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Modes of free convection in fractured low-permeability media

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[1] Abundant data indicate significant fluid and solute fluxes across low-permeability shales in the Gulf of Mexico, the precise mechanisms of which are poorly understood. In this study, we analyze possible modes of intrafracture and interfracture free convection that may occur across fractured low-permeability layers, such as shales. Fracture spacing, fracture aperture, shale thickness, and the density gradient across the shale unit are shown to play key roles in the governing fluid and solute transport processes. All modes of free convection (parallel to the fracture plane, perpendicular to the fracture plane, and convection between fractures on the larger layer scale) are theoretically possible for reasonable hydrogeologic parameters. Free convection parallel to the fracture plane is shown to be the dominant (and most likely) mode of free convection, requiring only very modest salinity differences for onset to occur. Least likely is convection perpendicular to the fracture plane. The results presented here suggest that free convection may not be uncommon in thick shale sequences, such as in the Gulf of Mexico Basin. An important consequence of these findings is that analyses that do not consider both interfracture and intrafracture convection modes may significantly underestimate the likelihood of the occurrence of free convection. These findings have important implications for the study of free convection and solute transport processes in fractured low-permeability media and associated numerical modeling analyses.

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1. Introduction

[2] The impetus for this study was the recognition of salinity inversions in the Gulf of Mexico Basin, which is dominated by low-permeability sediments. This basin and other similar sedimentary basins are major regions of energy resource production, including hydrocarbons, uranium, and lignite. *Morton and Land* [1987], *Land* [1991], *McKenna and Sharp* [1997], and *Sharp et al.* [2001] found that salinity inversions occur in the Gulf of Mexico Basin where more saline waters in formations (including brines in excess of 100,000 ppm total dissolved solids) overlie less saline formation waters (some about 20,000 ppm). This inversion creates a significant buoyancy gradient because the shallower waters are cooler and more saline than the deeper waters. Thus, density-driven (free) convection is a possible process for circulating fluids without large external inputs and outputs and has previously been suggested as a possible solute transport mechanism by several authors [e.g., *Wood and Hewett*, 1984; *Sharp et al.*, 1988; *Land*, 1991] to account for solute fluxes required for the observed levels of sediment diagenesis. *Bjorlykke et al.* [1988] calculated that free convection driven solely by normal temperature increases with depth in most basins was unlikely if laterally extensive shale layers were present. *Sharp et al.* [2001] noted that density effects induced by salinity variations are expected to dominate transport behavior. These studies suggest that free convection, should it occur, is expected to be driven mainly by salinity differences across shale layers and that temperature differences in these settings are generally less significant as a contributor to a combined thermohaline convective transport process. Because shales dominate the sedimentary sequence of many sedimentary basins, understanding their control on free convection is critical. As a first demonstration of free convection modes in a low-permeability layer, we restrict our analysis to solute-driven free convection only but note that these analyses can be extended to both thermal convection and combined thermohaline convection cases.

[3] In a numerical study of convection through large, basin-scale shale layers (assumed to be homogeneous), *Sharp et al.* [2001] showed that convection driven by salinity differences was possible when shale permeabilities are on the order of 10^{-15}–10^{-16} m^2, which is at the upper end of the expected range of values for clays and shales presented by *Neuzil* [1994] in a compilation of both laboratory data and regional modeling studies. Rayleigh stability criteria for the onset of convection in thick shales based on the theoretical work of *Lapwood* [1948] also indicate that free convection can occur in shales ranging in thickness from 10 to 200 m provided that shale permeabilities are near the upper end of the expected permeability range [*Sharp et al.*, 2001]. *Neuzil*'s [1994] data in low-permeability media (e.g., clays and shales) demonstrate that

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permeabilities range across many orders of magnitude. However, free convection analyses based upon homogeneous assumptions suggest that only a very small range of permeabilities at the highest possible end of the observed spectrum permit free convection driven by salinity inversions.

[4] It is appropriate to challenge the assumption of geologic homogeneity made in earlier analyses. Recent studies by *Schincariol et al.* [1997], *Simmons et al.* [2001], *Prasad and Simmons* [2003], *Simmons* [2005], and *Nield and Simmons* [2007] suggest that heterogeneity is critical in controlling the onset, growth, and decay of density-driven instabilities. Several studies have shown that the propensity for free convective transport is strongly influenced by heterogeneities that may otherwise be considered insignificant in regional flow calculations (that assume some average shale layer permeability) or in models of basin evolution. A number of studies have examined free convection processes in the presence of vertical conduits and fractures. *Sharp et al.* [2001] demonstrated numerically that density-driven flow through a shale layer can occur at significantly lower matrix permeabilities where higher-permeability vertical conduits (representing microfractures, fractures, or facies changes) that extended through the shale layer existed. *Ronen et al.* [1995] considered the buoyancy-induced flow of a tracer (both ^{18}O-enriched and ^{18}O-depleted waters) in vertical conduits. They simulated a section of a well with a 10 cm diameter tube and found that buoyancy-induced flow was detected in all experiments for a density difference whose equivalent total dissolved solids contrast was in the range 30–1000 mg L^{-1}. They concluded that buoyancy-induced flow may be an important groundwater transport and mixing mechanism in open conduits in aquifers. These observations are consistent with those of *Love et al.* [2007], who showed that double-diffusive convection induced by salinity and/or temperature variations is a plausible mechanism for heat and solute transport in a groundwater well. For a typical well geometry, it was shown that the geothermal gradient alone is unstable and can be further destabilized by the addition of salt in a destabilizing configuration, i.e., saltier water overlying fresher water. The important finding of these previous studies is that only very small to modest density differences (e.g., driven by salinity inversions on the order of several thousand milligrams per liter but often substantially less or by a typical geothermal gradient) are required for free convection phenomena to occur where vertical conduits exist. Previous studies have also suggested that fractures play an important role in free convection processes. In a numerical study of density-dependent solute transport in discretely fractured geologic media, *Shikaze et al.* [1998] found that fractures with aperture values as small as 25 μm can significantly enhance contaminant migration in the saturated zone relative to the case where the fractures are absent. *Graf and Therrien* [2007] used a numerical model to study dense plume migration in 2-D orthogonal and irregular fracture networks. Simulations in orthogonal networks containing both large- and small-aperture fractures showed that fractures with apertures smaller than 10 μm have no impact on density-driven flow when embedded in a network of larger fractures of aperture equal to 50 μm. Importantly, dispersive mixing in small fractures was not found to play an important role in controlling free convection processes. Most previous modeling studies [e.g., *Graf and Therrien*, 2007] were conducted in a vertical two-dimensional domain consisting of one layer of 3-D porous matrix blocks. In such a vertical slice, fractures are described by 2-D faces, which are essentially 1-D representations of fractures. As noted by *Graf and Therrien* [2007], this constraint in spatial dimensionality implies that previous numerical modeling analyses of free convection in a fracture network generally neglect convection processes within the fractures themselves and do not permit fluid flow in multiple directions within each fracture. This is an important and potentially very significant limitation in free convection analyses because intrafracture convection may be critical.

[5] Analytical stability analyses by *Murphy* [1979] and *Malkovsky and Pek* [1997] have shown that the critical Rayleigh number, Ra_{cr}, for the onset of thermal free convection in a fracture (convective circulation perpendicular to the plane of the fracture) can greatly exceed the critical value $Ra_{cr} = 4\pi^2$, the critical Rayleigh number determined by *Lapwood* [1948] for an infinite porous medium in which upper and lower boundaries are impermeable (for flow) and are of constant concentration (for solute). *Caltagirone* [1982] developed the conditions for the onset of convection for a porous layer bounded laterally by walls that are adiabatic and impermeable and showed that Ra_{cr} depends on the aspect ratios A and B in the two horizontal directions [*Caltagirone*, 1982; *Weatherill et al.*, 2004]. For a two-dimensional porous medium, Ra_{cr} is the minimum value of $4\pi^2$ when A is an integer. In the case of a fracture, we define $A = b/H$, where b is the fracture aperture and H is the fracture height. Usually H is much larger than b, and A approaches zero. For the case of convective flow perpendicular to the fracture plane, *Caltagirone* [1982] showed that Ra_{cr} is several orders of magnitude greater than the critical value $4\pi^2$.

[6] This study investigates all possible modes of free convection in a fractured low-permeability layer: both interfracture convection modes on the larger scale of the low-permeability layer and intrafracture convection modes perpendicular and parallel to the fracture plane. We define each possible convection mode and determine the quantitative conditions required for the onset of convection in each mode on the basis of a Rayleigh stability analysis. The critical onset conditions are compared to assess the likelihood of occurrence of each convective mode. While this study was motivated by a desire to understand transport processes within shales of the Gulf of Mexico Basin, the analysis and results presented here are generally applicable to free convective transport in fractured low-permeability media. Unlike previous analyses, we simultaneously consider all possible modes of convection (interfracture and intrafracture convection) within an idealized unified theoretical approach for convection in fractured low-permeability layers. Earlier analytical analyses have been restricted to a single fracture or box. Previous numerical modeling studies of free convection in a fracture network have not simultaneously considered both intrafracture and interfracture free convection modes. These models have been constrained by a fracture network created within a 2-D modeling framework where only interfracture convection could occur. We draw some general conclusions about the implications of this work

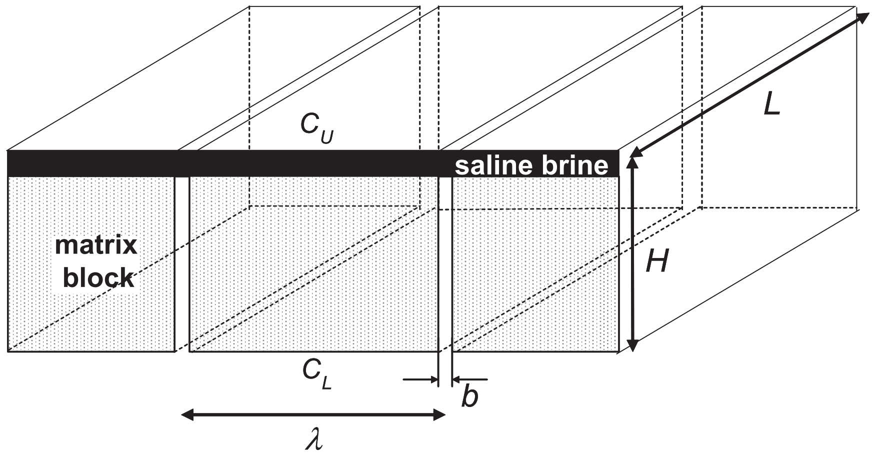

Figure 1. Conceptual model based upon work by *Sharp et al.* [2001] and *Simmons et al.* [2001]. A low-permeability layer of thickness H containing a periodic set of vertical fractures that traverse the vertical layer (e.g., vertically continuous fractures or faults). The lateral spacing between the vertical fractures is λ. The fractures have an aperture b and length L. A density gradient exists across the layer and is created by maintaining a higher concentration C_U on the upper boundary condition and a lower concentration C_L on the lower boundary condition. Between the upper and lower boundary conditions, a linear concentration gradient is assumed to exist across the fractured shale layer in the steady state Rayleigh stability analysis used to determine the onset conditions for free convection. Matrix blocks are impermeable to flow (zero fluid flux at fracture-matrix interface), but fracture-matrix diffusion is allowed.

for the future study of free convection and solute transport processes in fractured low-permeability systems and the numerical modeling of such phenomena.

2. Conceptual Model for Convection in Low-Permeability Shales

[7] In the south Texas portion of the Gulf of Mexico Basin, high-salinity fluids are commonly perched above or just below the top of overpressure within the transition zone [*McKenna and Sharp*, 1997]. The transition zone is where the fluid pressures become higher than hydrostatic pressure and less than about 0.7 of lithostatic pressure. In this region, pressures never reach lithostatic levels because the fracture pressure gradient is about 0.8–0.85 of the lithostatic pressure. There are few salt domes in this area, and data indicate that some of these brines are derived from the deep Mesozoic rocks of the basin. We infer that deep hot saline fluids are episodically expelled vertically upward along near-vertical fault zones. These fault zones may traverse the shale unit and may provide pathways for free convection across a shale layer, i.e., a solute transport mechanism to dissipate inversions once they are formed. It appears that the brines are not commonly found in the zone of extreme overpressures here (although deep salinity data are sparse) unless at depths not yet generally penetrated by drilling. Specifically, we are interested in exploring answers to questions such as the following: Does free convection occur in a low-permeability shale? What hydrogeologic conditions are required for free convection to occur? What are the possible modes of free convection? How likely is each mode of free convection? An understanding of solute transport processes in a fractured low-permeability layer is vital to understanding (amongst other processes) mechanisms of sediment diagenesis, the fate of disposed brines, and the evolution of hydrochemical systems.

3. Analytical Conditions for Free Convection Modes in a Fractured Low-Permeability Layer

[8] A conceptual model of a shale layer is a simplified layer cake stratification, based upon work by *Sharp et al.* [2001], of a sand layer overlying a shale layer, which in turn overlies a sand layer. Shale layers limit fluid flow because they are the lowest-permeability strata. On the basis of a review of earlier literature, *Simmons et al.* [2001] note that salinity inversions exist in the Gulf of Mexico Basin in which more saline waters in formations (including brines of >100,000 ppm total dissolved solids) overlie less saline formation waters (about 20,000 ppm) and present data in support of that conceptual model. The dense fluid overlies a fractured low-permeability shale layer in a sandy high-permeability unit above the shale layer. Free convection is gravity driven (and hence essentially a vertical phenomenon), and thus, vertical fractures are critical in controlling the onset conditions for free convection. A limited number of numerical modeling studies [e.g., *Graf and Therrien*, 2007] have also shown that the smaller-aperture fractures embedded within the network of larger-aperture fractures do not play a significant role in solute transport. We use these observations as the basis for defining the conceptual model employed in this analysis.

[9] We consider a low-permeability shale unit of thickness H containing a periodic set of vertical fractures that traverse the entire depth of the layer (e.g., vertically continuous faults) as shown in Figure 1. The lateral separation between the fractures is λ. The fractures have an aperture size b. In a time-dependent analysis, the initial condition would consist of a brine of higher concentration

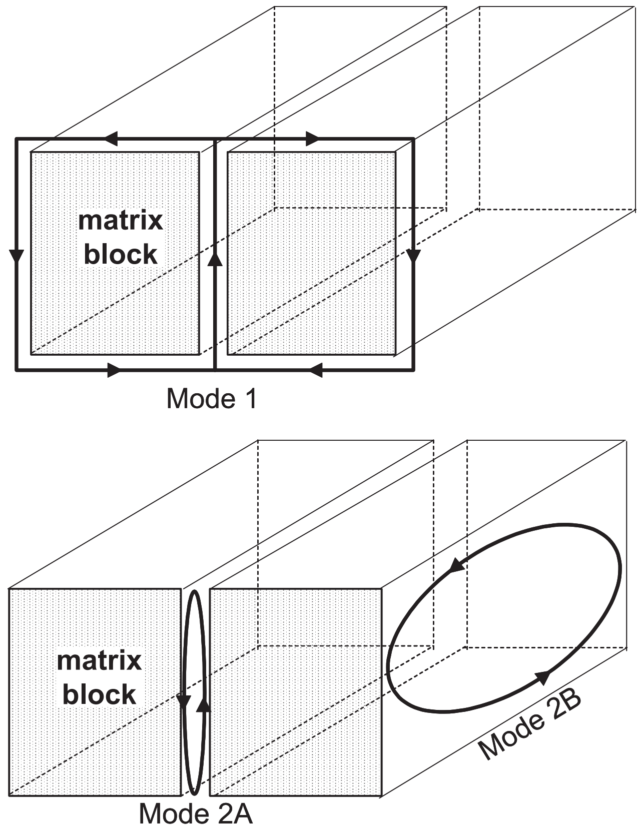

Figure 2. Convection modes in a fractured low-permeability layer: (top) large-scale interfracture convection with a horizontal length scale of order λ (mode 1) and (bottom) intrafracture convection perpendicular to the plane of the fracture (mode 2A) and intrafracture convection parallel to the plane of the fracture (mode 2B).

C_U which lies at the upper surface of the shale layer which would in turn overlie fluid of lower concentration C_L initially contained throughout and beneath the shale layer. However, as is traditional with Rayleigh stability theory approaches for determining the steady onset conditions for free convection, in the steady state analysis presented here a linear concentration gradient is assumed to exist across the fractured shale layer. As would be expected with a very low permeability shale layer, matrix blocks between fractures are assumed to approach zero permeability but not necessarily zero diffusivity. Thus, the fractures provide the only conduits for fluid movement, and the matrix acts to dissipate convection by diffusive exchange which may occur from the fracture to the matrix. This is an important difference from some previous studies [e.g., *Caltagirone*, 1982] which did not include diffusion across the vertical boundaries. A single horizontal layer that contains vertical fractures provides the most fundamental building block for a free convection analysis. It is possible that more complex orthogonal networks of fractures may be conceptualized as being built by stacking multiple layers of varying thicknesses.

[10] Our discussion of stability is made on the following basis. In order to establish the stability of a basic state, it is necessary to show that all disturbances from that state decay with time. In general, this is a complex task. On the other hand, in order to establish the instability of a basic state, it is sufficient to show that at least one disturbance from that state grows with time. The critical value Ra_{cr} of the Rayleigh number Ra is the minimum of Ra over all the possible disturbances. Each disturbance is characterized by a flow pattern. An upper bound on Ra_{cr} is obtained by taking the minimum over a subset of the possible disturbances, for example, those disturbances that are confined to two dimensions rather than three or those disturbances that are zero outside some subdomain of the flow domain. We refer to a subset of disturbance flow fields as a mode of instability.

[11] We consider the critical onset conditions for two possible modes of instability: (1) a large-scale interfracture convection, with a horizontal length scale of order λ (mode 1), and (2) intrafracture convection (mode 2). Figure 2 illustrates the modes of free convection that may occur in a three-dimensional low-permeability fractured layer.

[12] First consider mode 1. From the cubic law, the permeability of a fracture of aperture b is $b^2/12$. The average permeability of the medium is approximately

$$K_{\text{av}} = \frac{b^3}{12\lambda}. \tag{1}$$

[13] An effective solute Rayleigh-Darcy number for the medium is

$$Ra_D = \frac{gK_{\text{av}}H\beta\Delta C}{D_{\text{av}}\nu}, \tag{2}$$

where g is the gravitational acceleration, H is the layer height, $\beta = \rho_0^{-1}(\partial\rho/\partial C)$ is the coefficient of density variability, ΔC is the concentration difference between C_U and C_L, D_{av} is the average solute diffusivity, and ν is the fluid kinematic viscosity. The standard Horton-Rogers-Lapwood theory gives the critical value $4\pi^2$ for Ra_D. Hence an estimate of the critical concentration difference for mode 1 convection is

$$\Delta C_{\text{crit1}} = \frac{48\pi^2 D_{\text{av}}\nu\lambda}{g\beta b^3 H}. \tag{3}$$

[14] In mode 2, we apply the theory given in section 12 and, in particular, equation (12.21) of *Gershuni and Zhukhovitskii* [1976]. Their results are given in terms of a Rayleigh number based on fluid properties, the vertical gradient, and the half width of a plane vertical layer, so that here

$$Ra = \frac{g(b/2)^4\beta(\Delta C/H)}{D_{\text{av}}\nu}. \tag{4}$$

[15] It is appropriate to consider, in turn, the cases of convection perpendicular to the fracture plane (mode 2A) and convection parallel to the fracture plane (mode 2B).

[16] For the case of convection perpendicular to the fracture plane (mode 2A) the critical value $Ra_{\text{cr}} = \pi^4$ for the case of boundaries at constant concentration, and $Ra_{\text{cr}} = (2.365)^4 = 31.28$ for the case of zero fluid flux boundaries. In our case the latter boundary conditions are more appro-

priate because fluid flux across the vertical fracture–matrix boundary will be zero for an impermeable matrix block, but the vertical boundary will not necessarily be maintained at a constant concentration. These results lead to an estimate for the critical concentration difference for the onset of mode 2A convection

$$\Delta C_{\mathrm{crit}2A} = \frac{500.5 D_{\mathrm{av}} \nu H}{g \beta b^4}. \tag{5}$$

[17] The ratio of the two critical concentration differences is

$$\frac{\Delta C_{\mathrm{crit}1}}{\Delta C_{\mathrm{crit}2A}} = O\left(\lambda b / H^2\right). \tag{6}$$

[18] In the Landau order notation, we say $f(z) = O[g(z)]$ in some domain D if there exists a positive constant K such that $|f| \leq K|g|$ for all z in D. Thus, roughly speaking, f has the same order of magnitude as g. The ratio in equation (6) is expected to be small compared to unity. We conclude that mode 1 is more unstable than mode 2A, and thus the latter is generally of lesser importance.

[19] However, the situation is dramatically different for the case of convection parallel to the fracture plane (mode 2B). In this case, and for the subcase of boundaries at constant concentration, Ra_{cr} is given by

$$Ra_{\mathrm{cr}} = \left(\frac{\pi^2}{4} + \alpha^2\right)^2, \tag{7}$$

where α is the wave number in the horizontal direction parallel to the fracture boundary planes. Equation (7) is a monotonically increasing function of α and so attains its minimum as α tends to zero, the minimum value being $\pi^4/16$.

[20] On the other hand, for the subcase of constant flux (and in particular zero fluid flux) boundaries, Ra_{cr} is still attained as α tends to zero, but the minimum value is zero. In fact, for small values of α,

$$Ra_{\mathrm{cr}} = O\left(3\alpha^2\right). \tag{8}$$

[21] Thus the overall critical value can be arbitrarily small, being limited only by the horizontal extent of the fractures:

$$\Delta C_{\mathrm{crit}2B} = \left(\frac{16 D_{\mathrm{av}} \nu H}{g \beta b^4}\right) \alpha^2. \tag{9}$$

[22] For example, if the fractures extend a distance L, then one has $\alpha = 2\pi/(2L/b) = \pi b/L$, and so

$$\Delta C_{\mathrm{crit}2B} = \frac{16 \pi^2 D_{\mathrm{av}} \nu H}{g \beta b^2 L^2}. \tag{10}$$

[23] The ratio of the two critical concentration differences for mode 1 and mode 2B convection is

$$\frac{\Delta C_{\mathrm{crit}1}}{\Delta C_{\mathrm{crit}2B}} = O\left(L^2 / b^2\right). \tag{11}$$

[24] One would expect that this expression would be normally considerably greater than unity. Then the overall conclusion is that mode 2B (with a typical streamline lying in a plane parallel to the fracture plane boundaries) is the favored mode of convection. Hence, the appropriate estimate for the critical concentration difference is that given by equation (10) for the mode of convection most favored amongst possible intrafracture and interfracture convection modes.

4. Discussion and Conclusion

[25] It is apparent that the most likely mode of convection in a low-permeability layer is mode 2B, with free convection occurring parallel to the plane of the fracture. The critical concentration difference required for the onset of convection in each mode determines the likelihood of each mode occurring in realistic hydrogeologic settings. If we assume a matrix porosity of ε = 0.05 and an aqueous diffusion coefficient of $D = 10^{-9}$ m^2 s^{-1}, then the effective matrix diffusion coefficient (a reasonable approximation for the average solute diffusivity) is $D_{\mathrm{av}} = 5 \times 10^{-11}$ m^2 s^{-1}. Other relevant parameters are $\beta = 0.7$ and $\nu = 10^{-6}$ m^2 s^{-1}. In the following paragraph, we define minimum and maximum bounds on fracture aperture, fracture spacing, fracture length, and shale layer thickness to estimate the range of critical concentration differences that would be required for convection to occur in each mode. The parameters chosen here are intended to be illustrative and representative of those which may be encountered more generally in fractured rock hydrogeology rather than those which may specifically exist in the Gulf of Mexico Basin. The present simple and idealized analysis is intended to be generalized to low-permeability media rather than limited only to the specific case of the Gulf of Mexico Basin. The intention here is to demonstrate using reasonable parameters in a simplified analysis that free convection may be plausible in an otherwise low-permeability layer when suitable combinations of salinity differences, shale layer thickness, and fracture properties exist. These findings are therefore applicable to, but not limited to, the Gulf of Mexico case study.

[26] The ranges for these demonstrative and representative parameters are chosen as follows:

Fracture aperture

$$b_{\min} = 10^{-6}\ \mathrm{m}; \quad b_{\max} = 10^{-3}\ \mathrm{m}$$

Fracture spacing

$$\lambda_{\min} = 0.1\ \mathrm{m}; \quad \lambda_{\max} = 100\ \mathrm{m}$$

Fracture length

$$L_{\min} = 1\ \mathrm{m}; \quad L_{\max} = 100\ \mathrm{m}$$

Shale layer thickness

$$H_{\min} = 1\ \mathrm{m}; \quad H_{\max} = 100\ \mathrm{m}$$

[27] We begin by considering the dominant convection mode 2B. The critical concentration difference required for convection can be calculated using equation (10) for a large range of possible choices for the fracture geometry factor for mode 2B convection as $f_{\mathrm{mode}2B} = H/(b^2 L^2)$. Using the range of parameters stated above for shale layer thickness, fracture aperture, and fracture length, we find that fracture geometry factors f range between 10^2 and 10^{14} m^{-3}. Substitution into equation (10) gives critical dimensionless concentration differences required for convection in the approximate range 10^{-13}–10^{-1}. At the lowest end of the range, this is an extremely small concentration difference and suggests that convection would be expected to occur parallel to the fracture for almost any nonzero concentration difference. At the highest end of the range, this is equivalent to a salinity of 100,000 ppm, which is a realistic concentration difference, especially in the Gulf of Mexico setting. On this basis, we can conclude that mode 2B convection is likely to occur in most hydrogeologic settings. We do not have to "beg the data" for this mode of free convection to occur.

[28] The calculation of critical concentration differences for mode 1 and mode 2A convections may seem somewhat superfluous given the ease by which mode 2B convection is expected to occur and the fact that it is expected to be the dominant mode. However, it is useful to examine under what conditions other convective modes can occur and their associated likelihood. We calculate the concentration difference required for mode 1 convection. We now define a new fracture geometry factor for mode 1 convection as $f_{\mathrm{mode}1} = \lambda/(b^3 H)$. Using the range of parameters stated above for fracture separation, fracture aperture, and shale layer thickness, we find that fracture geometry factors f range between 10^6 and 10^{20} m^{-3}. Equation (3) then gives critical dimensionless concentration differences required for convection in the approximate range 10^{-9}–10^5. At the lowest end of the range, this is an extremely small concentration difference and suggests that interfracture convection would be expected to occur on the layer scale for almost any nonzero concentration difference. At the highest end of this range, the required concentration differences are not possible. Concentration differences in the range 0.3–0.5 represent the upper limit of those that are physically allowed. This is because the precipitation of many salts (such as chlorides, carbonates, and sulphates) occurs at or below this stated concentration range of 0.3–0.5. As a result, a substantial range of the required concentration differences for free convection is not realistic. Compared with mode 2B convection, the range of physically permissible concentration differences is substantially smaller for mode 1 convection. This result suggests that convection in mode 1 is likely in many (but not all) situations. When it occurs, it is likely to occur in combination with mode 2B convection.

[29] Finally, we calculate the concentration difference required for mode 2A convection. We define a fracture geometry factor for mode 2A convection as $f_{\mathrm{mode}2A} = H/b^4$. Using the range of parameters stated above for shale layer thickness and fracture aperture, we find that fracture geometry factors f range between 10^{12} and 10^{26} m^{-3}. Equation (5) then gives critical dimensionless concentration differences required for convection in the approximate range 10^{-3}–10^{11}. At the lowest end of this range, a very modest concentration difference of 1000 ppm is required for the onset of convection. This is a very small concentration difference and would additionally require, for example, that the low-permeability layer contain fractures with apertures of about 1 mm in a layer of about 1 m thickness. Low-permeability layers can be substantially thicker, and fracture apertures are often thinner, and free convection in this mode would require concentration differences far greater than that which is physically possible in many hydrogeologic systems. Compared with mode 2B and mode 1 convection, the range of physically permissible concentration differences is substantially smaller for mode 2A convection and suggests that mode 2A convection is the least likely mode to occur.

[30] The major outcome of this paper is a summary of plausible arguments for the likelihood of free convection, potentially occurring in even low-permeability sediments such as those in the Gulf of Mexico Basin. Although completely based on theoretical evaluations and assessments regarding criteria for the onset of convection by using known analytical relationships of critical Rayleigh numbers, the paper offers a number of theoretical findings. The important generalized finding of this study is that all modes of free convection (parallel to the fracture plane, perpendicular to the fracture plane, and convection between fractures on the larger layer scale) are theoretically plausible for reasonable hydrogeologic parameters but that the dominant (and most likely) mode of convection is expected to be intrafracture convection parallel to the fracture plane (mode 2B). The least likely mode is intrafracture convection perpendicular to the fracture (mode 2A). The results suggest that episodes of density-driven flow may not be uncommon in the thick shale sequences, such as in the Gulf of Mexico Basin. While *Sharp et al.* [2001] showed that convection driven by salinity differences was possible when shale permeabilities are near the upper end of the expected range of values, these new results suggest that density-driven flow may be significantly more widespread because of the presence of fractures in the shale layers and the ease by which mode 2B convection may be expected to occur. Shales with matrix permeabilities at the lower end of *Neuzil*'s [1994] range originally thought not to permit convection may contain the fracture aperture and separation spatial scales required for convection to be a plausible fluid flow and solute transport mechanism. Intrafracture convection parallel to fracture planes is likely to permit free convection at low concentration differences. The propensity for density-driven convection is strongly influenced by permeability heterogeneities that may otherwise be insignificant in regional flow calculations or in models of basin evolution. This analysis provides conditions for the onset of convection only; further analyses are required to examine the temporal patterns of the growth and/or decay of instabilities associated with free convection once they are established. The density contrast is ideally assumed to be stable over very long geologic time. Eventually, the convection processes will be very slow, and other dynamic processes will dominate them in the Gulf of Mexico Basin. However, this transient effect is also expected to depend upon other largely unknown factors such as the frequency and magnitude of episodic expulsion of saline brines from depth. The important consequence of these findings is that free convection is expected to be pervasive (at least at some

point in time) when subvertical/vertical fracturing is present in a low-permeability layer. Such fracturing is expected to eventually dissipate the salinity inversions over timescales that are presently unknown. Similarly, if a salinity inversion is observed above or across a low-permeability layer, it is likely to be either a short-lived (transient) phenomenon on geologic timescales or a saline fluid that lies above a low-permeability layer that does not contain significant vertical fracturing. These findings are important in understanding both salinity inversion data and the possible solute transport processes that may occur in low-permeability shales such as in the Gulf of Mexico Basin. Clearly, more work is required to understand the transient persistence of such phenomena as well as to more intimately and explicitly connect these phenomena with the Gulf of Mexico case study.

[31] Some general remarks can also be made about the numerical simulation of free convection in fractured low-permeability media. Our results suggest that analyses not conducted in three dimensions underestimate the likelihood of the occurrence of convection. The simulation of variable density flow phenomena in 2-D fracture networks [e.g., *Shikaze et al.*, 1998; *Graf and Therrien*, 2007] does not capture the dominant mode of convection expected to occur in the fractured rock system (i.e., parallel to the fracture planes themselves) since only the interfracture convection mode is usually considered in that network-modeling framework. This is an important finding that does not appear to have been stated explicitly in previous published numerical modeling literature. It was, however, demonstrated numerically by *Shi* [2005], who observed that when free convection initiates, convection cells occur on the fracture plane with axes parallel (normal) to the fracture plane. *Shi* [2005] concluded that two-dimensional numerical models of flow and transport normal to fracture strike are unable to capture the most important pattern of convective motion. This observation is consistent with the results of our present analysis.

[32] It is recognized that the explicit simultaneous simulation of free convective transport through a network of fractures, and within the fractures themselves, is computationally demanding for a fractured rock flow and transport numerical model. Although the results of this study are suggestive of the need for a fully 3-D schematization to model recirculating convection modes, this is not always necessary and may represent an overkill for a number of cases. It is therefore important to clearly discuss the consequences of modeling free convective phenomena in these systems with reduced spatial dimensions. Cellular convection can be modeled in two dimensions if the vertical fracture is discretized by 2-D fracture elements and the diffusive flux from the matrix is handled by a source condition in the direction normal to the fracture plane. This has previously been preferred in modeling hot dry rock recirculation currents in single fractures [e.g., *Kolditz and Diersch*, 1993] and can be analogously adapted to salinity-driven free convection in fractures. Indeed, a 2-D modeling approach similar to that presented by *Kolditz and Diersch* [1993] would be extremely useful for analyzing intrafracture convection (mode 2B). Furthermore, a network of fractures discretized by 1-D fracture elements in a 2-D vertical cross section would also be able to model the interfracture convection (mode 1). It is clear that only the combination of all free convection modes would require a full 3-D modeling approach. As an intermediate step, 2-D models with 2-D and/or 1-D fracture elements appear to be preferable for analyzing the most favored convection modes before a fully 3-D model is constructed. We therefore emphasize that the role of 2-D models should not be underestimated. This is especially important since intra-fracture convection parallel to the fracture plane (mode 2B) has been shown in this analysis to be the most likely convection mode and is only a 2-D process.

[33] Previous analyses by *Simmons et al.* [2001], *Simmons* [2005], and *Nield and Simmons* [2007] have suggested that heterogeneity in geologic properties is critical in controlling the onset, growth, and/or decay of free convection in a hydrogeologic setting. A particular controlling feature of that geologic heterogeneity is its structure and interconnectedness. Future work is required to examine the spatiotemporal patterns of dense plume migration through significantly more complex and realistic 3-D fracture networks. It is entirely plausible that heterogeneity at another level (for example, larger-scale variations in shale permeability and variability in fracture spacing, aperture, orientation, and connectedness) will be vital in controlling whether or not instabilities will occur in field-based settings and how persistent they may be over larger spatial scales and longer temporal scales. These are largely unresolved matters in current literature. These previous theoretical findings suggest that there is a strong interplay between geological heterogeneity and free convective phenomena that means that free convective processes are not easily amenable to prediction. This also suggests that the analysis presented here may best be used as a semiquantitative guide to which modes of free convection may be expected, a priori, to be dominant. What is critical here is that we demonstrate using analytical relations and plausible parameters for a fractured shale system that free convection is a theoretically possible solute transport process in thick-shale sequences and that the dominant and most likely mode of free convection is parallel to vertical fracture planes.

[34] As discussed previously by *Simmons* [2005], a critical challenge that remains for this field of research is that there are limited observations of unstable dense plume phenomena and free convection in field-scale settings. *Simmons* [2005] notes that research papers often quote secondary evidence for their existence (e.g., the salt deficit in the salt lake may be accounted for by the slow downward convection of dense water; numerical experiments demonstrate the existence of a convection cell; abundant data indicate high fluid fluxes consistent with density-driven flow; and the system Rayleigh number is greater than the critical value required and, therefore, convection is assumed to exist) rather than direct or primary evidence of the free convective processes themselves. We are acutely aware that a major challenge remains to verify theoretical findings such as those presented here but also more broadly throughout free convection research literature, in field-based settings. This is vital to confirming or refuting the free convective hypothesis in hydrogeologic settings. Underpinned by several decades of theoretical and modeling analyses, the free convection hypothesis has a solid theoretical basis but to date has been neither explicitly nor completely observed through robust data collection in hydrogeologic settings

using primary hydrogeologic evidence of groundwater flow and salinity (i.e., convective circulations by measurement of flow directions and rates, temporal measurements of descending dense plumes, or mapping of spatial fingering patterns by detailed measurements of groundwater salinity). Nonetheless, simple and idealized analyses based upon theoretical Rayleigh stability criteria such as those presented in this paper may provide useful guidance for both future numerical modeling and field-based experimentation.

Notation

- b fracture aperture (L).
- C fluid concentration expressed as a mass fraction (M_S M^{-1}).
- C_U maximum value of concentration expressed as solute weight relative to weight of solution (M_S M^{-1}).
- C_L minimum value of concentration expressed as solute weight relative to weight of solution (M_S M^{-1}).
- D apparent molecular diffusivity of solutes in solution (L^2 T^{-1}).
- D_{av} average solute diffusivity (L^2 T^{-1}).
- f fracture geometry factor (L^{-3}).
- g acceleration due to gravity (L T^{-2}).
- H shale layer thickness (L).
- K_{av} average intrinsic permeability (L^2).
- L fracture length (L).
- Ra Rayleigh number of the system (dimensionless).
- β coefficient of density variability, $\rho_0^{-1}(\partial\rho/\partial C)$ (dimensionless).
- ΔC concentration difference between C_U and C_L (M_S M^{-1}).
- α wave number in the horizontal direction parallel to the fracture boundary plane.
- ε porosity of matrix block (dimensionless).
- λ wavelength (spacing) of the periodic fracture set (L).
- ρ fluid density (M L^{-3}).
- ν kinematic viscosity of the fluid (L^2 T^{-1}).

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