Hydrogeological Modeling of Radionuclide Transport in Heterogeneous Low-Permeability Media: A Comparison Between Boom Clay and Ieper Clay

M. Huysmans and A. Dassargues

Abstract Deep low-permeability clay layers are considered as possible suitable environments for disposal of high-level radioactive waste. In Belgium, the Boom Clay is the reference host formation and the Ieper 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 parameters' heterogeneity is incorporated in the models using stochastic sequential simulation of hydraulic conductivity, diffusion coefficient and diffusion accessible porosity, using primary information and several types of secondary information, i.e. resistivity, gamma ray and grain size. The calculated radionuclide fluxes in the two clay formations are compared. Results show that in the Ieper Clay larger differences between the fluxes through the lower and the upper clay boundary occur than in the Boom Clay, larger total output radionuclide amounts are calculated than in the Boom Clay, and a larger effect of parameter heterogeneity on the calculated fluxes is observed, compared to the Boom Clay.

1 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 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 (ONDRAF/NIRAS, 2002).

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The aim of this study is 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; Huysmans and Dassargues, 2005b) 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.

2 Method

2.1 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. The Doel nuclear zone (province of Antwerp) is an alternative reference site for methodological studies regarding the Eocene Ieper Clay. 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.

2.2 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, measurements of secondary variables were also collected. Geophysical 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, 41 diffusion coefficient and diffusion accessible porosity measurements, 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

Table 1 Average and variance of Boom Clay and Ieper Clay parameters (Huysmans and Dassargues, 2006)

	Boom Clay	Ieper 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(1) (μm)	3.79	7.43
Grain size (d_{40}) variance(1) ($μm^2$)	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

(1) Grain size is expressed by the parameter d_{40}, the grain size for which 40% of the total sample has a smaller grain size.

depth of 688 m. Laboratory experiments on cores from the Doel boreholes provided 25 hydraulic conductivity values, 25 diffusion coefficient and diffusion accessible porosity measurements and 49 grain size measurements of the Ieper Clay. Geophysical logging provided logs of gamma ray and resistivity.

Comparison of the statistics of the parameters of the Boom Clay and Ieper 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 Ieper 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 35 m for the Boom Clay and 24 m for the Ieper Clay.

Table 2 Correlation coefficients between the parameters of the Boom Clay and the Ieper Clays (Huysmans and Dassargues, 2006)

	Boom Clay	Ieper Clays
$\log_{10} K_v$ - D	0.97	0.88
$\log_{10} K_v$ - η	0.44	0.81
$\log_{10} K_v$ - GR	−0.65	−0.53
$\log_{10} K_v$ - RES	0.73	0.41
$\log_{10} K_v$ - grain size	0.95	0.78
D - η	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 sills are fitted by the optimization program LCMFIT2 (Pardo-Iguzquiza and Dowd 2002).

2.3 *Stochastic Sequential Simulation of the Transport Parameters*

The Boom Clay and the Ieper 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 1 and 2 show examples of simulated fields of hydraulic conductivity, diffusion coefficient and diffusion accessible porosity in the Boom Clay and the Ieper Clay.

2.4 *Hydrogeological Models*

A local 3D hydrogeological model of the Boom Clay and a model of the Ieper Clay are constructed. Both models have the same size (20 m × 15 m × 102 m/104 m) and grid spacing (between 0.2 m and 1 m). 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 vertical hydraulic gradient is approximately 0.02 in the Boom Clay (Wemaere and Marivoet, 1995) and 0.25 in the Ieper Clay (ONDRAF/NIRAS 2002). The vertical hydraulic gradient in the Ieper Clay is more than twelve times larger

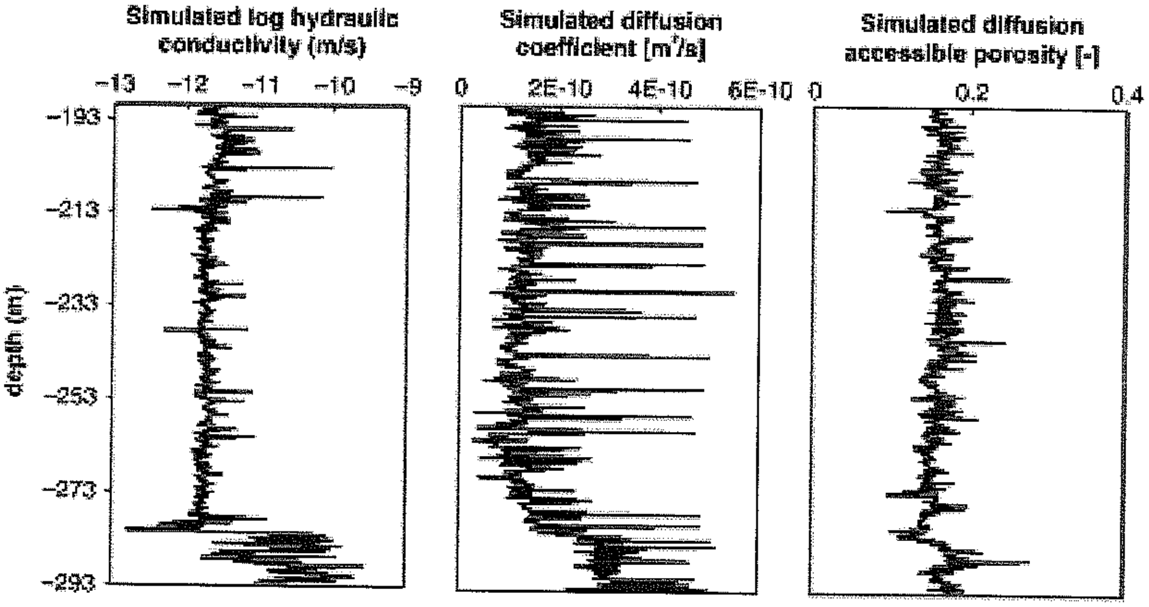

Fig. 1 Simulated hydraulic conductivity (m/s), diffusion coefficient (m^2/s) and diffusion accessible porosity (-) of the Boom Clay in the Mol-1 borehole (Huysmans and Dassargues, 2006)

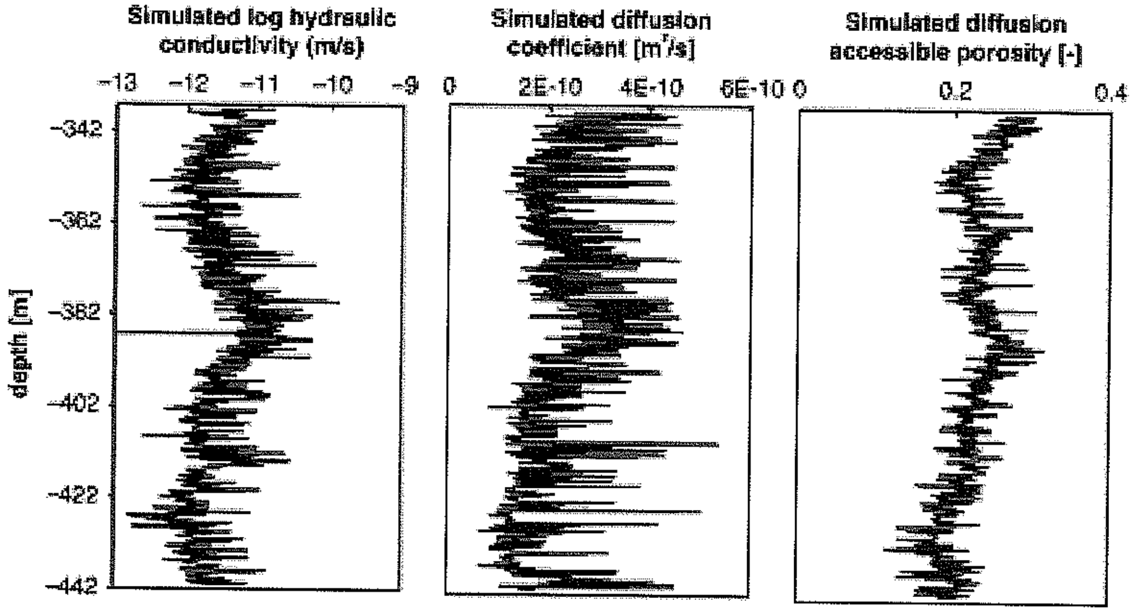

Fig. 2 Simulated hydraulic conductivity (m/s), diffusion coefficient (m²/s) and diffusion accessible porosity (-) of the Ieper Clay in the Doel borehole (Huysmans and Dassargues, 2006)

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 3 radionuclides: ^{79}Se, ^{129}I and ^{99}Tc. 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: an applied flux source or an applied concentration source depending on the effect of the solubility limit (Mallants et al., 1999). The initial transport condition is a zero concentration condition.

The 2 local 3D hydrogeological models are run with FRAC3DVS, a simulator for three-dimensional groundwater flow and solute transport in porous, discretely-fractured porous or dual-porosity formations (Therrien and Sudicky, 1996; Therrien et al., 2003). The models are run for 10 different random combinations of simulations of hydraulic conductivity, diffusion coefficient and diffusion accessible porosity.

3 Results

Figure 3 shows the computed total radionuclide fluxes through the lower and upper clay boundaries of the Boom Clay and the Ieper Clay for 10 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

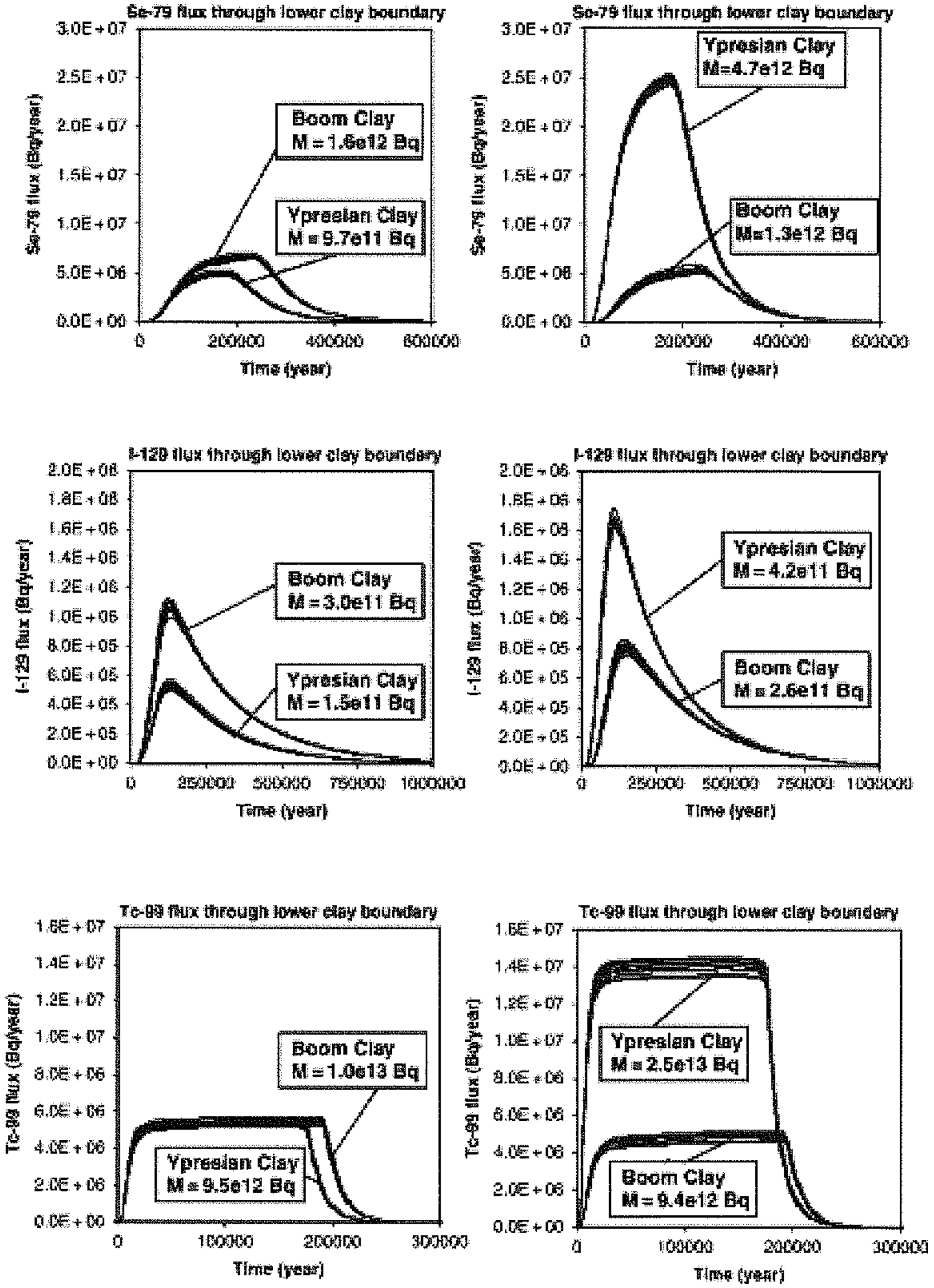

Fig. 3 Computed total radionuclide fluxes (Bq/year) versus time (year) through the lower and upper clay boundaries of the Boom Clay and the Ieper Clay for 10 different realizations of random fields of the transport parameters (Huysmans and Dassargues, 2006)

on Fig. 3. For the Boom Clay, 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 Ieper Clay, 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 Ieper Clay also shows that approximately twice as much radionuclides leave the Ieper Clay compared to the Boom Clay.

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. Arithmetic averages instead of effective parameters were chosen to compare the heterogeneous models of this study with earlier homogeneous models made by other agencies that used the arithmetic average. For the Boom Clay, there is a maximum difference of 27% between the radionuclide amounts calculated by the homogeneous and heterogeneous models. For the Ieper Clay, there is a maximum difference of 59% between the radionuclide amounts calculated by the homogeneous and heterogeneous models. These values show that incorporating parameter heterogeneity has a larger effect in the Ieper Clay than in the Boom Clay.

4 Discussion

In the Ieper Clay, larger differences between the fluxes through the lower and the upper clay boundary and larger total output radionuclide amounts are calculated than in the Boom Clay. 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 Ieper 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 Ieper 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 Ieper Clay than in the Boom Clay, in larger differences between the fluxes through the lower and the upper clay boundary and larger total output radionuclide amounts in the Ieper Clay than in the Boom Clay.

The larger effect of parameter heterogeneity in the Ieper Clay compared to the Boom 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 Ieper 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 Ieper Clay is therefore mainly a larger effect of hydraulic conductivity heterogeneity in the Ieper Clay compared to the Boom Clay. Since the hydraulic conductivity variation is not significantly larger in the Ieper 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 Ieper Clay. Therefore, the results are more sensitive to *K* heterogeneity.

5 Conclusions

In this study, the radionuclide fluxes that would migrate from a potential nuclear waste repository through the Boom Clay and the Ieper 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 Ieper Clay show that in the Ieper 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, compared to the Boom Clay. These results are explained by the larger and inversely oriented hydraulic gradient in the Ieper 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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