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M. ZEKİ ÇAMUR

FÜSUN CANPOLAT

HASAN YAZICIGİL

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Effects of Compaction Parameters on Heavy Metal Mass Transport in Ankara Clay Liner: Experimental Approach

M. ZEKİ ÇAMUR, FÜSUN CANPOLAT & HASAN YAZICIGİL

Middle East Technical University, Department of Geological Engineering, TR-06531 Ankara, Turkey
(E-mail: mzeki@metu.edu.tr)

Abstract: Mass transports of Cl, Cd, Cr, Cu, Fe, K, Mn, Ni, Pb and Zn ions through the Ankara clay liner prepared under two different compaction were studied experimentally on the bases of sorption and hydrodynamic dispersion processes. According to geotechnical tests, the Ankara clay material is highly plastic, inorganic clay having a liquid limit of 73, a plastic limit of 29 and a plasticity index of 44. Optimum compaction of the material requires 32% water content and 1.413 g/cm³ dry density. Two materials, prepared using half of the optimum water content but optimum energy conditions (I) and optimum water content but half of the optimum energy conditions (II), have the effective porosities of 0.35 and 0.40, respectively. Hydraulic conductivities of these materials (I and II) were 3.1–4.38x10⁻⁸ cm/s and 916–1057x10⁻⁸ cm/s, respectively. Dispersivities determined through column experiments were 0.88 cm and 1.78 cm in materials I and II, respectively. Mass transports of the ions were minimum in the material that were prepared using optimum conditions (diffusion dominant mass transfer) but maximum in the material that were prepared using optimum water content but half of the optimum energy conditions (advection dominant over diffusion type of mass transfer). The Ankara clay material is suitable for a liner design in waste disposal sites as long as it is compacted under optimum energy conditions.

Key Words: diffusion, dispersivity, hydrodynamic dispersion, clay liner, Ankara clay

Ankara Kili Taban Örtüsünde Sıkıştırma Parametrelerinin Ağır Metal Taşınımı Üzerindeki Etkileri: Deneysel Yaklaşım

Özet: Katı-sıvı iyon etkileşimi (sorbsiyon) ve hidrodinamik yayılım (dispersiyon) mekanizmaları kapsamında Cl, Cd, Cr, Cu, Fe, K, Mn, Ni, Pb and Zn iyonlarının Ankara kili taban malzemesindeki taşınımı, farklı sıkıştırma koşullarında deneysel olarak çalışılmıştır. Jeoteknik testlere göre Ankara kili, sıvı limiti 73, plastik limiti 29 ve plastisite indeksi 44 olan yüksek plastik özellikli bir malzemedir. Malzemenin optimum koşullarda sıkıştırılabilmesi için %32 su içeriği ve 1.413 g/cm³ kuru yoğunluk gerekmektedir. Biri optimum su içeriğinin yarısı fakat optimum enerji koşullarında (I) diğeri de optimum su içeriği fakat optimum enerjinin yarısı koşullarında (II) olmak üzere, etkin gözenekliliği sırasıyla 0.35 ve 0.40 olan iki malzeme ile kolon deneyleri gerçekleştirilmiştir. Bu malzemelerin (I ve II) hidrolik iletkenliği aralığı sırasıyla 3.1–4.38x10⁻⁸ cm/s ve 916–1057x10⁻⁸ cm/s'dir. Malzeme I ve II de kolon deneyleri ile belirlenen yayılımlar sırasıyla 0.88 cm ve 1.78 cm dir. İyonların kütleli taşınımı optimum koşullarda hazırlanan malzemede en az ve optimum su içeriği fakat optimum enerjinin yarısı koşulları ile hazırlanan malzemede en fazladır. Ankara kili malzemesi optimum enerji koşullarında hazırlanması durumunda atık depolama alanı taban örtüsü olarak kullanılabilir.

Anahtar Sözcükler: kirlenici taşınımı, yayılma (dispersivite ve difüzyon), kil taban malzemesi, Ankara kili

Introduction

Natural clay liners have been extensively used to prevent leakage of effluents in disposal areas (for summary see Christensen *et al.* 1994). The suitability of a clay liner has been conventionally evaluated on the basis of hydraulic conductivity to control the advective mass transport. However, under optimum compaction conditions dominant mass transfer in liners is by diffusion (Quigley *et al.* 1987; Rowe 1994). Departure from the optimum conditions increases hydraulic conductivity and causes

advective mass transfer to become the dominant mass transfer process. Two major compaction parameters, energy and water content of the liner material, must be well controlled for the proper applications.

The natural liner applications indicate that the desired hydraulic conductivity conditions are generally not achieved in practical field applications (Farquhar 1994). In addition, in cases where the desired conditions are achieved, the liners are subject to hydraulic conductivity changes at the disposal operation stage (Peirce *et al.*

1987; Quigley *et al.* 1988; Broderick & Daniel 1990; Farquhar 1994). As a result, hydraulic properties of liners generally deviate from optimum conditions in different degrees under field applications. Hence, the mass transfer of contaminants in liners is subject to a range of changes from diffusion-controlled to advection-controlled cases.

The mass transfer evaluation in these cases requires transport parameters data for ions covering the range of interest. However, some available data were collected using intact soils (which were not subject to different compaction conditions). In addition, most of the available data related to mass transport in clay liners are based on experiments that were performed using solutions with a limited number of species (binary or ternary systems) (e.g., Carroll & Starkey 1971; Gordon & Forrest 1981; Quigley *et al.* 1987; Peirce *et al.* 1987; Rowe *et al.* 1988; Barone *et al.* 1989). Although multi-component system related diffusion data applicable to the transport processes in liners are presented by Çamur & Yazıcıgil (2005), dispersion data is not available. Çamur & Yazıcıgil (2005) have experimentally estimated the effective diffusion coefficients for heavy metals (Cd, Cr, Cu, Fe, K, Mn, Ni, Pb, and Zn) in a multi-component system using Ankara clay material compacted under optimum conditions.

The purpose of this study is to determine the impacts of diffusive to advective multi-component mass transports on the Ankara clay liner via estimation of hydrodynamic dispersion data experimentally. The liner of interest was compacted under two different conditions; optimum energy but half of the optimum water content and optimum water content but half of the optimum energy. These conditions correspond to advection-diffusion-controlled type of mass transfers and advection dominant over diffusion type of mass transfers, respectively. For the optimum conditions (diffusion dominant mass transfer), data of Çamur & Yazıcıgil (2005) was utilized. These conditions will demonstrate the increased mass transport resulting from the improper handling of a liner design in the field for multi-component mass systems.

Material and Geotechnical Tests

The suitability of the Ankara clay as a liner material was investigated. Detailed geotechnical characteristics of the

Ankara clay were reported in Çamur & Yazıcıgil (2005) and Met *et al.* (2005). Reddish-brown mudstone layers in the Upper Pliocene sediments around Ankara plain, Turkey is called 'Ankara clay' (Birand 1963; Ordemir *et al.* 1965). XRD analyses of collected samples indicate that the Ankara clay is composed of about 37% calcite, 32% quartz, 12% smectite, 10.5% illite, 6% feldspar, 2.5% chlorite and kaolinite minerals. Distribution of clay, silt, sand, and gravel size particles is 50%, 34%, 11%, and 5%, respectively. According to geotechnical tests (ASTM D4318), the material is a highly plastic, inorganic clay having a liquid limit of 73, a plastic limit of 29 and a plasticity index of 44. The compaction test results are shown in Figure 1. The compaction tests based on ASTM D698 indicate that the optimum compaction of the material can be achieved under 32% water content and 1.413 g/cm³ dry density conditions. The compacted material under optimum conditions has bulk porosity of 0.48 and effective porosity (volumetric water content) of 0.32. Hydraulic conductivity of the compacted material is 1.3x10⁻⁹ cm/s as measured using triaxial permeability test with two back pressure systems.

On the bases of these compaction tests, optimum energy but half of the optimum water content compaction condition corresponds to 1.38 g/cm³ dry density and 16% water content. As measured from geotechnical tests (ASTM D698, ASTM D854 and ASTM D2434), the material compacted under these conditions has bulk porosity of 0.49, effective porosity (volumetric water content) of 0.35, and hydraulic conductivity of 3.1x10⁻⁸ cm/s. Optimum water content but half of the optimum energy compaction condition corresponds to 1.21 g/cm³ dry density and 32% water content. The material compacted under these conditions has bulk porosity of 0.55, effective porosity (volumetric water content) of 0.40, and hydraulic conductivity of 9.2x10⁻⁶ cm/s.

Column Experiments

Two column experiments were carried out. A schematic figure of the column apparatus made of plexiglas used in the experiments is given in Çamur & Yazıcıgil (2005). In Column I, the Ankara clay was compacted under optimum energy but half of the optimum water content conditions. In Column II, the clay was compacted under optimum water content but half of the optimum energy conditions.

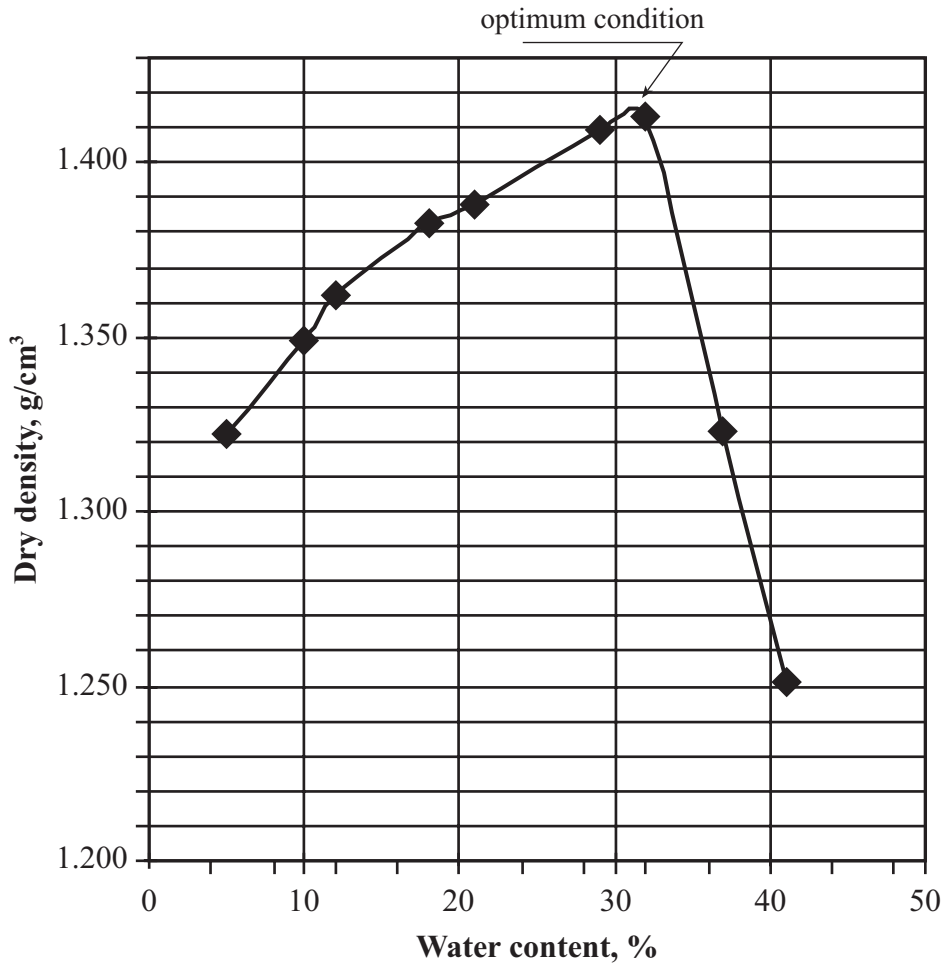


Figure 1. Compaction curve for the Ankara clay used in the experiments.

In each column, the clay was compacted using standard *Proctor method* into the lower part of the column after a polyethylene plate (porosity= 0.38) of 1 mm thickness was placed at the bottom of the sample. The porous polyethylene plate at the bottom of the sample was later used to set the bottom boundary condition. The lower part of the column was placed onto the plexiglas base plate with a 5 mm hole in the middle. The boundary between the base plate and the lower part of the column was sealed using commercial transparent silicon from outside. A plastic tube was mounted to the base plate through which the effluent (if present) could be drained into the collection bottle. Once the sample was in place, an effective pressure was applied onto the sample with a perforated plexiglas disk having 0.38 porosity and 10 mm thickness. The boundary between the plexiglas disk and the lower part of the column was also sealed. The

porous plexiglas disk at the top of the compacted sample was used to ensure that the permeating solution entered the sample surface evenly. Then, the upper part of the column (solution reservoir) was placed onto the lower part and the boundary in between was sealed from outside. The upper part of the column was filled by pre-prepared solution representing waste leachate and 22 cm solution height was obtained. The solution including average waste leachate concentrations (mg/l) of Cl (4500), Cd (0.234), Cr (1.750), Cu (0.180), Fe (6.97), K (618.2), Mg (187.7), Mn (16.77), Ni (0.580), Pb (1.28), and Zn (21.5) was prepared in the laboratory using pure standards and chemicals. pH of the solution was adjusted to about 6.5 using NaOH. The waste leachate chemical composition is approximately based on the average concentrations of leachates reported from the waste disposal sites in the European section of

İstanbul city (Mimko 1995). Finally, the upper part was closed using a plexiglas lid. The lid had a hole on it to insert either a mixer or a sampling pipette. The solution was mixed periodically with a polyethylene mixer to maintain a relatively uniform concentration distribution throughout the reservoir depth during experiment.

Total fluid flow through the sample into the collection bottle was monitored. During the experiment, a 1 ml solution sample was collected from the reservoir at specified times (see Figure 2) through the sampling port for monitoring Cl concentration. To prevent a drop in height of solution in the reservoir, the volume of distilled water equal to the volume of reservoir solution and effluent (if present) collected was added after each monitoring period. Thus, the height of solution in the reservoir remained relatively constant. The dilution

resulting from the addition of distilled water was taken automatically into consideration by the theory used for the back-analysis of parameters during data evaluations (for detailed information see Rowe *et al.* 1988). During the experiment, the lowest and the highest laboratory temperatures recorded were 23 °C and 29 °C, respectively. Possible evaporation was minimized by the lid on the reservoir. After the termination of the experiment, the compacted Ankara clay sample was sectioned to determine the vertical distribution of the selected chemical constituents in pore water which was extracted from the sections using a pneumatic pore-water squeeze apparatus at a pressure of 25 MPa for 3 hr.

In Column I, the solution started to leak into the collection bottle in the third day. The experiment was carried out for 21.93 day. Based on collected data, the

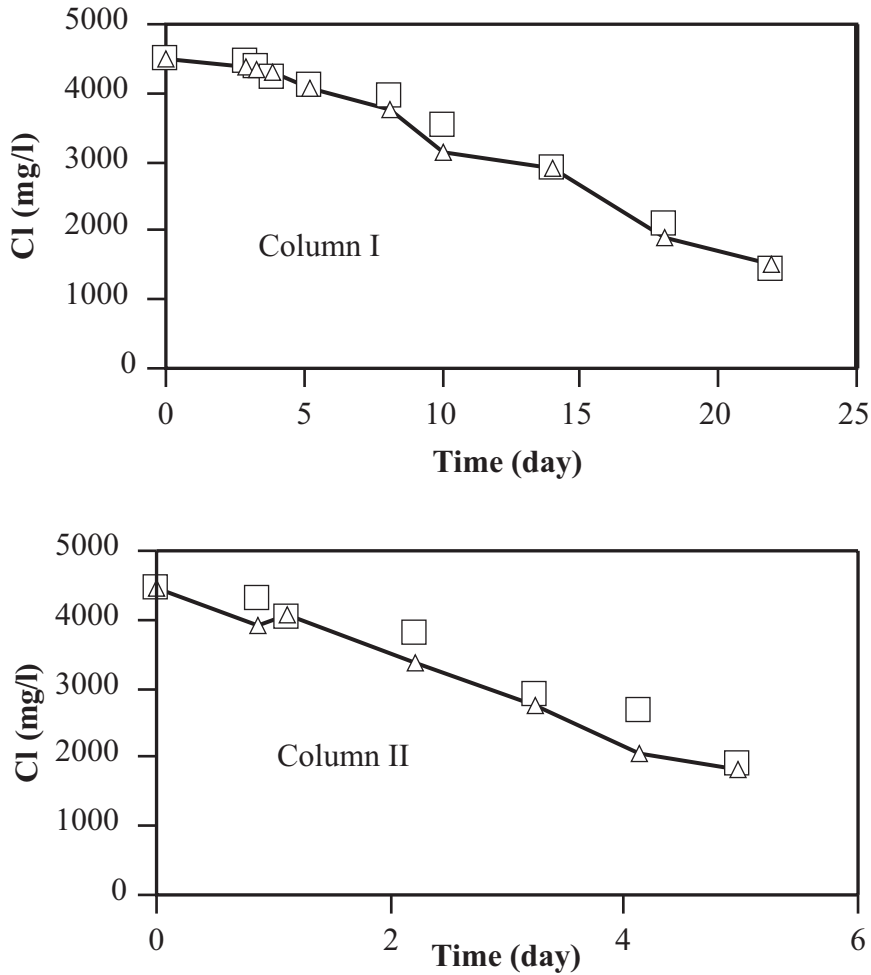


Figure 2. Comparison of theoretical (triangle, line) and experimental (square) Cl concentrations in the reservoir.

seepage velocity, hydraulic conductivity and effective porosity (volumetric water content) were determined as 3.62×10^{-7} cm/s, 4.4×10^{-8} cm/s, and 0.35, respectively. In Column II, the solution started to leak into the collection bottle in the first day. The experiment was carried out for 4.96 day. According to the collected data, the seepage velocity, hydraulic conductivity and effective porosity were 773.4×10^{-7} cm/s, 10.5×10^{-6} cm/s, and 0.40, respectively. Column I hydraulic conductivity result indicates that Column I conditions correspond to both advection and diffusion-controlled type of mass transfers. Column II result suggests that the conditions correspond to advection dominant over diffusion type of mass transfers (Rowe 1994).

Data Analysis

Contaminant transport through liners (soils) can be modelled using advection-dispersion equation which, for one dimensional condition can be expressed as

$$\partial c / \partial t = [1 / (1 + \rho_b K_d / n_e)] D_h (\partial^2 c / \partial z^2) - v_s (\partial c / \partial z)$$

where

c: ion concentration at a given depth z and time t,

ρ_b : dry density of the soil,

K_d : distribution or partitioning coefficient,

n_e : effective porosity,

D_h : hydrodynamic dispersion coefficient,

v_s : seepage velocity ($=v_a/n_e$), and

v_a : advective (Darcy) velocity

If the values of ρ_b , K_d , n_e , v_s and D_h parameters are known, the concentration distribution of any ion at a given time and space can be calculated using above equation for a given porous material and boundary conditions. The equation can also be used to calibrate hydrodynamic dispersion coefficient for an ion if the ion concentration, effective porosity, seepage velocity, dry density and distribution coefficient are known at a given time and space (as in the case of experiments explained in the previous section) (Rowe *et al.* 1988; Domenico & Schwartz 1999). This type of hydrodynamic dispersion coefficient determinations is commonly adopted by other workers (e.g., van Genuchten 1978; Rowe *et al.* 1988; Barone *et al.* 1989).

The mass transport equation given above was solved for different values of hydrodynamic dispersion coefficient (D_h) for Cl ion until the match was obtained between the observed and theoretical concentrations in the source solution as a function of time using the known dry density of the clay, seepage velocity, effective porosity, distribution coefficient, and initial source solution concentration. The mass transfer equation is solved using POLLUTE software of Rowe *et al.* (1994). Finite mass top boundary condition was used in the calculations. The top boundary condition in theory of POLLUTE is flux controlled. Conservation of mass (the mass of ion in the source solution at time, t_i , is equal to the initial mass of the ion minus the mass has been transported into the liner material up to this time, t_i .) can be stated algebraically:

$$C_{top}(t_i) = C_o - (1/H_f) \int_0^{t_i} f_f(t) dt$$

At any given time, t_i , the concentration of the ion of interest in the column solution, $C_{top}(t_i)$, is related to initial concentration of the ion, C_o , in the column (source) solution, the height of solution in the column, H_f , and the mass flux of this ion (f_f) into the liner at time t. The bottom boundary was specified as fixed outflow.

The estimated hydrodynamic dispersion coefficients for Cl are 9.83×10^{-6} cm²/s for Column I material and 146.8×10^{-6} cm²/s for Column II material. Theoretical Cl concentrations in the reservoir calculated using these hydrodynamic dispersion coefficients are compared with the experimentally measured values in Figure 2 where correlation coefficients between experimental and theoretical values are 0.99 and 0.97 for Column I and II, respectively.

By definition the hydrodynamic dispersion coefficient may be treated as the sum of the effective diffusion coefficient and the mechanical dispersion coefficient ($D_h = D_e + D_m$). The mechanical dispersion coefficient, dispersivity and seepage velocity relation ($D_m = \alpha v_s$) result in dispersivities of 0.88 cm and 1.78 cm for Column I and II materials, respectively, based on effective diffusion coefficient value of 9.51×10^{-6} cm²/s measured for Cl by Çamur & Yazıcıgil (2005).

Hydrodynamic dispersion coefficients for Cd, Cr, Cu, Fe, K, Mg, Mn, Ni, Pb, and Zn ions were calculated for two column materials separately using the mechanical dispersion coefficients determined in this study and the

effective diffusion coefficients reported for these ions in the Ankara clay by Çamur & Yazıcıgil (2005) (Table 1). Theoretical pore water concentrations of the ions in the materials calculated using these estimated hydrodynamic dispersion coefficients and sorption isotherms of Çamur & Yazıcıgil (2005) are compared with the experimentally measured values in Figures 3 & 4. It was not possible to obtain concentration for all the ions because the volume of extracted pore fluid amount from each compacted clay section was very little. Those that were measured are shown in the figures. It should be kept in mind that effective diffusion coefficients determined by Çamur & Yazıcıgil (2005) under optimum compaction conditions were assumed to be the same in this paper in the derivation of dispersivity and hydrodynamic dispersion coefficients for all the ions but chloride.

Application

In order to show the effects of different compaction conditions on ion mass transport, the following three cases were examined with computer simulations using experimentally determined mass transfer values. In case 1, the liner was prepared using optimum compaction conditions (diffusion-dominant mass transfer). In case 2, optimum energy but half of the optimum water content was used in the preparation of the liner (both advection and diffusion-controlled mass transfer). In case 3, optimum water content but half of the optimum energy was used for the liner (advection dominant over diffusion

type of mass transfer). In all the cases, it was assumed that the liner thickness was 1 m and compacted on a 1000 m x 1000 m disposal site. The underlying aquifer was assumed to have a 0.38 porosity and a 15 m thickness. The groundwater advective velocity was set to 6.34×10^{-5} cm/s for case 1 and case 2 and was set to 2.12×10^{-3} cm/s for case 3 in the aquifers below the liners during simulations in order to meet continuous flow conditions (any contaminant reaching to the aquifer would be removed by groundwater flow). The simulations were performed for the constant source concentrations given earlier for a 50 year period.

Calculated pore water concentration distributions for some ions in all the cases are shown in Figures 5 & 6. According to the results, minimum and maximum mass transports occurred in case 1 and case 3, respectively. Under optimum compaction conditions, Cd, Cr, and Ni after 10 cm, Cu, Fe, Mn, Pb, and Zn after 20 cm and K after 60 cm would reach to natural background pore water concentrations in the liner at the end of 50 years. The results of case 2 are close to those of case 1 suggesting relatively lower mass transport impact of water content than of energy. On the other hand, if compaction energy improperly applied (when advection dominates), all the initial concentrations present in the leachate could be transported to groundwater. Thus, the application of the proper compaction energy under field conditions becomes more important than the water content in producing an effective clay liner.

Table 1. Calculated hydrodynamic dispersion coefficients based on mechanical dispersion coefficient values of 3.17×10^{-7} cm²/s and 1.40×10^{-4} cm²/s for column I and Column II, respectively.

Ion	Effective Diffusion Coefficient* (cm ² /year) x 10 ⁻⁶	Hydrodynamic Dispersion Coefficient (Column I) (cm ² /year) x 10 ⁻⁶	Hydrodynamic Dispersion Coefficient (Column II) (cm ² /year) x 10 ⁻⁴
Cl	9.51	9.83	1.468
Cd	2.54	2.85	1.398
Cr	2.22	2.54	1.395
Cu	3.14	3.17	1.401
Fe	2.22	2.54	1.395
K	7.93	8.24	1.452
Mn	3.08	3.39	1.403
Ni	1.59	1.90	1.389
Pb	3.17	3.49	1.404
Zn	2.54	2.85	1.398

* Values from Çamur & Yazıcıgil (2005).

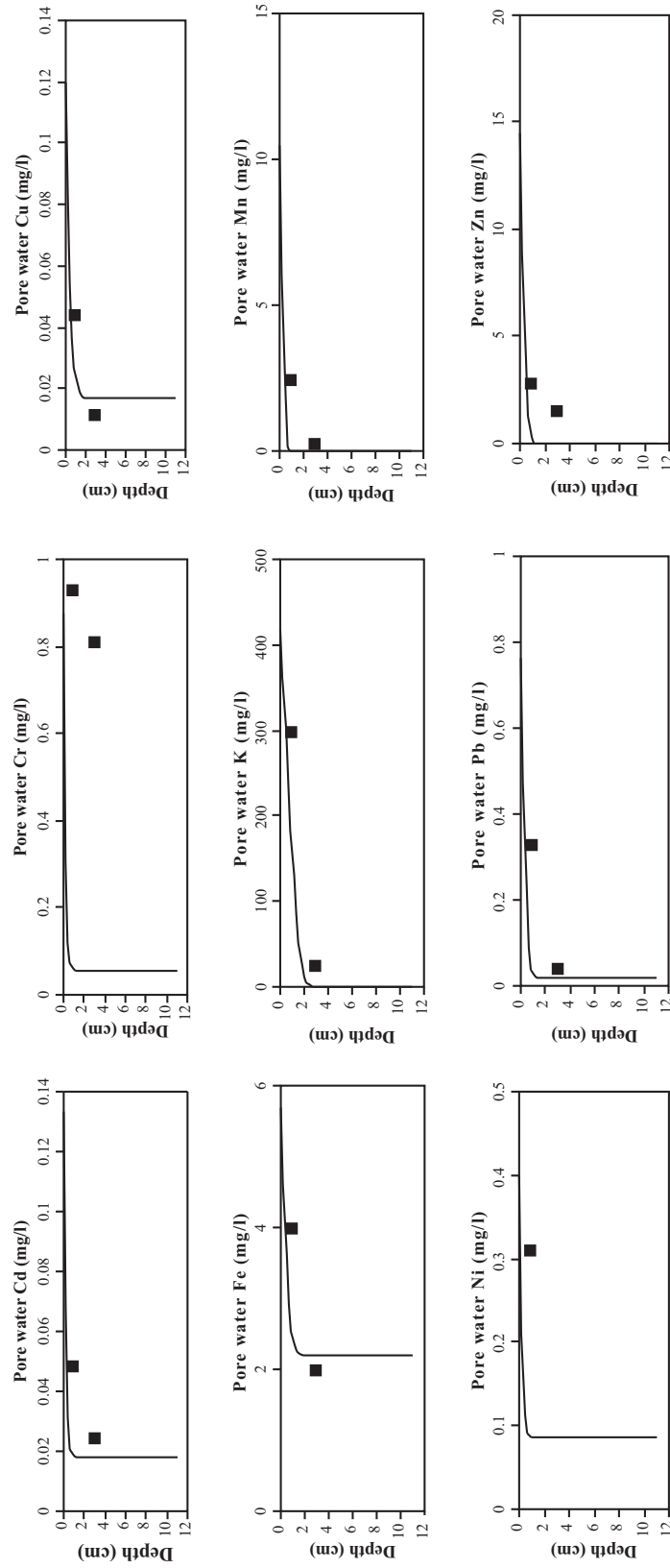


Figure 3. Comparison of theoretical (line) and experimental (square) concentrations in the pore water of Column I after 21.93 day..

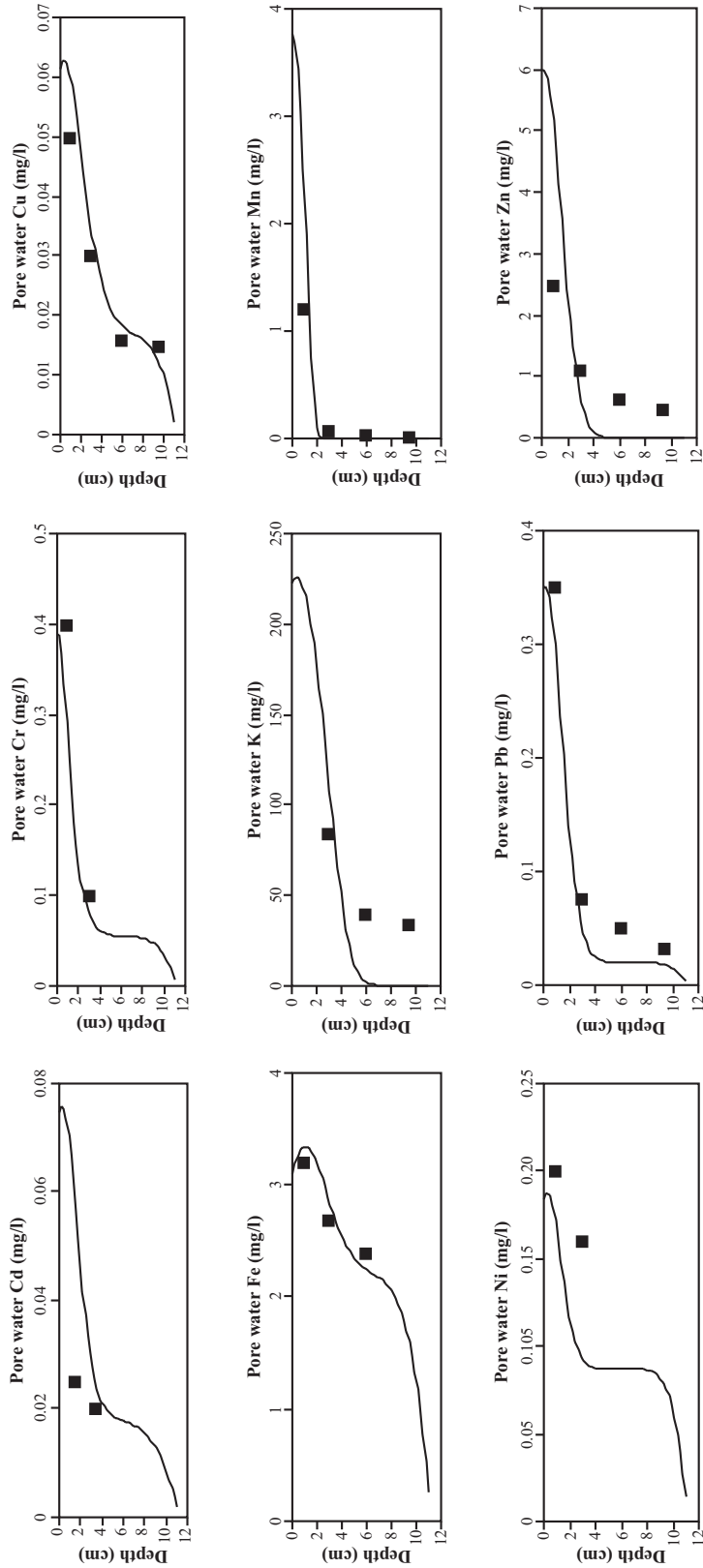


Figure 4. Comparison of theoretical (line) and experimental (square) concentrations in the pore water of Column II after 4.96 day.

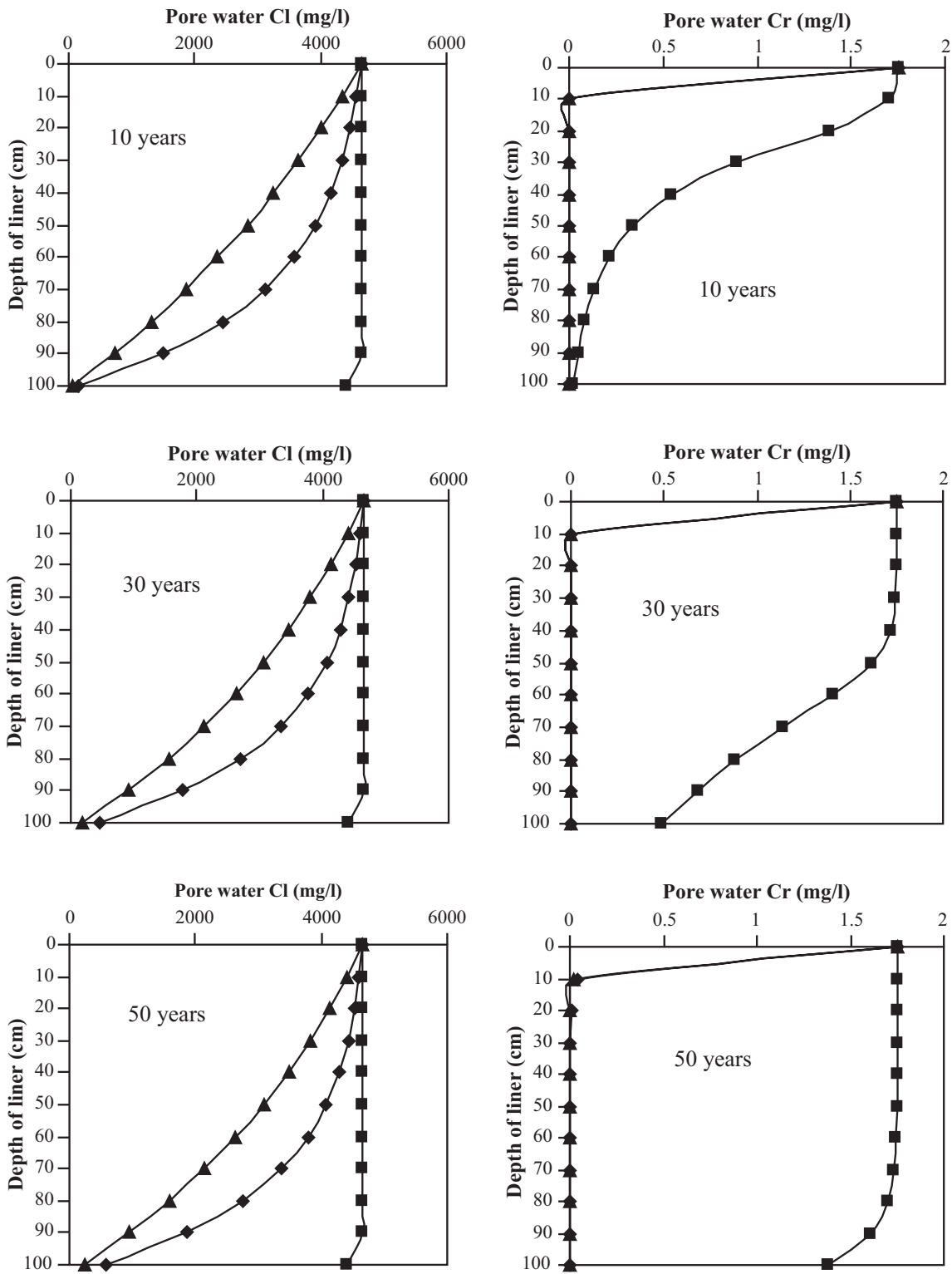


Figure 5. Pore water concentration distributions of Cl and Cr ions after 10, 30, and 50 years for three different cases explained in the text. Triangle– case 1, diamond– case 2, square– case 3.

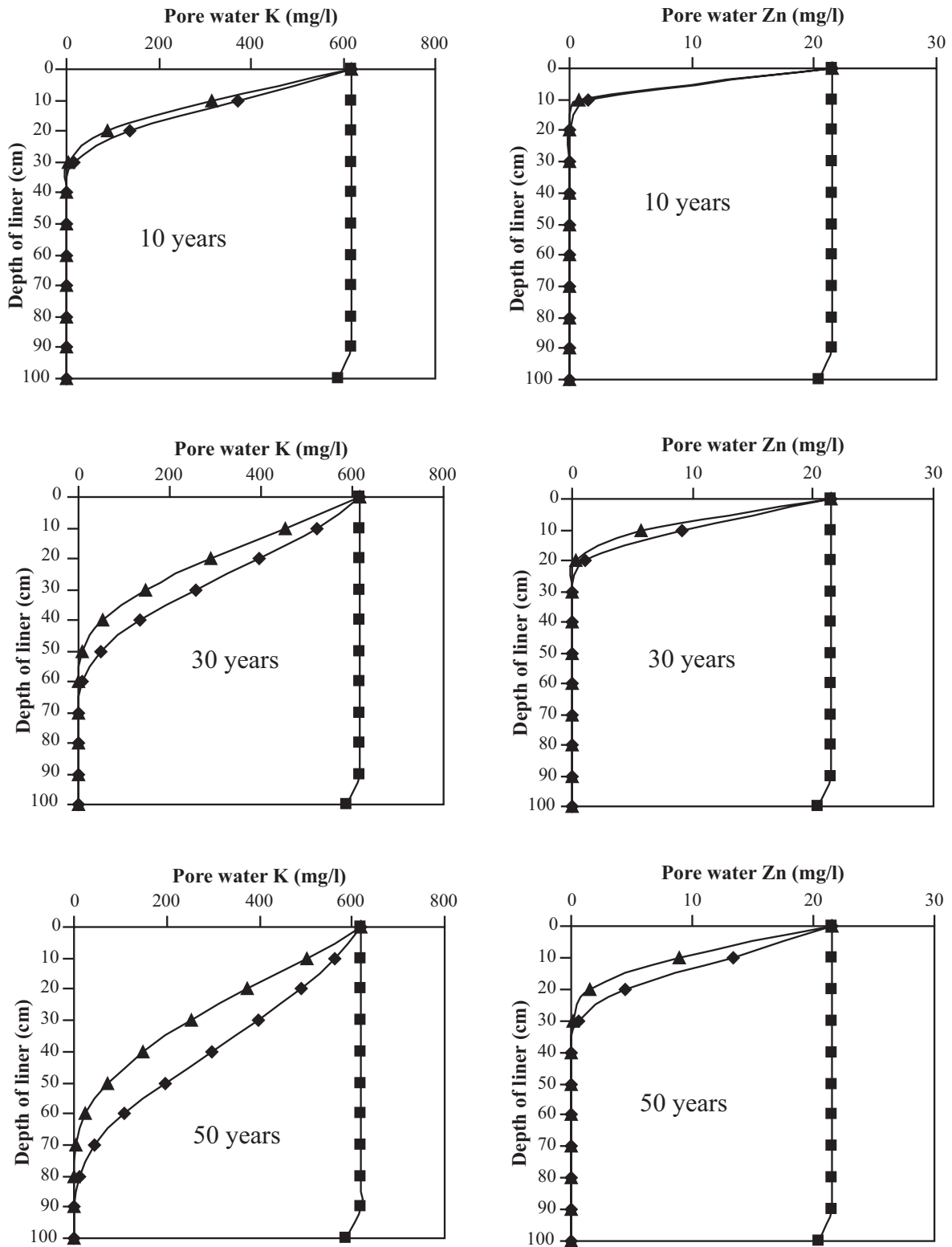


Figure 6. Pore water concentration distributions of K and Zn ions after 10, 30, and 50 years for three different cases explained in the text. Triangle– case 1, diamond– case 2, square– case 3.

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