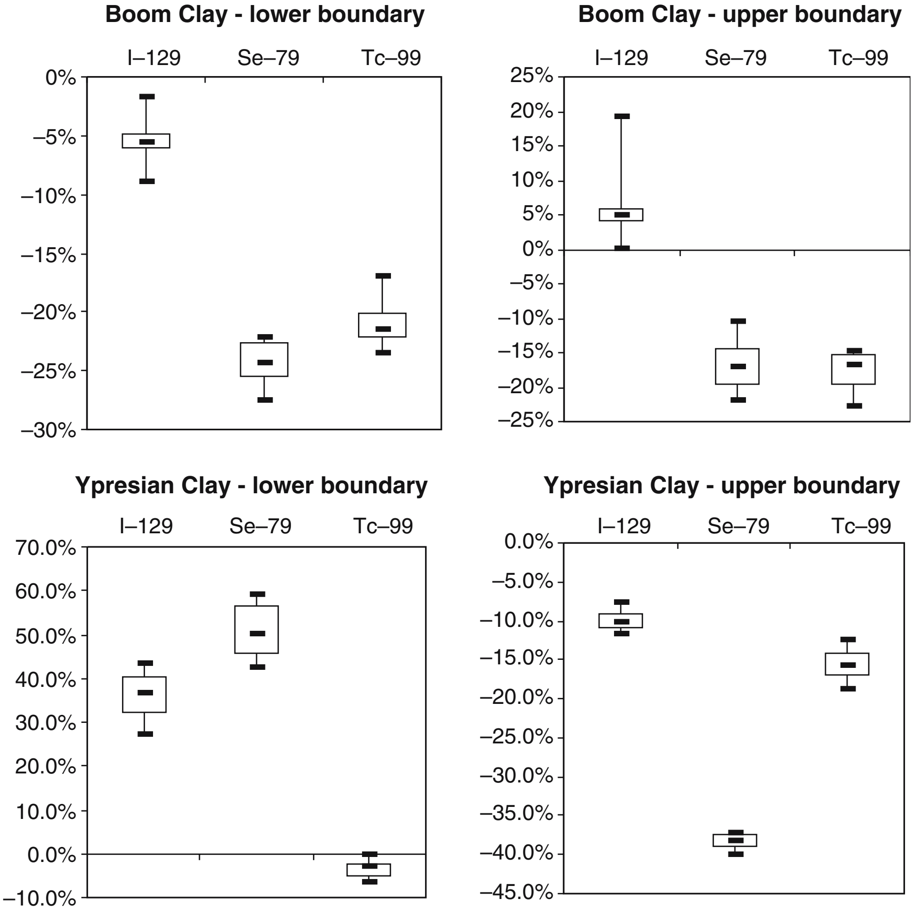

Fig. 8 Boxplots of the difference in percentage of total radionuclide activity between the heterogeneous models and the homogeneous model with a homogeneous hydraulic conductivity, diffusion coefficient and diffusion accessible porosity equal to the arithmetic averages of the measurements

Boom Clay and the Ypresian Clay were modeled and compared. Two hydrogeological models were built to calculate the radionuclide fluxes through these two clay formations. Transport parameter heterogeneity was incorporated in the models using geostatistical co-simulations of hydraulic conductivity, diffusion coefficient and diffusion accessible porosity. The calculated radionuclide fluxes in the two clay formations were compared with the results from homogeneous models and with the results of the other clay formation.

A first conclusion of this study is that differences of up to 59% of the calculated output radionuclide amounts between heterogeneous and homogeneous models are observed. This study thus demonstrates that parameter heterogeneity can have an important effect on the results and should be incorporated in transport studies in low permeability media.

Comparison of the results of the Boom Clay and the Ypresian Clay show that in the Ypresian Clay (1) larger differences between the fluxes through the lower and the upper clay boundary occur, (2) larger total output radionuclide amounts are calculated and (3) a larger effect of parameter heterogeneity on the calculated fluxes is observed. These results are explained by the larger and inversely oriented hydraulic gradient in the Ypresian Clay that results in a larger importance of transport by advection in this clay. Since both the radionuclides fluxes and the effect of heterogeneity on these fluxes are largely affected by the direction and magnitude of the hydraulic gradient and since the gradient in nuclear waste disposal studies is subject to large uncertainty due to the large time periods considered, this study illustrates the importance of using a range of possible hydraulic gradients as input for safety studies.

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References

Huysmans M, Dassargues A (2005a) Stochastic analysis of the effect of heterogeneity and fractures on radionuclide transport in a low permeability clay layer. Environ Geol 48(7):920–930

Huysmans M, DassarguesA (2005b) Stochastic analysis of the effect of spatial variability of diffusion parameters on radionuclide transport in a low permeability clay layer. In: Proceedings of ModelCARE2005, the 5th international conference on calibration and reliability in groundwater modelling: from uncertainty to decision making, The Hague (Scheveningen), The Netherlands, 6–9 June 2005

Isaaks EH, Srivastava RM (1989) An introduction to applied geostatistics. Oxford University Press, New York

Mallants D, Sillen X, Marivoet J (1999) Geological disposal of conditioned high-level and long lived radioactive waste: Consequence analysis of the disposal of vitrified high-level waste in the case of the normal evolution scenario. ONDRAF/NIRAS report R-3383, Brussel, Belgium

Mallants D, Marivoet J, Sillen X (2001) Performance assessment of vitrified high-level waste in a clay layer. J Nuclear Material 298(1–2):125–135

ONDRAF/NIRAS (2002) Safety assessment and feasibility interim report 2 - SAFIR 2. NIROND 2001 - 06 E, Brussel, Belgium

Oz B, Deutsch CV, Tran TT, Xie Y (2003) DSSIM-HR: A FORTRAN 90 program for direct sequential simulation with histogram reproduction. Comput Geosci 29(1):39–51

Pardo-Iguzquiza E, Dowd PA (2002) FACTOR2D: a computer program for factorial cokriging. Comput Geosci 28(8):857–875

Therrien R, Sudicky EA (1996) Three-dimensional analysis of variably-saturated flow and solute transport in discretely-fractured porous media. J Contaminant Hydrol 23(1–2):1–44

Therrien R, Sudicky EA, McLaren RG (2003) FRAC3DVS: An efficient simulator for three-dimensional, saturated-unsaturated groundwater flow and density dependent, chain-decay solute transport in porous, discretely-fractured porous or dual-porosity formations, User's guide

Van Marcke Ph, Laenen B (2005) The Ypresian clays as possible host rock for radioactive waste disposal: an evaluation. ONDRAF/NIRAS document in-press

Wemaere I, Marivoet J (1995) Geological disposal of conditioned high-level and long lived radioactive waste: updated regional hydrogeological model for the Mol site (The north-eastern Belgium model). ONDRAF/NIRAS Report R-3060, Brussel, Belgium