



A novel approach for characterizing the mixing zone of a saltwater wedge

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[1] The mixing zone between freshwater and seawater in a coastal aquifer is an important transition region that controls regional groundwater flow dynamics and reactive transport processes. While experimental procedures have been developed to study the movement of salt wedges in aquifers, so far no one has attempted to exclusively map the mixing zone associated with the wedge in a laboratory-scale aquifer model. We propose a colorimetric experimental method, which employs the reaction between alkaline freshwater and acidic saltwater, to map the mixing zone. The experiment was completed in a two-dimensional aquifer model to simulate a steady saltwater wedge. We used the density-dependent flow model SUTRA to simulate these experimental results and to estimate the mixing parameters, i.e., dispersivity coefficients. The results show that the proposed colorimetric technique is a useful experimental approach to visualize and quantify the level of mixing in a saltwater intrusion experiment.

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1. Introduction

[2] In coastal aquifers, the mixing occurring between regional-freshwater and recirculating-saltwater flows poses significant environmental and economical challenges. Understanding the extent of mixing between these two waters has important water management implications since the presence of even a small fraction of seawater (about 1%) would make a freshwater system unfit for drinking. The mixing process also forms a transition zone that controls many natural biological and chemical reactions [Charette and Sholkovitz, 2002; Spiteri et al., 2008]. Although the importance of this mixing zone has been widely recognized, making a realistic prediction of the extent of this mixing zone, using numerical groundwater models, is still a significant challenge. This is because the values of longitudinal and transverse dispersivity coefficients currently used in aquifer-scale models are often determined by numerical modeling constraints rather than actual measurements. Recent studies have identified that the transverse dispersion is the key driving mechanism that controls seawater recirculation patterns in coastal aquifers [Abarca et al., 2007].

Therefore, a fundamental understanding of dispersion mechanisms is critical for evaluating the seawater recirculating flow, which can be a significant component of the overall submarine groundwater discharge.

[3] Understanding the saltwater movement in a field-scale aquifer is a complex task [Michael et al., 2003; Taniguchi et al., 2006]. Therefore, experimentalists have instead studied (and visualized) saltwater wedges in laboratory-scale physical models using different colored waters. For example, researchers have used various inert dyes to differentiate the saltwater and freshwater regions in laboratory scale experiments [Mualem and Bear, 1974; Goswami and Clement, 2007]. While the location of the saltwater wedge was perfectly mapped in these experiments, the studies encountered some difficulties in characterizing the extent of the mixing zone. To the best of our knowledge, so far, no one has attempted to exclusively map the extent of the mixing zone in a saltwater intrusion experiment.

[4] In other scientific areas, researchers have developed noninvasive optical measurement techniques to map the mixing dynamics of different fluids. Most of these studies have focused on mixing under turbulent flow conditions [Webster et al., 2003]. One of the most promising visualization techniques used in turbulent flow studies is the laser-induced fluorescence method [Fountain et al., 1998; Catania et al., 2008]; however, this method requires complex experimental setups [Zhang et al., 1995]. A simpler alternative is to employ the colorimetric reaction method that induces a color change in the system by allowing two reactive tracers to mix. These reactive tracers could be pH indicators, redox indicators, or complexometric indicators (color change in presence of specific metal ions, e.g., EDTA). Redox indicators such as Fe(II) have been used to study the effects of convection on chemical waves [Pojman et al., 1991]. Starch has been used as an indicator to study propagating reaction fronts [Forstova et al., 2002]. However, colloidal indicators, such as starch, tend to flocculate and filter and introduce additional complexities. Complexometric indicators such as EDTA [Gramling et al., 2002] and Tiron [Oates and Harvey, 2006] have been proposed to study reactive transport in porous media. However, the high molecular weight of these complexes can change the density of the solution thus preventing their use to study density dependent flow systems. The pH indicators have been used to study mixing in stirred vessels [Nere et al., 2003] and also to study mixing in groundwater flow systems [Theodoropoulou et al., 2003; Cirpka et al., 2006; Bauer et al., 2007]. In particular, phenolphthalein has been widely used as a pH indicator to study various turbulent mixing problems [Breidenthal, 1981; Menisher et al., 2000]. However, none of these studies have analyzed mixing in porous media under density dependent flow conditions.

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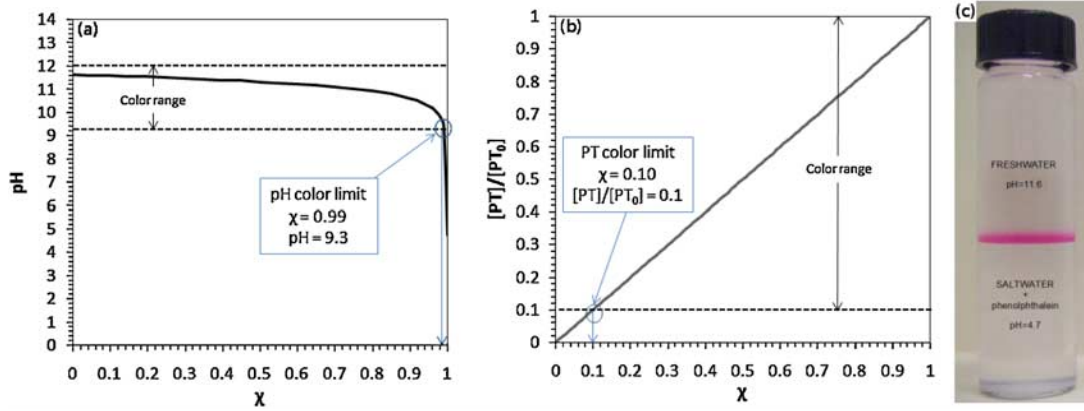


Figure 1. (a) Changes in pH computed using (3) for different saltwater mixing ratios (this data defines the pH controlled boundary), (b) changes in phenolphthalein (PT) concentration for different mixing ratios (this data defines the PT-concentration controlled boundary), and (c) observed interface between acidic saltwater (bottom) and alkaline freshwater (top) in a batch reactor.

[5] The objective of this work is to visualize and characterize the mixing zone between salt and fresh waters in a saltwater-intrusion experiment. We have designed an experimental methodology that employs a colorimetric technique to directly map the mixing zone between these two waters. The location and the observed thickness of the mixing zone were reproduced by a numerical model. The model was also used to establish the upper and lower bounds of the values of dispersion parameters that control the mixing process.

2. Materials and Methods

2.1. Chemical Procedures

[6] We adapted a colorimetric reaction to map the extent of mixing between freshwater and saltwater near a salt wedge. Our method employs the reaction between a strong acid and a strong base that can be visualized using a pH indicator. When a strong-acid solution is mixed with a strong-base solution, the pH variation resulting from the reaction can be modeled using the expression [Ricci, 1952]:

$$[H^+] - [OH^-] = [A^+] - [B^-] \quad (1)$$

where $[H^+]$ and $[OH^-]$ are the molar concentration of the protons and hydroxide ions and $[A^+]$ and $[B^-]$ the molar concentration of the strong acid and base, respectively. $[A^+]$ and $[B^-]$ can be expressed in terms of the initial concentration of acid $[A_0]$ and base $[B_0]$ and the volumetric ratio χ in which they are mixed:

$$[A^+] = \chi[A_0^+] \quad [B^-] = (1 - \chi)[B_0^-] \quad (2)$$

χ is the mixing ratio, which is defined as $V_A/(V_A + V_B)$ where V_A is the volume of the initial acidic solution and V_B the volume of the basic solution. Substituting (2) in (1) after expressing $[OH^-]$ in terms of $[H^+]$ and water dissociation constant (K_w), the following expression is obtained:

$$[H^+] - \frac{K_w}{[H^+]} = \chi[A_0^+] - (1 - \chi)[B_0^-] \quad (3)$$

The above equation can be used to relate the mixing ratio χ with pH variations.

[7] About forty liters of colorless acidic-saltwater and alkaline-freshwater solutions were prepared. The freshwater solution was prepared by adding a sufficient amount of NaOH to de-ionized water to raise the pH of the solution to 11.6. The saltwater solution was prepared by adding commercial salt to de-ionized water to increase the density to 1025 kg/m^3 (density was measured using ASTM 11H hydrometer). HCl was then added to lower the saltwater solution pH to 4.7. Finally 10 ml of 1% phenolphthalein-indicator solution was added to this acidic saltwater. The computed pH values for different mixing proportions of these saltwater and freshwater solutions, calculated using (3), are shown in Figure 1a. The χ values shown in Figure 1a were defined in terms of the volume of acid and hence they represent the volumetric fraction of the saltwater. We also computed the pH variations after accounting for ionic strength effects with PHREEQC [Parkhurst and Appelo, 1999] and the results were almost identical to the values shown in Figure 1a. Therefore, we assumed the effects of ionic strength to be negligible in our experiments.

[8] We selected phenolphthalein (PT) as the indicator because changes in salinity and temperature have negligible effect on PT colorimetric reaction [Kolthoff, 1937]. PT changes from colorless to pink between the pH values of 8.2 and 10 [Kolthoff, 1937]. We define pH_{IN} as the value of the pH at which bright pink color is obtained and the estimated value of pH_{IN} is 9.3; this is the typical value assumed in standard titration experiments and in other visualization experiments [Breidenthal, 1981]. Using (3), we estimated that the pH_{IN} value of 9.3 would correspond to a mixing ratio (χ) of ~ 0.99 . This implies that if pH is the limiting condition then the color change would occur at about 1% of mixing level. Note even if the range of uncertainty in the pH_{IN} values is assumed to be the entire color-change range (of 8.2 and 10), which is a worst case scenario, the corresponding mixing ratios would only vary within 0.973 to 0.995; i.e., the maximum error in χ would be only about 2%. Therefore, the effects of pH_{IN} variations should be negligible.

[9] There are two factors that limit the color of PT in an aqueous solution. The first and the most important factor is pH (as explained above), and the second factor is the concentration of PT. A minimum concentration of PT is necessary to obtain a visible color intensity level. Based on preliminary experiments, we estimated that the visibility threshold level for [PT] should be about 2×10^{-6} M, i.e., 10% of the initial concentration [PT]₀ in our saltwater solution (see Figure 1b). This implies that at least 10% of the saltwater solution (acid + PT) is needed for the reaction to be visible. This threshold value was estimated by completing several screening experiments with different amounts of PT in the basic solution and observing the intensity of the color change by naked eye. We estimated a maximum error of about 5% (of our PT₀ value) in the evaluation of PT concentration that is required to meet the visibility threshold level. Since there is a linear relationship between [PT] and χ , the corresponding uncertainty in defining the mixing region under PT limiting condition should be about $\pm 5\%$.

[10] The values of pH and [PT] will define the limits of the colored region expected to be observed in our saltwater intrusion experiment. According to our design, the mixing zone visible in our experiment should correspond to the values of χ ranging from 0.1 to 0.99. Notice in Figure 1 that the pH values vary logarithmically whereas the PT concentrations vary linearly. Therefore, we expect a sharp transition region when the interface's extent is controlled by the value of pH, and a more diffused transition region when its extent is controlled by the concentration of PT.

[11] An interface was setup in a test tube reactor (without any porous media) to test the feasibility of the proposed visualization technique. Figure 1c shows the interface between saltwater (at the bottom) and freshwater (at top) present in the reactor. The interface is clearly defined by the pink region that is present in between the two colorless solutions. The lower limit of the colored region (corresponding to $\chi = 0.99$), which is limited by the value of pH, is sharp. On the other hand, the upper limit (corresponding to $\chi = 0.1$) of the colored region, which is limited by the concentration of PT, is more diffused.

2.2. Details of the Experimental Set-Up

[12] A laboratory-scale aquifer model similar to the one described by *Goswami and Clement* [2007] was used in this study (see the auxiliary material for a conceptual figure).¹ A two-dimensional tank was constructed using 1-cm thick Plexiglas. The central porous chamber (0.5 m \times 0.3 m \times 0.027 m) was packed under wet conditions with 1.1 mm diameter glass beads to a height of 0.195 m. We consider the glass-bead system to be homogeneous and isotropic. Two constant head containers were used to deliver the saltwater and freshwater solutions. The tank was initially flushed with a freshwater solution of pH around 10, flowing from right to left, until all the water used to pack the tank was fully replaced. Then the acidic saltwater solution from a constant head chamber was fed into the left inlet chamber. The height of the saltwater level (h_s) in the left chamber was fixed at 0.182 m, and the height of the freshwater level (h_f) in the right chamber was fixed at 0.191 m. Previous

experiments have indicated that a steady-state salt wedge can be established in this system within an hour. Therefore, the system was run for about 90 minutes. After this period, the solution in the freshwater chamber was replaced with the alkaline freshwater solution of pH 11.6. When the alkaline freshwater reached the interface it reacted with the acidic saltwater and formed a bright pink mixing zone. The system was continuously run well over an hour and no further changes were observed in the shape and extent of the mixing zone; this confirmed that the system had reached steady-state conditions. The flow and transport parameters indicated that the experiments were conducted in a dispersion-controlled regime ($300 < Pe < 105$); therefore, we could safely disregard the effects of molecular diffusion. In the above analysis, we estimated the value of Pe as the ratio between the characteristic freshwater flow rate (q) and the molecular diffusion factor ($\phi D_m/L$). Since all the transported species (including NaCl, PT, OH⁻ and H⁺) are conservatives, we expect the colorimetric-reaction zone to represent the saltwater mixing zone. Discrepancies may occur for small groundwater flows that approach a diffusion-dominated regime. At steady state, low flow ($Pe < 0.3$) conditions (or no flow condition similar to the batch experiment), the differences in the values of molecular diffusion coefficients of the transported species could lead to discrepancies in the saltwater mixing zone and the observed colorimetric reaction zone. However, the high rate of flow used in our experiment would prevent this effect.

3. Results

3.1. Visualization of the Saltwater-Freshwater Mixing Zone

[13] Figure 2a shows the location of the saltwater wedge mapped in our two-dimensional experimental aquifer. The pink region shown in Figure 2a identifies the dispersion driven saltwater-freshwater mixing zone. The observed mixing zone is thin near the base of the aquifer and becomes progressively thicker as the interface approaches the discharge zone. The toe of the interface penetrated about 15 cm into the freshwater system. At the discharge boundary (point of contact near the left chamber), the thickness of the mixing zone was about 1.5 cm (measured along the vertical direction).

3.2. Characterization of the Mixing Zone

[14] Numerical simulations were completed using the finite element, density-dependent flow and transport code SUTRA [*Voss and Provost*, 2002]. The problem domain (0.5 m \times 0.91 m) was discretized using a quadrilateral grid with 511 \times 195 elements. A transient simulation ($\Delta t = 1$ s) was run until the steady state was reached. A grid refinement study was completed to ensure convergence (see the auxiliary material for details). The boundary conditions used are: (1) a prescribed freshwater head of 0.191 m with density set to 1000 kg/m³, and (2) a prescribed saltwater head of 0.182 m with a salt-mass fraction value of 0.0357 (density of 1025 kg/m³). The hydraulic parameters used in the simulations are: equivalent freshwater hydraulic conductivity = 1000 m/d, porosity = 0.39, fluid viscosity = 0.001 kg/m-s, and molecular diffusion coefficient = 1.477×10^{-9} m²/s. The porous media properties were evaluated using the experimental methods described by *Goswami*

¹Auxiliary materials are available in the HTML. doi:10.1029/2008GL036995.

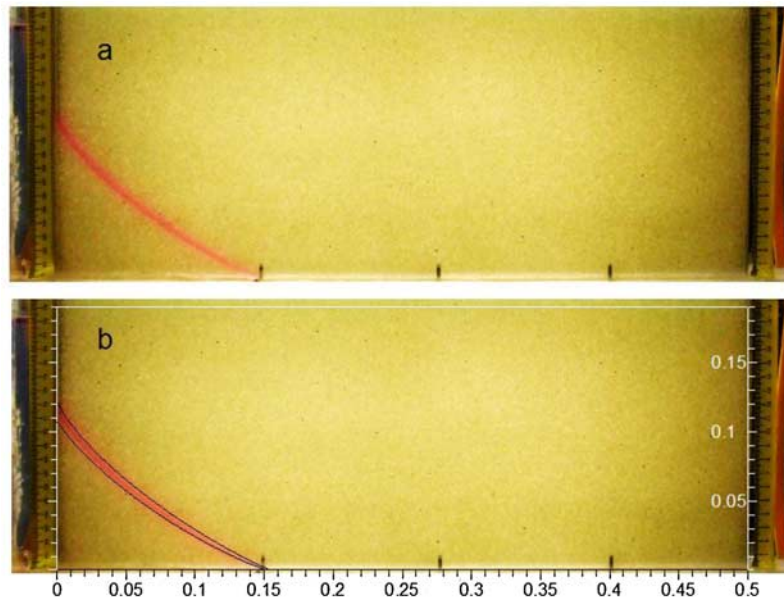


Figure 2. (a) Mixing zone between saltwater and freshwater regions mapped using the colorimetric method and (b) mixing zone simulated by the SUTRA model. The two solid lines represent the 10% and 99% saltwater isolines. Length unit is in meters.

and Clement [2007]. The longitudinal (α_L) and transverse (α_T) dispersivity coefficients were estimated by fitting model simulation results to the observed data.

[15] Dimensionless analysis of a similar type of saltwater intrusion problem had indicated that the bottom extent of the salt wedge (also known as the toe location) would depend solely on the hydraulic gradient, density contrast, and dispersivity values [Dentz *et al.*, 2006; Abarca *et al.*, 2007]. Since the hydraulic conductivity and density are known for our system (from measured data), the model predicted toe location would primarily depend on the assumed values of dispersivities. Furthermore, the thickness of the mixing zone would solely depend on the dispersivity values [Abarca *et al.*, 2007].

[16] We completed fourteen numerical simulations by systematically varying α_L and α_T values within an expected range (see auxiliary material for details). Based on a literature review of the values of dispersivity coefficients, we varied α_L values within a range of 0.0001 m to 0.002 m and the ratio of α_L/α_T within a range of 2 to 40 [Johannsen *et al.*, 2002; Oswald and Kinzelbach, 2004; Benekos *et al.*, 2006; Goswami and Clement, 2007].

[17] Three metrics that characterize the spatial locations of the salt wedge boundaries and the thickness of the mixing zone near the discharge boundary (see auxiliary material for more details) were used together with a visual inspection to evaluate the goodness of the model fit. Although our calibration procedure is approximate we were able to establish the upper and lower bounds of the values of the dispersivity coefficients that characterize the system. A good match between the experimental data and the numerical model result was obtained for $\alpha_L = 0.0005$ m and $\alpha_T = 0.00005$ m (Figure 2b). Also, an alternate simulation with the same α_T value, but with a higher α_L value ($\alpha_L = 0.001$ m) offered a good match. The model simulation results clearly show (see

Figure S3 and Table S1 in the auxiliary material) that the value α_L can be bounded (definitely higher than 0.00025 m and smaller than 0.002 m). Therefore, the value of α_T for our system was estimated to be ~ 0.00005 m, and the value of α_L is expected to be in the range of ~ 0.001 to 0.0005 m. These dispersivity estimates are within the range of values measured for a similar type of porous medium [Benekos *et al.*, 2006], and are in agreement with the results reported by Grane and Gardner [1961], Harleman and Rumer [1963], and Eberhardt and Grathwohl [2002].

4. Conclusions

[18] In this study we have successfully applied, for the first time, a colorimetric method to map the extent of mixing between freshwater and saltwater in a laboratory-scale saltwater intrusion experiment. The width of the observed mixing zone allowed us to quantify the values of dispersion coefficients. Studying the mixing zone between saltwater and freshwater regions using the proposed experimental procedure would help better understand saltwater mixing patterns in coastal aquifers. The main advantage of the proposed technique is that it allows exclusive visualization of the mixing zone. The applicability of this technique is restricted to advection-dominated systems in which the effects of differences in molecular diffusion coefficient values can be disregarded. The main limitation of this study is that our analysis was restricted to a steady state system. Future applications should focus on extending the proposed method to study saltwater intrusion dynamics under transient conditions.

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