# Release of clay particles from an unconsolidated clay–sand core: experiments and modelling

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## Abstract

This work identifies the main phenomena that control the peptisation and transport of clay particles in a sand core. Clay can be dispersed into small particles in an aqueous solution of low ionic strength. This property is used to generate clay particles with NaCl concentration varying from 0.5 M to 0.015 M. For this purpose, a chromatographic column is initially packed with a 5% clay–sand mixture. The monitored decrease of the NaCl concentration of the feed solution allows the control of transport of the particles without plugging the porous medium. In this condition it is shown that in a column of a given length, the amount of clay particles released into solution and available to transport, depend only on NaCl concentration. Some clay particles are available to migration when the NaCl concentration of the feed concentration is between 0.16 M and 0.05 M (first domain) or between 0.035 M and 0.019 M (second domain). An empirical function, $P_d$([NaCl]), accounts for this particle generation. Transport is mainly dependent on the hydrodynamic characteristics of the porous medium that vary during the elution, probably owing to the particle motion inside the column. A phenomenological modelling is derived from these results, coupling the particle generation term, $P_d$([NaCl]), with an adapted nonequilibrium transport solute model. Similarly to the solute, particles were attributed a characteristic time of mass transfer between mobile and immobile water zones. This is sufficient to take into account the kinetic limitations of particles transport. The values of the parameters are determined by independent experiments. Finally, breakthrough curves of clay particles are predicted when a column of a given length is flushed by a salinity gradient of NaCl in various conditions. © 1997 Elsevier Science B.V.

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## 1. Introduction

Particles are suspected to be involved in the transport of pollutants in soils (Avogadro and De Marsily, 1984). The possible phenomena involved in the control of particle transport are linked to the physical properties of the particles in solution (flocculation or dispersion according to the solution), the interaction between the particles and the porous medium (mechanical retention (filtration), adsorption (DLVO theory or sorption reaction)) and of course, the hydrodynamic properties of the porous medium (porosity, permeability, dispersion · · · ).

It is well known that, exchanging the salt water present in a sand core with a fresh water, reduces the permeability of the core owing to clay particles migration (Van Olphen, 1977). This problem is of interest for petroleum engineers and agricultural researchers. They usually focus their studies on the evolution of the permeability of the sand core until plugging occurs. Few quantitative studies exist on the clay migration (Khilar and Fogler, 1987). These works show that the behaviour of a sand core is dependant on the type of clay, and on the nature of the salt. They show that release of particles occurs below a critical salt concentration (CSC) (Khilar and Fogler, 1984).

Here, rather than decreasing salinity to reach fresh water and clogging the porous medium by a release of a large amount of particles, the salinity decrease is stopped just below the CSC. This decrease must be large enough to generate particles, but avoids the plugging of the column. One of the preliminary works has been to perform well controlled and reproducible experiments. It is necessary to develop a phenomenological modelling and to test numerical code predictions. The modelling approach is to first establish, from experiments, a relationship between the salinity in the porous medium and the amount of particles released then, to develop a transport model taking into account the observed behaviours (released amount, kinetic effects) and finally, to compare the simulated and experimental results and to test the limits of the model (flow rate, pH, percent of clay, column length).

This work is part of a study (Fauré, 1994) on the modelling and experimental investigation of the coupled transport of radionuclides and particles. A general article was presented earlier (Fauré et al., 1994), in which a particle transport model is used to simulate the coupled transport of caesium and particles in a salinity gradient. In the present contribution, we want to focus the methodology and the experimental results which have led to this model of particle transport.

## 2. Experimental set-up and method

We use a standard liquid chromatographic system. Two pumps, one fed by distilled water and the other by a NaCl solution, monitored by means of a gradient performer, allow us to feed the column at constant flow rate (usually 1 ml min$^{-1}$) with a linearly decreasing NaCl concentration with time. At the outlet of the column, we monitor the variations of NaCl concentration with a conductimetric cell. A UV (280 nm) continuous detector measures the particle concentration in the effluent. On a large range of particle concentrations, the response of UV detector is linear. The porous medium consists in a

mixture of 5% weight Foca3 bentonite clay and sand (99% $SiO_2$). A 16 mm diameter column is packed with 8 g of the mixture. The column is purged of air with $CO_2$ and then saturated with an upward flow of sodium chloride (0.5 M) at pH = 6.5.

## 2.1. Reproducible experiments

The NaCl concentration of the feed solution of the column is decreased from the initial NaCl concentration, $C_i$, to the final, $C_f$, then increased to $C_i$. This cycle is repeated several times to observe the influence of these signals on the particle breakthrough curves. Progressively, the amount of particles eluted during a cycle decreases to zero, though there is still clay in the column. For a given decrease of concentration from $C_i$ to $C_f$, the amount of particles capable of being released from the column is limited. The history of the column cannot be ignored and we will refer to reproducible experiments only for a fresh column. Preliminary results showed that step changes in the NaCl concentration at the inlet of the column led to large perturbations (inducing possible clogging) in the column and non reproducible experiments from one fresh column to another. In a typical experiment, the NaCl concentration of the feed solution is decreased from $C_i$ to $C_f$ within a volume, $V_g$, of solution, then the concentration of the feed solution is maintained constant at $C_f$ as long as some particles leave the column. This typical experiment is reproducible from one fresh column to another with identical physical characteristics: length ($\approx 3$ cm), composition (5% clay) of porous medium and flow rate (1 ml $min^{-1}$). Important parameters are: $C_i$, $C_f$ and $V_g$. We limit the study with 0.015 M $< C_f <$ 0.5 M to avoid plugging of the porous medium. In this case, the amount of particles leaving the column is negligible compared with the amount of clay staying in it. We shall now vary them to characterise some aspects of the particle transport.

## 3. Experimental results and interpretation

### 3.1. Typical experiment

At the beginning of the experiment, the salinity of the feed solution decreases and no particles are detected in the effluent. Then, the particles begin to leave the column and the concentration of particles in the effluent increases up to a maximum value and finally decreases. Let us focus on three points: the shape of the particle breakthrough curve, the time when particles begin to leave the column and the total amount of particles released.

### 3.2. Shape of the particle breakthrough curve

The amount of particles available for migration, during the decrease of the NaCl concentration of the feed solution, is clearly limited. However, even when the NaCl concentration decrease stops (the column is then fed with a solution at constant NaCl
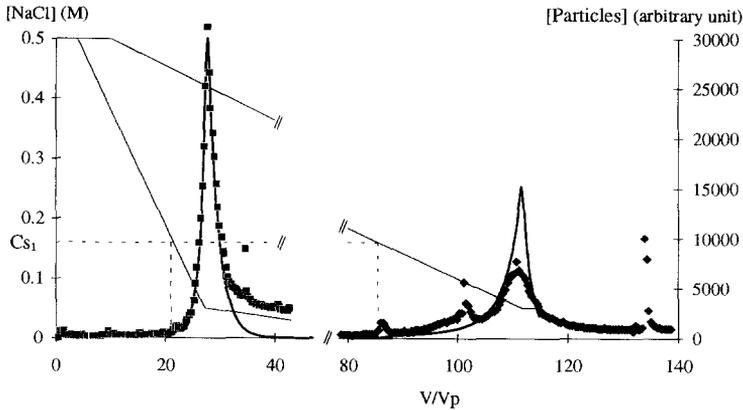
Fig. 1. Comparison of particle release between simulation (————) and typical experiments with small and large $V_g$ values. The NaCl concentration (————) of the feed solution is decreased from $C_i = 0.5$ M to $C_f = 0.05$ M with $V_g = 23.3$ $V_p$ (■) or with $V_g = 107$ $V_p$ (◆).

concentration), particles continue to leave the column. The system is not at equilibrium and we deduce that some kinetic limitations occur in the particle transport.

### 3.3. Measurement of the particle output threshold

We look for a NaCl CSC below which the particles begin to release. Several typical experiments are performed with the NaCl concentration decreasing from 0.5 M to 0.05 M and from 0.05 M to 0.015 M, and the effect of the parameter $V_g$ is studied. Finally, for each domain of NaCl concentration variation, a threshold concentration is obtained,
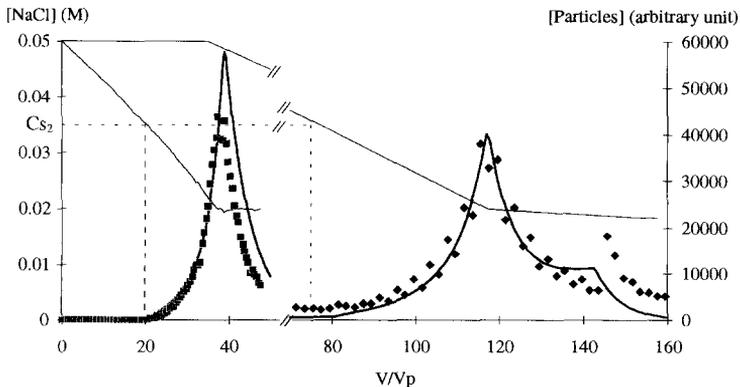


Fig. 2. Comparison of particle release between simulation (————) and typical experiments with small and large $V_g$ values. The NaCl concentration (————) of the feed solution is decreased from $C_i = 0.05$ M to $C_f = 0.02$ M and $V_g = 37.5$ $V_p$ (■). In the other experiment, the NaCl concentration of the feed solution is decreased from $C_i = 0.05$ M to $C_f = 0.02$ M with $V_g = 80$ $V_p$ then NaCl is decreased to 0.01 M with $V_g = 254$ $V_p$ (◆).

independently of $V_g$: $C_{s1} = 0.16$ M and $C_{s2} = 0.036$ (Figs. 1 and 2). For a naturally consolidated sandstone (8% clay content, mainly kaolinite), Khilar and Fogler (1984) have determined a CSC equal to 0.07 M NaCl. This value is in the same order of magnitude as $C_{s1}$ and $C_{s2}$. The difference can be attributed to the fact that particle release is dependant on clay type, the composition of sand and the structure of porous medium.

## 3.4. Amount of particles released

Two columns are fed with a NaCl solution, the concentration of which is decreased from $C_i$ to $C_{f1}$. Then, we continue to decrease the concentration to $C_{f2}$ in the first column, whereas we feed the second with a constant NaCl concentration solution. The breakthrough particle curves are identical as long as the NaCl concentrations of the feed solution are the same (Fig. 3). The results of several typical experiments from $C_i(C_i = 0.5$ M or 0.05 M) to $C_f$ (0.05 M or 0.015 M) with different $V_g$ values are considered. The plot of the amount of particles that leave the column during an experiment, $Q_P$, shows that $Q_P$ is only dependent on $C_i$ and $C_f$, but not on $V_g$. However, $V_g$ has an effect on the concentration of the particles in the effluent of the column. Keeping $C_i$ and $C_f$ constant, the larger the required volume $V_g$ to elute the same amount of particles, the lower the concentration of particles in the effluent.

## 3.5. Verification of the existence of several CSC

$C_{s1}$ and $C_{s2}$ have been determined from distinct experiments, decreasing NaCl concentration from 0.5 M to 0.05 M, keeping the NaCl concentration constant as long as
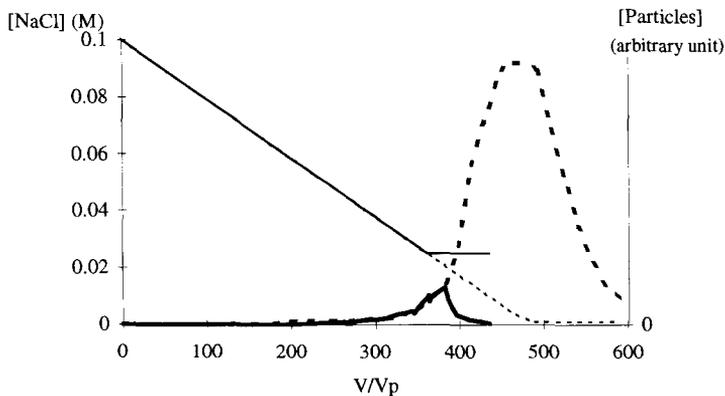


Fig. 3. Effect of the final concentration, $C_f$, on the amount of particles leaving the column. The NaCl concentration of the feed solution is decreased to 0.001 M in column 1 ($\cdots$) while in column 2, the NaCl concentration is decreased from 0.1 M to 0.025 M (————). As long as the NaCl concentration is the same, the particle breakthrough curves are identical (column 1 — — — —, column 2 ————).
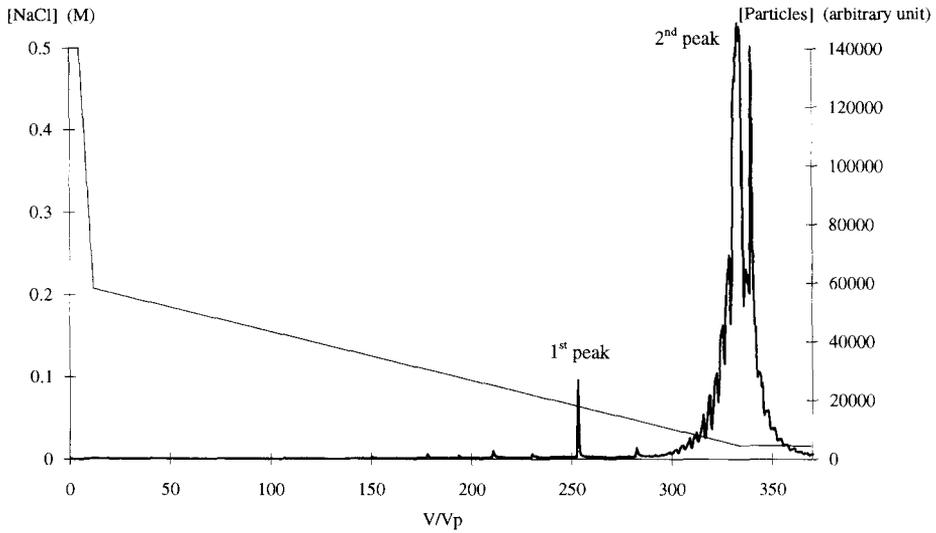
Fig. 4. Particle elution profile (————) from a column where the NaCl concentration of the feed solution (————) is decreased from 0.2 M to 0.015 M with $V_g = 336\ V_p$.

particles are eluted for the column, and decreasing it again from 0.05 to 0.015 M. To test if $C_{s2}$ is not a kinetic effect owing to the plateau at 0.05 M, a column is fed with a NaCl solution decreasing from 0.2 M to 0.015 M without the plateau at 0.05 M (Fig. 4). We detect, as previously assumed, two peaks of particles. The particle release begins when the concentration reaches 0.16 M, then stops and resumes when the concentration drops below 0.036 M. This confirms the previous observations: two domains of NaCl concentration occur in which some particles are able to migrate in the column. The upper limits of these domains are the threshold concentrations. The lower limits, $C_{1s1}$ and $C_{1s2}$, below which the particles are no longer released into solution, are 0.05 M and 0.019 M respectively. These are determined from experiments with large $V_g$ volumes to limit the kinetic effects. In each of these, it has been checked that $Q_P$ is the same for experiments with $C_f$ equal to $C_{1s2}$ or slightly lower than $C_{1s}$. This experiment, revealing

Table 1
Empirical function $P_d([NaCl])$: amount of particles available to migrate as a function of NaCl concentration. $P_d$ is in (D.O. unity)/(Kg of porous medium). This function is plotted with experimental points in Fauré et al. (1994)

| [NaCl] domain | Equation of $P_d([NaCl])$ |
|---|---|
| [NaCl] > 0.16 M | $P_d([NaCl]) = 0$ |
| 0.16 M $\geq$ [NaCl] $\geq$ 0.05 M | $P_d([NaCl]) = X_1\ (\exp([NaCl]^n) - \exp(0.16^n))$ |
| 0.05 M $\geq$ [NaCl] $\geq$ 0.035 M | $P_d([NaCl]) = X_1\ (\exp(0.05^n) - \exp(0.16^n))$ |
| 0.035 M $\geq$ [NaCl] $\geq$ 0.019 M | $P_d([NaCl]) = X_2\ (\exp([NaCl]^n) - \exp(0.035^n)) + P_d(0.05)$ |
| 0.019 M $\geq$ [NaCl] | $P_d([NaCl]) = X_2\ (\exp(0.019^n) - \exp(0.035^n)) + P_d(0.05)$ |

$n = 0.6,\ X_1 = 75500,\ X_2 = 7970$

the two domains for particle release, is very tricky. Indeed, we must choose a convenient intermediate value of the volume $V_g$. If $V_g$ is too small, the two particle peaks of the two domains interfere. On the contrary, if $V_g$ is too large, the concentration of particles in the effluent will be below the detection limit of the UV cell. This means that some sub-domains of particle output, that we have not detected, could exist. The distinct threshold concentrations might be related to the size or to the nature of the clay.

In order to model the particle release, we need an equation to account for the amount of particles available for migration. An empirical function, $P_d([NaCl])$, is fitted on previous experimental plots, $Q_P$ versus NaCl concentration. $P_d(C_f) - P_d(C_i)$ is the amount of particles that leave the column when decreasing the NaCl concentration of the feed solution from $C_i$ to $C_f$. This relationship is given in Table 1.

### 3.6. Hydrodynamics

We have shown earlier (Fauré et al., 1994) that initially, the water flow is near that of a piston flow. However, immediately prior to the detection of the first eluted particles from the output of the column, preferential pathways and dead zones are created; their size increases as the NaCl concentration is decreased. This is attributed to particle transport and reordering inside the column.

## 4. Modelling the transport of particles in a salinity gradient

### 4.1. Hydrodynamics modelling

To model the hydrodynamics of the porous medium in the different situations, we used the classical description of one-dimensional convective–dispersive solute transport with dead end pore volume (Coats and Smith, 1964). A mobile water zone, in which the transport is controlled by convection and dispersion, is assumed to diffuse through the immobile water zone located in dead end pore volume. This description is applied to the discrete model of mixing cells in series (Villermaux, 1974). The column, of porous volume $V_P$, is represented by $J$ identical cells in series. Equivalence between this discrete approach and the classical continuous model has been shown under these conditions (Sardin et al., 1991), the relationship between $J$ and the axial dispersion

Table 2
Hydrodynamic characteristics of the porous medium

|  | Domain 1 [NaCl] 0.16 M to 0.05 M | Domain 2 [NaCl] 0.035 M to 0.019 M |
|---|---|---|
| $J$ | 51 | 15 |
| $\theta_{im}$ | 0.125 | 0.285 |
| $t_M$ | $0.05\,\tau$ | $0.09\,\tau$ |
| $t_{Mp}$ | $12\,\tau$ | $20\,\tau$ |

Table 3
Mass transfer equations of NaCl solute and of particles used in the model

| | |
|---|---|
| Tracer in mobile fluid zone | $QC_{m,k-1} = QC_{m,k} + \theta_m \dfrac{V_p}{J} \dfrac{dC_{m,k}}{dt} + \Phi_{e,k}$ |
| Tracer in immobile fluid zone | $\theta_{im} \dfrac{V_p}{J} \dfrac{dC_{im,k}}{dt} = \Phi_{e,k}$ |
| Tracer flux between zone | $\Phi_{e,k} = \dfrac{\theta_m V_p}{J} \dfrac{C_{m,k} - C_{im,k}}{t_M}$ |
| Particles in mobile fluid zone | $QP_{m,k-1} = QP_{m,k} + \theta_m \dfrac{V_p}{J} \dfrac{dP_{m,k}}{dt} + \Phi_{e,P,k}$ |
| Particles in immobile fluid zone | $\theta_{im} \dfrac{V_p}{J} \dfrac{dP_{im,k}}{dt} + \Phi_{s,P,k} = \Phi_{e,P,k}$ |
| Particle flux between zones | $\Phi_{e,P,k} = \dfrac{\theta_{im} V_p}{J} \dfrac{P_{m,k} - P_{im,k}}{t_{M,P}}$ |
| Particle flux source | $\Phi_{s,P,k} = -\dfrac{m}{J} \dfrac{dP_d}{d[NaCl]} \dfrac{d[NaCl]}{dt}$ |

coefficient, $D$, being $Pe = uL/D = 2(J - 1)$ ($Pe$ = Peclet number, $L$ = length of column, $u$ = interstitial velocity). Each cell contains a mobile fluid zone of volume $\theta_M V_p/J$ and an immobile fluid zone of volume $\theta_{im} V_p/J$. The mass balance equations in the $k$th cell, for a tracer, are listed in Table 2. $C_{m,k}$ and $C_{im,k}$ are the concentrations of the tracer in the mobile and the immobile fluid zones. $\Phi_{e,k}$ is the flux between the mobile and the immobile fluid zones. As a first approximation, this flux is assumed to be controlled by a linear first order kinetic law (Table 3, 3rd Eq.) and $t_M$ is a characteristic mass transfer time. According to the nature of the mass transfer resistance (film model, internal diffusion in the aggregates $\cdots$), the expression of $t_M$ can be calculated from the main characteristics of the grain packing (Sardin et al., 1991).

## 4.2. Particle transport modelling

We apply the description of the tracer transport to the particles by adding a term for the generation of particles, $P_d([NaCl])$. As for the solute, a characteristic mass transfer time for the particles, $t_{Mp}$, is introduced. No other term of kinetic limitation is added. We first want to verify if $t_{Mp}$ accurately takes into account all the kinetic limitations found earlier in the experimental study of particle transport. We assume that the immobile fluid zone is in contact with the porous medium that produces particles according to the NaCl concentration. The mass balance equations for the particles, the flux between the mobile and the immobile fluid zones and the source term of particles from the porous medium to the immobile fluid zone are in Table 3 (4th–7th Eqs.).

## 4.3. Determination of the parameters and model verifications

Table 3 (1st–3rd Eqs.) describe the NaCl transport, considered as a tracer, (4th–7th Eqs.) describe the transport of particles. The parameters ($V_p$, $J$, $\theta_{im}$, $t_M$) are determined

from experimental measurements of residence time distribution (Fauré, 1994). The hydrodynamic characteristics change continuously as NaCl concentration is decreased, but we only know these values for a few NaCl concentrations. So, during simulation, we keep these parameters constant. For each domain of destabilisation, the hydrodynamic characteristics of the porous medium used are in Table 2. To determine $t_{Mp}$ (Table 2), we fit it with two breakthrough curves in each domain. The model is then tested on a set of a dozen of experiments of clay destabilisation, four of which are shown here (Figs. 1 and 2). These figures are representative of the two NaCl concentration domains studied and of the values of $V_g$ used. For typical experiments from 0.5 M to 0.05 M, or from 0.05 M to 0.015 M, the agreement between experiments and simulation is good when $V_g$ is not too large ($< 150 \, V_p$). When $V_g$ increases, the simulation anticipates the particle breakthrough curve. These differences are possibly owing to the approximation on the invariance of the porous medium hydrodynamic characteristics. During the simulation, we keep $\theta_{im}$, $t_M$, $t_{Mp}$ constant, whereas actually, they increase. The increase of $\theta_{im}$ and $t_{Mp}$ involves a retardation of the particle migration. Another reason could be the insufficient knowledge of the $P_d([NaCl])$ function.

## 5. Meaning of the characteristic parameters for the particles: $P_d$, $t_{Mp}$

To simulate the transport of particles in a porous medium, a filtration term is often introduced. We do not use this approach. This term of filtration is implicitly enclosed in the $P_d$ function. We have made typical experiments with a long column ($V_p = 11.8$ ml instead of 3 ml usually). The amount of particles (referred to the mass of porous medium) that leave this column is less than that from a column of smaller length. It would be interesting to get a flocculation curve of clay particles, that is, for a given time, the rate of flocculation of the clay as a function of the NaCl concentration.

$t_{Mp}$ is the characteristic mass transfer time. Additional experiments with different flow rates and a constant $V_g$, show that the particle concentration in the effluent increases as flow rate decreases (the total amount of particles that leave the column being constant). The flux between the mobile and the immobile water zones in a mixing cell is then probably in diffusive mode.

## 6. Conclusion

The transport of clay particles, caused by a decrease of the salinity of the feed solution, in an unconsolidated sand core is modelled by coupling a non-equilibrium transport model with a term of particles generation, $P_d([NaCl])$. The hydrodynamic properties of the porous medium vary as soon as the particles are in movement inside the column. The kinetic limits of particle transport are simply taken into account by the hydrodynamic model. $P_d([NaCl])$ is linked to the properties of flocculation of the clay

particles and of the filtration in the porous medium. This model is tested on a set of a dozen of experiments.

| | |
|---|---|
| $C_i$, $C_f$ | Initial and final NaCl concentration of the fed solution (M) |
| $C_s$ | Threshold concentration (M) |
| $C_{m,k}$, $C_{im,k}$ | NaCl concentration in the mobile and immobile fluid zone (M) |
| $J$ | Number of cells |
| $P_d([NaCl])$ | Particles available to migration as a function of NaCl concentration ((number of particles)/(mass of clay), arbitrary unit) |
| $Q$ | Row rate $(1\ s^{-1})$ |
| $Q_p$ | Amount of particles that leaves the column (number of particles, arbitrary unit) |
| $V_g$ | Volume of the gradient, volume of eluent to decrese the concentration from $C_i$ to $C_f$(1) |
| $V_p$ | Porous volume (1) |
| $\Phi_{e,k}$, $\Phi_{e,P,k}$ | Solute and particle flux between the mobile and immobile fluid zone (M $s^{-1}$) |
| $\Phi_{s,P,k}$ | Particle fux between immobile fluid zone and the porous medium in the $k$th cell ((number of particles) $1^{-1}\ s^{-1}$, arbitrary unit) |
| $\theta_m$, $\theta_{im}$ | Fraction of mobile and immobile fluid zone, $\theta_m + \theta_{im} = 1$ |
| $\tau$ | Mean residence time in porous medium, $\tau = V_p/Q$ (s) |

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