# 16. INTERSTITIAL WATER STUDIES, LEG 78A<sup>1</sup>

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#### ABSTRACT

Studies of interstitial water carried out during Deep Sea Drilling Project Leg 78A (Sites 541, 542, and 543) have revealed that alteration of volcanic material dispersed in the sediments is largely responsible for increased concentrations of dissolved calcium and decreased concentrations of dissolved magnesium and potassium. This conclusion is supported by studies of the  $^{87}$ Sr/ $^{86}$ Sr ratio of dissolved strontium, as well as by studies of the oxygen isotopic composition of interstitial waters. At Site 543 a large part of the  $\delta^{18}$ O decrease in the interstitial waters must result from diffusive exchange with underlying basement rocks. Data from studies of interstitial water do not indicate upward advection of fluids at Sites 541 and 542.

### **INTRODUCTION**

During Leg 78A holes were drilled at three sites with the aim of studying the geophysics of a seismically defined deformation front associated with the Lesser Antilles forearc (Fig. 1). At Sites 541 and 542 holes were drilled into the seaward edge of the Barbados Ridge complex, and at Site 543 holes were drilled into the adjacent Atlantic ocean crust. Penetration was not great at either Site 541 or Site 542, because of the drill was unable to penetrate a major deformation zone at 431 m sub-bottom at Site 541 and because the pipe stuck at Site 542. Valuable information was gained, however, on the nature of tectonic processes associated with the subduction of the Atlantic Plate under the Barbados Ridge complex (Moore and Biju-Duval, this volume).

At all three sites, interstitial water samples were obtained throughout the drilled sections, and at Site 541 the *in situ* interstitial water sampler was successfully deployed three times. Here we wish to report both the shipboard geochemical data and the data obtained in our own laboratories.

#### METHODS

For major and minor chemical analyses, the methods described by Gieskes (1974), Gieskes and Lawrence (1976), and Gieskes and Johnson (1981) were employed. Shipboard analyses included Ca, Mg, pH, alkalinity, chlorinity, and salinity. The data are presented in Table 1.

Chloride contents are generally within 1% of the bottom-water value (~19.3 g/kg), except for lower values in one *in situ* sample (P.W. 2) and in a few samples collected at greater depths (541-43-5, 135-150 cm; 541-48-4, 135-150 cm; 543A-7-2, 0-10 cm). These lower values could result from slight contamination with distilled water during the squeezing process. Yet in only one sample (541-43-5, 135-150 cm) is the chlorinity too low by ~ 4%, compared to the bottom water value, and this is

also reflected in low salinity. Concentrations of dissolved ammonia are rather scattered, which may result in part from oxidation of  $NH_4^+$  during storage. The  $^{87}Sr/^{86}Sr$  ratio of dissolved strontium was determined using

The  $\frac{87}{Sr}$  ratio of dissolved strontium was determined using the methodology described by Hawkesworth and Elderfield (1978). The  $\frac{18}{O}$  composition of interstitial waters was determined using the CO<sub>2</sub> equilibration technique of Epstein and Mayeda (1953).

The lithologies of the sites are presented in Figure 2, as well as in the Figures 3, 4, and 5, which contain the interstitial water data.

#### RESULTS

DSDP interstitial water studies have revealed, at sites away from areas of rapid sediment accumulation, a general correlation between observed increases in concentrations of dissolved calcium in the interstitial waters and decreases in the  $\delta^{18}$ O of these waters (Lawrence and Gieskes, 1981; Gieskes and Lawrence, 1981). At many sites linear correlations between increases in calcium content and decreases in magnesium content are observed. Mc-Duff (1981) showed that generally in such cases the source of dissolved calcium and the sink for magnesium are located in the basalts of Layer 2, rather than in the sediments. When correlations between calcium and magnesium contents are non-linear, reactions in the sediment column must be responsible; these reactions will then usually superimpose signals in calcium contentbut particularly in magnesium content-on the signals generated in the underlying basement (Gieskes and Lawrence, 1981). Using analyses of both the <sup>18</sup>O/<sup>16</sup>O composition of the interstitial waters and the 87Sr/86Sr ratio of dissolved strontium, it has been shown that a major contributor to reactions in the sediment column is the alteration of volcanic matter (Gieskes and Lawrence, 1981; Hawkesworth and Elderfield, 1978; Elderfield and Gieskes, 1982; Gieskes, 1983). Considerations of mass balance, based on oxygen isotopic compositions of the interstitial waters, indicate, however, that much of the signal in  $\delta^{18}$ O and presumably in dissolved-calcium content are associated with alteration reactions in Layer 2 of the

 <sup>&</sup>lt;sup>1</sup> Biju-Duval, B., Moore, J. C., et al., *Init. Repts. DSDP*, Vol. 78A: Washington (U.S. Govt. Printing Office).
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Figure 1. A. Regional location map. Note position of deformation front defining eastward boundary of Barbados Ridge complex and Tiburon Rise underthrusting deformation front near Sites 541, 542, and 543 (inset). Water depths in km. B. Detailed map (inset in Fig. 1A) showing positions of Leg 78A sites. Note position of deformation front and locations of seismic reflection lines used for site survey. Water depths in m.

#### Table 1. Analyses of interstitial water, Leg 78A.

Hole	Sample (interval in cm)	Sub-bottom depth (m)	pH	Alk. (meq/L)	S (g/kg)	Ca (mM)	Mg (mM)	Cl (g/kg)	Sr (µM)	<sup>87</sup> Sr/ <sup>86</sup> Sr	Li (µM)	K (mM)	SO4 (mM)	NH4 (μM)	Si (µM)	δ <sup>18</sup> OSMOW ‰
541	2-5, 140-150	8.5	7.40	2.91	36.6	18.5	40.2	19.29	132	0.70852	15.1	11.0	25.9	19	453	-
	5-1, 140-150	31.0	7.49	1.57	36.0	24.1	36.3	19.73	207	0.70832	37.3	9.3		355	375	-0.79
	P.W. 1 <sup>a</sup>	48.5	7.65	2.57	35.5	27.5	32.9	19.46		-		-			· ·	-0.66
	8-4, 140-150	64.0	7.03	1.60	36.6	25.0	31.2	19.66	249	-	22.5	8.4	25.7	199	247	-
	P.W. 2	95.0	8.94	1.70	33.8	24.7	34.6	17.97	220	0.70840	22.5	7.3	23.4	97	118	-0.56
	13-5, 140-150	113.0	7.44	0.73	35.8	26.8	29.1	19.53	279	0.70836	22.0	7.4		80	167	-1.71
	P.W. 3	143.5	8.50	1.23	35.8	32.4	29.5	19.32	326	0.70834	37.5	4.5	24.8	191	153	-1.08
	18-4, 140-150	159.0	7.69	0.53	35.2	31.5	25.8	19.32	326	0.70824	22.0	6.5	-	301	147	-1.34
	23-3, 140-150	205.0	7.58	0.55	38.0	33.5	25.8	19.29	256	0.70841	22.0	5.3	-	27	164	—
	33-4, 138-150	301.5	8.03	0.31	34.9	39.3	16.4	19.19	407	0.70824	51.4	5.0	25.2	231	149	-1.31
	38-5, 135-150	350.5	8.20	1.05	35.8	41.2	19.2	19.27	220	0.70746	37.5	4.4		246	119	(-1.13)
	43-5, 135-150	398.5	8.26	1.00	33.6	42.1	18.0	18.40	190	-	60.0	3.9		280	97	-1.84
	48-4, 135-150	450.0	7.00	1.55	35.2	42.9	20.9	18.94	166	0.70787	134.9	7.3	24.8	240	443	—
542	H1-6, 0-07	0-88	7.47	0.99	36.8	12.0	48.5	19.51	218	0.70844	22	8.7	25.4	135	184	—
	H2-3, 142-150	88-154	7.45	0.79	35.2	30.0	29.9	19.48	303	0.70836	37	7.0	25.7	308	169	—
	H3-2, 125-135	154-202	7.90	0.38	36.3	37.0	25.2	19.51	355	0.70812	37	6.5	-	125	154	—
	1-5, 125-135	209	7.72	0.50	35.8	35.7	28.1	19.51	333	0.70803	37	5.5		201	147	-1.34
542A	3-2, 140-150	252	7.78	0.91	35.2	41.6	27.3	18.97	196	0.70746	53	4.1		21	147	(-3.59)
	8-4, 135-150	312	7.82	0.49	34.9	42.9	27.3	19.10	204	0.70750	74	4.6	25.4	227	117	-2.31
543	2-4, 140-150	16	7.37	2.69	36.3	13.9	46.1	19.48	114	0.70881	15	10.6	29.2	38	315	0.00
	5-3, 140-150	43	7.60	1.94	36.0	17.4	44.3	19.38	113	0.70835	15	9.4	31.4	129	182	-0.09
	9-5, 140-150	84	7.44	1.79	36.0	24.2	38.4	19.34	113	0.70812	15	7.6	-	97	172	-0.062
	19-4, 140-150	178	6.55	1.34	35.8	28.7	36.0	19.31	112	0.70801	82	7.5		149	927	-1.09
	26-4, 140-150	244	6.45	1.21	35.8	30.8	40.3	19.27	125	0.70877	120	7.0	28.9	113	1045	-1.52
	29-4, 140-150	273	6.13	0.85	36.0	32.6	39.1	19.27	112	0.70820	134	7.2	_	61	1000	-1.97
543A	2-1, 140-150	333	6.86	1.46	36.6	32.7	41.9	19.39	131	0.70821	105	6.9	-	198	619	-1.63
	7-2, 0-10	381	7.05	1.49	36.0	41.0	39.2	18.90	125		105	4.4	25.7	22	257	- 3.01
	10-1, 37-47	408	7.14	1.07	36.3	39.2	39.4	19.34	130	0.70795	97	5.5	27.8	91	368	-2.41

Note: — indicates no data; H indicates wash core; data in brackets are of uncertain quality. <sup>a</sup> P.W. = in situ





Figure 2. Lithostratigraphy and structural geology at Sites 541, 542, and 543.

ocean crust, and that the overprint resulting from alteration of volcanic matter in the sediments is relatively small (Lawrence et al., 1979; Lawrence and Gieskes, 1981).

The sediments cored during Leg 78A contain fairly large amounts of volcanic material. Shipboard interstitial water data on concentrations of dissolved calcium and magnesium indicate a nonlinear correlation between these elements at all three sites drilled (Fig. 6). This led us to decide on a concentrated program of study, not only of the major constituents of the interstitial waters, but also of the oxygen isotopic composition of the interstitial waters and the isotopic composition of dissolved strontium ( $^{87}$ Sr/ $^{86}$ Sr). This study constitutes the first intensive effort using this combined approach. In the following sections we discuss our observations at each site, and then conclude with more general discussions of the implications of the data.

#### Site 541

Notwithstanding fairly high recent rates of sediment accumulation, little sulfate reduction has occurred in the sediments at this site; this is also attested by the relatively small increases with depth in the concentration of dissolved ammonia. Alkalinity values show decreases with depth (Fig. 3), suggesting removal of bicarbonate from the interstitial waters, presumably as calcium carbonate. The relatively small decrease in alkalinity, however, would not noticeably affect trends in dissolved-calcium content. Of some interest is the consistent difference between in situ alkalinities and the alkalinity data from shipboard squeeze tests, the latter of which show lower values. This difference may result in part from calcium carbonate equilibria, as was shown for surface sediments by Emerson et al. (1982) and Jahnke et al. (1982). For the other components (Ca, Mg, Sr, 87Sr/86Sr, Si, Li, and K), the agreement between in situ values and shipboard data is generally very good; the data on  $\delta^{18}$ O (cf. correlation of Ca and  $\delta^{18}$ O, Fig. 6) may perhaps be slightly displaced.

The most important feature of Hole 541 is the rapid increase in dissolved-calcium content with depth in the upper 50 m (Fig. 3). With this are associated rapid decreases in dissolved-magnesium content and in the  ${}^{87}$ Sr/  ${}^{86}$ Sr ratio, as well as in the  $\delta^{18}$ O of the interstitial waters. The correlation between changes in concentrations of dissolved calcium and magnesium (Fig. 6) is almost lin-



Figure 3. Interstitial water profiles, Site 541. Age units: I = Quaternary, II = Pliocene, III = lower Miocene, IV = middle Miocene, V = lower Miocene; see Figure 2. Dashed line = contemporaneous seawater. Open symbols = *in situ* data.

ear (note that sampling artifacts do not necessarily imply an exact seawater end-member). The correlation between changes in dissolved-calcium content and  $\delta^{18}$ O is linear, within the accuracy of the data. There are two mechanisms by which these data can be explained: (1) the pore-water profiles are dominated by reactions involving alteration of volcanic matter, which is an important component in the upper sediment section; or (2) the curvature in the various concentration-vs.-depth profiles results from upward advection of interstitial waters from lower horizons. In a subsequent section we will argue against the second interpretation.

Evidence that alteration of volcanic matter contributes to the observed increases in dissolved-calcium content and decreases in magnesium and potassium contents comes from the sharp decrease in the isotope ratio of dissolved strontium. This rapidly decreases to a value of ~0.7083, well below the contemporaneous seawater value. The low ratio implies exchange with volcanic matter (Hawkesworth and Elderfield, 1978, Elderfield and Gieskes, 1982). In a subsequent section we shall return to this subject.

### Site 542

Hole 542 was "washed" to  $\sim 200$  m sub-bottom, so that sediments retrieved from the core barrel could have come from anywhere in the washed interval (indicated by bars in Fig. 4). The concentration-vs.-depth profiles,

therefore, are tentative and drawn by analogy with those for Hole 541.

Again, relatively large overall decreases in <sup>87</sup>Sr/<sup>86</sup>Sr with depth indicate that substantial exchange with volcanic material must have occurred. Of some interest are the lower <sup>87</sup>Sr/<sup>86</sup>Sr values in the lower Miocene section of Hole 542A (cf. the low value for Hole 541 in sediment of the same age). Perhaps more active exchange occurs in these sediments, or perhaps carbonate contributions are smaller—this problem will be addressed in a subsequent section.

The major-constituent profiles are very similar to those for Hole 541.

#### Site 543

Site 543 is the so-called Oceanic Reference Site, and was drilled to basaltic basement.

Throughout the Site 543, alkalinity decreases with depth, and the profiles of calcium and magnesium concentrations (Fig. 5), which are nonlinearly correlated, clearly indicate that, especially for magnesium (minimum at ~100 m sub-bottom), but also for calcium, reactions involving volcanic material must occur in the sediments. This again is indicated by the low  $^{87}$ Sr/ $^{86}$ Sr values for the upper sediment column. The correlation between calcium and magnesium contents is nonlinear, and is quite different from those for Sites 541 and 542. If relatively less removal of magnesium occurs in the up-



Figure 4. Interstitial water profiles, Site 542 (combining data from Table 1 for Holes 542 and 542A). Age units: I = Quaternary, II = Pliocene, III = lower Miocene; see Figure 2. Dashed line = contemporaneous seawater.



Figure 5. Interstitial water profiles, Site 543 (combining data from Table 1 for Holes 543 and 543A). Age units: I = Quaternary, II = Pliocene, III = lower Miocene, IV = middle Miocene, V = lower Miocene, VI = Oligocene, VII = Eocene, VIII = Paleocene, IX = Upper Cretaceous; see Figure 2. Dashed lines = contemporaneous seawater (1: Hart and Staudigel, 1978; 2: Burke et al., 1982).

per sediment column, and if a substantial contribution to the calcium signal originates in the lower-lying sediments or basalts, the observed correlation between calcium and magnesium contents could be expected. The Ca- $\delta^{18}$ O correlation (Fig. 6) does indicate curvature, with values of  $\delta^{18}$ O near the bottom of Hole 543A being  $\sim -3\%$ . This nonlinear correlation between Ca concentration and  $\delta^{18}$ O suggests production of calcium in the sediments, with a relatively small associated signal in  $\delta^{18}$ O. We shall return to this problem in the following section.

Data for dissolved strontium suggest little carbonate influence. Dissolved-silica content reflects biogenic silica, and lithium appears to be released in the siliceous zone. Uptake of potassium is indicated in the upper sediments, as well as in the basal sediments or underlying basalts.

# ALTERATION OF VOLCANIC MATERIAL

The distribution of <sup>87</sup>Sr/<sup>86</sup>Sr at all three sites clearly demonstrates that at Sites 541 and 542 all ratios are below contemporary seawater values, as they are in the upper 200 m of sediments at Site 543. At Site 543 no significant increases in dissolved-strontium content with depth are observed, so we can estimate the volcanic contribution to the <sup>87</sup>Sr/<sup>86</sup>Sr ratio. For this we will take <sup>87</sup>Sr/<sup>86</sup>Sr to be 0.708 at 100 m sub-bottom (for contemporaneous seawater, <sup>87</sup>Sr/<sup>86</sup>Sr  $\approx$  0.709). If volcanic material were to have an <sup>87</sup>Sr/<sup>86</sup>Sr ratio of 0.703 before alteration, then the contribution by volcanic material to the <sup>87</sup>Sr/ <sup>86</sup>Sr ratio of dissolved strontium would be ~17%. This should be a minimum estimate because it ignores diffusion and assumes a relatively low ratio in volcanic matter (Elderfield and Gieskes, 1982). Thus, exchange with volcanic material in the sediments must be substantial, though of course in the absence of data for strontium in the solid phase we cannot estimate the amount of volcanic matter involved.

In Hole 541 at ~150 m sub-bottom, the ratio  ${}^{87}\text{Sr}/{}^{86}\text{Sr}$  is ~0.7083. At this depth the concentration of dissolved strontium is ~300  $\mu$ M, and contemporaneous seawater has an  ${}^{87}\text{Sr}/{}^{86}\text{Sr}$  ratio of ~0.709. Allowing for input from contemporaneous carbonate, (i.e., with regard to the increase of dissolved strontium), one calculates that volcanic material has contributed at least 30% of the observed  ${}^{87}\text{Sr}/{}^{86}\text{Sr}$ . Of particular interest, of course,



Figure 6. Correlations between Ca and Mg (A) and  $\delta^{18}$ O (B) for Sites 541 ( $\bullet$ ), 542 ( $\blacktriangle$ ), and 543 ( $\blacksquare$ ).  $\odot = in situ$  values.

is that changes in the concentration of dissolved magnesium over depth are also larger at Site 541 ( $\Delta$ Mg ~ 25 mM), compared with those at Site 543 ( $\Delta$ Mg ~ 17 mM). These changes are approximately proportional to the volcanic contributions to the <sup>87</sup>Sr/<sup>86</sup>Sr signal, and would therefore support the contention that magnesium uptake is associated with alteration of volcanic matter. Of course, we realize that comparisons of this nature are not based on extensive information.

The lower <sup>87</sup>Sr/<sup>86</sup>Sr ratios in lower Miocene sediments at Sites 541 and 542 can be understood in terms of lesser contributions from contemporaneous carbonates (lower dissolved-strontium concentrations).

Previously we observed that at Site 541 a substantial portion of the  $\delta^{18}$ O must have been generated in the sediments, presumably as a result of alteration of volcanic matter. With a mean value of  $\delta^{18}$ O of -1%, one estimates that in a closed system about 5-15 m of ash must be responsible for the decrease in  $\delta^{18}O$  (Lawrence and Gieskes, 1981). This estimate could be increased when the possibility of diffusive exchange with the overlying ocean is also considered. However, this estimate would require alteration of  $\sim 20\%$  of the sediments in the upper 150-200 m of the sediment column. From the data on the 87Sr/86Sr ratio, we estimated that volcanic contributions to the changes in magnesium and calcium at Site 543 were substantially less than at Site 541. For this reason the expected  $\delta^{18}$ O signal would also be much less, so that the reactions in the upper 100 m at Site 543 would cause only a minor perturbation in the  $\delta^{18}$ O-vs.-depth profile, which at this site seems to be governed mainly by reactions occurring in the underlying basalts. The nonlinearity in the Ca- $\delta^{18}$ O correlation for Site 543 (Fig. 6) is caused mainly by abundant production of calcium in the upper sediment column, accompanied by a relatively small depletion in  $\delta^{18}O$  and a significant decrease in <sup>87</sup>Sr/<sup>86</sup>Sr. Though basalt alteration causes a substantial depletion in  $\delta^{18}$ O (Lawrence and Gieskes, 1981), the decrease in <sup>87</sup>Sr/<sup>86</sup>Sr associated with this process appears very small (Elderfield and Gieskes, 1982). This makes <sup>87</sup>Sr/<sup>86</sup>Sr a very sensitive indicator of contributions by volcanic matter dispersed in the sediments to changes in calcium and magnesium concentrations at many DSDP sites (Gieskes and Lawrence, 1981).

# **CONTEMPORANEOUS** <sup>87</sup>Sr/<sup>86</sup>Sr CURVE

Recent studies of the 87Sr/86Sr ratio in carbonate rocks as a function of age (Burke et al., 1982) have revealed a curve that largely deviates from previous estimates, particularly for Tertiary and Cretaceous rocks (Peterman et al., 1970; Dasch and Biscaye, 1971; Veizer and Compston, 1974). In a recent paper, Elderfield and Gieskes (1982) reviewed the available body of data on the 87Sr/ <sup>86</sup>Sr ratio of dissolved strontium in interstitial waters and showed that at many sites alteration of volcanic matter leads to 87Sr/86Sr ratios considerably lower than contemporaneous seawater values. In their study, use was made of the polynomial developed by Hart and Staudigel (1978) for the contemporaneous 87Sr/86Sr curve. With the proposed new curve of Burke et al. (1982), many sites still show deviations below contemporaneous <sup>87</sup>Sr/<sup>86</sup>Sr values. Thus, even at sites with carbonates, the influence of alteration of volcanic matter may not be negligible, as was suggested by Elderfield and Gieskes (1982) and Elderfield et al. (1982).

Hole 543 is relatively free of carbonate, and in Figure 5 we present the contemporaneous seawater curves of Hart and Staudigel (1978) and of Burke et al. (1982). Of interest is the close agreement of our data, below 300 m sub-bottom, with the curve of Hart and Staudigel. Of course, little carbonate is present at this site, with the possible exception of the basal sediments. Deviations of our data from the curve of Burke et al. (1982) may well result from diffusion and from reactions in the upper sediment column having gained recent significance; this could cause a non-steady-state profile in  ${}^{87}\text{Sr}/{}^{86}\text{Sr}$ , leading to the minimum at ~100 m sub-bottom (cf. magnesium minimum at same depth). At present we are undertaking a study of carbonate sediments with the specific purpose of investigating further how the alteration of volcanic matter may affect the distribution of  ${}^{87}\text{Sr}/{}^{86}\text{Sr}$  in both the interstitial waters and carbonates.

# **ADVECTION IN THE SEDIMENTS?**

Drilling at Site 542 was terminated because of inadvertent "packing" of Hole 542A, with clear evidence of overpressure at the base of the hole, possibly associated with migration of interstitial fluids (see the Site Report for Site 542, this volume. In addition, temperature measurements during the logging program (Davis and Hussong, this volume) indicated that relatively warm waters did flow up the hole after penetrating or contacting the downgoing sediment slab. This again raises the question of possible fluid migration through the sediments before drilling was initiated. The further question remains, however: in what direction would such migration take place upward or lateral?

In situ temperature data (Davis and Hussong, this volume) for Site 541 did indicate a possible linear temperature gradient with depth, though the instrumental data are of very low quality. Linear temperature gradients would rule out any significant upward advective flow (Crowe, 1981), though some such flow might occur. We therefore decided to examine our concentration-vs.-depth profiles for indications of upward migration of fluids. Because diffusion coefficients of salts are much lower than diffusion coefficients of heat curvature in interstitial water, gradients might offer evidence of upward advective flow, albeit at low rates (Crowe, 1981).

Neither the distribution curves of dissolved strontium nor the <sup>87</sup>Sr/<sup>86</sup>Sr profiles suggest significant upward advection at Sites 541 and 542. This is especially clear from a comparison with the profiles for Site 543, the Oceanic Reference Site. At this site no advection should occur, and we have already shown that concentration gradients for calcium and magnesium can best be understood in terms of alteration of volcanic matter in the sediment column. As noted before, the calcium-magnesium correlation is nonlinear; that is, reactions must be at least partly responsible for observed concentration changes in these components. Profiles for dissolved lithium suggest no significant upward advection either at Site 541 or at Site 542.

We conclude that if advection of interstitial fluids is associated with the subducting sediment slab, this advection is not occurring in an upward direction.

#### ACKNOWLEDGMENTS

This research was supported by grants NSF-OCE-80-23966 (JMG), NERC GR3/4444 (HE), and NSF-OCE-80-24044 (JRL).

We thank the shipboard chemist Dr. David Ripley for his diligent work, Dr. J. Natland for discussion, and Drs. R. McDuff and F. L. Sayles for their reviews of the manuscript.

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Date of Initial Receipt: February 18, 1983 Date of Acceptance: August 10, 1983