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Key Points:

- There is insufficient topography to drive connate brines from many sedimentary basins
- The ratio of topographic relief to basin depth predicts the presence of stagnant brines
- Basins that do not contain brines either have high topographic gradients, shallow extents, or never contained brines

Supporting Information:

- Supporting Information S1

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The Persistence of Brines in Sedimentary Basins

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Abstract Brines are commonly found at depth in sedimentary basins. Many of these brines are known to be connate waters that have persisted since the early Paleozoic Era. Yet questions remain about their distribution and mechanisms for retention at depth in the Earth's crust. Here we demonstrate that there is insufficient topography to drive these dense fluids from the bottom of deep sedimentary basins. Our assessment based on driving force ratio indicates that sedimentary basins with driving force ratio > 1 contain connate waters and frequently host large evaporite deposits. These stagnant conditions appear to be relatively stable over geological time and insensitive to factors such as glaciations, erosion, compaction, and hydrocarbon generation.

Plain Language Summary Brines that are millions of years old are present at the bottom of many sedimentary basins. Regional groundwater flow should have flushed these brines out of these systems if not for the presence of some trapping mechanism. Here we demonstrate that there is insufficient topography to drive dense brines out of the bottom of many deep sedimentary basins. We provide geochemical evidence that brines within these basins originated as ancient seawater that was trapped during deposition of the sediments in these basins. Our findings may have applications in determining where stagnant groundwaters are present in sedimentary basins.

1. Introduction

Salinity tends to increase with depth in sedimentary basins, with dense brines present in the deepest parts of many sedimentary basins (Dickey, 1969). In most cases, these waters are relict seawater trapped since sediment deposition (i.e., connate waters). The persistence of these brines in the presence of regional groundwater flow systems has been the subject of some debate (Bethke & Marshak, 1990; Hanor, 1994; Kreitler, 1989). The median global groundwater turnover since the last glacial maximum is 5 ± 3 times, but deeper parts of flow systems turnover less quickly (Befus et al., 2017); for example, most groundwaters stored more than 100–400 m below the land surface tend to be more than 10,000 years old (Jasechko et al., 2017). Numerical simulations have suggested that flushing of large sedimentary basins requires timescales on the order of several millions of years (Deming & Nunn, 1991; Ranganathan, 1993). However, data suggest that dense brines can be stagnant over even longer timescales. Multiple lines of evidence, including Na:Cl, Cl:Br, stable isotopes of the water molecule, and radiogenic noble gases, demonstrate that brines are tens to hundreds of millions years old in numerous basins (Carpenter, 1978; Darrah et al., 2015; Hanor, 1994; Ma et al., 2009). Evaporite dissolution has been invoked as a mechanism to maintain high salinities during flushing in some environments (Grasby & Chen, 2005; Gupta et al., 2012; McIntosh et al., 2011). However, dissolution rates are insufficient to explain the persistence of high salinity brines in the presence of regional flow (Ranganathan, 1993). Instead, variations in fluid density may contribute to the presence of stagnant brines in the deepest areas of sedimentary basins (Bachu & Hitchon, 1996; Palombi, 2008). Elucidating such controls on brine stagnation over geologic timescales thus becomes a factor in predicting long-term stability of waste material stored in sedimentary basins. Here we show that the driving force ratio (*DFR*) is an effective means for first-order evaluation of the persistence of connate brines in basins spanning across North America.

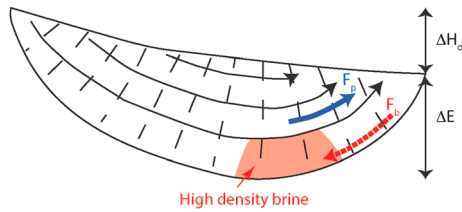


Figure 1. Conceptual model of high density brine trapped where buoyancy forces (F_b) exceed topographic driving forces (F_p) due to lack of ∇H relative to ∇E . The dashed black lines represent freshwater equipotentials, and the solid black lines represent flowpaths for a freshwater system.

where g is acceleration due to gravity; ρ is fluid density; $\Delta\rho$ is the difference between fluid density and reference density; ρ_o is reference density; H_o is equivalent freshwater head; E is the topography of the aquifer, which we also refer to as the structural gradient; F_p is the force due to pressure and topographic differences; and F_b is the force due to buoyancy differences (Figure 1). Bachu (1995) defines H_o as

$$H_o = \frac{P}{\rho_o g} + z \quad (2)$$

where P is fluid pressure and z is elevation. Where aquifers have sufficiently high dip angles or low hydraulic gradients, F_b can exceed F_p , causing brines to flow downdip against equivalent freshwater head gradients. To determine when downdip flow of dense brines is possible, Bachu (1995) defined DFR as

$$\text{DFR} = \left(\frac{\Delta\rho}{\rho_o} \right) \frac{|\nabla E|}{|\nabla H_o|_h} \quad (3)$$

At $\text{DFR} > 0.5$, buoyancy effects are important in determining groundwater flow patterns (Bachu, 1995; Davies, 1987). In situations where fluid flow is driven by topography, downdip flow driven by negative buoyancy will dominate where $\nabla H_o < \frac{\Delta\rho}{\rho_o} \nabla E$ (i.e., $\text{DFR} > 1$).

3. Methods

DFR was calculated for 31 sedimentary basins in North America that had at least 20 water chemistry samples available in the U.S. Geological Survey Produced Water Database (Blondes et al., 2014) or documented water chemistry from another source for basins located primarily within Canada (Connolly et al., 1990; Grasby et al., 2012). Basin dimensions were estimated using Tellus sedimentary basin outlines (CGG Geoconsulting Robertson, 2016), thickness of sediment cover from CRUST1.0 (Laske et al., 2013), and topography from GMTED2010 (Danielson & Gesch, 2011). The maximum topographic gradient across the basin is used to estimate the hydraulic gradient (∇H), assuming that regional groundwater flow is driven by topography. The difference between the minimum surface topography and the lowest elevation of the sedimentary basin is used to calculate the minimum structural gradient (∇E) of the basin. Both calculations are assumed to apply over the widest dimension of the basin. The use of the same width to estimate both structural and hydraulic gradients results in a simplified DFR that includes the ratio of maximum structural drop (ΔE) to maximum topographic drop (ΔH) of a basin:

$$\text{DFR} = \left(\frac{\Delta\rho}{\rho_o} \right) \frac{|\Delta E|}{|\Delta H_o|_h} \quad (4)$$

This approach will underestimate ∇E in most cases, particularly in the case of sag basins where the maximum depth is near the center of the basin. DFR could be underestimated by a factor of 2 for sag basins where the maximum structural gradient occurs over half the length of the topographic gradient, but the maximum and minimum topographic elevations exist at the edges of the basin. Overestimation of ∇H is probable for many basins. The ground surface can serve as a rough approximation of the water table in flatter, humid basins but the relationship between topography and water table position is weaker in areas where greater topographic relief or low groundwater recharge rates are present (Gleeson et al., 2011). In

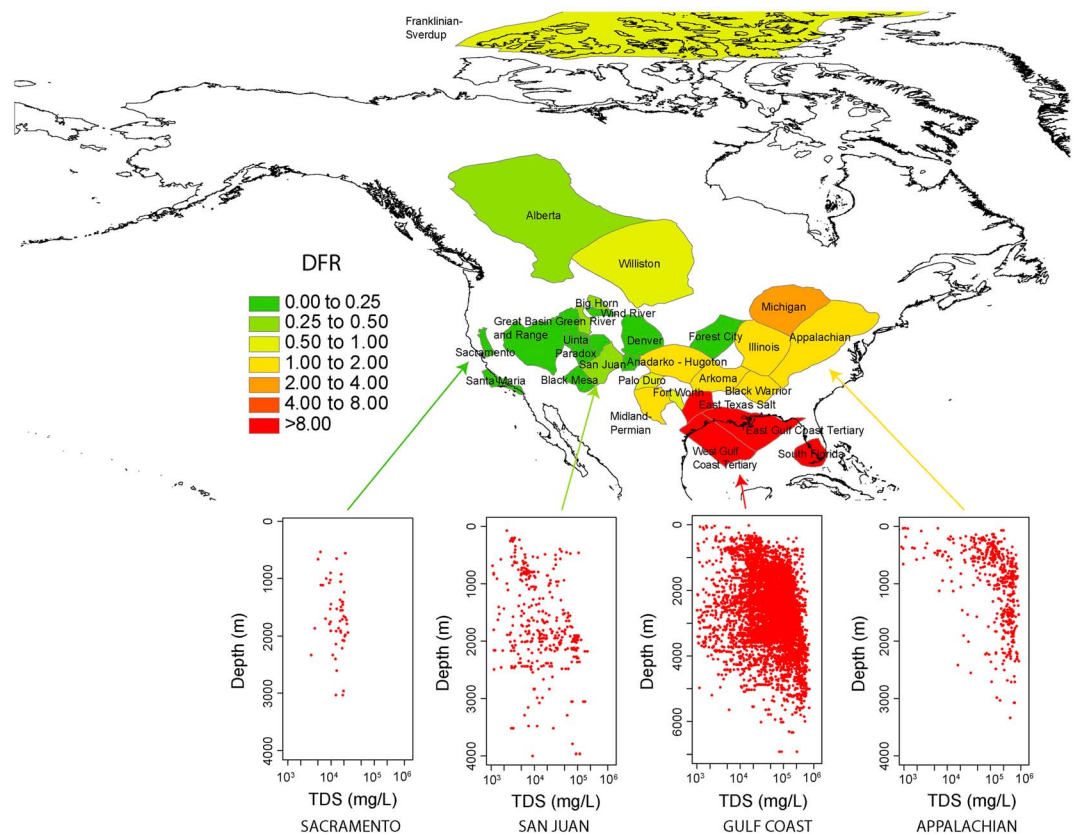


Figure 2. DFR for selected sedimentary basins in North America. Sedimentary basins in western North America are more likely to have been flushed and have lower salinities, similar to those observed in the Sacramento and San Juan basins. Basins in eastern North America tend to have higher DFR are likely to host stagnant brines and have TDS distributions similar to those found in the Gulf Coast and Appalachian basins.

other instances, areas of higher elevations may form local fault-bound systems that are separate from basin-scale systems (Grasby & Hutcheon, 2001). The net result of the underestimation of ∇E and overestimation of ∇H will be underpredicted DFRs. Changes in basin geometry and boundary conditions resulting from deposition, erosion, and glaciations may also affect the DFR value over time.

Density values were estimated from maximum total dissolved solid (TDS) values obtained from the U.S. Geological Survey Produced Water Database (Blondes et al., 2014) as well as additional studies in Canada (Connolly et al., 1990; Grasby et al., 2012). These TDS values were used to estimate fluid density with the following equation (Collins, 1987):

$$\rho = 1 + \text{TDS} \times 6.95 \times 10^{-7} \quad (5)$$

where ρ is in kg/L and TDS is in mg/L. Other more complex equations for estimating brine density that account for temperature and pressure effects exist (Adams & Bachu, 2002). However, the temperatures, pressures, and presence of Ca-Cl brines in many of the deeper basins of the study are beyond the range of applicability for these equations. The temperature increases with depth are associated with decreases in fluid density, while pressure increases cause increases in fluid density. These effects tend to counteract each other, leaving TDS as the dominant control on fluid density. Additional DFR values based on densities estimated with the relationship provided by Batzle and Wang (1992) are presented in Table S1.

4. Trapped Brines in North American Sedimentary Basins

We analyzed a selection of North American sedimentary basins to determine the relationship between basin geometry and water chemistry. Of the 31 sedimentary basins considered, conditions necessary to trap high-

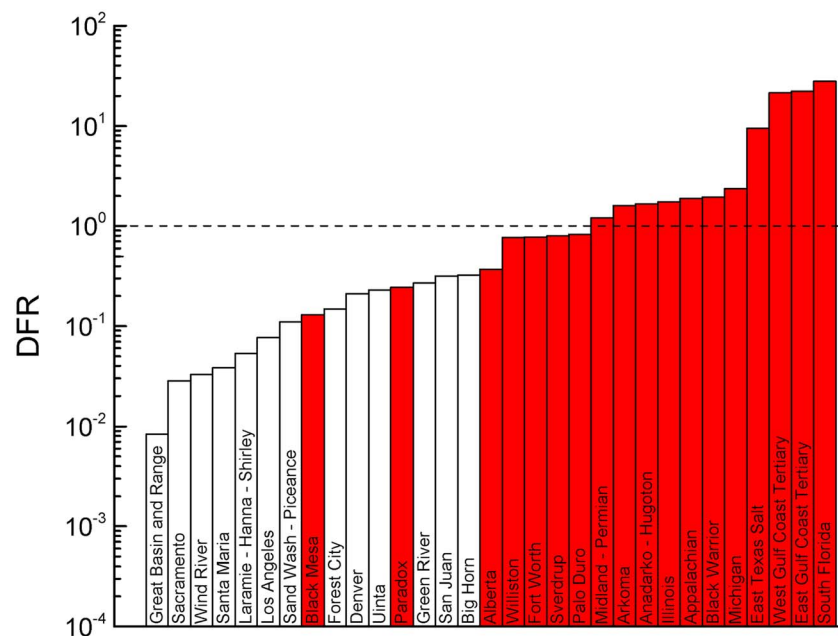


Figure 3. DFR for selected basins in North America. The red bars indicate basins where brines have been characterized as connate water based on Na:Cl, Cl:Br, or deuterium excess in the U.S. Geological Survey Produced Waters Database (all basins in the United States; see the supporting information for Na:Cl) or other studies (Connolly et al., 1990, for Alberta Basin and Grasby et al., 2012, for Sverdrup Basin).

density fluids in their deepest extents were present in many sedimentary basins in North America (Figure 2). DFR is >1.0 in 11 of the 31 sedimentary basins examined (Figure 3). These basins all have depths exceeding 3,000 m and topographic relief of less than 1,500 m and include some basins with offshore components. All

basins with $DFR > 1$ contain brines with TDSs $> 100,000$ mg/L and have structural gradients at least 4 times greater than hydraulic gradients; the presence of connate brines (discussed below) in basins with high DFR values (Figure 3) further supports our main finding: deep basins with low topographic relief contain stagnant brines. Calculations using an alternate equation for estimating fluid density resulted in similar results (Table S1).

Nearly all basins with $DFR > 0.37$ are sag basins, foreland basins, or passive margins (Table S1) that contain sediments deposited in marine environments. The presence of connate water in these basins was also assessed using the presence of evaporites, Ca:Cl, Na:Cl, $\delta^2\text{H}$, and $\delta^{18}\text{O}$ (Blondes et al., 2014; Connolly et al., 1990; Grasby et al., 2012). Evaporites, in particular extensive amounts of halite (NaCl), were found in most basins with $DFR > 1.0$ (Figure 4). Ca-Na-Cl type brines, which are typically associated with precipitation of halite, were found in the Alberta, Michigan, Williston, East Texas, and Gulf Coast basins, indicating that waters have been present since the formation of evaporites in those basins. Brines with Na:Cl molar ratios less than 1 (equivalent to seawater ratios) are typically interpreted as connate waters, whereas brines with Na:Cl molar ratios equal to 1 arise from the dissolution of halite (Carpenter, 1978). Basins with low Na:Cl brines include the Anadarko, Appalachian, Arkoma, Black Mesa, Black Warrior, Fort Worth, Illinois, Palo Duro, Paradox, Permian, South Florida, Mississippi Embayment, and Sverdrup basins (see the supporting information), plus the aforementioned basins with Ca-Na-Cl brines. These basins all have

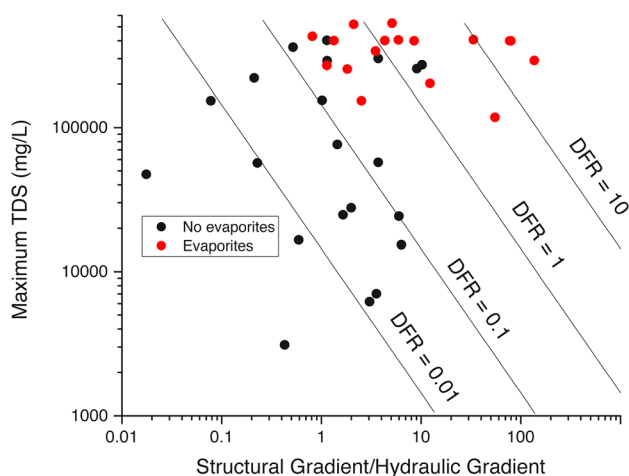


Figure 4. Relationship between DFR, maximum documented salinity, and the ratio of the structural gradient to the topographic gradient for selected North American sedimentary basins. High DFRs tend to occur where connate water and evaporites are present. In particular, halite beds, which have been documented in the Alberta Basin (Connolly et al., 1990), Palo Duro Basin (Eastoe et al., 1999), Williston Basin (Wittrup & Kyser, 1990), Anadarko Basin (Perry, 1989), East Texas (Jackson & Seni, 1983), Michigan Basin (McIntosh et al., 2011), Paradox Basin (Hite, 1968), Gulf Coast Basin, Permian Basin, Appalachian Basin (Smosna & Patchen, 1978), and South Florida (Pierce & Rich, 1962), are associated with high DFRs.

DFR > 0.37 with the exceptions of the Paradox Basin (DFR = 0.24) and Black Mesa Basin (DFR = 0.13). The Paradox Basin contains thick bedded salt formations with associated connate brines at depth (Hanshaw & Hill, 1969). The evidence for connate water in the Black Mesa basin is less extensive. Salinities are lower and Na:Cl ratios > 1.0 exist for some deeper samples, and evaporites are present in some areas (Lessentine, 1965). Noble gas analyses in the Appalachian Basin (Darrah et al., 2015) and Michigan Basin (Ma et al., 2009) also indicated that connate water is present.

The presence of connate water in sedimentary basins with DFR < 1 can be explained by some of the simplifications we made in our large-scale analysis. While we estimate a DFR of 0.37 for the Alberta Basin, Bachu (1995) provides estimates of DFR > 2 exist locally. Our regional estimates likely overestimate the topographic drop driving regional flow in deeper parts of the Alberta Basin by including areas higher in the Rocky Mountains that contain local fault-bound systems (Ferguson & Grasby, 2011; Grasby & Hutcheon, 2001). If recharge to the regional flow system occurs in the front ranges and foothills at lower elevations, DFR will approach 1 for the basin. In other cases, correcting the structural gradient to account for the position of the basin's depocenter can explain the presence connate waters where we estimate DFR < 1. Applying such a correction to the Williston Basin results in DFR ~ 1.5. It is not obvious what corrections would be required to arrive DFR > 1 for the Black Mesa and Paradox basins although this could be related to the structural complexity of these wrench basins or low recharge rates that create deep water tables that are disconnected from topography.

Basins with DFR < 0.37 generally have lower salinity than high DFR basins. Low DFR sedimentary basins have a range of depths and topographic gradients but tended to be located in areas with greater topographic relief (>1,500 m within a given basin) in western North America. Most of these basins have been classified as wrench basins (CGG Geoconsulting Robertson, 2016; Table S1) that never contained high salinity waters (Kingston et al., 1983). In cases where these basins contained marine environments, they are typically dominated by Cretaceous and younger sediments, which contained water with salinity similar to modern seawater (Petersen et al., 2016). Ten basins have structural gradients < 1.0, which would require unrealistically high fluid densities of 2,000 kg/m³ to create a situation where buoyancy-driven downdip flow would dominate over topography-driven flow. In cases where Paleozoic sediments are present, such as the Denver, San Juan, and Big Horn basins, waters have TDS exceeding 100,000 mg/L ($\rho \sim 1,070$ kg/m³) in some areas and variable chemistries with Na:Cl > 1.0. Brackish water (TDS between 1,000 and 10,000 mg/L) is found at depths of up to 1,000 m over much of western North America, while brackish water only extends to depths of ~500 m in the midcontinent (Stanton et al., 2017). Deep fresh groundwater has been found at depths of up to 3,000 m in Central Valley of California (Kang & Jackson, 2016) and at a depth of over 4,500 m in the Powder River Basin (Lynds, 2013).

DFR would have varied over time. Basins have been affected by uplift, erosion, deposition, sediment compaction, and hydrocarbon generation. For example, regional hydraulic gradients in basins in western North America would have experienced higher topographic gradients during the Cretaceous Period, prior to extensive erosion in the region (Garven, 1995; Gupta et al., 2012). However, connate brines persist in the Alberta Basin. Similarly, hydraulic gradients in the Gulf Coast are significantly affected by compaction of recently deposited sediments (Hart et al., 1995; Mello et al., 1994). Some basins would have been affected by Pleistocene continental glaciations (Person et al., 2007). The Williston, Alberta, Michigan, and Appalachian basins were all covered by ~2,000 m of ice in their northern extents at the last glacial maximum (Marshall et al., 2002). Ice sheet thickness near modern discharge areas increased to the point that it exceeded the elevation of ice free areas in the south. The increase in hydraulic head associated with the ice sheet caused a reversal in flow direction across these basins (Ferguson et al., 2007; Grasby et al., 2000; Grasby & Chen, 2005). During this reversal of flow direction, DFR would have increased in the Williston and Alberta basins at the basin scale. The difference in elevation between the elevation of the ice sheet in the north and the Black Hills and Rocky Mountains on the other side of these basins would have been less than the modern topographic drop across the basin. The DFR would have decreased across the Michigan Basin during the Pleistocene because the 2,000 m of ice sheet thickness exceeds the modern topographic drop across the basin (McIntosh et al., 2011). The Illinois Basin would have been covered by ~1,000 m of ice in its northern extent, exceeding the modern topographic drop across the basin. This decreases our estimated DFR to values of slightly less than 1 in each of these basins. Subglacial recharge has been noted in each of these basins (Grasby et al., 2000; Grasby & Chen, 2005; McIntosh et al., 2011; Person et al., 2007). However, brines with

chemistry indicative of an evaporated marine source remains in the deeper reaches of these basins (Connolly et al., 1990; Grasby et al., 2000; Stueber & Walter, 1991; Wilson & Long, 1993) where DFR values are highest, suggesting that changes in DFR were not large enough in magnitude or duration to displace connate brines.

The long-term presence of trapped brines in sedimentary basins is important for the distribution of salinity throughout these basins. Ranganathan and Hanor (1987) suggest that the Michigan, Alberta, Illinois, and Gulf Coast basins have salinity profiles that reflect diffusion. More detailed models of diffusion have recently been produced for the Michigan Basin (Al et al., 2015) and Williston Basin (Hendry et al., 2013). Regional flow in high permeability zones associated with shorter-term events such as glaciations will manifest as perturbations to the longer-term diffusive salinity distributions. Nonetheless, the presence of stagnant deep brines provides a continuous source of diffusive solutes that controls the salinity of water in basins and that will limit the depth at which freshwater can be found that can be used for consumption, irrigation, or other uses.

5. Conclusions

DFR exerts a strong control on the mobility of brines in sedimentary basins. In North American sedimentary basins examined, where $DFR > 0.37$ all contain connate brines that have been stagnant over geologic time-scales. Basins with $DFR < 0.37$ contain a range of salinities due to a combination of flushing by regional groundwater flow and low initial salinities. More detailed examination of selected basins with $0.37 < DFR < 1.0$ indicates that these basins likely have $DFR > 1.0$ throughout some portion of their extent, allowing for trapping of brines. Basins with $DFR < 0.37$ that still contain connate brines can be explained by locally high DFR or very slow flushing rates. The findings of this study provide new insights into the distribution of connate brines and regional groundwater flow systems in North America. DFR and its control on groundwater flow and brine distribution should be considered, along with other factors, in waste isolation projects. Studies seeking to understand the distribution of fresh and saline waters may also benefit from this understanding of persistent brines.

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