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The fate of chloride anion passing through columns of montmorillonite clay layers (A pilot study)

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ABSTRACT

Soil serves as a puros medium through which pollutants can be transported top to the deeper layers. Additionally, the movement and dispersion of salts and pollutants within the soil are influenced by soil moisture conditions, such as saturated or unsaturated, as well as the velocity of the flow. The present research aimed to monitor the behavior of water and chlorine in the soil columns containing montmorillonite clay layers and to simulate salt movement across the soil profile under the pilot conditions. To this end, in columns characterized with 150 cm in height and 15 in diameter containing loamy sand soil, treatments with and without clay layers with thicknesses of 13 and 20 cm were embedded. The columns were washed with ammonium nitrate, applying 13 and 20 mEq/l potassium chloride, then the volume and concentration of chlorine drainage, breakthrough curves and simulation of chlorine concentration in soil profile were performed. The results of this study showed that increasing the thickness of the clay layer was well prevented the drainage. In addition, the concentration of chloride in the outlet stream positively correlated with the thicker clay layer and higher salt concentration. The predicted results showed similar results in comparison with the observed data. In charts with a thicker clay layer, chlorine concentrations reached the rates much faster. For the other two treatments, this increase was lower. According to the results, the coefficient of determination (R^2) for chlorine in the soil column was between 0.87 and 0.92, indicating the ability of the model to yield an acceptable simulation.

Keywords: Chloride transport fate, clay layer, HYDRUS-1D, Potassium chloride.

1. Introduction

Avast part of Iran like other arid climates face challenges related to soil salinity. Generally, water in these areas is saline, and when used for irrigation, it exacerbates soil salinity. With each irrigation cycle, a greater amount of salt is brought into the groundwater, rendering it unsuitable for drinking and leading to a loss of a viable water source for the community. Studies have focused on investigating the impact of salt intake concentration and clay layer thickness, as these factors play a significant role in groundwater and soil salinity dynamics. The soil porous medium is a substrate for moving and transporting pollutants from surface to depths. Soil moisture characteristics including saturated and unsaturated flow and flow velocity, are important in how the salts and pollutants move in soil. When saturated, the macrospore flow becomes significant in the transport of the ions , leading to a substantial salt accumulation. This increase is directly related to the saturation level of the soil and the velocity flow of water, consequently affecting the flow of solutes (Bejat et al., 2000; Doustaky et al., 2023). In this regard, the use of modern irrigation methods and new sources such as the unconventional (gray) waters (saline water, sewage sludge) are important management strategies (Doustaky et al., 2023; Akhavan et al., 2019). Utilizing modeling techniques enables the comprehension of various experimental phenomena such as water and solution flow through the application of mathematical formulas. This approach offers the advantage of eliminating the necessity for costly repetitive field experiments. (Khayamim and Gohari, 2005; Ebrahimi et al., 2010; KAboli et al., 2019; Pazhman et al., 2021).

The spatial variability of soils (e.g. texture, soil type) presents a significant challenge for scientists in the field of soil science (Seyed Alikhani et al., 2011). Analyzing water flow in the soil, particularly on a large scale like a relatively shallow basin, becomes difficult due to this variability. However, an effective solution to address these variability issues is the application of scaling methods. These methods, developed based on Miller's theory of similar environments, provide a means to overcome the challenges posed by spatial variability and facilitate the analysis of water flow in diverse soil conditions, even at larger scales (Rezai Rashti et al., 2008; Karimpoor et al., 2013). Scaling methods have various applications, one of which is the provision of general solutions for the flow,, such as the Richards Equation. These general solutions can be applied to different soil types and conditions. Particularly in the case of the highly nonlinear Richards' Equation, which necessitates complex numerical methods for solving, scaling methods prove to be highly efficient. They significantly reduce the computational complexity and volume, particularly when dealing with spatial variability (Rezai Rashti et al., 2008). An advanced model often utilized in studying water, salt, and heat movement in soil is the HYDRUS model (Tazangi et al., 2020; Karimpoor et al., 2022). This model has been extensively employed in laboratory and field studies to simulate and analyze soil hydraulic properties or to inversely estimate them (Rezai Rashti et al., 2008; Fazlali et al., 2015; Tazangi et al., 2020; Karimpoor et al., 2022). The HYDRUS model, initially developed by Simunek et al. in 1998 at the American Salinity Laboratory, is a powerful tool for simulating water and salt flow in both saturated and unsaturated conditions. The model takes into account the transport of solutes, considering both physical and chemical equilibrium, and allows for the expression of non-equilibrium conditions. By utilizing the Marquardt-Levenberg parameter estimation technique, the HYDRUS model can inversely estimate and optimize soil hydraulic parameters as well as parameters related to the absorption, storage, and transfer of salts. Furthermore, the model provides graphical illustrations of the simulation results for water and salt movement in the solid and solution phases. (Simunek et al., 2006., Akhavan et al., 2018; Ebrahimi, 2022; Taghdisi et al., 2020). The performance of the HYDRUS-1D software has been extensively used in various research projects conducted both in Iran and worldwide. One such investigation involved studying the active adsorption of ammonium in a sandy column using the HYDRUS-1D model (Source ???). Additionally, the risk associated with using sewage for groundwater recharge was evaluated by studying NH4+ adsorption through a laboratory-made solution. Comparing the use of sewage to the laboratory solution, it was found that the ammonia uptake time decreased by up to 7 times due to higher concentrations of competing ions like calcium and magnesium in the sewage. The application of the HYDRUS-1D model demonstrated its ability to effectively simulate the transfer of ammonium in the soil column, highlighting its usefulness in such studies (Jallali et al., 2010). Another study focused on investigating the rate of water infiltration in a 5-layer soil column with a height of 300 cm. The study compared the performance of the Green Amp model with the HYDRUS-1D model. The results revealed that the HYDRUS-1D model successfully simulated the water infiltration process, and the cumulative infiltration depth was accurately predicted by the model in the soil layers (Ying et al., 2009). The modeling of nitrogen leaching from a field experiment using the HYDRUS software demonstrated that in soils characterized by a coarse texture, such as sandy loam, nitrogen leaching increased as the water discharge increased. This resulted in the transportation of nitrogen to deeper soil layers, which subsequently led to the pollution of underground reserves. This finding highlights the importance of managing nitrogen fertilization and water discharge rates in sandy loam soils to mitigate the risk of groundwater contamination (Azhdary et al., 2008). During a test conducted on a soil sample at a depth of 3 meters, the Green Amp model was employed to obtain measurements of momentum penetration, cumulative penetration, and the depth of waterfront infiltration. These measurements were then compared with the results obtained from the HYDRUS-1D model. The comparison revealed that the HYDRUS-1D model exhibited greater accuracy in simulating momentum and cumulative penetration rates compared to the depth of waterfront infiltration. This suggests that the HYDRUS-1D model is particularly effective in accurately predicting the movement of water through the soil (Ying et al., 2010). The objective of the study was to investigate the behavior of chloride ions and water as they flow through columns containing clay layers. Specifically, the researchers aimed to simulate the movement of chloride ions in soil columns that contained trapped layers of montmorillonite clay using the HYDRUS-1D model under pilot conditions. By utilizing the HYDRUS-1D model, the researchers aimed to gain a deeper understanding of the transport and behavior of chloride ions in the presence of clay layers.

2. Materials and Methods

In this study, the sandy loam soil used was collected from the Ziarat village, located at coordinates 36°42'8" E and 54°28'29" N. Additionally, clay soil was obtained from the outlet of Shirvan city in North Khorasan province, with coordinates 37°24'48" E and 57°53'26" N. Once collected, the soil samples were air-dried and subsequently sieved through a 2 mm mesh to remove any larger particles or impurities. This preparation process ensured that the soil samples used in the study were consistent and free from any unwanted debris. Based on the provided information, the measured physical properties of the studied soils are presented in Table 1. It was observed that the soil with a coarse texture contained 75% sand, while the soil with a fine texture consisted of 46% clay. In terms of acidity, both soils were found to be neutral within the measured range. The electrical conductivity of both soils indicated that they were in the non-saline soil category. To create the soil columns, pillars with a diameter of 16 cm and a height of 150 cm were prepared.. The tubes were cut and a lid was placed on one side, while a drain outlet was created at the other end of the column. The inner wall of the column was covered to prevent the formation of preferential flow paths. To improve water drainage, a small simple funnel covered with fine mesh was installed at the bottom of the columns. Additionally, a small amount of fine stones and

Table 1. Coarse and fine grained soil characteristics used for the experiment

	EC	pH	Prticle density (g/m^3)	Bulk density (g/m^3)	Clay (wt%)	Silt (wt%)	Sand (wt%)	Texture (wt%)
Coarse	1.56	7.23	2.62	1.24	8	17	75	Sandy loam
Fine	2.2	7.35	2.65	1.3	46	34	20	clay

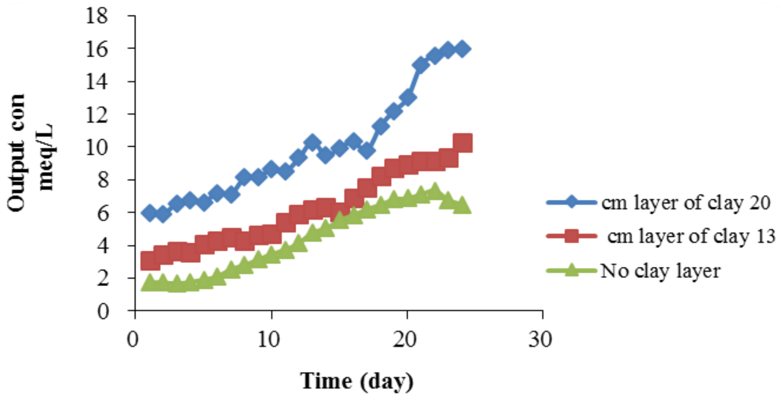


Fig. 1. Chlorine concentration variation over time in soil column drain with different clay layers fed with a concentration of 20 mEq/l potassium chloride

gravel were placed at the bottom of the columns to facilitate better dranage . The soil was carefully poured into the columns and gently compacted using compressed iron cylinders to achieve a bulk density similar to the actual excavated soil. A total of 18 columns were filled for the experiment. For the clayey layer, three treatments were employed: one with a 20 cm clay layer, another with a 13 cm clay layer, and a third treatment without any clay layer (clay-free treatment). To begin the experiment, the primary chlorine was washed out using ammonium nitrate salt. The initial concentration of chlorine in the columns was around 10-15 mEq/l, which was then leached with one normal concentration of ammonium nitrate to reduce it to approximately 0.5 to 1 mEq/l. Ammonium nitrate was used as the fertilizer in the experiment. Chloride, serving as a tracer, was added to the soil in two treatments: 13 mEq/land 20 mEq/l. In all the columns, a water load of 20 cm was applied to the top of the soil. The amount of water output from the columns was measured twice daily at around 03:30 AM and 03:30 PM. Then, the soil samples were transferred to the laboratory and stored in refrigerator. Once the experiments were completed and soil information was collected, the data was processed and analyzed using the software. The HYDRUS-1D model, was used to simulate the movement of chlorine ions in the soil and predict the chloride concentration in the soil columns.

3. Results and Discussion

This section initially presents the findings regarding the measured characteristics of the examined soils. Subsequently, the results pertaining to soil column data on the drainage curves of treatments are provided. Statistical comparisons are conducted for both cases. Following that, the curves for various salt concentrations are presented based on the results of plotting. Finally, the results derived from simulating HYDRUS-1D are presented, which include diagrams displaying the chlorine concentration in soil profiles for columns featuring clay layers with varying salt input concentrations. Additionally, a statistical comparison is performed for the simulation results.

3.1. Soil characterization

The characterization results of both fine and coarse-texture soils are presented in Table 1.

3.2. Soil column data comparison

In this section, the outcomes derived from the soil column experiments are presented and compared. Specifically, the variations in output drain concentration and volumetric flow over time are analyzed and discussed. Figure 1 reppresents the results of soil column drain concentrations when 20 mEq/lof salt was applied, considering different depths of clay loam layers. The data reveals a significant disparity between the clay layer treatments for both salt concentrations. The clay layer with a thickness of 20 cm exhibited the highest concentration of chlorine in the drainage, while the

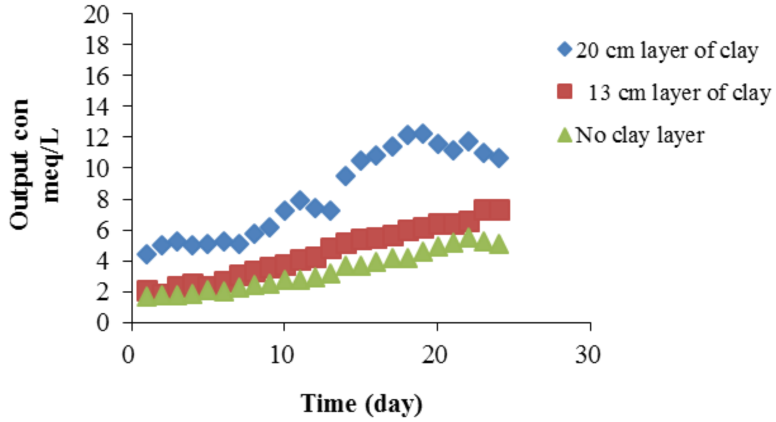


Fig. 2. Chlorine concentration variation over time in soil column drain with different clay layers fed with a concentration of 13 mEq/l potassium chloride

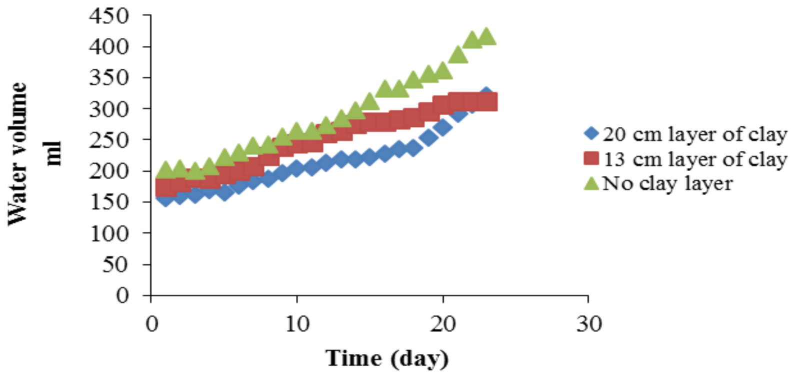


Fig. 3. The water volumetric flow variation over time in soil columns with different clay layers fed with a concentration of 13 mEq/l of potassium chloride

control sample, lacking a clay layer, exhibited the lowest concentration. This discrepancy can be attributed to the anionic decomposition property of the clay layer, leading to a thicker layer and consequently a higher chlorine concentration in the drainage. On the other hand, the control treatment, consisting of lightweight clay-free material, had the ability to absorb chlorine, resulting in a lower concentration in the drainage. In the column with a thick clay layer, the concentration of salt in the outlet stream surpassed that of the inlet, possibly due to effective groundwater dehydration processes. In Figure 1, which represents a higher salt concentration (20 mEq/l) in the soil columns, the treatment with a thicker clay layer exhibited the greatest disparity, similar to what was observed in Figure 2 with the 13 mEq/l treatment. The discrepancies between the three treatments can be attributed to the varying thicknesses of the clay layer and the anionic clay repellency. The treatment with the thicker clay layer experienced a higher concentration of chlorine in the drainage due to these factors. A comparison between Figures 2 and 1 indicates that as the salt concentration increased, the differences between the treatments decreased, and the behaviors of the different treatments became more similar, albeit with slight variations in soil response In this section, the volumetric flow curves of the treatments using soil column data were analyzed. The drainage from columns without clay and columns with varying clay layers were measured and plotted for treatments with 13 and 20 mEq/l concentrations. The diagram illustrates that the treatment with a 20 cm clay layer exhibited the lowest drainage output. This can be attributed to the presence of small pores in the clay layer, which restricts the passage of drainage. In contrast, the treatment without clay showed the highest drainage output, as it had a more permeable texture compared to the clay columns.

In this section, the treatments were compared based on statistical data obtained from the soil column experiments, specifically focusing on volume and output drainage concentration. The volume comparison of drainage between treatments with and without clay layers, at a concentration of 13 mEq/l, was evaluated using the Tukey test at a significance level of 0.05. As depicted in Figures 3, 4 and 5 and Table 2, while there

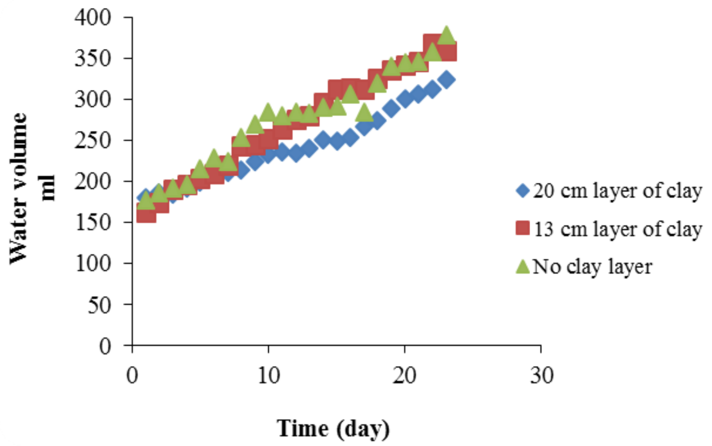


Fig. 4. The water volumetric flow variation over time in soil columns with different clay layers fed with a concentration of 20 mEq/l of potassium chloride

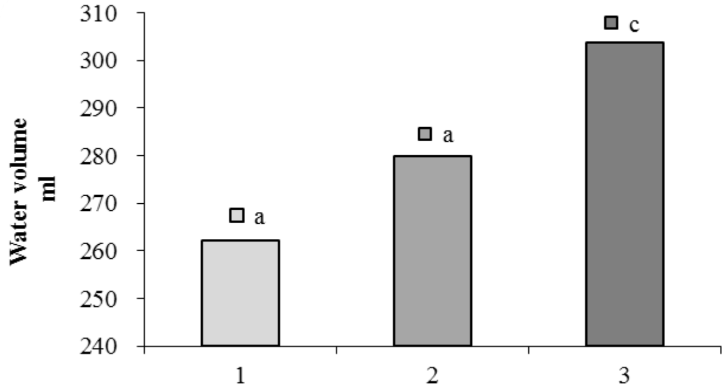


Fig. 5. The ratio of the volume of water outlet in columns with and without clay layer fed with a concentration of 13 mEq/l of salt (similar letter represent non significancy at 0.05 level by the Tukey test)

Table 2. Comparison of the average output of drainage water at concentrations of 13 and 20 mEq/l

Salt Concentration	Without clay	13 cm clay	20 cm clay
13 mEq/l	303.79 b*	279.92 a	262.12 a
20mEq/l	324.19 c	270.56 b	240.92 a

*data with similar letter represent non significancy at 0.05 level by the Tukey test

was no significant difference observed among the clay layers in terms of drainage output ($P < 0.05$), a significant difference was found between the columns with clay layers and the control treatment.

Additionally, in the analysis of volumetric results at a concentration of 20 mEq/l, the treatments were statistically evaluated using the Tukey test at a significance level of 0.05. Figure 6 and Table 3 demonstrate a significant difference among the treatments at the 0.05 level ($P < 0.05$). Notably, there was a significant difference observed among the treatments with clay layers, indicating the impact of concentration on the drainage output within the clay layers.

The statistical comparisons of drainage concentration between the control treatment and the clay layers, when fed with a salt concentration of 13 mEq/l, were performed using the Tukey test at a significance level of0.05. Based on Figures 7 and 8 and Table 3, all three treatments exhibited a significant difference when compared to each other ($P < 0.05$). The treatment with the thicker clay layer demonstrated the highest concentration of chlorine in the water drainage, which

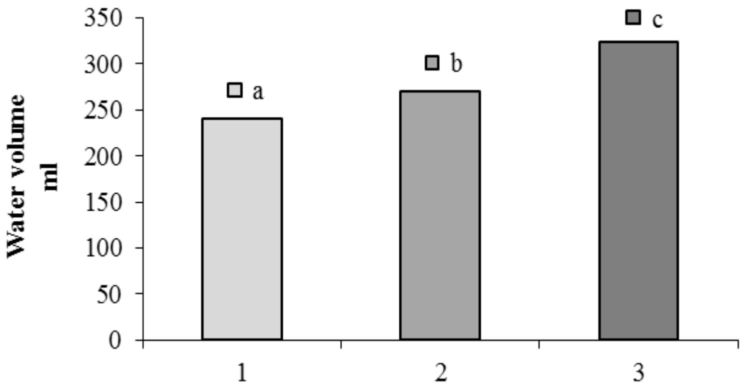


Fig. 6. The ratio of the volume of water outlet in columns with and without clay layer fed with a concentration of 20 mEq/l of salt

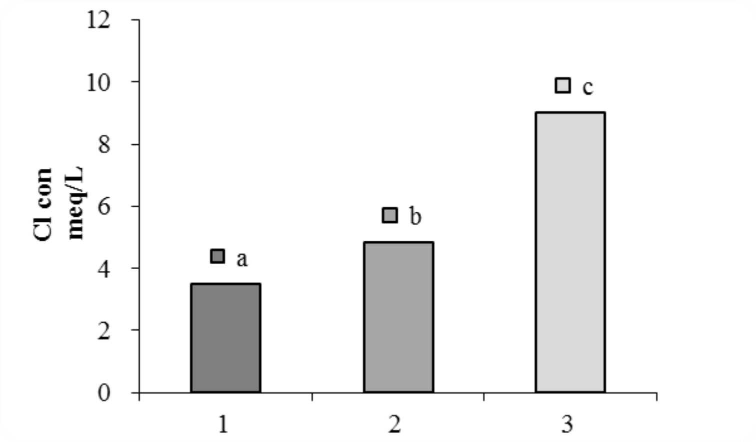


Fig. 7. The ratio of water outflow of columns with and without clay layer fed with a concentration of 13 mEq/l of salt

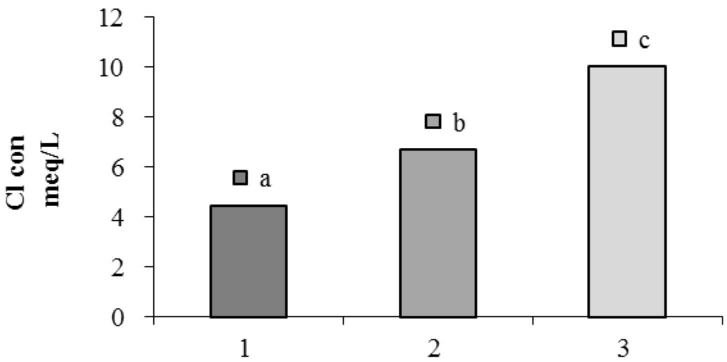


Fig. 8. The ratio of water outflow of columns with and without clay layers with a concentration of 20 mEq/l of salt

Table 3. Comparison of the average drainage concentration at concentrations of 13 and 20 mEq/l

Salt Concentration	Without clay	13 cm clay	20 cm clay
13 mEq/l salt	9.00 c	4.81 b	3.5 a
20 mEq/l salt	10.02 c	6.69 b	4.46 a

*data with similar letter represent non significancy at 0.05 level by the Tukey test

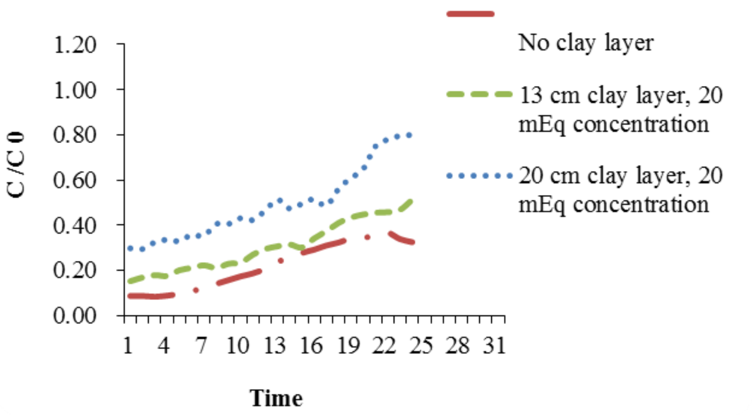


Fig. 9. Intrusion curve for bellows and clay layers for 20 mEq/l of salt

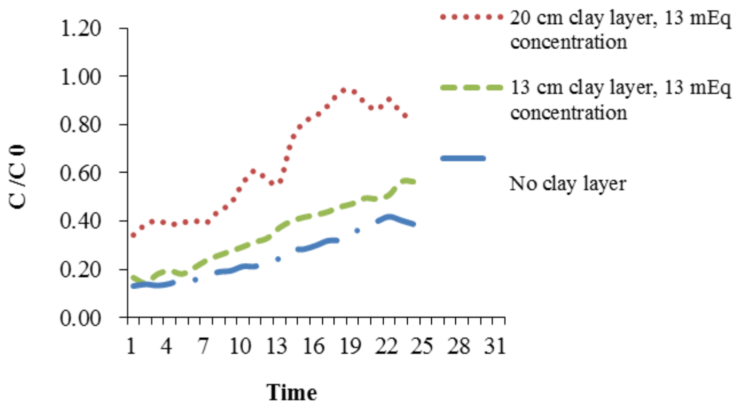


Fig. 10. The penetration curve for bell-free columns and clay layers for a concentration of 13 mEq/l of salt

can be attributed to the anionic decay properties of clay. Conversely, the soil with a lighter texture was capable of absorbing a substantial amount of chlorine at its surface, resulting in the lowest chlorine output in the treatment.

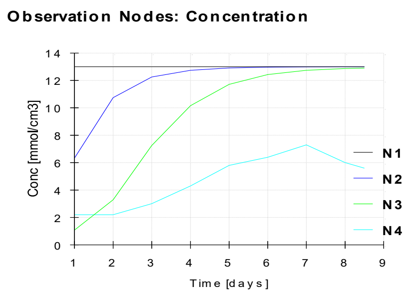
The study's results include a statistical comparison of the drainage concentration between different soil columns when supplied with a salt concentration of 20 mEq/l. The statistical analysis, conducted using the Tukey test with a significance level of 0.05, indicated a significant difference among the treatments. The highest concentration of drainage was observed in the clay layer at a depth of 20 cm. When comparing the two graphs for each treatment, it was found that increasing the salt concentration from 13 to 20 mEq/l did not show a significant difference in the increase of concentration. This suggests that the concentration increase had a somewhat limited effect in this range. These results are summarized in Table 3.

3.3. Investigation of the results of infiltration curves for b and clay treated with different concentrations of salt

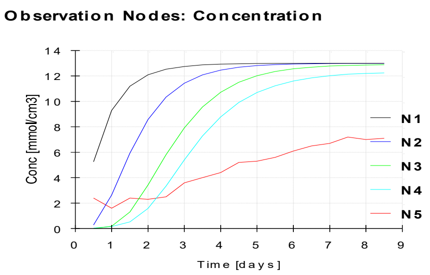
The penetration curves for bell-shaped columns with different clay layers for the concentration of 13 and 20 mEq/l of nutritional salt are shown in Figures 9 and 10. By increasing the thickness of the clay layer at a similar concentration, the concentration of chlorine passed through the initial chlorine increased.

3.3. HYDRUS-1D model results evaluation

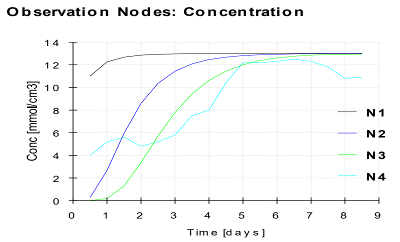
In Figures 11 and 12, the chlorine concentration is depicted for the control point (column without a clay layer) and columns with clay layers, all of which were fed with a concentration of 13 mEq/l. There is a notable difference in the behavior of chlorine concentration at the control point compared to the column with the clay layer. This discrepancy arises due to the presence of a clay layer of certain thickness. In the graph featuring a 20 cm clay layer, the concentration reaches its peak much faster compared to the other two columns. Additionally, the presence of chlorine ions is evident as the input concentration of chlorine increases. In the other two columns, the increase in concentration is slower, indicated by a gentler slope. The presence of a thicker clay layer results in higher anion excretion, contributing to this variation in behavior.



(a)

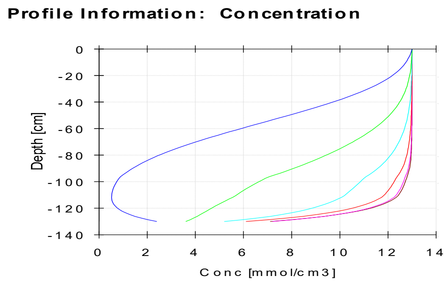


(b)

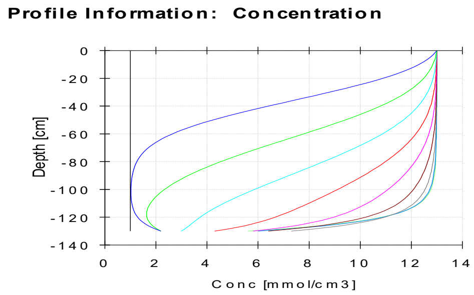


(c)

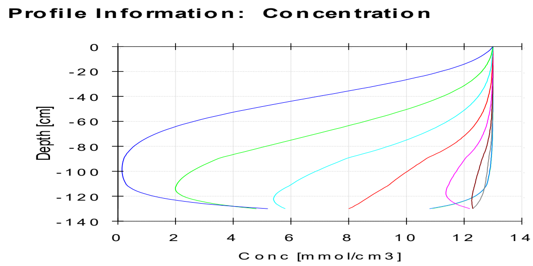
Fig. 11. Concentration of chlorine at the control points, a column without clay layer (a), column with with 13 cm (b) and 20 cm (c) clay layer with an initial salt concentration of 13 mEq/l



(a)



(b)



(c)

Fig. 12. The concentration of chlorine in the soil profile for a column without clay layer (a), column with with 13 cm (b) and 20 cm (c) clay layer with an initial salt concentration of 13 mEq/l

As it can be seen, with increasing the thickness of the clay layer, at longer time intervals, the lines of the diagram are closer to each other, which is due to the behavior of anionic clay repellency.

In Figure 13, the chlorine concentration at the control points is illustrated for columns with varying clay layer thicknesses, all initially fed with a salt concentration of 20 mEq/l. As depicted in the Figure 13, there is a discernible difference in the behavior of chlorine concentration at the control points due to the variation in clay layer thickness. In the graph featuring a 20 cm clay layer, the concentration reaches its peak much faster compared to the other two columns. Additionally, the presence of chlorine ions is evident as the input concentration of chlorine increases. However, in the other two columns, the increase in concentration is slower.

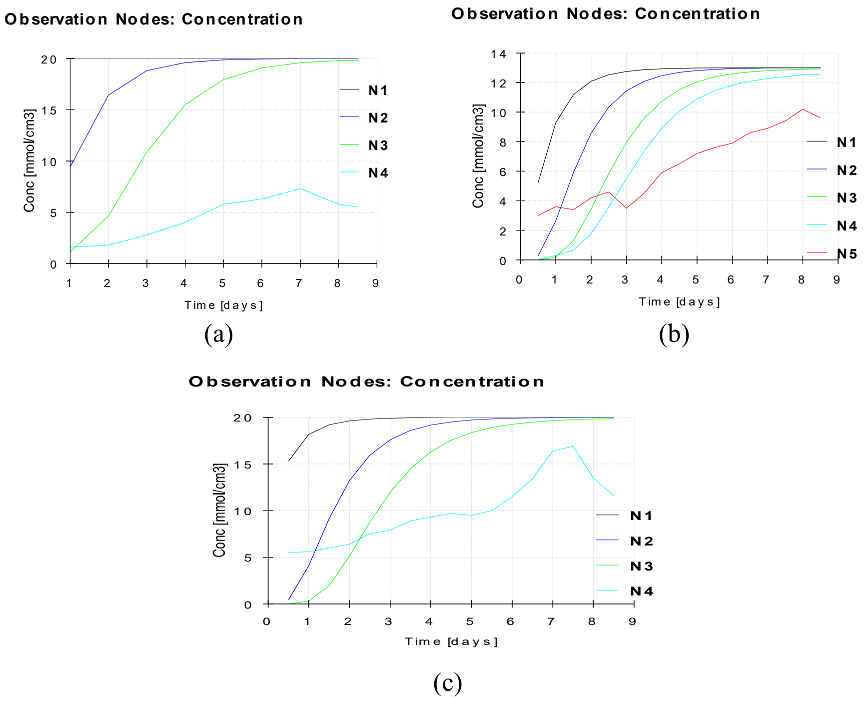


Fig. 13. Concentration of chlorine at the control points, a column without clay layer (a), column with with 13 cm (b) and 20 cm (c) clay layer with an initial salt concentration of 20 mEq/l

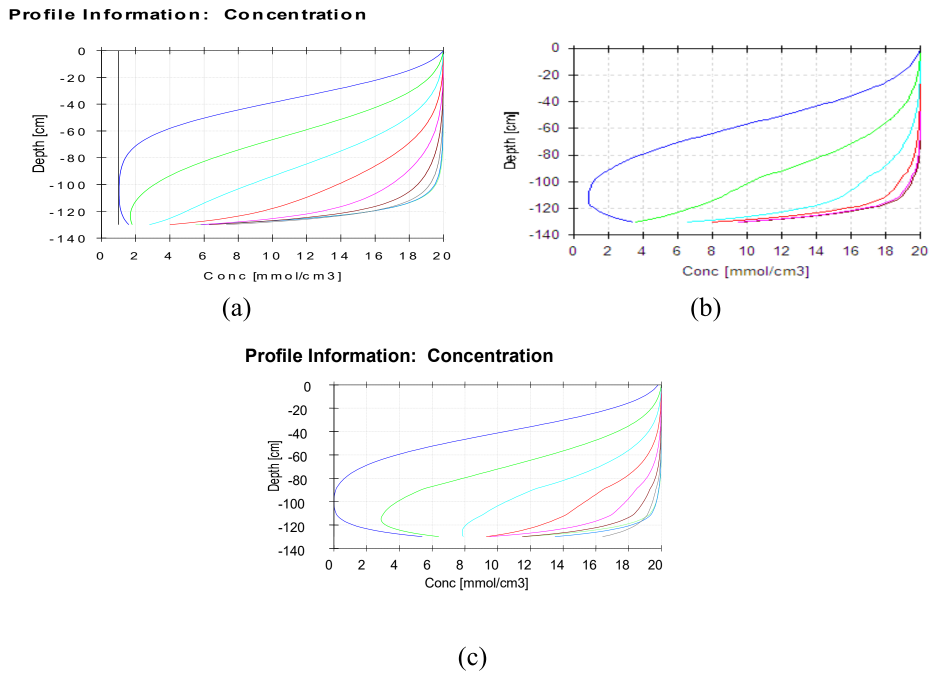


Fig. 14. The concentration of chlorine in the soil profile for a column without clay layer (a), column with with 13 cm (b) and 20 cm (c) clay layer with an initial salt concentration of 20 mEq/l

As observed, the lines in the diagram tend to converge or become closer to each other at longer time intervals as the thickness of the clay layer increases (Figure 14). This behavior can be attributed to anionic clay repellency. Anionic clay repellency refers to the tendency of clay layers to repel or hinder the movement of anions, such as

Table 4. Statistical Comparison of HYDRUS-1D Simulation

Control 13	Control 20	Layer 13 cm, con 13 mEq/l	Layer 13 cm, con 20 mEq/l	Layer 20 cm, con 13 mEq/l	Layer 20 cm, con 20 mEq/l	
0.92	0.88	0.88	0.87	0.90	0.89	R^2

chlorine ions, within the soil profile. As the clay layer becomes thicker, it exerts a stronger influence on the movement of anions, resulting in reduced differences in chlorine concentration over time between the different columns. This phenomenon causes the lines in the diagram to converge, indicating a more consistent behavior of anionic clay repellency as time progresses.

Table 4 presents the statistical comparison of the HYDRUS-1D simulation results. The calibration steps for the HYDRUS-1D model involved adjusting coefficients such as longitudinal dispersivity, Henry, Beta, Frac, and Alpha to achieve the best match between the measured values of moisture and salinity and the mean simulation values of the model. This calibration process ensures that the model outputs align closely with the observed data, improving the accuracy and reliability of the simulation results.

The coefficient of determination (R^2) value indicates the effectiveness of the HYDRUS-1D model in simulating the chlorine concentration along the soil column. The results indicate that the column with a thicker clay layer exhibited the highest chloride concentration in the drainage for both salt input concentrations. This can be attributed to the increased thickness of the clay layer within the column. The presence of more clay enhances anionic clay repellency, leading to a greater likelihood of anionic extraction. When an anion is introduced to soil colloids with a net negative charge at the soil pH, the process of anion removal, or negative absorption, occurs. This occurs because the soil has a limited capacity for anion adsorption, resulting in anion excretion. Several factors influence the anion recovery process, including electrical charge and anion concentration, exchangeable cationic species, soil pH, presence of other anions, and the nature and type of charge on the colloid surface (Fazlali et al., 2015). The results also indicate that the column without clay effectively absorbed chlorine. In the control column, with an inlet salt concentration of 13 mEq/l, an inoculum concentration of 20 mEq/l, and a drainage concentration of 7 mEq/l, a barley concentration of 5 mEq/lwas observed. To study salt propagation, leaching experiments were conducted on soil pillars in the Azizabad region of Bam. The results revealed that coarse-grained soils had a greater impact on ion transfer compared to fine-grained soils. Salts were found to enter the drainage more readily in less compacted soils. A study by Shirani et al. (2015) on anion bromide leaching in soil columns supported these findings, demonstrating that fine-grained soils absorb a greater amount of anions on their surface, while coarse-grained soils exhibit anion excretion properties, which aligns with the results obtained in this study.

The study's findings indicate that as time progresses, the volume of drainage water increases. However, the presence of a clay layer causes a delay in water movement compared to the clay-free treatment. There was no significant difference observed between the 13 cm and 20 cm clay layer treatments when the salt concentration was 13 mEq/l. However, both clay layer treatments showed a significant difference compared to the clay-free treatment. When the salt concentration was increased to 20 mEq/l, all three treatments (clay and different clay layer thicknesses) exhibited a significant difference, with the physical characteristics of the clay layer contributing to the delayed water movement.

The complex structural properties of clay layers have been found to be effective in various applications, such as the containment of different pollutants like organic hydrophobic contaminants, heavy metal cations, and radioactive materials (Zhu et al., 2016). Esmaeili et al. (2005) concluded that the presence of a clay layer can have an impact on solute transfer when lime is used. Lime has a positive effect in preventing the movement of clay and salts in the soil.

These findings highlight the importance of considering the presence and characteristics of clay layers in understanding water and solute movement in soil systems and suggest potential strategies for managing and mitigating the effects of clay layers on solute transport.

4. Conclusions

The diffusion rate of pollutants through different soil thicknesses plays a crucial role in the management and protection of groundwater aquifers and controlling groundwater pollution. This study aimed to investigate the performance of clay layers embedded in columns of coarse-textured soil, specifically focusing on water flow behavior and salt concentration in the drainage. The HYDRUS-1D software was employed to simulate salt movement in the soil. The findings of the study demonstrated that increasing the thickness of the clay layer effectively hindered the transfer of drainage volume. Moreover, the concentration of chloride in the outlet stream exhibited an increasing trend, with the highest concentration observed in the column with a thicker clay layer and higher salt concentration. These results underscore the significance of clay layers in regulating water flow and solute transport in soil systems. Understanding the behavior of clay layers can contribute to developing strategies for maintaining and safeguarding groundwater quality and mitigating pollution risks.

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