

## 29. COMPOSITION OF SEDIMENTS AND PORE FLUIDS ON THE PERUVIAN CONTINENTAL MARGIN FROM GEOCHEMICAL AND GEOPHYSICAL WELL LOGS, SITE 679, LEG 112<sup>1</sup>

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### ABSTRACT

Geophysical logs measure variations in physical properties in a well, such as electrical resistivity, sonic velocity, and bulk density. Newly developed geochemical logs determine variations of elemental concentrations of silica, calcium, iron, aluminum, titanium, potassium, sulfur, gadolinium, uranium, thorium, hydrogen, and chlorine in a borehole. In turn, these analyses allow one to describe changes in bulk-rock chemistry that correspond to physical changes measured on the Peruvian continental margin. In Ocean Drilling Program (ODP) Site 679, Leg 112, sedimentological as well as lithological changes resulting from climate change, diagenesis, and ground-water hydrology were observed. Uranium and gamma-ray cycles were recorded that might be attributable to climate changes on the continent. Pyrite and uranium-rich layers indicated phosphorite deposits. Aluminum and the heavy rare-earth element gadolinium helped to locate volcanic-ash layers. Magnesium and calcium variations were observed that determine the locations and extent of dolomitization. A pore-water salinity inversion was measured using the chlorine/hydrogen ratio, with freshwater found in permeable sands beneath a calcium-cemented cap that forms a prominent seismic reflector in the area.

### INTRODUCTION

The geochemistry of sediments and pore fluids from a continental margin record the history of subsidence and uplift, diagenesis, and fluid migration in a distinctly different way than do physical-property changes within those same sediments. For example, velocity or density changes with depth may reflect compaction, whereas uranium and sulfur changes within the same sediments may result from diagenetic reactions that are caused, in turn, by hydrological processes. The combination of geophysical well logging, which determines physical-property variations with depth, and geochemical well logging, which measures elemental compositions of sediments and pore fluids, provides a more complete geological description of processes that have occurred in the past along a continental margin than either set of information taken alone.

Next, we describe geochemical, as well as the more standard geophysical, logging results from an ODP drill site into the continental margin: Site 679, off the Peruvian coast of South America (Fig. 1). This hole was drilled during Leg 112 as part of a transect across the margin to determine the effects of subduction of the Nazca Plate on the depositional history of the accretionary margin of the South American Plate. In addition, the past effects of climate change were investigated along this eastern boundary of the Pacific Ocean circulation system (Suess, von Huene, et al., 1988).

### GEOPHYSICAL WELL LOGS FROM SITE 679

The suite of geophysical logs from Site 679 provides an excellent framework within which to interpret the lithological contrasts observed in the sparsely recovered core from this hole (Fig. 2). Both core and geophysical logging results define three major units in the logged interval of the well from 75 to 330 m below the seafloor (mbsf).

Above an unconformity between the upper Miocene and lower Pliocene, lithologic Unit II (Unit I was not logged) consists of siliceous oozes and mudstones having low natural radioactivity (gamma ray), very high porosities (>60%) that cause low electrical resistivities, sonic velocities that approach seawater values, and low densities. The location of the unconformity separating Units II and III was placed at 103 m, based upon core recovery of a sandstone layer at that depth. Geophysical logs show that the boundary between changes in physical property from Units II to III occurs at 92 m (Fig. 2).

Within Unit III, increases in gamma ray, resistivity, velocity, and density can be seen at 175 mbsf, where a transition occurs from siliceous oozes and mudstones mixed with siltstones above, to predominantly siltstones below (Fig. 2). The overall increase in resistivity, velocity, and density observed in this unit was caused by normal, load-induced compaction.

The upper part of Unit III contains five depositional cycles, best seen on the gamma-ray curve (Fig. 2). Peaks in gamma-ray values occur at 107, 121, 132, 144, and 157 m. Steadily decreasing gamma-ray values below each peak suggest that some sort of slow depositional change caused a decrease in relative burial rate of the radioactive elements potassium, uranium, and thorium. Variations in a windblown or river-carried, clay-rich component would produce such an effect. The sedimentation rate in this interval was as much as 80 m/m.y., from age-dating using nannoplankton and diatoms (Suess, von Huene, et al., 1988). Thus, the average duration of these cycles was approximately 150,000 yr, and these cycles might be related to Milankovitch climate changes.

Unit IV was observed below another unconformity at 255 m. Here, increased gamma-ray, resistivity, velocity, and density values indicate increased mudstone, sandstone, and dolomite and decreased siltstone contents. Porosities decreased from 55% in Unit III to 45% in Unit IV (Fig. 2).

The Unit III/IV boundary provides the acoustic impedance contrast that results in the prominent seismic reflector seen in Figure 1. A synthetic seismogram produced from velocity and density logs reproduces the seismic-reflector sequence.

<sup>1</sup> Suess, E., von Huene, R., et al., 1990. *Proc. ODP, Sci. Results*, 112: College Station, TX (Ocean Drilling Program).

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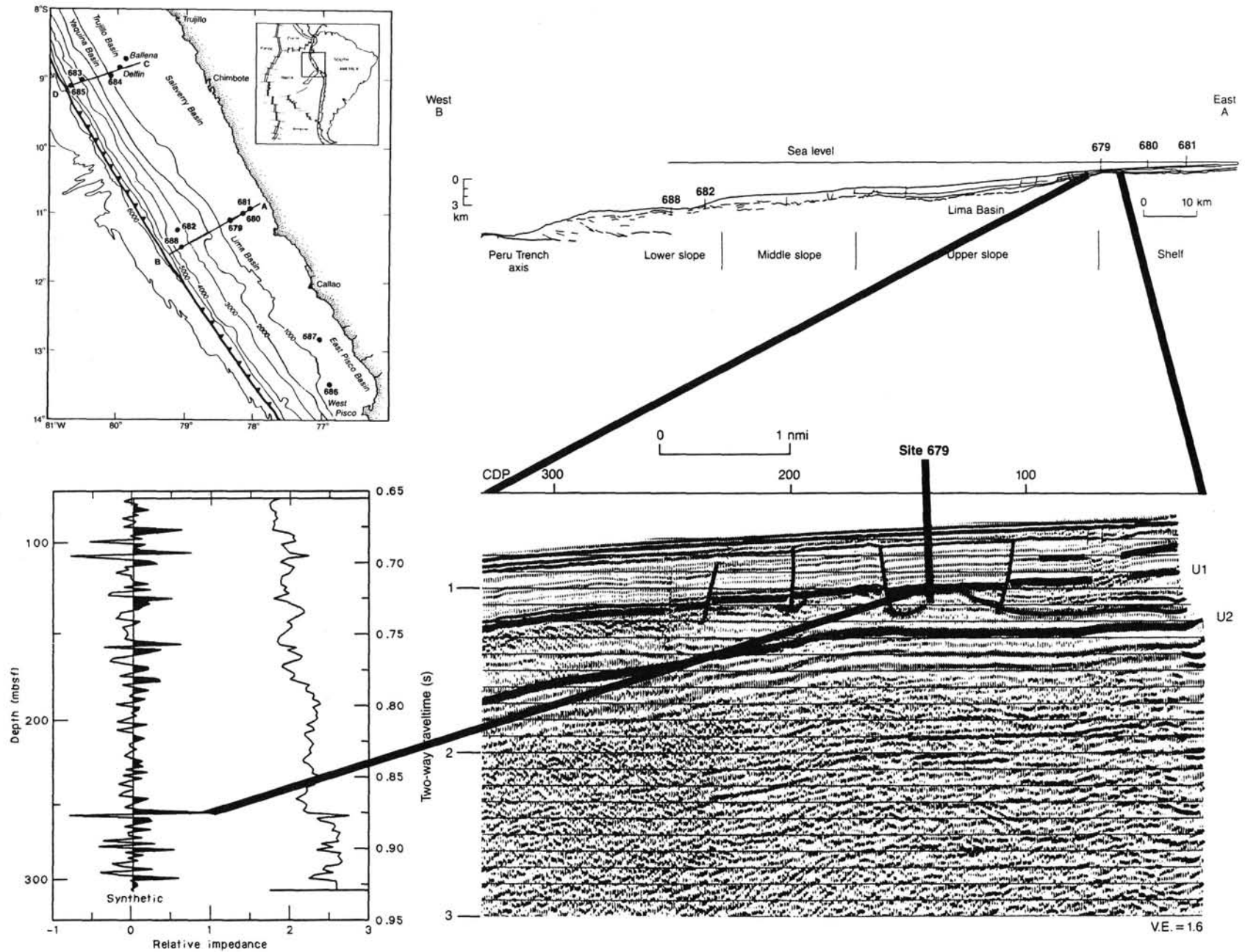


Figure 1. Seismic-reflection profile (right, top) from east to west across Peruvian continental margin and Hole 679 drilled during Leg 112 (left, top). Synthetic seismogram (left, bottom) generated from velocity and density logs in Hole 679 shows lithology change at 255 mbsf that is responsible for prominent reflector in the subsurface (right, bottom) (Figure from Suess, von Huene, et al., 1988).

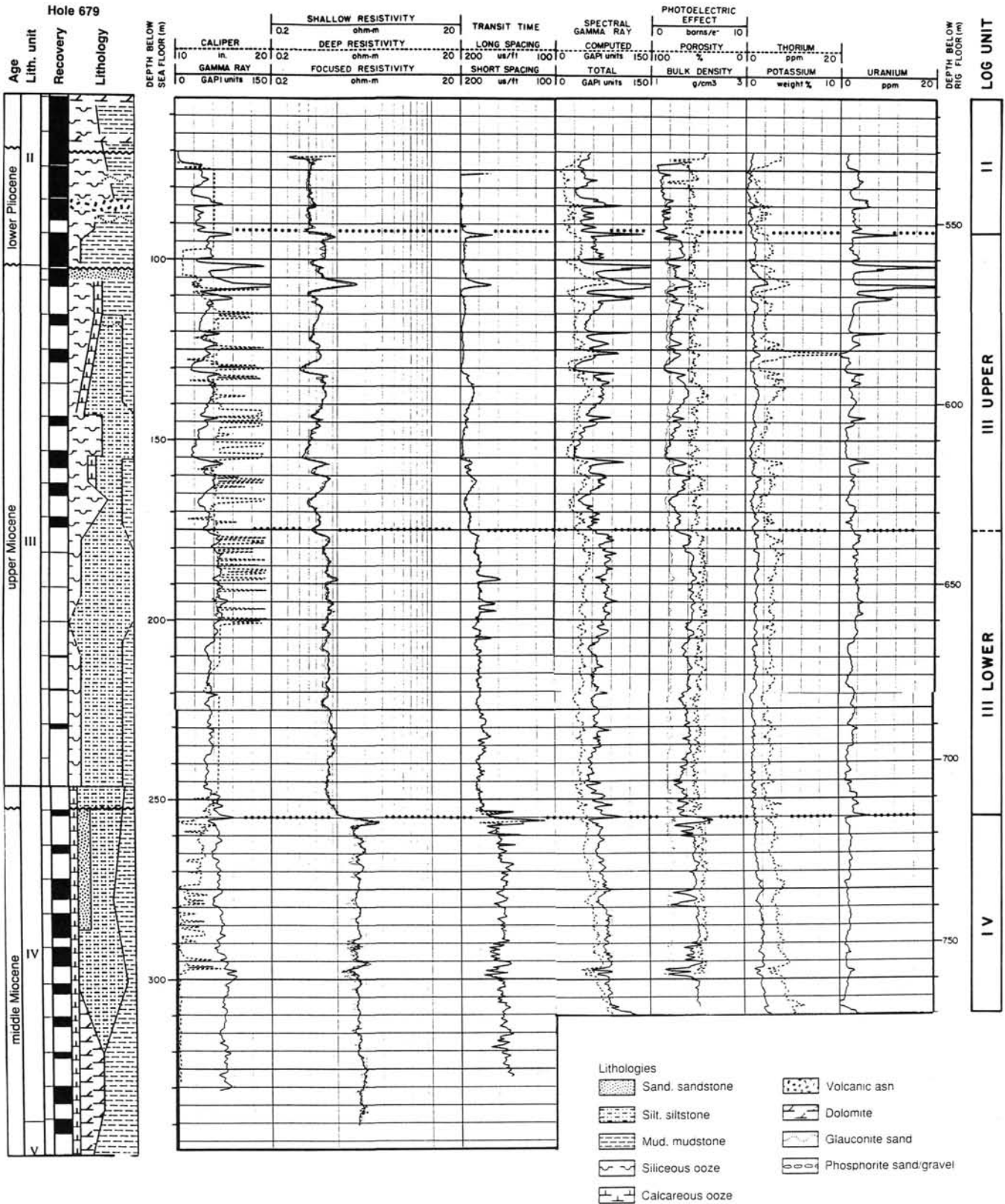


Figure 2. Lithologic column (left) and geophysical logs (right) from Hole 679. Note physical-property changes in logs at lithologic unit boundaries.



## GEOCHEMICAL WELL-LOGGING TECHNIQUE

Geochemical well logging was developed by Schlumberger Well Services for use in sedimentary basins by the oil industry. Scientists are beginning to use this technique for science in drill holes through sedimentary, igneous, and metamorphic rocks (Anderson et al., 1988a, 1988b, 1989). The geochemical logging tool measures the relative concentrations of 11 major and minor elements *in situ*. Precision of geochemical logging measurements within a well is excellent, although accuracy can be considerably poorer than that of analyses performed on core samples in a laboratory (Anderson et al. 1988a, 1988b, 1989).

A geochemical logging tool carries four measurement devices into a borehole: (1) A NaI crystal, natural gamma-ray spectral analyzer detects energy levels of naturally emitted gamma rays from the formation. The weight percent of potassium (K), as well as abundances of thorium (Th) and uranium (U), in parts-per-million, are determined; (2) A californium neutron source activates aluminum atoms in the formation and a NaI scintillation detector records the emitted gamma rays. The weight percent of aluminum (Al) in the rock is subsequently calculated; (3) A pulsed neutron, capture gamma-ray spectrometer measures the abundances of silicon (Si), calcium (Ca), iron (Fe), sulfur (S), and titanium (Ti), as well as the trace abundance of the heavy rare-earth element (HREE) gadolinium (Gd). Hydrogen (H) and chlorine (Cl) contents of the wellbore fluid and formation are also measured. The combination of these analyses with the measurement of (4) the photoelectric capture cross section of the formation using a cesium source of gamma rays allows one to estimate the total concentration of magnesium (Mg) plus sodium (Na) in the rock by a differencing technique (Hertzog et al., 1987).

Gamma-ray spectroscopy measurements performed by the geochemical logging tool detect a fraction of the total spectrum that is linearly proportional to the concentration of each element within the spherical volume of the measurement (approximately 1 m<sup>3</sup>). The relative yields from the measured elements are renormalized to elemental oxides, which are summed to 100% at each sampling depth interval (Hertzog et al., 1987). Since the elements measured by either capture activation or natural spectroscopy make up almost all of the significant oxides that are contained in the rock, calculations should be reliable in all geological formation (Hertzog et al., 1987; Anderson et al., 1988a).

Schlumberger has demonstrated that geochemical logs in sediments yield measurements that can be converted to dry weight percent of elemental oxides, with accuracies of 5% to 20% (Hertzog et al., 1987). Precision of these elemental yields has been tested by up to 10 separate recordings within a well, with individual elemental abundances reproducible to better than 5%. Anderson et al. (1988a, 1988b) showed that geochemical log measurements in basalts, granites, and metamorphic rocks are less accurate (10% to 30%), although precisions remain excellent.

In spite of the poorer accuracy relative to laboratory analyses, geochemical well logs add to the geological reconnaissance capabilities within a drill hole because these logs provide analyses that are fast and continuous over the logged interval of the well. As opposed to the many months required for laboratory analyses of partial sections of a borehole that are recovered during most coring operations, continuous records of relative elemental yields are derived in real-time while logging, and weight percentages of the major elemental oxide and trace-element abundances are available within days at the well site.

## GEOCHEMICAL WELL LOGS IN SITE 679: SOLID PHASES

The geochemical logging tool was run over the interval from 70 to 310 m of Hole 679 (Fig. 3). Major elemental variations further differentiate between lithological units recorded in the well by geophysical logs and the recovered core. Unit II contains lower Si and Al and higher Ca and Mg than in the upper part of Unit III, indicating that there are fewer clays (high Al content) and sands (high Si), and more abundant carbonates (both Ca- and Mg-rich). Dolomite stringers are particularly abundant from 80 to 90 mbsf and can be located on the logs by their low porosities. In both Unit II and the top of Unit III, Fe correlates with sulfur (S), indicating the presence of pyrite.

Several phosphorite-rich zones were recovered in the core (Suess et al., 1987). These phosphorites were deposited by currents that reworked surface sediments during vigorous circulation periods of poleward flow along the margin. Assuming uranium also is concentrated in the phosphorites, several possible phosphorite-rich zones evident in the U log were not recovered in Hole 679 cores; particularly enriched horizons are found at 110, 125, 135, and 160 m. Volcanic-ash layers are distinguished at 85, 95, and 138 m by peaks in Fe, Ti, Mg, Al from clay-rich constituents and by abundant Gd, a heavy-rare earth element.

The lower section of Unit III is distinguished from the upper part of Unit III (boundary at 175 m) by higher Si and Ca contents (more sand and carbonate) and lower Fe, Mg, and Al contents (lower clay content). This was the most poorly recovered unit in the well. The average Ca-content of the core samples was <25%, but the geochemical logging results indicate a somewhat greater abundance of Ca *in situ*.

Unit IV contains higher Si, Fe, and K contents than the lower segment of Unit III, indicating higher sand, feldspar or illite abundances. An interesting peak in Al at 275 m corresponds to low Si and Fe, suggesting the abundant presence of an Al-rich mineral. High permeability sandstone stringers found in the geophysical logs can be seen as Si-peaks that do not correlate with Fe or Al abundances (therefore, they are not clays). The uranium content of these sands is low, possibly because of significant hydrological flow along these high-permeability pathways. Uranium is a highly mobile component of low temperature, relatively fresh, groundwater flow-systems (Williams et al., 1988). The seismic reflector at 255 m is a layer enriched in Ca, Fe, and, particularly, U. This reflector may be a calcite-cemented, permeability cap to this hydrological system. If so, the U was precipitated during the deposition of the calcite-rich cements.

The proposed mobility of U discussed above leads to an important point about the interpretation of geochemical logging results. Any bulk-chemical changes detected by the tool generally are measurements of the contrast in chemistry of the rock, not of the fluid in pore spaces. The pore-water-chemistry changes observed in fluids (like those squeezed from core samples) should be the opposite of the bulk-rock changes measured by the geochemical logging tool because the predominant numbers of cations irradiated by the tool reside in the rock, not in the pore fluid. In analyses of fluid samples from Site 679, Ca<sup>2+</sup> increases and Mg<sup>2+</sup> decreases with depth (Suess, von Huene, et al., 1988). In the geochemical log-derived analyses, CaO weight percentages decrease and MgO increases. Both these changes are consistent with dolomitization occurring in Site 679. The bulk chemistry of the rock reflects the addition of Mg to carbonates, whereas the chemistry of the pore fluids shows the removal of Mg from the fluid and the consequent release of Ca from the rock into the fluid.

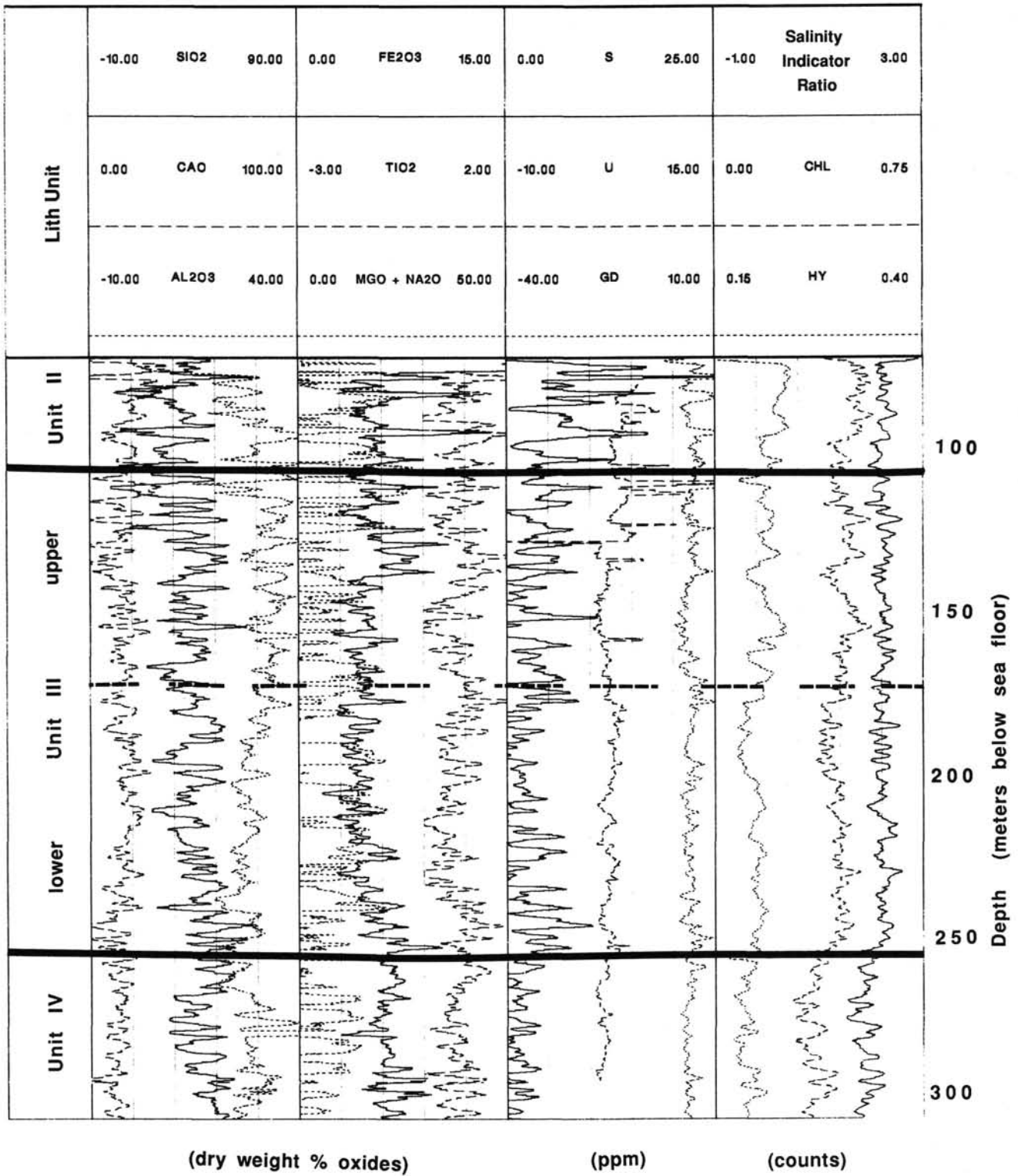


Figure 3. Geochemical logs from Hole 679. Unit boundaries are marked here by changes in composition of the sediments, rather than by physical-property changes, as in Figure 2.

## PORE-FLUID SALINITIES

The abundances of two elements measured by the geochemical logging tool reflect the composition of the pore fluid, rather than that of the rock. Chlorine and hydrogen contents are predominantly from the pore fluid because little H (only as hydroxyls in clays) and no Cl is contained in the rock in Site 679. However, variations occur in the amounts of H and Cl measured by the logging tool at different depths if changes either in the borehole size or the composition of the drilling fluid occur. Consequently, we must correct the elemental abundances for changes in borehole size and carefully monitor the salinity of the drilling fluid.

The caliper curve displayed in Figure 2 is from a mechanical arm that tracks changes in the size of the hole during logging operations. This mechanical-caliper can be unreliable, however, because it can be clogged by formation mud. For this study, we developed a technique for determining hole size more accurately by using traveltimes from the multichannel sonic logging tool. The total traveltime of a sonic pulse from source to receiver is the sum of three traveltimes: (1) that of a wave through the drilling fluid from the source on the logging tool to the borehole wall, (2) that of a refracted wave through the rock along the wellbore, and (3) that of a wave through the drilling fluid, again, to the receiver on the logging tool. Velocities of the refracted wave are determined precisely by the sonic log itself, and drilling fluid velocities are estimated from measurements of temperature, pressure, and mud resistivity from the auxiliary measurement sonde at the top of the tool string. Snell's law defines the geometry of the travel path, so that the distance between the tool and the borehole wall can be calculated at all depths. The sonic-caliper measures average hole diameter much more accurately than the mechanical-caliper (compare Fig. 2 with Fig. 4).

Another unknown is the variation in compositions of Cl and H in the borehole fluid. ODP used no drilling mud of any kind in Hole 679E, largely because the well was washed to within 100 m of total depth before any coring was done. Logging began 24 hr after coring. Seawater was the only drilling fluid used. To verify this, the electrical resistivity of the borehole fluid was measured throughout the logged interval by the auxiliary measurement sonde.

Analyses from the geochemical logging tool were reconstructed using sonic-caliper and mud resistivities to correct for borehole effects (Fig. 3). Abundances of H and Cl then represent concentrations in the formation pore fluid.

Two other difficulties arise when determining accurate pore-fluid chlorinities in the formation. First, downhole changes in porosity result in large variations in the total amounts of H and Cl measured by the geochemical logging tool. The variation in porosity with depth is measured by no less than four different geophysical logs. Neutron porosity (Fig. 2), and sonic, resistivity, and density porosities (Fig. 4) all show similar decreases in porosity with depth in Site 679, although these decreases differ somewhat in detail.

To compensate for these porosity changes, we considered the variation in the Cl/H ratio, termed the "salinity-indicator-ratio" by Schlumberger (Fig. 3), although "chlorinity-indicator-ratio" might be more accurate. If the composition of the formation pore fluid were to remain constant with depth and only porosity were to change, then this ratio would remain constant. Indeed, within Unit III, the Cl/H ratio remains relatively constant (Fig. 3), even though porosity decreases from >60% near the top, to 50% at the bottom of the unit (Fig. 4).

The Cl/H ratio is only a qualitative measure of changes in chlorinity of formation pore fluids because each curve is

derived from a count of emitted gamma rays of appropriate energies received at the detector after each burst of neutrons by the accelerator. The capture cross section of Cl is much larger than that of H, and thus, the count rate of Cl is higher, even though there is much more H than Cl in the pore waters. The "salinity-indicator-ratio" was calibrated by using chloride concentrations (in mM) measured on pore fluids squeezed from the core samples of Hole 679 (Fig. 5).

Another important effect that must be accounted for before pore fluid salinities can be examined is a possible variation with depth in the amount of H that is chemically bound as hydroxyls in clays of the formation. The geochemical logging tool measures not only the amount of clay present, but the composition of the clay because the abundances of the elements K, Fe, and, particularly, Al are largely controlled by clay content in continental margin sediments. An increase in clay content should lower the "salinity-indicator-ratio" by increasing the amount of H present in the formation without increasing Cl in the formation waters. Further, a change in clay composition should change the ratio as well, since the hydroxyl contents of illites, smectites, and kaolinites are different.

From the decrease in Al-content that occurs below the porosity change in Unit III (Fig. 3), there may be a decrease in the total volume of clay of about 10%. Opposing this decrease in the total amount of H bound in the formation are increases in K and Fe contents that suggest the presence of more illite (which contains more H) in the deeper Unit IV. Neither of these changes can significantly affect the Cl/H ratio of the pore fluid because less than 20% of the total clay volume is hydroxyl (and 20% of a 10% change is not significant).

## RESULTS

The Cl/H variation recorded by the geochemical logging tool in Hole 679E (Fig. 3) indicates that pore fluid salinities are near the salinity of seawater throughout Units II and III. However, below the major seismic reflector at 255 mbsf, pore fluids are significantly fresher than seawater (Fig. 5). The porosity change across this boundary, though significant, does not lower the Cl/H ratio (as pointed out previously). The lowered Cl/H ratio is caused by a much greater decrease in Cl than that occurring in H caused by lowered porosity (Fig. 3). Thus, geochemical logging measurements corroborate results from the sparse fluid sample analyses that indicate a salinity inversion exists in this well (Suess, von Huene, et al. 1988). Further, these log-derived results place the transition abruptly at the boundary between Units III and IV; that is, at the impermeable, diagenetic boundary discussed previously (Fig. 5).

In contrast, the pore-fluid salinity pattern observed in analyses of fluids squeezed from cores from Sites 680 and 681, landward of Site 679, indicates the presence throughout each hole of heavily saline brines (Suess, von Huene, et al. 1988). This complex pore-water salinity distribution on the Peruvian margin was attributed by the Leg 112 scientists to three possible sources (Suess, von Huene, et al., 1988): (1) the freshwaters and brines are a remnant of a fossil, Miocene hydrological system formed when the margin was subaerial (freshwater system was isolated geographically from the desiccation of the Salaverry Basin by the topographic high that currently separates them); (2) the freshwaters and brines flow today from the continent as two separate groundwater hydrological systems; or (3) the freshening of groundwaters occurs chemically from the natural dissociation of interstitial chloride during advection upsection from the accretionary prism to the west.



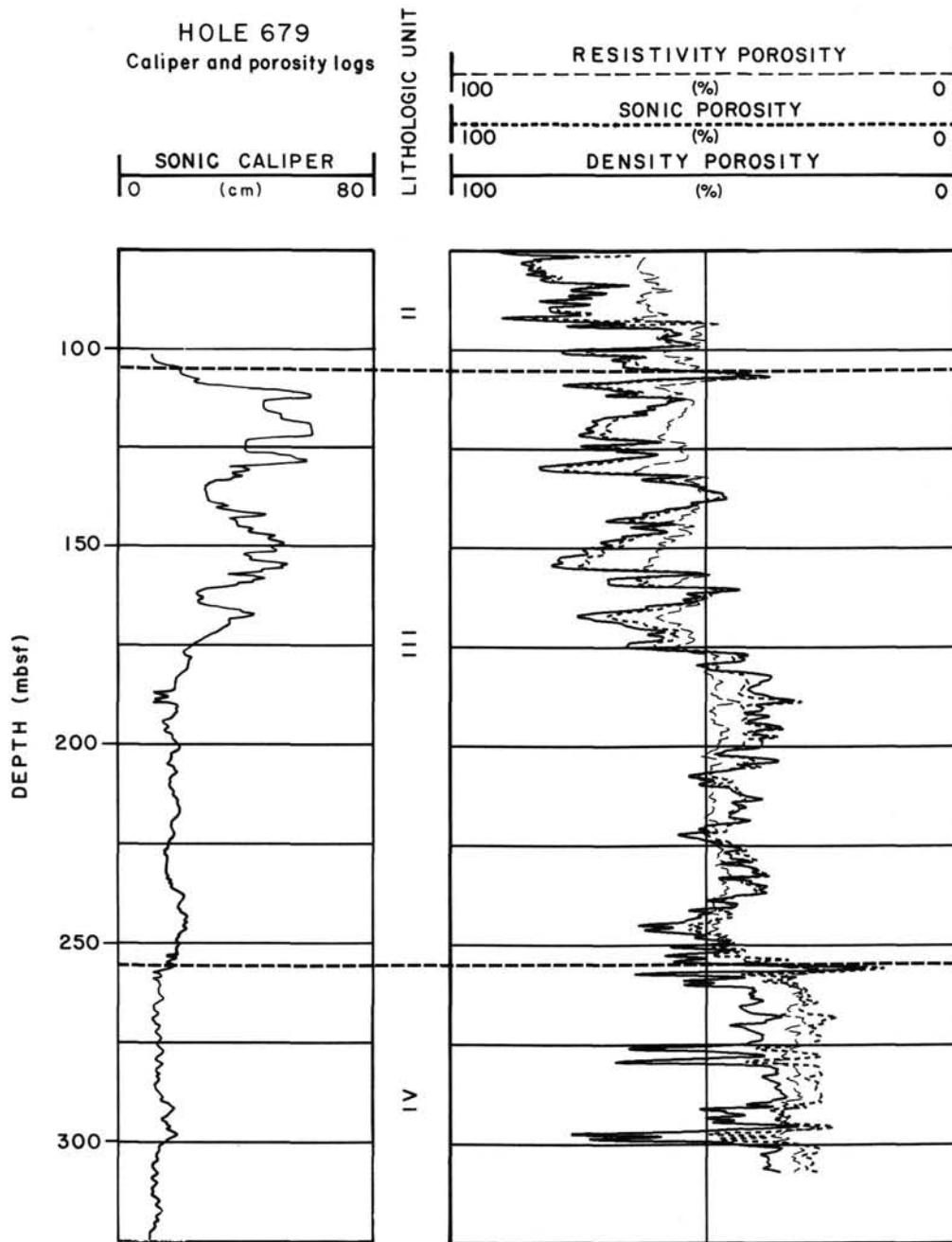


Figure 4. Hole size (left) determined from sonic caliper as in text. Porosity curves (right) determined from sonic, resistivity, and density logs. See Anderson et al. (1988c) for methods of calculation. In brief, the sonic porosity measures the interconnectedness of grains; resistivity porosity is controlled by the contrast in conductivity between water and rock; and density porosity is controlled by the total atomic weight of the formation.

The hydrodynamics of the salinity inversion observed in the geochemical logs from Hole 679 place significant constraints on which of these hypothetical scenarios is more likely to be correct. Preservation of a salinity inversion over significant geological time requires an impermeable cap to prevent the dense, saline fluids above from mixing with the light, fresh pore waters below. It is unlikely that such a disequilibrium system was preserved since the Miocene in actively compacting and dewatering continental margin sediments. In addition, the high-permeability pathways indicated by the geochemical (uranium-loss) and porosity logs suggest

that an active hydrological system is present on the Peruvian margin today. The brines were probably derived from evaporite deposition in the Salaverry Basin in late Miocene time (Suess, von Huene, et al., 1988). These brines must have isolated Site 679, which is considerably seaward of and separated from the Salaverry Basin by a topographic high, from any fresh pore fluid that might flow from the continent (Fig. 5).

The most likely explanation for the occurrence of fresh waters beneath saline waters at Site 679 thus is the freshening of interstitial waters during fluid flow upsection from deeper in

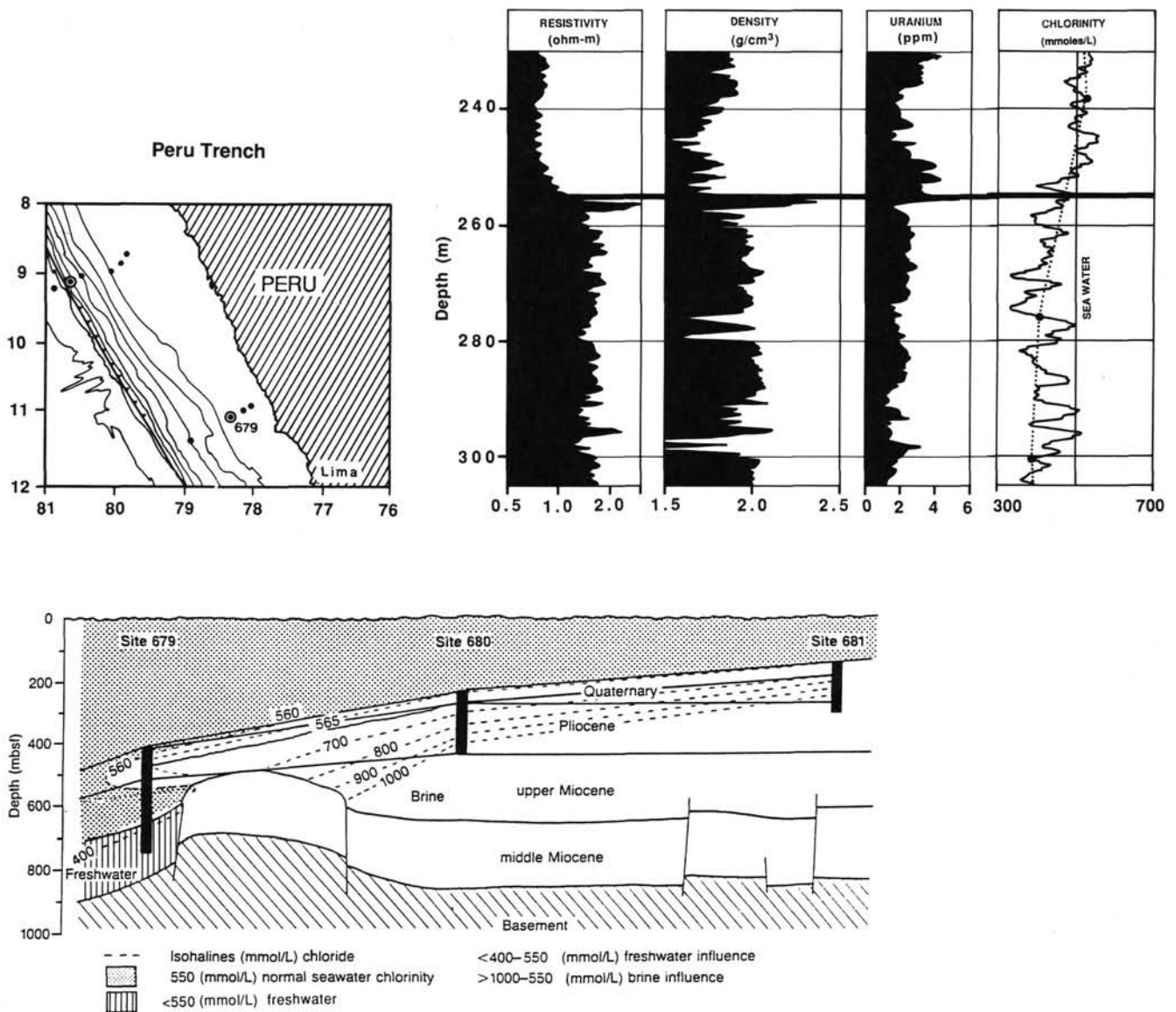


Figure 5. A. Geophysical (resistivity, and density) and geochemical (uranium and salinity from Cl/H ratio) logs in Hole 679, across major lithological and hydrological boundary at 255 mbsf. The salinity inversion from denser, saline pore fluids above, to less dense, fresher pore waters below this boundary was confirmed by analyses of salinity in waters squeezed from core samples (solid dots). B. Cross section through transect of wells drilled during Leg 112. Shading indicates saline pore fluids. Inversion in Hole 679 is unlikely to have been caused by groundwater invasion from the continent because of the topographic ridge and the heavy brine pore fluids of the Salaverry Basin. More likely, dewatered pore fluids from downslope in the accretionary prism invaded the continental margin. These pore fluids would then be fresher than seawater because of scavenging of chloride during advective movement (figure from Suess, von Huene, et al., 1988).

the accretionary prism. Dissociation of Cl during formation of gas hydrates, or clathrates, has been shown to cause large-scale freshening of groundwaters (Hesse and Harrison, 1981). Clathrate deposition was found in deeper sites during Leg 112 (Suess, von Huene, et al., 1988). "Ultra-filtration" during flow of pore waters across clay membranes has also been shown to cause freshening of fluid flow systems in the subsurface (Hanshaw and Coplen, 1973; Marine and Fritz, 1981). Another mechanism that would freshen pore waters is the addition of newly released hydroxyls into the existing saline pore fluid during dehydration of clays at greater depths in the accretionary prism (von Huene and Lee, 1982).

In any event, the effects of differing pore fluid salinities on diagenesis in margin sediments is significant. Fresh pore fluids are depleted in U, organic carbon, and phosphorites, whereas

brines promote the formation of dolomites (Baker and Kastner, 1981) and the deposition of U and organic carbon-rich phosphorites. Consequently, a strong diagenetic boundary exists between the fresh and saline pore fluids (at 255 m in Hole 679).

### CONCLUSIONS

Adding geochemical logs to the standard geophysical logging suite allows one to compare physical-property changes vs. chemical changes that occur in the subsurface in response to the same geological processes. In Hole 679, we found that fresher pore fluids existing below 255 m caused chemical changes in the sediments. Geophysical porosity responses of these same sediments indicated that thin sand layers within the freshwater interval are highly permeable. Geochemical



logs indicated that the boundary between these two hydrological systems is a Ca-cemented, U-enriched layer. Geophysical logs detected the increased velocity, density, and resistivity that produced a prominent seismic reflector at this boundary. This reflector was then mapped away from the drill hole. This drill site provided an excellent example of how geophysical and geochemical logging methods complement each other to provide an extended understanding of the subsurface.

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