Contribution to the simulation of nitrates transport in a double-porosity aquifer

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Abstract

Nitrates pollutions of the groundwater occur more and more often, especially in intensive agricultural areas. The aquifer of "Hesbaye" constitutes the main drinking water supply of the whole city of Liège (Belgium). This Cretaceous chalky aquifer is concerned with increasing concentrations of nitrates. One of the major difficulties to describe accurately the transport of nitrates in this aquifer, comes from the effect of the two porosities: the primary porosity due to the pores in the chalk matrix, and the secondary porosity due to fractures and fissures. To represent and simulate the contamination of the fracture water, the working assumption of a Representative Elementary Volume (REV) is used and the model is governed by classical advection-dispersion equations. Inside the REV, the contamination of the pore water is taken into account considering a transfer coefficient from the mobile water to the immobile water in the pores, representing diffusion processes into the pores of the chalk matrix. Experimental data have been collected from various laboratory tests, providing among others, the molecular diffusion coefficient and dispersivity values, needed to determine the transfer coefficient in the REV. Then, simulations using a finite element code with an objective upwind method and a special time integration scheme, have been tested. The model can solve a transient 2D convection-dispersion equation taking into account additional processes as linear adsorption on the solid, and immobile water effect.

Introduction

Few transport models have been performed for the simulation of the nitrates contamination in aquifers, objecting that nitrates transfer is complicated by complex chemical influences in the soils (non linear and non isotherm adsorption, biodegradation, ...). Those phenomena can be depicted with special chemical models.
In the "Hesbaye" aquifer (near Liège in Belgium), we have considered for the study described here, that nitrates ions become chemically stable when they reach the Cretaceous chalk aquifer. This water table aquifer is in a double porosity chalk: as well porous as fissured. The groundwater flow and transport parameters depend strongly on the scale of observation. The pore permeabilities have been evaluated by laboratory tests. The results provide a pore permeability coefficient of 10^{-8} m/s. In another hand, global permeability coefficients (taking into account heterogeneities, fissures,...) are available from pumping tests. Those coefficients vary from 10^{-5} to 10^{-3} m/s, so that it can be assumed that pore water is "immobile" water. Finally, the aquifer is far larger than a representative element of volume (REV). Hence, it is possible to describe the pollution with the "determinist" system of advection-diffusion equations, applied to an "equivalent" porous medium (Biver, 1993):

$$R_{d_m} \cdot \frac{\partial C_m}{\partial t} + div(v . C_m) - div(\underline{D_h} . gradC_m) + A_m . C_m - \alpha_m . C_{im} = 0 \qquad (1)$$

$$R_{d_{im}} \cdot \frac{\partial C_{im}}{\partial t} + A_{im} . C_{im} - \alpha_{im} . C_m = 0 \qquad (2)$$

$$F + K_d . (C_{im} + C_m) = 0 \qquad (3)$$

(1), (2) and (3) are the mass balance equations respectively in the mobile water, immobile water and solid, with coefficients defined as following :

$R_{d_{im}} = 1 + \frac{\theta_s}{\theta_{im}} . \rho_s . (1 - p) . K_d$ the retardation factor of immobile water

$R_{d_m} = 1 + \frac{\theta_s}{\theta_m} . \rho_s . p . K_d$ the retardation factor of mobile water

$A_{im} = k_{im} + (R_{d_{im}} - 1) . k_s + \alpha_{im}$

$A_m = k_m + (R_{d_m} - 1) . k_s + \alpha_m$

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$\alpha_{im} = \alpha_d^* / \theta_{im}$
$\alpha_m = \alpha_d^* / \theta_m$
v = average effective velocity
$\underline{D_h}$ = hydrodynamic dispersion tensor
C_m = mobile water volumic concentration
C_{im} = immobile water volumic concentration
θ_x = volumic part of phase x (mobile water, immobile water or solid)
k_x = degradation constant of pollutant in phase x (mobile water, immobile water or solid)
p = part of solid-liquid contact surface concerning mobile water
α_d^* = transfer constant between mobile and immobile water
ρ_s = specific mass of the solid

This system takes into account immobile water effect, linear adsorption and degradation, convection, diffusion and mechanical dispersion; but it needs to be considered at the representative elementary volume (REV) scale, and many parameters are to be determined.
In Cretaceous chalk, we may assume that $A_{im} = \alpha_{im}$, $A_m = \alpha_m$, $R_{d_{im}} = R_{d_m} = 1$ (as degradation and adsorption are considered as low at this stage of the study); v is given by a 3D groundwater finite element model (Dassargues *et al.*, 1988, and Dassargues, 1994) ; D_h is obtained by in situ tracer tests (Biver, 1994). The last parameter to evaluate is α_d^* . At this stage, one must notice that this coefficient allows the representation of a porous matrix effect, even if the permeability values of the porous chalk are much lower than those of the fissures. Hence, a detailed study of the transfer processes in the non-fissured porous chalk was necessary to prove that the proposed theoretical concept is more or less equivalent to the physical processes.

Experimental work

a. Test on the porous matrix

Two series of tests were performed with a system which allows constant concentration boundary conditions by circulating fluids. The first tests are only diffusive with various length-times (from 8 hours to 1 week), the second ones are dispersive with various pore pressure gradients (from 0 to 0.875). All the results are presented on figures 1 and 2. Nitrates contents in the chalk are determined by chemical analysis. The sample is sliced, each slice is crushed and leached with water, and finally the nitrate concentration of the obtained solution is measured with a specific electrode. Those tests have been analytically and numerically interpreted. Considering that no adsorption occurs in porous chalk, the molecular diffusion coefficient *Dm* of this medium has been evaluated to 4.5 10^{-10} m^2/s (diffusive tests).

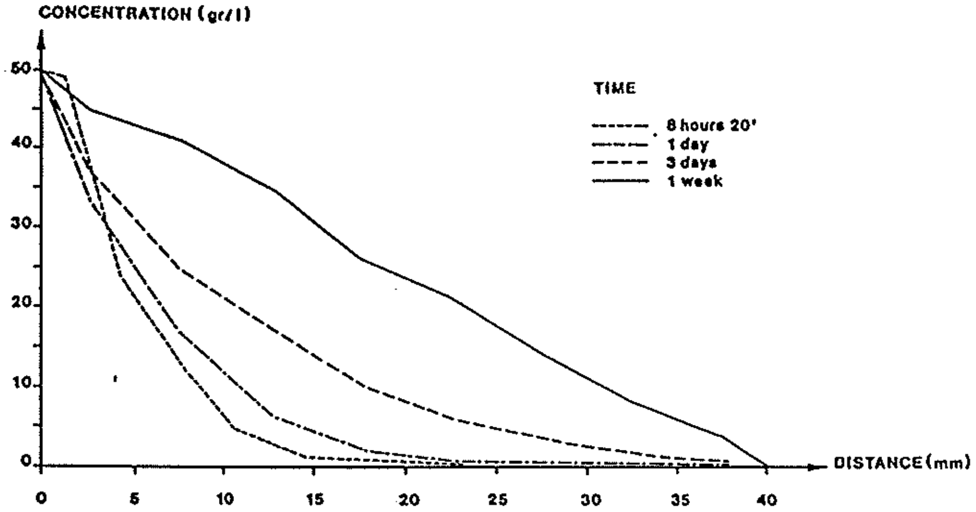

Figure 1: Molecular diffusion in the chalk matrix

When a fluid velocity is acting (dispersive tests), it has been also possible to determine a longitudinal dispersivity a_L equal to 0.11 m. Those values are comparable with those obtained by Porel (1988) with laboratory uranine tracer tests in chalky samples: 3.4 10^{-10} m^2/s for molecular diffusion coefficient and 0.02 to 0.2 m for longitudinal dispersivity.

To be sure that the measured longitudinal dispersion is an intrinsic parameter, a correlation of longitudinal hydrodynamic coefficient with effective velocity has been done (Fig. 3). A quasi linear regression is possible except for two values. This can be explained if we know that distinction between pore and fissure is not always very strict; in fact, micro fissures may often be detected at the scale of the sample.

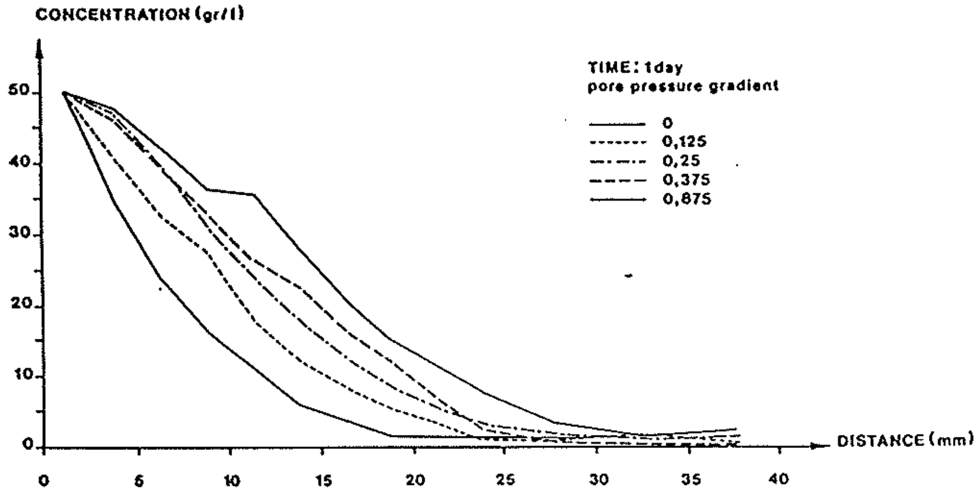

Figure 2: Dispersion tests in the chalk porous matrix

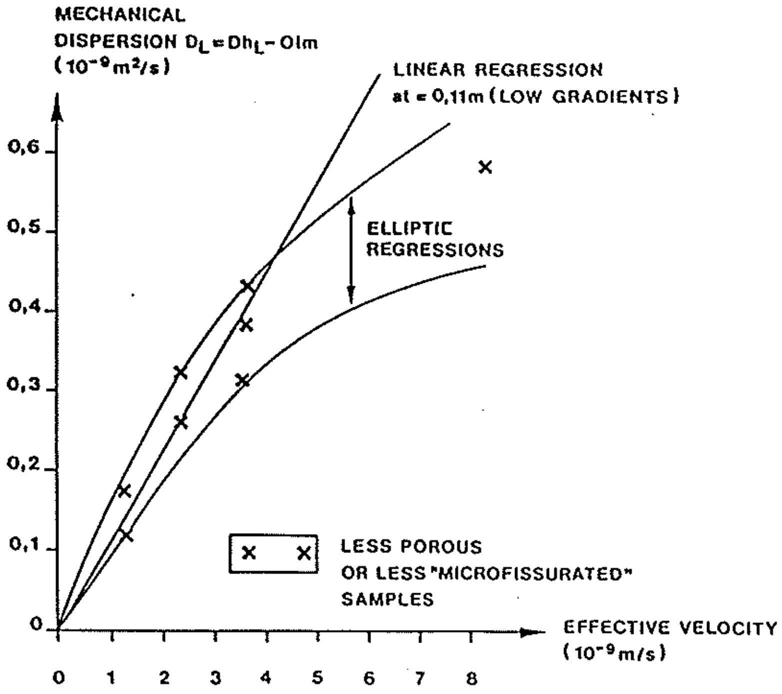

Figure 3: Mechanical dispersion correlated to effective velocity

b. Determining a transfer coefficient of the immobile water

Let us consider that main part of immobile water is pore water (42% of the total volume against less than 2% in the fissures). By simulating the pollution of a block matrix with constant boundary concentration, it is possible to evaluate the time evolution of the pore water average concentration and to deduce the value of α_d^* . The mass balance equation in immobile water, without degradation and adsorption, is written :

$$\frac{\partial C_{im}}{\partial t} + \frac{\alpha_d^*}{\theta_{im}} . (C_{im} - C_m) = 0 \tag{4}$$

if C_m does not vary with time, this equation may be integrated as follow :

$$C_{im} = C_m . (1 - e^{(-\alpha_d^*/\theta_{im}).t}) \tag{5}$$

as θ_{im} ($\theta_{pores} = 42\%$), C_m , t are known, if C_{im} is given by numerical simulation, α_d^* is determined. Such a numerical simulation has been performed. The α_d^* value deduced at each time has been plotted (Fig. 4). It does not vary a lot (from 6.8 10^{-8} s^{-1} to 2.4 10^{-7} s^{-1}) and that's why this parameter can be considered as an intrinsic characteristic of the equivalent porous medium. For this first approach, mechanical dispersion has not been taken into account.

Another simulation could be done including this second phenomenon, but it is certainly not very important because the maximum hydraulic gradient acting in the aquifer is about 0.5 10^{-2} (Dassargues *et al.*, 1991); the corresponding mechanical dispersion in the porous block matrix represents only 5 percents of the total hydrodynamic dispersion (the transport is mainly diffusive in the matrix).

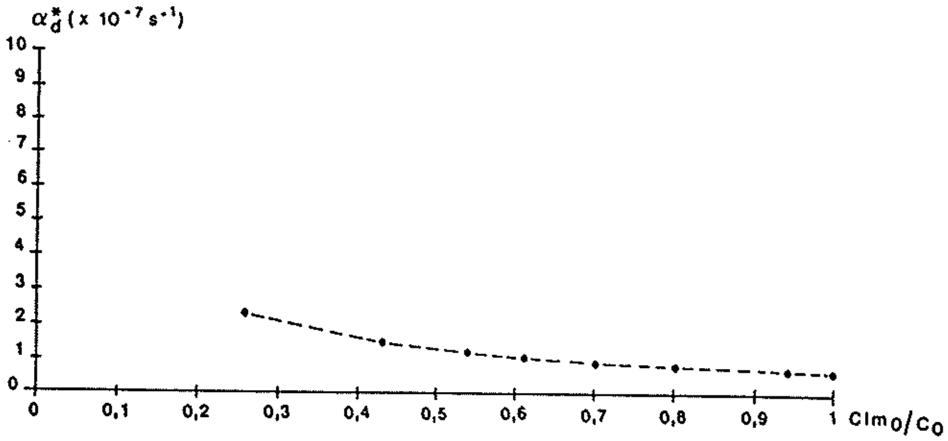

Figure 4: Evolution of α_d^* when pollution is acting on the block of chalk matrix

Numerical considerations

a. Basic concepts of the Full Upwind Petrov Galerkin method (FUPG)

The classical finite element method are not efficient to solve advection diffusion equation, and a new integration method is necessary to evaluate a correct average equilibrium. Suppose that *L(Cm) = 0* is the local equilibrium equation. The average equilibrium is obtained as follow :

$$\int_{\Delta t} \int_{\Delta V} L\Big(N_1(t').N_i(\underline{x}).C_{m_i}^{t_n} + N_2(t').N_i(\underline{x}).C_{m_i}^{t_{n+1}}\Big).W_i(\underline{x},t').dV.dt' = 0 \tag{6}$$

for *i = 1,N* where $N_1(t') = (1 - t')/\Delta t$

$N_2(t') = t'/\Delta t$ are classical temporal interpolation functions

$N_i(\underline{x})$ are the spatial interpolation functions

C_{m_i} are nodal concentrations

$W_i(\underline{x}, t)$ are the test functions or weight functions.

The test functions may vary in time and space. In classical finite element method, $W_i(\underline{x}, t') = N_i(\underline{x}).W(t')$; it is the Bubnov-Galerkin formulation. In the FUPG method established by Yu and Heinrich (1986), other test functions are used :

$$W_i(\underline{x}, t) = N_i(\underline{x}).F(t') + \alpha.\frac{\|\Delta x\|}{2}.\frac{\underline{v}}{\|\underline{v}\|}.\underline{grad}(N_i(\underline{x})).F(t')$$
$$+\beta.\frac{\|\Delta x\|}{2}.\frac{\underline{v}}{\|\underline{v}\|}.\underline{grad}(N_i(\underline{x})).\frac{\Delta t}{2}.\frac{\partial F(t')}{\partial t'} \qquad (7)$$

where $F(t') = 4.N_1(t').N_2(t')$, α and β are upwind parameters chosen to obtain exact nodal solution for a one dimensional problem.

b. Modelling secondary process of the nitrates transport

(1) Degradation and adsorption

As the FUPG method is a consistent integration, it is not difficult to take into account those phenomena in the model if the same interpolation is used for those terms. Results prove good accuracy for a pulse migration with degradation or adsorption. Concerning linear adsorption, it is not surprising as it only induces a scalar constant multiplication of the concentration time derivative. For degradation Codina *et al.* (1990) have proved that with a consistent upwind method it is possible to keep good accuracy for linear degradation terms.

(2) Immobile water effect

If time discretization of C_m is introduced in the mass balance equation of immobile water, it can be written as follow :

$$R_{d_{im}}.\frac{\partial C_{im}}{\partial t'} + A_{im}.C_{im} - \alpha_{im}\left[N_1(t').C_m^{t_n} + N_2(t').C_m^{t_{n+1}}\right] \qquad (8)$$

This equation is analytically solved and time variation of immobile water is found to be :

$$C_{im}(t') = \left[C_{im}^{t_n} - \alpha_{im}.\frac{R_{d_{im}}}{A_{im}}.(\frac{C_m^{t_n}}{R_{d_{im}}} - \frac{\dot{C}_m^{t_n \rightarrow t_{n+1}}}{A_{im}})\right].e^{-A_{im}t'/R_{d_{im}}}$$
$$+\alpha_{im}.\frac{R_{d_{im}}}{A_{im}}.\left(\frac{C_m^{t_n}}{R_{d_{im}}} - \frac{\dot{C}_m^{t_n \rightarrow t_{n+1}}}{A_{im}}\right) \qquad (9)$$

where $\dot{C}_m^{t_n \rightarrow t_{n+1}} = \frac{C_m^{t_{n+1}} - C_m^{t_n}}{\Delta t}$

The equation (9) is introduced in the mass balance equation of mobile water. The resulting equation involves only concentration in mobile water. It can be solved numerically by the SUPG method (Spatial Upwind Petrov Galerkin method). As the SUPG test function vary with time, the immobile water effect is taken into account over all the time step by the time function $F(t')$ and its time-derivative. Without modifying the values of the upwind parameters α and β, good results are obtained either in 1D or 2D problems (Biver, 1993).

Conclusions

A determinist approach for modelling pollutant transport in a double porosity medium has been presented. The first stage of the experimental study has provided characteristics of the immobile water effect. The immobile water transfer coefficient can be considered as an intrinsic characteristic of the porous medium. This parameter has been evaluated to $2\ 10^{-7}\ s^{-1}$ for a fissured Cretaceous chalk.

The numerical development needed when taking into account this immobile water effect are presented in the context of the finite element method. Without modifying the amount of upwind, it is possible to consider a wide range of values for immobile water transfer coefficient. More details about the results of the method can be found in Biver & Dassargues (1994).

These developments have been revealed very useful to interpret the measured non symmetric breakthrough curves of in situ tracer tests and particular concentration profile of dispersion measured in laboratory tests on large fissured samples.

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