2 DEPOSITIONAL ENVIRONMENT AS INDICATED BY CHLORINITY OF INTERSTITIAL WATER IN CORES FROM KARNES COUNTY, TEXAS

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BY

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Abstract

Natural-state chlorinity of interstitial water in Eocene shale and permeable sandstone of Jackson age in uranium terrain in Karnes County, Texas, obtained from cores cut with oil-base mud to a depth of several hundred feet, varies as depositional environment from marine to non-marine.

Chlorinity in one formational unit reflects mainly progressive change of depositional environment with depth, and, in another, oscillation between marginal marine and fresh water deposition.

The overall relation between chlorinity and depositional environment is not obscured by inverse variation between chlorinity and saturation in intervals that are not completely water saturated, among which are the principal uranium-bearing zones. Considerably greater variability in saturated rock is shown by core water chlorinity than by equivalent sodium chloride content as obtained from drill hole resistivity and core porosity.

A satisfactory explanation is not evident for retention of connate chloride in shallow permeable Eocene sandstone that has been long subject to flushing by fresh meteoric ground water. Among possible explanations are: retention of chloride by ionic adsorption on sand grains, as noted

by Alger, Sarma, and Rao; or, immobility of a significant fraction of

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Introduction

In a study of uranium occurrence in shallow and generally unconsolidated Eocene sandstone of Jackson age in Karnes County, Texas (Manger and Eargle, 1967), the possible relation of interstitial water chlorinity to depositional environment was investigated.

Cores were obtained in the extreme western part of Karnes County,
Texas, about 50 miles southeast of San Antonio and south of the abandoned settlement of Deweesville. The locations of the drill holes

(K1, K2, and K3) are shown in figure 1. Oil-base mud was used for core
drilling in order to obtain cores where interstitial water would be
preserved in natural-state amount and salinity.

Water content of the cores was obtained by modification of a method described by Rall and Taliaferro (1946). Immediately after removal from sealed plastic tubes, core samples were crushed, weighed, and extracted with toluene for 24 hours. Vapor from boiling toluene distilled the core water which was collected in a calibrated glass tube. The oil-base mud was dissolved in condensed hot toluene as it flowed through the sample on its return to the boiling flask.

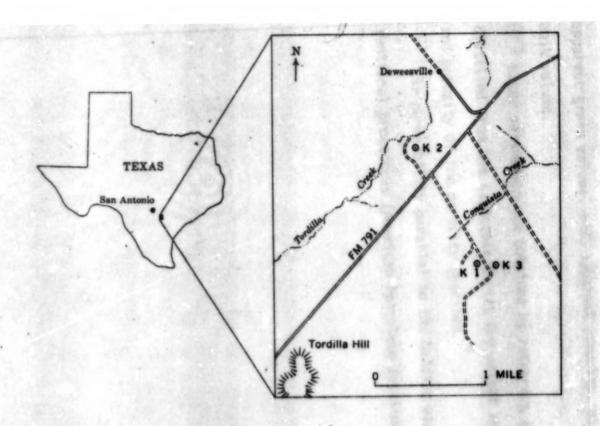
Following toluene extraction, the core samples were extracted with hot distilled water for an additional 24 hours to remove the salts.

The chloride content of the water solution was determined by potentiometric titration with silver nitrate, using a silver electrode.

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Bulk density of adjacent core was obtained by dividing the weight of an extracted and oven-dried specimen by its bulk volume as determined by displacement of mercury from a pycnometer. Apparent grain volume of the bulk density specimen was determined by a pressure-volume relation-ship by use of a helium gas porosimeter (Rall, Hamontre, and Taliaferro, 1954). Apparent pore volume was determined as excess of bulk volume over apparent grain volume, and, apparent porosity, as this excess per unit bulk volume.

Water saturation was obtained by referring the volume of water distilled from the sandstone to its dry weight and to the bulk density and porosity of the porosity specimen.

Air permeability parallel to the bedding was obtained using conventional procedures (American Petroleum Institute, 1960, pp. 33-36).

General relations of chlorinity to depositional environment

A general correspondence of mean chlorinity of interstitial water with depositional environment of members of the Eocene Whitsett and McElroy Formations is shown in table 1.

Lowest mean chlorinity as 167 mg/leas found in the generally shaly nonmarine part of the Dubose Member of Whitsett Formation below the Tordilla Sandstone bed. The next lowest mean chlorinities as 211 mg/l and 242 mg/l were found in moderately permeable sandstones from the upper part of the Dilworth Member and the Manning Clay Member of the McElroy Formation, respectively. Depositional environment of the Dilworth is described as marine only at its base, and, of the Manning Clay, as non-marine or lagoonal. Slightly higher chlorinity was obtained for the Conquista Clay, which is described as marine or lagoonal above the pasal few feet. Thus, chlorinity varies with depositional environment although it is much less than the connate water chlorinity of marine sediments.

Volcanic ash, tuffaceous material, and bentonitic clay are frequently sufficiently abundant to characterize the lithology of the cored beds, as is shown in table 1. In the permeable sandstone the percentage of clay-size grains is low, but Bargle and Snider (1957, p. 17) state that in many beds of sand montmorillonitic clay forms a matrix for the sand grains.

coras of the Jackson (Eocene) Group, drill holes K1, K2, and K3, Karnes County Texas

(Stratigraphy, depositional environment, and lithology from Eargle (1959). Chlorinity by W.T. Wertman, T.E. Sterner, R.B. Lowe, and W.M. Smith, U.S. Bur. Mines

Pormation	Member	Unit	Depositional	mintinetive Thick-	pept of	Peracabili Lyoniami	71 710 .	The state of the s	Range	190. 0
	Dubose	Tordilla Sandstone bed	Marine, a temporary return of marine conditions		16-59 (KI, KS)	404	The second second second	3,020		34
Whitsett	1		Nonmarine	Highly 70	45-129 (KI, K3)	110	10	167	8-404	33
	Stones Switch Sandstone	the tribe	Locally marine, transitional from marine	Zi+	16- 37 K2, K2A	63	17	1,751	500-4680	38
	1		Conquista to nonmarine Dubose	555	(KI, K3)	72	22	533	76-1200	71
	Conquista	man and all	Marine or lagoonal above the basal few feet	Bentonitic and lignitic clay 48	37-98 (M2) 170-218 (K1)	60	3	493 416	105-795 213-802	20 15
HcElroy	Dilvorth		Marine at base, grades into non- marine	75	\$8-175 (K 2)	74 48	12	211 413	60-598 206-1 <i>p</i> 60	20 25
	Manning Clay		Nonmarine or lagoonal	Much volcanic ash and ben- tonitic clay	The state of the s	And in contrast of the last of	177	242	70-438	22

Chlorinity in formational units

Tordilla Sandstone

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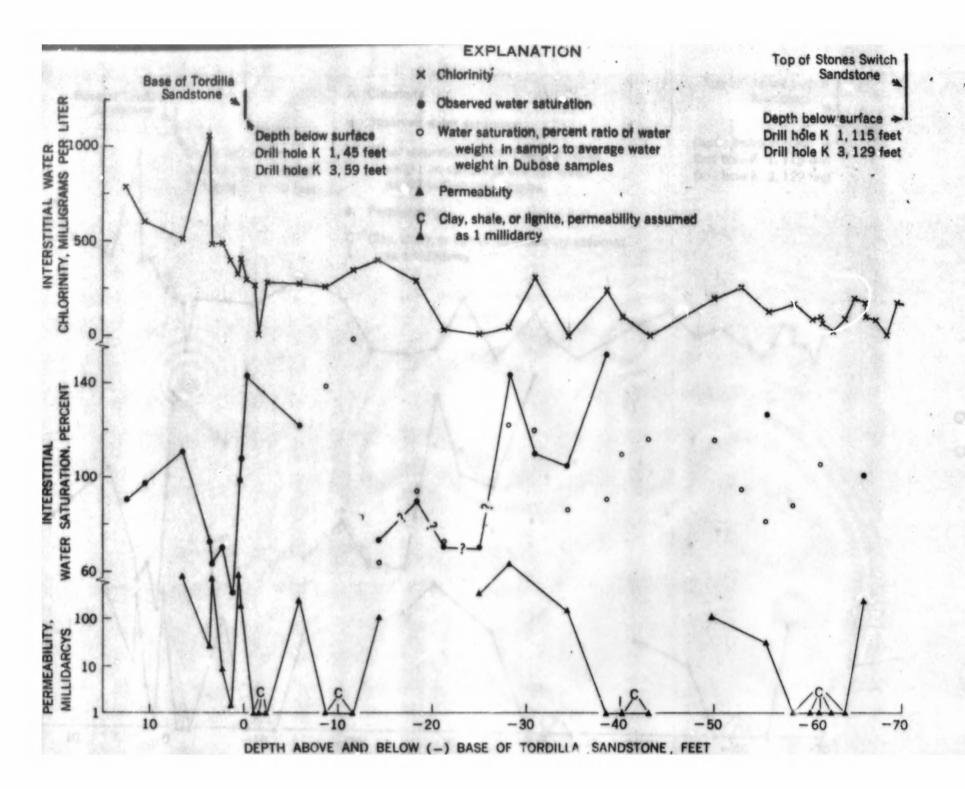
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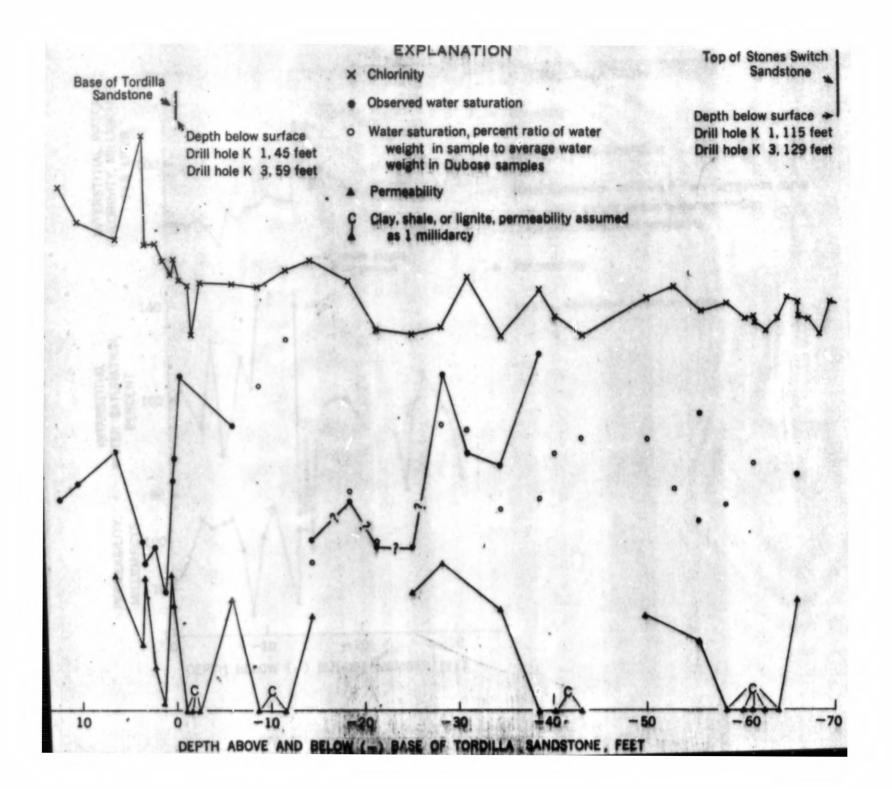
Chlorinity in the marine Tordilla Sandstone generally is high, as is seen in table 1. Exceptionally high chlorinity in partly saturated core obtained at very shallow depth probably is due to atmospheric evaporation. Chlorinity near the base of the Tordilla or at a depth of 40 feet is about 1,000 mg/l (fig. 2) and approximately equals the maximum chlorinity shown by water-saturated Stone Switch Sandstone at a depth of 85 feet (fig. 3).

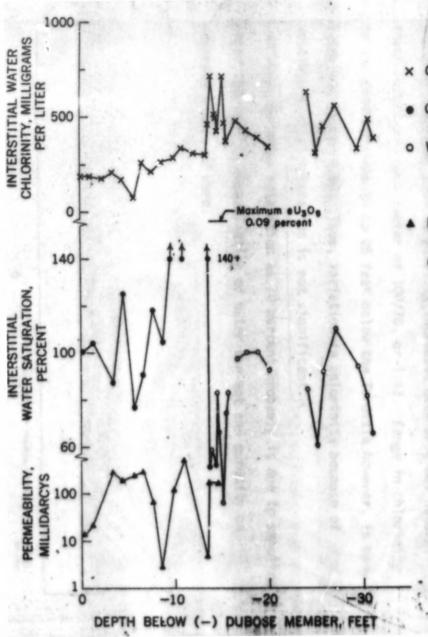
Dubose below the Tordilla Sandstone

Chlorinity in the nonmarine part of the Dubose, or the part of the Dubose that lies below the Tordilla Sandstone, generally is low (fig. 2), but increases irregularly upwards towards the marine Tordilla Sandstone. As based on 33 samples taken from depths to 70 feet, the coefficient of correlation (\underline{r}) between depth and chlorinity is -0.42, and, the coefficient of determination or variance (\underline{r}^2), 0.18. About 18 percent of the variation of chlorinity in the Dubose Member therefore is considered due to an irregular upwards recovery toward marine conditions of deposition.

Within the Dubose chlorinity at six depths is less than 40 mg/l (fig. 2). A marked decrease in chlorinity to 76 mg/l also occurs at a depth of 5 or 6 feet below the base of the Dubose or top of the Stones Switch Sandstone at drill hole K 3 (fig. 4). Chlorinity varies independently of permeability, as is seen in figure 2.







EXPLANATION

- × Chlorinity
- Observed water saturation
- Water saturation estimated from regression curve of water weight versus water saturation for unconsolidated sandstone
- ▲ Permeability

eU3Oa, equivalent uranium oxide



The few determinations of water saturation for samples of the nonmarine portion of the Dubose Member (fig. 2) suggest that the Dubose generally is saturated. In order to supplement observed saturations, computations were made of the percent ratio weight of water in a sample to the average weight of water in all of the samples. A similarity of spread of ratios and observed saturations with depth around the 100-percent value for most samples (fig. 2) suggests a saturated condition throughout the Dubose.

Partial water saturation is doubtfully indicated in the Dubose in the interval from 14 to 25 feet below the Tordilla Sandstone bed. Four samples show observed mean saturation as 76 percent and minimum saturation for the Dubose (25 feet below the Tordilla) as 70 percent. Observed chlorinty thus is possibly increased over original natural-state chlorinity by a factor of 100/70, or 1.43. Range in chlorinity in the interval from 14 to 25 feet below the Tordilla however, is many times the factor 1.43. Thus, variation in chlorinity because of possible partial saturation is not significant.

The low water saturation as 70 percent probably is due to sampling error involved in determination of water content and porosity on separate pieces of core.

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Near-minimum chlorinity as 43 and 11 mg/l in the noted Dubose interval from 14 to 25 feet below the Tordilla and permeability probably in excess of 100 millidarcys suggest a good quality aquifier but one of low yield. Maximum induction log resistivity for the Dubose as 13.6 ohm-meters in this interval also suggests the occurrence of water of very low salinity (Alger, 1966).

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Variation of chlorinity of interstitial water in the Dubose, irrespective of permeability, suggests that observed chlorinity reflects depositional environment. As a 50-fold variation in chlorinity, from 8 to 404 mg/l, is far greater than observed variation in chlorinity in present-day ground water, variation from sample to sample seems to be a reflection of original differences of chlorinity and hence of differences in depositional environment.

Chlorinity minima in the Dubose and at the top of the Stones

Switch are interpreted to indicate that a deluge of fresh water marked

the close of deposition of Stones Switch beds, and that gradual

recovery towards marine conditions of sedimentation in the Dubose pro
ceeded through six cycles characterized by minimum chlorinity within

a cycle as less than 40 mg/l and higher chlorinity at a cycle boundary.

These cycles suggest oscillations between marginal marine and fresh

water sedimentation. Recovery culminated in the deposition of the

marine Tordilla Sandstone.

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Stones Switch Sandstone

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Chlorinity in the Stones Switch Sandstone Member of the Whitsett Formation varies as depositional environment, which is transitional upwards from marine beds of the Conquista Clay Member of the McElroy Formation to nonmarine Dubose beds of the Whitsett Formation (table 1). Upward although irregular decrease in chlorinity (figs. 3 and 4) agrees with lithologic and fossil evidence of an upward approach to nonmarine conditions. Maximum chlorinity in the Stones Switch apparently occurs several feet above the base of the member.

Increased chlorinity with depth in the Stones Switch Sandstone at drill holes K 1 and K 3 is expressed by the coefficient of correlation (\underline{r}) and variance or the coefficient of determination (\underline{r}^2) , as follows:

Drill hole	Thickness of sampled interval, feet	Number of samples	r	r2	2
K 1	49.0	36	0.85	0.72	
К 3	30.8	32	.58	.34	

The coefficient \underline{r}^2 for the longer K l section shows that about two-thirds of variation in chlorinity reflects upward transition from marine to nonmarine conditions of deposition. For the shorter K 3 section about one-third of the variation in chlorinity reflects upward transition. Thus, dependence of chlorinity upon depth, and presumably upon change of environment of deposition with depth, is more pronounced in the Stones Switch Sandstone Member than in the Dubose Member, where, as was noted, the value of \underline{r}^2 was 0.18.

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Surface	Dubose Member		Standard deviation	Mean	Standard deviation		
			Drill h	ole K	1		
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142.3	27:3	61.2	16.4	832	129	9	-0.47
0149.9	to 34.9				THE COLUMN		200
156.5	41.5	82.5	14.0	896	231	9	46
0164.1	to49.1						
			Dill ho	le K 3			
142.7	13.7	63.3	16.6	523	128	8	65
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Thus, the	precedin	g 26 sa	mples toget	her sh	ow r as 0.0	54 and r2	as 0.
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82.5 pe	ercent and	63.3 p	ercent at d	rill ho	oles K 1 an	d K 3, re	specti
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Zones of increased electrical log resistivity in the Stones Switch
Sandstone and other formational units usually include or are co-extensive
with zones of partial saturation. As these zones are very porous but
contain more saline water, the increased resistivity reflects partial
unsaturation.

material

Saturation as affected by tuffaceous/and sampling error

Mean saturation in excess of 100 percent was observed for watersaturated samples from the top of the Stones Switch Sandstone. The excess probably is due to highly tuffaceous beds that are particularly abundant in the Dubose, as noted in table 1, and extend into the top of the underlying Stones Switch. For samples from the top of the Stones Switch where porosity specimens were only oven dried mean saturation as 97.4 present in able 2 shows a standard error for sampling as 7.6 percent saturation. Where adjacent porosity specimens from the same samples were prepared according to the regular method of extraction by toluene and hot water before oven drying, mean saturation calculates as 115.2 percent. The excess over 100 percent saturation apparently is due to a physical decrease in porosity imposed during extraction. Thus saturation as 115.2 percent multiplied by the porosity ratio 37.4/42.5 reduces to saturation as 101.4 percent. By comparison, for samples from the Dilworth Sandstone and top of the Manning Clay, where the amount of tuffaceous material is diminished, mean saturation as seen in table 2 is 99.2 percent.

As chlorinity is calculated as chloride assumed to be in water distilled from a sample, spurious values of porosity and consequent spurious saturation values do not affect chlorinity. Chlorinity in partly saturated rock, however, varies inversely with saturation, as was noted above. According to standard deviations shown in table 2 a saturation value less than about 70 percent probably reflects partial saturation. According to standard errors, mean saturation less than about 90 percent, except for a very small statistical sample, probably

reflects partial saturation.

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Table 2. - Saturation sampling error for saturated moderately permeable sandstone, drill holes K1, K2, and K3, Karnes County, Texas

[Data by W.T. Wertman, T.E. Sterner, R.B. Lowe, and W.M. Smith, U.S. Bureau of Mines. Mean permeability as antilog of logarithmic mean, as permeability tends to follow log normal distribution (Law, 1944).]

Dilworth Sandstone and Manning Clay Members of the McElroy Formation, 25 samples -----

Permeability millidarcys	Water	saturati	on, percent		Porosity	percent
<u>Mean</u>	Mean	Strait S	tandard eviation	Standard	Mean	Standard deviation
34	99.2	2.2	11.2	1.2	37.7	6.0
Top of Stones Switch Sandstone	e Member of	the Whi	tsett For	mation, 12	samples	
Usual extraction method for	llowed					
67 and permeability	115.2	13.2	45.6	13.2	37.4	6.5
Porosity specimens oven-dr	ied only				1 1	
80	97.4	7.6	26.2	0.0	42.5	6.2

Summary

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Interstitial water in crushed rock was vaporized in a flow of toluene vapor at about 110°C. The water vapor was condensed and water was collected and measured in a calibrated glass tube. After toluene extraction, recycling of many pore volumes of hot water as condensed steam recovered chloride and salts that were precipitated during evaporation of the interstitial water. Natural-state chlorinity was calculated as chloride per unit volume of interstitial water.

Natural-state interstitial water saturation was calculated as water volume per unit pore volume. The use of different portions of a sample to obtain water content and pore volume (or porosity) resulted in significant standard error for saturation. For some samples pore volume apparently was physically reduced during extraction by toluene and distilled water, as evidenced by mean saturation that significantly exceeded 100 percent. Mean saturation as about 100 percent, however, was indicated as based on the porosity of adjacent specimens of the same samples that remained unextracted and were only oven dried. Some samples from above a depth of about 200 feet were only partly saturated.

Mean chlorinity for saturated formational units varies as original chlorinity, or the chlorinity of the depositional environment, although presently observed chlorinity is much less than original chlorinity.

Within formational units chlorinity varies with progressive change of depositional environment with depth. In partly saturated intervals chlorinity varies inversely as saturation, but such variation is not so extensive as to obscure variation of chlorinity with depositional

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environment. The principal uranium-bearing zones occur in the partly
A low-yield aquifer of fairly fresh water in
saturated intervals./ probably saturated Dubose sandstone is indicated
by electric log resistivity and core porosity.
Discussion

Original chlorinity apparently was reduced because of flushing by fresh meteoric water but about in proportion to original chlorinity. As noted by Anders (1962, p G 12): "The water-bearing formations in Karnes County are being replenished continually by a small part of the precipitation on their outcrop areas. Most of the rainfall in and near Karnes County runs off in streams, evaporates, or is transpired by vegetation. Water that reaches the zone of saturation moves slowly through the rocks until it is discharged through some natural outlet, is intercepted by wells, or escapes by slow movement into overlying beds downdip from the outcrop. Most of the formations in the county must have contained salty water at one time, either because they were deposits in the sea or in brackisha water zones near the sea, or because the sea flooded the area shortly after their deposition. In Karnes County some beds of sand downdip from the outcrop are filled with fresh water, indicating that fresh water absorbed by the sand at the outcrop moved downdip and flushed out the salty water. At present, most of the sand beds contain fresh water near the outcrop and, generally, for some distance downdip. Farther downdip the water contains more mineral matter, the saline water being only partly flushed. Still farther downdip, the beds contain connate water, presumably water trapped in the sediments when they were

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deposited (Winslow and others, 1957, p. 387)."

Reduction of chlorinity was a shallow-depth process, as can be inferred from Anders' remarks. The present core samples are from the extreme western part of Karnes County where a sucession of unconformities (Anders, 1962, p G 10) is observed in a thin section of generally unconsolidated sediments extending from the top of the Eocene through the Younger Tertiary. The unconformities obviously represent depositional hiatuses approximately along ancient shorelines of the Gulf Coast geosyncline. Thickness of overburden on the top of the (Jackson) Eocene probably never was more than about 2,100 feet (Anders, 1962, table 2, p. G 10). Thiness of overburden during geologic time is attested by the extreme softness (unconsolidation) of many sand (sandstone) samples that precluded obtaining measurements for porosity and permeability. Thus reduction in original formation or interstitial water chlorinity probably is not attributable to generally recognized processes (Jones, 1969)/that result in changes in salinity/in deeper beds of the Gulf Coast geosyncline, such as ion filtration and osmotic salt concentration as dependent on clay membrane effects.

Reasons for reduction in chlorinity in proportion to original

chlorinity are not evident. The postulated proportionate reduction of chlorinity because of flushing by fresh meteoric ground water evidently was continued during laboratory extraction. Recycling of many pore volumes of hot water perhaps removed virtually all chloride remaining after ground water flushing. Greater reduction in original chlorinity would be expected in permeable sandstone because of more extensive flushing by ground water, but observed chlorinity is independent of permeability. Some moderately permeable samples from marine formational units show about maximum observed interstitial water chlorinity.

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A possible explanation for proportionate reduction in chlorinity
by some mechanism
perhaps is that in the natural state chloride was trapped / in moderately
permeable as well as poorly permeable rocks but was virtually completely
released when the rock speciman was flooded

by innumerable pore volumes of distilled water.

Retention of chloride ion by adsorption apparently is possible even in saturated clay-free permeable sandstone. Alger (1966, p. 16) noted that sand grains as well as shale particles adsorb ions and develop double-layer or surface conductance, the effect of which is evidenced where the resistivity of a saturating solution is high. For a sand pack composed of grains 0.375 mm in diameter and containing a NaCl solution of 44.2 ohm-meters resistivity, Sarma and Rao (1962, p. 474; 1963, p. 311) found that the resistivity of the saturated pack was 34.5 ohm-meters.

Immobility of a significant fraction of pore water in saturated permeable rock is suggested by Seevers' (1966) measurements of nuclear "fine to medium grained friable magnetic resonance. For saturated clay-free permeable sandstones"

Seevers (1966, p. 12) found that pore volume significantly exceeded the volume of mobile water. An excess surface area indicated by gas adsorption over the Kozeny (1929) surface area, as found by Brooks and Purcell (1952) for various sedimentary rocks, was considered by Seevers (1966, p. 2 and 3) to indicate that much of the total surface area is contained in a small fraction of the total pore volume and is not involved in fluid flow.

Immobility of pore water in partly saturated sandstone and consequent retention of chloride apparently/ due to virtually zero effective

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permeability for the flow of water where water saturation is low. Virtually zero effective permeability, for example, perhacs occurs in the Stones Switch Sandstone at drill hole K-3 at a depth of about 144 feet, where saturation is 36.5 percent, intrinsic permeability, about 100 millidarcys, and ch lorinity, 718 mg/l. The relatively high chlorinity is due (to probably) removal of interstitial water possibly by evaporation or by formation of hydrated minerals.

Retention in saturated sandstone of a fraction of Eocene connate water is suggested also by a comparison of average salt content as equivalent sodium chloride as obtained from induction electrical log sandstone zones with resistivity and core porosity for several / chlorinity for the zones as obtained from core samples (Manger and Wertman, 1967). Equivalent sodium chloride varies by a factor of 3, but, chlorinity, by a factor of 20. The greater variation in core chlorinity is thought to be a reflection of differences in original composition of the connate water.

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The present results yield relative estimates of paleosalinity.

By comparison, Johns (1963) found that chlorine is incorporated as insoluble magnesium hydroxy chloride in chlorite, and the amount of chlorite in the clay fraction of sediments increases with approach to a marine environment of deposition. Nelson (1967) noted a widespread occurrence of small amounts of the phosphates of iron and calcium in argillaceous sediments and found that the relative proportions of the phosphates reflect original salinity of the water at the site of deposition. Estimates of salinity were obtainable throughout the range from fresh water to marine for argillaceous rocks from the Paleozoic to the Holocene. Paleosalinity thus can probably be better defined by methods other than those used in the present study. The present results, however, suggest persistence of the relative chlorinity connate water over geologic time in variably saturated rocks.

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