

RELATIONSHIP BETWEEN CALCITE SOLUBILITY AND CHLORIDE CONTENT OF GROUND WATER IN THE PALEOZOIC AQUIFER, ALCORN COUNTY, MISSISSIPPI

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Introduction

Chloride contents of ground water produced from the Paleozoic aquifer in Alcorn County, Mississippi, are significantly higher than in ground waters from the overlying and hydraulically-connected Cretaceous aquifer system. Although the chloride content of the Paleozoic aquifer, typically greater than 100 mg/l, is not a problem from the standpoint of water quality, its origin is problematic. Chloride content at this level generally comes from the "flushing out" of trapped connate water or the dissolution of evaporite minerals present in the aquifer (Feth 1981; Hem 1985). The Paleozoic aquifer was deposited more than 300 million years ago, and the present fresh ground-water system has probably existed for at least 10 million years. In this situation, one would expect easily extractable chloride sources to have been removed long ago. For example, much younger carbonate formations, such as the Tertiary limestones that comprise the Floridian aquifer of Georgia, Alabama, and Florida, generally have Cl contents less than 20 mg/l (Back and Hanshaw 1970).

Chloride is the most conservative major ions in solution; it does not enter into redox reactions, does not form complexes with other ions unless present at elevated levels, is not easily adsorbed on mineral surfaces, and does not form salts of low solubility (Hem 1985). Because of these properties, and assuming no human-derived sources (e.g., road salt), the chloride content of the ground waters in the Paleozoic aquifer in Alcorn County is not easily explained. For example, Feth (1981) states: "The continuing release of Cl at rates that cause modest and virtually constant concentrations in some natural waters pose problems of supply mechanism that remain unanswered--indeed largely unexplored."

Regional Geology

Alcorn County is within the outcrop belts of the Late Cretaceous Eutaw Group, Coffee Sand, Demopolis Chalk, and Ripley Formation (Figure 1). The Cretaceous section consists predominately of sand, clay, and chalk with subordinate amounts of limestone. The relatively unconsolidated Cretaceous sediments rest unconformably on an eroded surface consisting of consolidated sediments of Paleozoic age.

The Paleozoic section crops out in the eastern extreme of the adjoining Tishomingo County and was described by Morse (1929) and more recently by Merrill and others (1988). The outcropping Paleozoic section was assigned to various Devonian and Mississippian units by Merrill and others (1988). The oldest exposed unit is the Devonian Ross Formation and the youngest exposed Paleozoic unit is the Hartselle Formation.

Examination of well cuttings from the Alcorn Oil Company's Matthews No. 1 well demonstrated that the Paleozoic section in the Alcorn County area consists almost exclusively of limestones, dolostones, and chert. In Alcorn County, the lithology typically encountered at the top of the Paleozoic section is a fractured, sparsely fossiliferous, light gray to black, chert. Most authors to date have refrained from assigning this chert to a stratigraphic unit and refer to them only as the "Paleozoic chert" or "Paleozoic undifferentiated." Exceptions are Boswell (1963) who suggested a Devonian age and Madlinger (1982) who assigned the unit to the luka Formation which is approximately equivalent to the Mississippian Fort Payne Formation. Lithostratigraphic correlations suggest the chert is equivalent to the Mississippian section cropping out in Tishomingo County. The contact with the overlying Cretaceous sediments

appears sharp, typically with a discontinuous dark- to medium-gray clay directly above the contact.

The Cretaceous section crops out in Alcorn County with the Eutaw Group in the extreme eastern portion of the county, the Coffee Sand and the chalks of the Selma Group in the center, and the sands and clays of the Ripley Formation (Selma Group) in the west (Figure 1). The Tuscaloosa group is locally present in the subsurface of Alcorn County and consists of a gravelly unit with fine- to coarse-grained sand as a matrix.

In the subsurface, the Eutaw Group is typically a fine- to medium-grained sand with thin, carboniferous, clay beds. Some sand beds contain significant amounts of fine-grained glauconite. Where the Tuscaloosa is not present, the Eutaw rests on the Paleozoic section. In Tennessee, Russell and Parks (1975) assign the Eutaw formational ranking and divide it into two members. Both upper and lower contacts of the Eutaw are considered by Merrill and others (1988) to be unconformable in Tishomingo County, and they list the maximum thickness as 200 feet.

The Coffee Sand crops out throughout eastern Alcorn County and typically consists of fine- to medium-grained sands with beds rich in glauconite. In the subsurface, the Coffee Sand has fewer clay beds and a more uniform sand section than does the Eutaw. Merrill and others (1988) note the Coffee Sand is as much as 150 feet in thickness in Tishomingo County and is approximately 230 feet thick at Corinth.

The Demopolis Formation overlies the Coffee Sand and consists of a thick argillaceous chalk with subordinate limestones. Russell and Parks (1975) consider the lower Demopolis contact as conformable in Tennessee. The upper section of Demopolis becomes sandy and argillaceous. This zone is often referred to as the "transitional zone" and is included in the Ripley Formation.

The Ripley Formation of Mississippi consists of a number of members which are assigned formational ranking in Tennessee. The basal Coon Creek and McNairy Sand members are both represented in Alcorn County. The "transitional zone" is usually included in the Coon Creek Member; however, others would assign the zone to the Demopolis and retain only the well sorted, fine-grained, marine sands in the Coon Creek. The McNairy Sand is composed of fine- to coarse-grained, typically crossbedded, sand with subordinate clay beds. The McNairy Sand can be divided into a lower sand section, a middle

interbedded sand and clay section, and an upper sand section. Only the lower and middle sections are well exposed in Alcorn County. Russell and Parks (1975) note that the lower sand is finer grained than the upper sand section. They also note an unusually thick development of the middle section in western Alcorn County where more than 30 feet of the middle unit are exposed.

Hydrogeologic Setting

Seldom do stratigraphic units and hydrogeologic units match. Such is the case in the Alcorn County area. The Paleozoic section, unlike Kermode's investigation in Lee County, does not function as a confining unit but as an aquifer. The lower Paleozoic confining unit in the Alcorn County area is virtually unknown. Most investigators attribute the water producing capability of the Paleozoic aquifer to secondary permeability developed in the brittle chert. Assuming this hypothesis is correct, the base of the Paleozoic aquifer may correspond to the base of the highly fractured chert zone. Examination of cuttings from the Alcorn Oil Company's Matthews No. 1 well from Corinth reveals little evidence for primary permeability below the upper chert beds. Within the Tuscumbia and Fort Payne outcrop belts in Lauderdale (Harris and others 1963a) and Colbert (Harris and others 1963b) Counties, Alabama, water production is attributed to fracture permeability and openings resulting from dissolution. Merrill and others (1988) report transmissivity values from 800 to 52,000 gallons per day per foot (gpd/ft.) and yields from wells in excess of 800 gallons per minute (gpm) in Tishomingo County from the Paleozoic aquifer.

The Paleozoic-Mesozoic contact is of special interest in that extensive regolith development along the unconformity, such as described by Mellen (1937) in Tishomingo County, could effectively separate the Paleozoic aquifer from the overlying Cretaceous aquifers (e.g. Wasson and Tharpe 1975). Examination of well cuttings from the Corinth area supports the hypothesis that hydraulic connections exist. No evidence of oxidation was noted in well cutting from Corinth as might be expected from regolith development as described by Mellen (1937). The Corinth cuttings and well logs did indicate a discontinuous clay bed overlying the Mississippian erosional surface. This bed is considered a unit of the overlying Eutaw Group and not a weathering product. The basal clay is typically dark- to medium-gray and contains sparse, green, glauconite grains and carbonized plant material. We suggest that any regolith development as described by Mellen (1937)

was largely removed by the Cretaceous transgression. Since the basal Eutaw clay bed is discontinuous, we support the conclusions of several previous authors that there are hydraulic connections between the Paleozoic aquifer and the overlying Eutaw. These hydraulic connections may allow the Paleozoic aquifer to be recharged not only from the unit's recharge area in outcrop but also from the Cretaceous aquifers.

The Eutaw and Coffee Sand are distinguished by the higher percentage of clay beds within the Eutaw which are hydraulically connected. Newcome and Callahan (1964) state that the Eutaw will yield water sufficient only for domestic supplies. Wasson and Tharpe (1975) acknowledge hydraulic connections between the Eutaw and Coffee Sand except where the Mooreville Chalk is present to the south. A transmissibility of 6,000 gpd/ft² was noted by Newcome and Callahan (1964) for the Coffee Sand in western Alcorn County. Gandl (1982) reports Coffee Sand transmissivities from 930 to 1,200 gpd/ft² and states aquifer recharge is from precipitation within the recharge area. Gandl also suggests leakage occurs between the Coffee Sand and the Eutaw.

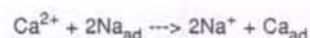
The Demopolis Chalk forms an effective confining unit above the Coffee Sand due to its low hydraulic conductivity. For example, Kernodle (1981) estimated a hydraulic conductivity value of 4.0×10^{-10} feet per second in his hydraulic modeling in Lee County. Keady (1962) reports a coefficient of permeability of 4.46×10^{-8} centimeters per second (cm/sec.) and a porosity of 30 percent. Figure 2 is an illustration of the aquifer system in the Alcorn County area.

Water Quality And Geochemical Modeling

Ground waters from the Coffee Sand and Eutaw Group typically have total dissolved solid (TDS) contents of less than 300 mg/l (Wasson and Tharpe 1975, and Table 1) with Ca and bicarbonate as the major ions and only minor amounts of Na and Cl. Iron contents in the 1-10 mg/l range are common, which is true of calcium-bicarbonate ground waters from other Coastal Plain aquifers (e.g., Lee 1985; Lee and Strickland 1988). In contrast, ground water from the Paleozoic aquifer typically contains >300 mg/l TDS, typically has iron contents in the 0.X mg/l range, and is also a calcium-bicarbonate type (Table 1). In addition, sodium and chloride are also present in appreciable amounts.

Many Coastal Plain ground waters have elevated Na contents due to a cation exchange process where clays (and zeolites?) exchange adsorbed Na for Ca in

solution (Lee 1985; Lee and Strickland 1988) as in the following reaction:



However, in these Coastal Plain ground waters, chloride is only present in minor amounts, typically <5 mg/l. In contrast, chloride contents of >100 mg/l are common in the Paleozoic aquifer in Alcorn County. A plot of Na vs. Cl content for ground waters from the Paleozoic aquifer indicates that there is a general trend of increasing chloride paralleling sodium content (Figure 3). This general trend roughly approximates the seawater dilution line (Figure 3) which represents the Na:Cl ratio of seawater that has been diluted with pure water to the salinity range of ground waters from the Paleozoic aquifer. Also shown is a slightly different trend of Na:Cl ratios also resulting from the progressive dissolution of halite (NaCl) by pure water. On the other hand, a plot of sulfate vs. chloride (Figure 4) indicates that the Paleozoic aquifer ground waters are generally depleted in sulfate compared to diluted seawater.

The parallel increase in sodium and chloride in ground waters from the Paleozoic aquifer indicates that both were contributed by a single process or source. Coastal Plain aquifers in Mississippi commonly contain saline waters in their down-dip extensions, and Lee (1985) proposed that local elevated Na and Cl contents in these aquifers are due to mixing. On a more local scale, Wasson and Tharpe (1975) interpreted the high chloride content of ground water from the Paleozoic aquifer in Alcorn County to be the result of pumping-induced leakage of saline waters from deeper levels. Currently, there are no available data on the chemical composition of ground waters below the Paleozoic aquifer or the nature of the basal confining layer in Alcorn County, all of which would be required to evaluate Wasson and Tharpe's hypothesis. However, the close proximity of the Paleozoic formations in Alcorn County to their outcrop area in adjacent Tishomingo County (Figure 1) suggests that these formations should contain low chloride water.

In order to evaluate possible water-aquifer chemical reactions that may have played a role in the observed major ion concentrations in Alcorn County ground waters, the computer program SOLMINEQ.88 (Kharaka and others 1988) was employed. SOLMINEQ.88 is a solution chemical model that calculates activities of dissolved species from the input analytical concentrations. The activities are then used to calculate saturation indices (SI) for various minerals of possible geochemical importance using established

thermodynamic stability data for those minerals. The SI is defined as $\log(IAP/Kt)$, where IAP is the ion activity product (calculated from the input water analysis) and Kt is the mineral stability constant at the input solution temperature. A solution that is theoretically saturated with respect to a given mineral will have a SI of 0, whereas a negative SI value indicates undersaturation, and a positive value indicates supersaturation.

SI values for the minerals calcite, dolomite, and halite are listed for the 6 representative water analyses in Table 1. Analyses D12 and D03 from Cretaceous formations are greatly undersaturated with respect to calcite and dolomite, although J75 is approximately saturated with respect to both minerals, as are the 3 analyses from the Paleozoic aquifer (Table 1). In addition, the higher Na-Cl contents of ground water from the Paleozoic aquifer is reflected in SI values for halite that are 2-3 orders of magnitude greater than in ground water from the Cretaceous formations. Similar observations are apparent when considering the larger data base of ground water analyses for Alcorn County. Figure 5 is a plot of calcite SI vs. chloride content which shows that the low-Cl waters from the Cretaceous formations are generally undersaturated with respect to calcite, whereas the Paleozoic aquifer waters are typically saturated to supersaturated with respect to calcite. Local calcite saturation within the Cretaceous formations is probably due to equilibration with calcareous fossils, whereas calcite saturation in the Paleozoic aquifer is probably due to limestone dissolution at the base of the chert.

The hydraulic connection between the Cretaceous formations, which typically contain calcite-undersaturated waters, and the Paleozoic aquifer with its calcite-saturated ground water, suggests a possible ground water chemical evolution process. Downward movement of ground water into the Paleozoic aquifer would cause calcite dissolution along the top of the limestone at the base of the chert. This process can be modelled using SOLMINEQ.88, which can calculate the result of mineral dissolution or precipitation on the input chemical analysis. For example, the proposed calcite-dissolution process can be modelled by using a representative water analysis from the Cretaceous aquifer such as D12 of Table 1. SOLMINEQ.88 can be used to calculate a theoretical chemical analysis after D12 dissolves calcite to equilibrium. Results indicate that the pH jumps to 7.8, Ca content increases from 15 to 33 mg/l, and bicarbonate increases from 63 to 120 mg/l. These trends are consistent with observed chemical differences between the Cretaceous and Paleozoic ground waters.

The chemical modeling, along with the geologic and hydrogeologic relationships, give support to the idea that the calcite-undersaturated waters are recharging the Paleozoic aquifer causing calcite dissolution. In fact, the data points shown in Figure 5 may be defining a mixing line between low-Cl waters with calcite SI of about -1 and higher Cl waters closer to saturation with respect to calcite.

If calcite dissolution is occurring along the base of the Paleozoic aquifer, then the process can also explain the source of chloride in the water. Seawater trapped as fluid inclusions or along grain boundaries in the limestone would slowly be released as calcite is dissolved, causing Na and Cl to increase in the ground water by amounts related to the Na:Cl molar ratio in seawater. This process offers a plausible explanation to Feth's (1981) questions about constant and continuing sources of Cl in some natural waters.

Discussion

The role of saline fluid inclusions in the chemical evolution of ground waters has been recently documented by Nordstrom and others (1989) for ground water in crystalline rocks at Stripa, Sweden. In this case, fluid inclusions with up to 25 wt. % NaCl have been released to the slow-moving ground waters due to water-rock reactions and locally cause chloride contents in excess of 1000 mg/l. By comparison, it would seem that even small amounts of fluid inclusions of seawater (NaCl content of 3.5 wt. %) in a much more soluble host mineral (calcite) would be more than capable of releasing Na and Cl levels observed in ground water from the Paleozoic aquifer.

Additional evidence of the role of seawater comes from the sulfur balance. As discussed previously, the sulfate content of waters from the Paleozoic aquifer are depleted with respect to seawater. A possible explanation for the depletion (assuming that seawater was the source of sulfur) is that seawater-sulfate sulfur has been incorporated into the abundant authigenic pyrite and marcasite that are present along fractures in the chert. Two samples of the iron sulfide minerals were analyzed for their sulfur isotopic content. Results ($\delta^{34}\text{S}$ contents of +17.3 and +18.6 per mil CDT, respectively) compare favorably to Mississippian seawater sulfate with a $\delta^{34}\text{S}$ value of approximately +18 per mil (Holser and Kaplan 1966). Significant reduction of sulfate to sulfide at temperatures <50°C can only be accomplished by sulfate reducing bacteria (Ohmoto and Rye 1979). For this setting the following reaction approximates the process:



where the sulfate-reducing bacteria use the organic matter as a nutrient source. Because of the gradual release of sulfate due to calcite dissolution, reduction would take place under conditions that are closed with respect to sulfate, meaning that the rate of sulfate reduction is faster than the rate at which sulfate is supplied (Ohmoto and Rye 1979). On the other hand, the system is open to H_2S because any available sulfide sulfur would be rapidly precipitated as pyrite due to the high iron content of the water in the Paleozoic aquifer. Under these conditions, the results of bacterial sulfate reduction can produce reduced sulfur with close to the same isotopic composition as the seawater sulfate (Ohmoto and Rye 1979). Lignite and wood fragments, commonly replaced by pyrite, are very abundant in the Cretaceous formations and provide additional evidence for the general process in this setting. However, the source of the organic matter required for sulfate reduction in the chert is problematic. Perhaps dissolved organic carbon from the lignite diagenesis process is carried downward with the moving ground water; or alternatively, perhaps there is a source of reactive organic carbon in the dissolving limestone. At any rate, there can be little doubt that the source of sulfur in the authigenic pyrite was from seawater sulfate which further implicates Mississippian seawater as the source of chloride in the Paleozoic aquifer.

Conclusions

The processes responsible for controlling the chemical evolution of ground water in the Paleozoic aquifer appear to be somewhat unique, with chemical constituents being added from above and below a major geologic unconformity. Mixing of these constituents appears to control the chloride content of ground water from the Paleozoic aquifer. Major components of the proposed mixing model include:

1. Downward movement of calcite-undersaturated recharging water from the Cretaceous formations into the Mississippian chert section;
2. Dissolution of calcite in Mississippian limestone at the base of the chert;
3. Release of Mississippian seawater trapped along grain boundaries or in fluid inclusions from the dissolving calcite;
4. Reduction of seawater sulfate causing a depletion in dissolved sulfate and pyrite precipitation.

Therefore, the composition of water from the Paleozoic aquifer in Alcorn County, Mississippi, appears to be best explained as the consequence of natural water-mineral reactions as opposed to recent pumpage-induced leakage.

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Table 1
Groundwater Geochemistry, Corinth Area, Mississippi

WELL	D12	D03	J75	COR17	COR16	COR13
Unit	Coffee	Eutaw	Coffee	Paleozoic	Paleozoic	Paleozoic
Depth (m)	44	107	166	173	151	152
pH	6.7	6.1	7.4	7.9	8.1	8.1
Ca	15	8.0	48	43.2	31.2	25.6
Mg	3.1	1.9	9.0	11.2	10.7	15
Na	3.9	3.9	3.3	60.7	61.7	31.1
K	2.0	1.8	4.2	6.4	5.4	4.6
Fe (tot)	5.5	2.5	0.56	0.25	0.18	0.27
Mn	ND	ND	0.13	0.052	0.127	0.168
Cl	1.5	3.6	2.6	110	98	58
F	0.1	0.1	0.1	0.3	0.45	0.17
SO ₄	5.0	7.6	7.8	9.2	8.6	15.1
HCO ₃	63	32	196	148	138	146
SiO ₂	17	20	22	ND	ND	ND
TDS	116	81	293	390	353	296
SI _{cal}	-1.73	-2.87	-0.05	0.25	0.17	0.16
SI _{dol}	-2.83	-5.05	0.45	1.24	1.15	1.41
SI _{hal}	-9.78	-9.39	-9.63	-6.76	-6.78	-7.32

Note: Analyses J75, D12, and D03 are reported in Wasson and Tharpe (1975), for Alcorn County, Mississippi. Analyses COR13, COR16, and COR17 are from Corinth municipal supply wells and were provided by the Corinth Gas and Water Department. Analyses were conducted by the Mississippi State Department of Health.

Abbreviations: ND= not determined; SI= saturation Index ($\log(IAP/Kt)$); cal= calcite; dol= dolomite; hal= halite.

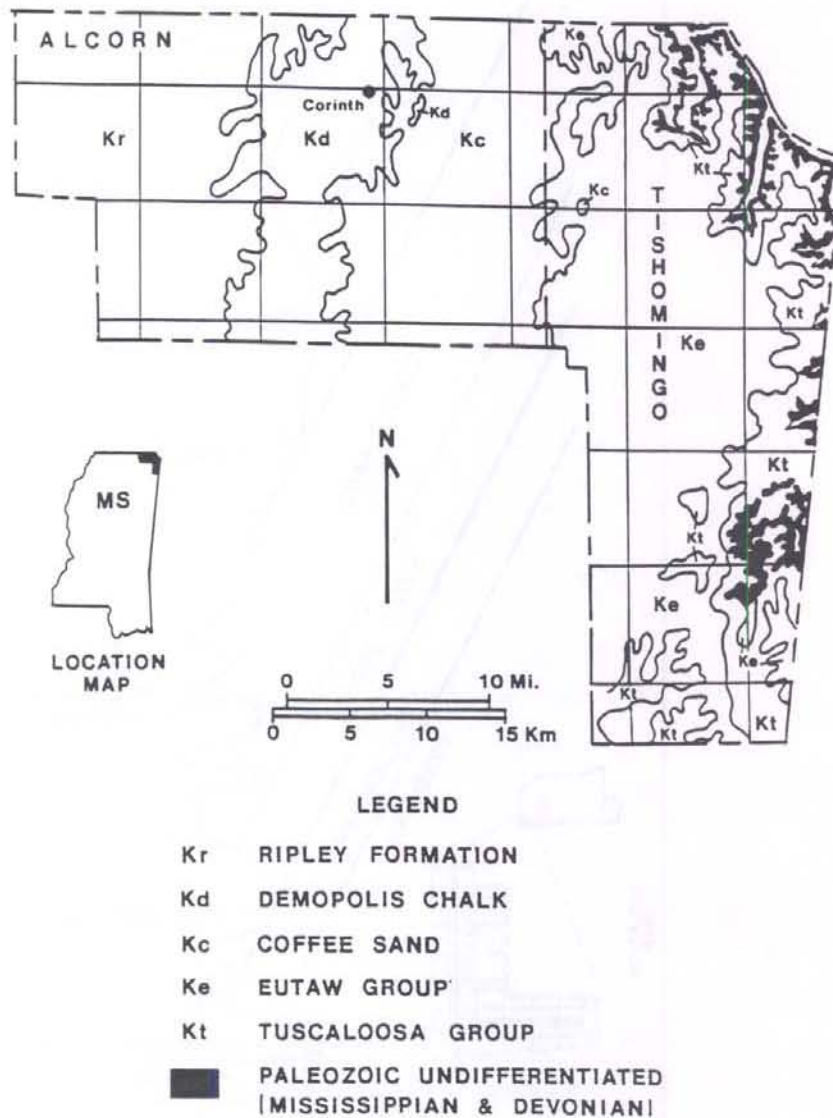


Figure 1: Geologic map of Northeast Mississippi (modified from Mississippi Geological Survey, 1969).

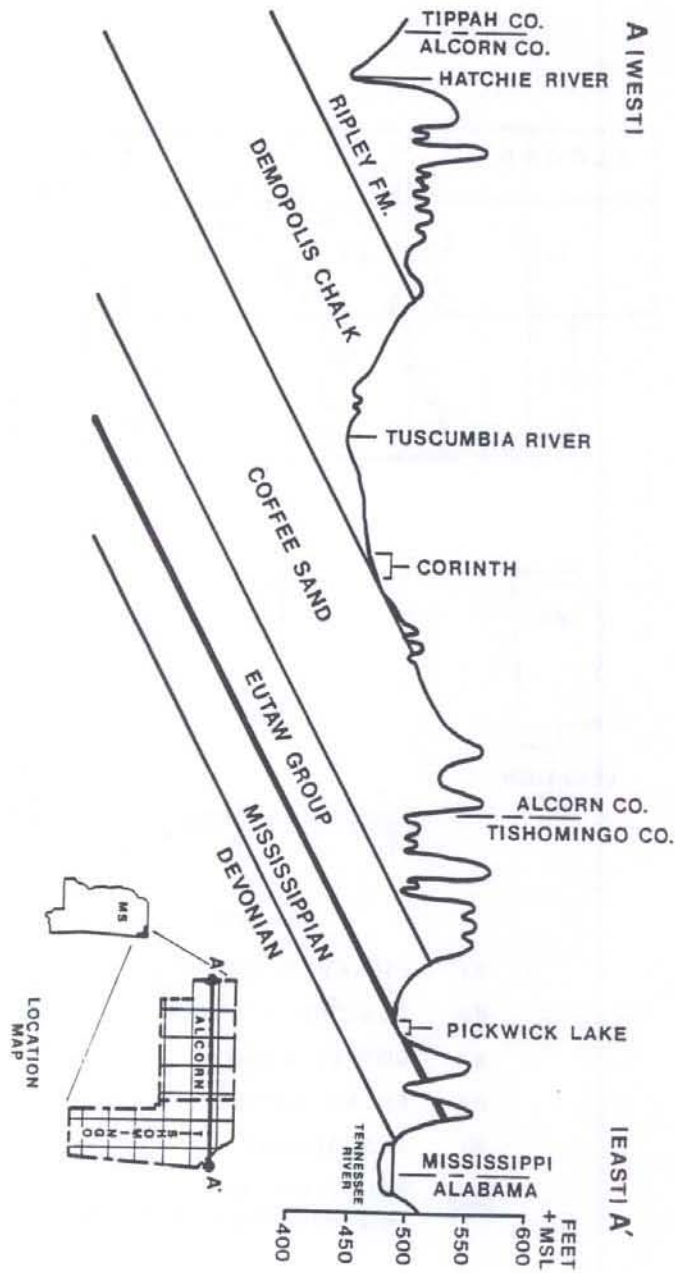


Figure 2: Geologic cross section through Tishomingo and Alcorn counties, Mississippi.

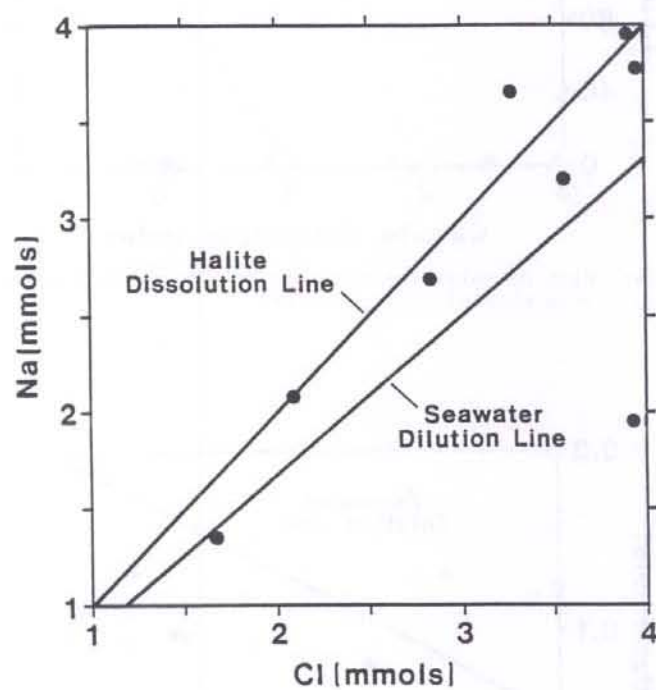


Figure 3: Plot of molar sodium and chloride content of ground waters from the Paleozoic aquifer.

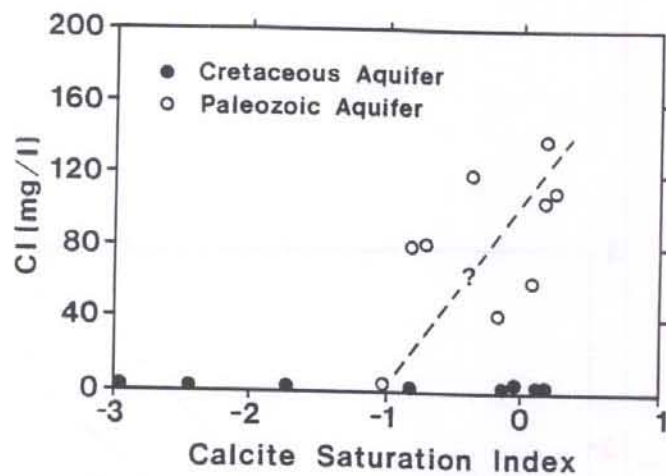


Figure 4: Plot of molar chloride and sulfate content of ground waters from the Paleozoic aquifer.

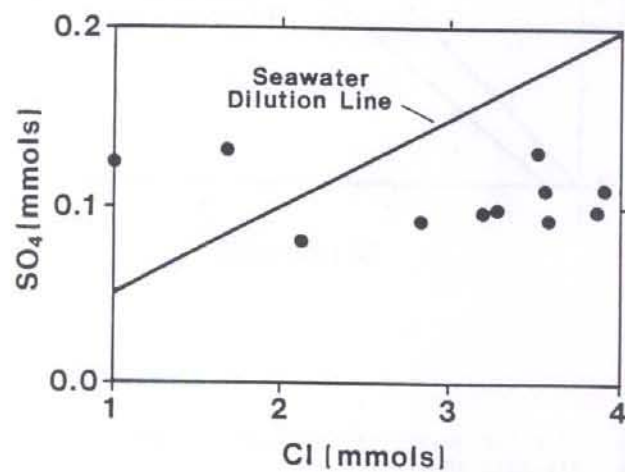


Figure 5: Plot of chloride content vs. calcite saturation indices for ground water in Cretaceous formations (dots) and the Paleozoic aquifer (open circles). Dashed line represents a possible mixing line.