18. THE CONCENTRATION AND ISOTOPE COMPOSITION OF SULFUR FROM THE GALAPAGOS MOUNDS AREA SEDIMENTS, LEG 70, DEEP SEA DRILLING PROJECT[1]

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INTRODUCTION

Data obtained while investigating the mounds area near the Galapagos Spreading Center demonstrate the direct influence of solutions derived from the interaction of seawater and young oceanic crust on the sedimentary cover. Investigation of metalliferous sediments from the mid-oceanic ridges, the Galapagos mounds, and the FAMOUS-area zone formations have shown that this influence and the resulting products are dependent on composition, temperature, and conditions of solution input.

The study of sulfur in upwardly migrating solutions and the interaction of these solutions with sediments is of great interest. Investigations of different types of hydrothermally derived formations (Edmond, et al., 1979; Spiess et al., 1980; Styrt et al., 1981; Rosanova 1976; Grinenko et al., 1978) have shown the significant role of sulfur-bearing minerals in deposits formed from high-temperature solutions. In contrast, the addition of hydrothermal sulfur is negligible in those metalliferous sediments that precipitated as a result of the interaction between the solutions and open seawater (Bonatti et al., 1972, 1976; Gordeev et al., 1979; Migdisov, Bogdanov, et al., 1979). For example, sulfides are absent in clearly oxidized metalliferous sediments from the East Pacific Rise (EPR). Barite sulfur from these sediments is identical with seawater sulfate sulfur in isotope composition (Grinenko et al., 1978). Gurvich and Bogdanov (1977) have suggested that barium from EPR metalliferous sediments results completely from biological activity and from the components of ocean waters.

Edmond et al. (1979) report that low-temperature springs from the Galapagos Rift axis contain two types of solutions: those with and those without H_2S.

Cores obtained during Leg 70 of the Deep Sea Drilling Project (DSDP) present useful material for studying the role of sulfur in the formation of mounds, which are assumed to result from the interaction of hydrothermal solutions with sediments. These cores contain sedimentary sequences from mounds and off-mounds that are almost undisturbed by drilling. Sediments from low heat-flow areas, which lacked the influence of hydrothermal solutions, were also sampled in detail. This permitted us to compare sulfur concentration and sulfur-isotope composition in sediments and pore waters from both low heat-flow areas and the mounds region. The occurrence of worm burrow tubes of pyrite (Fig. 1) in the lower mounds sediments (compare hydrothermal smectites in burrows within the lower mounds pelagic oozes) also provided a reason for searching for hydrothermal sulfur in the mounds sections.

ANALYTICAL PROCEDURE

The separation of sulfur from sediments and determination of pyrite and monosulfide were performed using the method suggested by Volkov and Zhabina (1977), with a determination limit of 0.001%.

Separation of sulfate from pore waters as $BaSO_4$ was carried out, the error being less than 0.01% (±0.006% from the data of parallel, duplicate analyses). Isotopic analysis of SO_2 was performed by means of the MI-1305 mass spectrometer, following the procedure suggested by Ustinov and Grinenko (1965); the experimental error was 0.3‰. $\delta^{34}S$ data were expressed as parts per mil relative to the meteorite standard.

RESULTS

The data obtained on pyrite sulfur in sediments and sulfate sulfur in pore waters, as well as on the isotope composition of the sulfur, are shown in Tables 1 to 4 and Figures 2 to 5. In plotting the graphs, we also used the results of C_{org} determinations performed on board the *Glomar Challenger*.

Sulfide and Sulfur Sediments

The content of sulfide sulfur was determined in 56 samples from 11 hole sections. Monosulfide occurs in amounts below the limits of determination. The contents of pyrite sulfur varied significantly in analyzed samples.

In green clays from the hydrothermal mounds, the content of pyrite sulfur was very low. In nine samples, which occurred mainly as pure granulated smectite, these clays were too poor in sulfur (<0.001%). In weight samples (mainly fine grained and mixed with pelagic-ooze smectites) the amounts of pyrite sulfur in smectite varied from 0.005% to 0.039%, with an average of 0.019%. Averaging all determinations from 17 samples of green clays yields a pyrite-sulfur content of 0.009%.

Upper pelagic sediments from the mounds sections were practically lacking in pyrite sulfur (<0.001%). Alternating with smectite, pelagic oozes from hydrothermal units varied in pyrite-sulfur concentration by three orders of magnitude (<0.001%–0.156%), the average being 0.200%. Great variation was also observed in pyrite-sulfur concentrations in the lower pelagic sediments

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[1] Honnorez, J., Von Herzen, R. P., et al., *Init. Repts. DSDP*, 70: Washington (U.S. Govt. Printing Office).

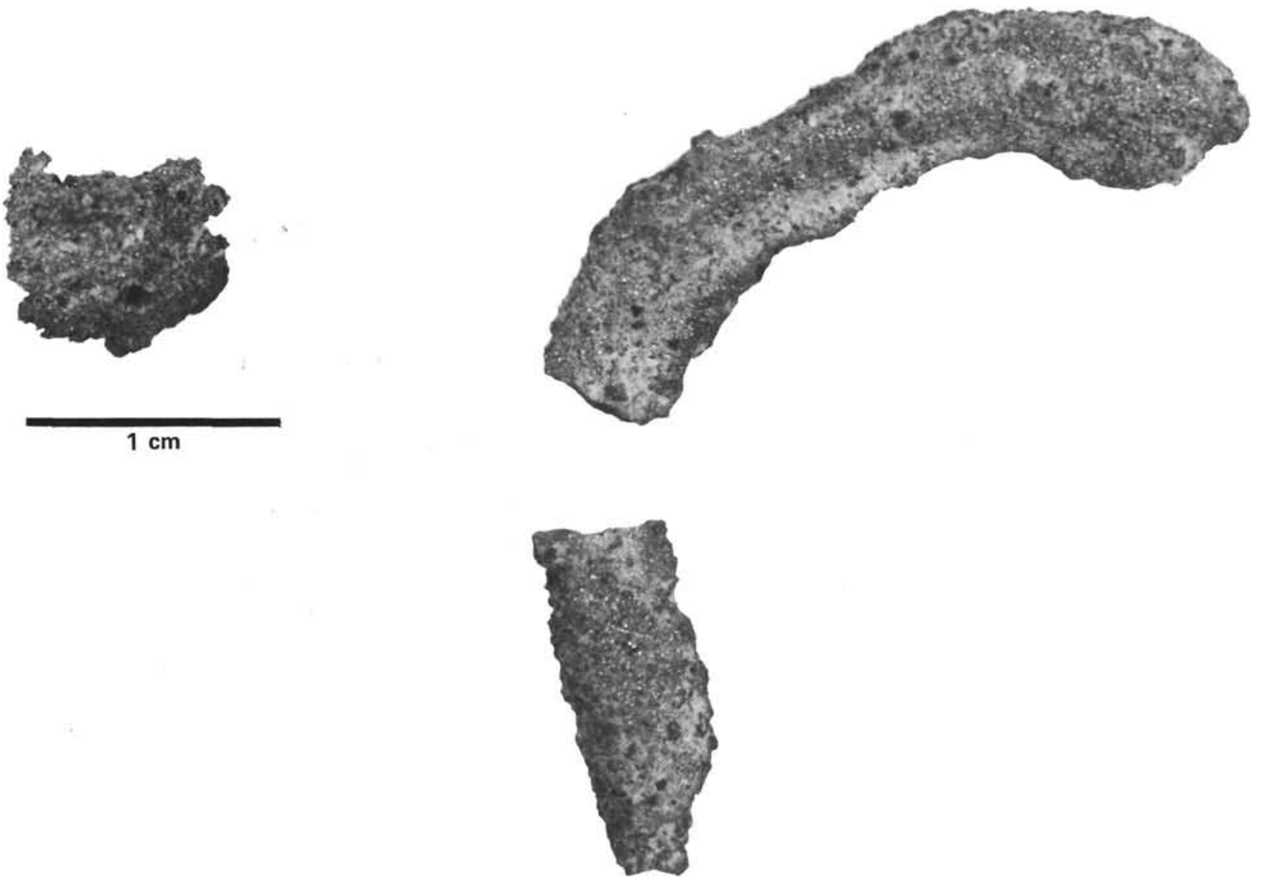

Figure 1. Pyrite worm tubes from the lower pelagic sediments, Hole 506C.

of the mounds (0.103%-1.196%). In these layers occurs the maximum concentration of S_{pyr}, and this is also where sulfide worm tubes were found, which measured up to 5 cm in length (Fig. 1).

The highest dispersed pyrite concentrations were localized in some burrows. The parts of the burrows with high pyrite concentrations were also enriched by a brown opaque material assumed to be organic carbon.

Off-mounds sediments (Holes 507C, 507H, and 509) were characterized by lower amounts of pyrite-sulfur content with wide fluctuations (0.011%-0.389%). Minimum concentrations in off-mounds sections were found in the deepest sediments. In other parts of the sections, pyrite distribution is more homogeneous (0.101%-0.389%). All but the deepest low heat-flow area sediments (Holes 508 and 510) revealed rather small S_{pyr} content variations (0.079%-0.135% and 0.109-0.422% for Holes 508 and 510, respectively). Some depletion in pyrite in the upper oxidized layer (S_{pyr} = 0.049%) should also be mentioned. The basal horizon of Hole 508, which belongs to a lithified layer of partly recrystallized foraminiferous sediment, was practically lacking in S_{pyr}. Low concentrations of S_{pyr} in basal sediment of other holes from that site (e.g., Hole 508C) have been confirmed as a regular characteristic in this region.

The isotope composition of pyrite sulfur in analyzed samples is fairly homogeneous and characterized by a high accumulation of light-sulfur isotopes ($\delta^{34}S$ = −40%). Only three exceptions can be noted. In one sample, from an alternating layer of smectite pelagic ooze, slightly lower ^{32}S accumulation is observed ($\delta^{34}S$ = −37.6‰). The same situation is peculiar to samples from the deep horizons of Hole 508 ($\delta^{34}S$ = −35.8 to −32.9‰).

Sulfate Sulfur of Pore Waters

Sulfate concentrations in pore waters from mounds-area sediments vary within narrow limits which are close to the values in seawater (0.086%-0.112%). In different types of mounds sediments, the average contents of sulfate sulfur are practically identical (0.093%-0.095%). The average sulfate-sulfur concentration of pore waters from off-mounds sediments is very close to these values (0.100%).

The isotopic composition of sulfate sulfur from pore waters from the mounds formation also varied slightly ($\delta^{34}S$ = 18.2-20.6‰). The average isotope composition of sulfate sulfur from different types of mounds sediments was similar and close to that of seawater ($\delta^{34}S$ = 19.2-19.8‰). In sediments from low heat-flow areas, occurrences of H_2S were observed in both Hole 508 (with 35 m of sediments) and, especially, Hole 510 (with 110 m of sediment cover). In pore waters from both holes, high NH_3 content was detected as a result of microbiological sulfate reduction; in pore waters from Hole 510 high concentrations of H_2S (up to 360 m) were

Table 1. Concentration and isotope composition of sulfur in sediments, Site 506.

Sample (interval in cm)	Pyrite Sulfur (wt.%)	Pyrite Sulfur $\delta^{34}S$ (‰)
Hole 506		
1-1, 45–47	<0.001	n.d.
2-1, 56–60	<0.001	n.d.
3-1, 52–54	<0.001	n.d.
3-2, 143–146	<0.001	n.d.
4-3, 123–126	<0.001	n.d.
6-1, 46–48	0.008	n.d.
6-2, 62–65	0.005	n.d.
6-3, 47–49	0.122	−46.6
7-3, 53–57	0.561	−37.6
8-1, 3–5	0.001	n.d.
8-1, 98–100	0.030	−46.3
Hole 506C		
2-2, 42–44	0.018	n.d.
3-1, 33–39	0.039	n.d.
6-2, 107–109	0.181	−42.6
6-2, 113–115	0.296	−44.8
6-2 pyrite worm tube		−43.0
6-3, 64–69	0.068	−43.7
8-1, 91–93	0.014	n.d.
8-1, 102–104	0.026	n.d.
8CC pyrite worm tube		−42.5
Hole 506D		
1-1, 99–103	0.036	−41.2
2-1, 81–85	0.207	−48.3
3-2, 54–57	0.103	−46.2
6-1, 39–45	1.196	−43.4
8-1, 98–105	0.142	−45.5

Note: n.d. = not determined.

Table 2. Concentration and isotope composition of sulfur in sediments and pore water, Site 507.

Sample (interval in cm)	Pyrite Sulfur		Sulfate of Pore-Water		Sulfur
	(wt.%)	$\delta^{34}S$(‰)	(no.)	(wt.%)	$\delta^{34}S$ (‰)
Hole 507C					
1-3, 121–125	0.165	−41.8			
Hole 507D					
3-3, 59–63	<0.001	n.d.			
3-3, 78–89			64	0.089	n.d.
4-1,			65	0.092	20.6
5-1, 75–80	<0.001	n.d.			
5-3,			69	0.096	18.2
6-3, 117–121	0.029	−45.2			
8-3, 56–60	0.182	−48.9			
8-3,			73	0.098	19.7
9-3, 8–10	0.121	−45.2			
9-3, 100–110			75	0.092	19.8
10-1, 12–14	0.017	n.d.			
Hole 507F					
1-1, 89–91	<0.001	n.d.			
1-1, 140–150			76	0.092	19.7
3-1, 54–58	0.298	−50.6			
4-2, 56–60	0.365	−46.4			
4-3			82	0.090	18.6
7-1, 139–150			87	0.097	19.7
Hole 507H					
6-1, 53–55	0.101	−45.1			
8-1, 115–119	0.011	n.d.			

Note: n.d. = not determined.

Table 3. Concentration and isotope composition of sulfur in sediments and pore water, Site 509.

Sample (interval in cm)	Pyrite Sulfur		Sulfate of Pore-Water Sulfur		
	(wt.%)	$\delta^{34}S$(‰)	(no.)	(wt.%)	$\delta^{34}S$ (‰)
Hole 509B					
1-1, 150			118	0.086	18.6
3-1, 150			119	0.095	20.5
3-2, 50			120	0.103	19.5
3-2, 68–72	<0.001	n.d.			
4-2, 7–12	<0.001	n.d.			
5-1, 18–20	0.454	−48.1			
6-2, 28–30	0.115	−48.1			
7-3, 70–74	0.164	−45.4			
Hole 509					
2-3, 139–150			124	0.111	19.0
3-3, 50–52	0.151	−40.3			
3-3, 130–140			127	0.092	18.3
4-3, 110–125			130	0.097	19.2
4-3, 80–82	0.359	−49.3			
6-2, 91–95	0.273	−48.1			
6-2, 140–150			135	0.092	18.4
8-1, 19–21	0.042	−45.3			
8-1, 139–150			139	0.112	19.7

Note: n.d. = not determined.

also determined (Bender, this volume). Sulfate sulfur concentrations in pore waters from these sediments reflect less variation than can be expected as a result of active sulfate reduction. In Hole 508, sulfate-sulfur concentration was 0.088% to 0.098%, and $\delta^{34}S$ was 18.1 to 20.00‰. Limits of sulfur-content variation in Hole 510 waters are 0.079% to 0.090%, and $\delta^{34}S$ values of this sulfate varied from 21.5 to 25.4‰. The occurrences of hydrogen sulfide in the pore waters suggest that sulfide may be partly oxidized into sulfate either in sediments or during pore-water sampling and analysis.

DISCUSSION

The data presented here suggest that the pure hydrothermal smectites are practically lacking in reduced sulfur. There is a tendency for sulfide to increase with depth, which correlates roughly with an increase in reduced iron content in smectites. Low S_{pyr} content in the green clays is clearly related to transition of the pure granulated smectites to fine-grained smectites mixed with pelagic oozes. For this reason, the admixture of pelagic sediment is thought to be responsible for the conservation of sulfide sulfur from parent pelagic oozes

Table 4. Concentration and isotope composition of sulfur in sediments and pore water, Holes 508 and 510.

Sample	Pyrite Sulfur		Sulfate of Pore-Water Sulfur		
(interval in cm)	(wt.%)	$\delta^{34}S$(‰)	(no.)	(wt.%)	$\delta^{34}S$ (‰)
Hole 508					
1-2, 48–50	0.049	−46.6			
1-2,			91	0.089	n.d.
3-3, 14–18	0.117	−41.1			
4-1,			99	0.088	18.6
5-3, 120–124	0.079	−47.7			
8-1, 82–84	0.121	−35.8			
8-2, 33–35	0.135	−32.9			
8-2,			113	0.098	18.1
8-3,			114	0.094	20.4
8-3, 63–64	0.001	n.d.			
Hole 508C					
1-1, 132–136	0.016	n.d.			
	0.011	n.d.			
Hole 510					
2-1, 133–137	0.187	−47.4			
	0.143	n.d.			
3-1,			163	0.085	21.5
3-1, 134–138	0.422	−44.6			
4-2, 133–137	0.304	−46.3			
4-2,			165	0.090	22.2
5-2,			167	0.079	24.0
6-2,			169	0.086	25.4
6-5, 135–139	0.181	−49.0			
8-1, 83–87	0.109	−46.3			

Note: n.d. = not determined.

in hydrothermal sediments. This hypothesis is confirmed by the isotope composition of pyrite sulfur from the hydrothermal smectite (Sample 507D-6-3, 117–121 cm). The value obtained ($\delta^{34}S$ = −45.2‰) is considered identical to those from the mounds and off-mounds pelagic oozes ($\delta^{34}S$ = −40 to −51‰), which are characterized by extremely high accumulations of the light-sulfur isotope.

The isotope compositions of sulfide sulfur from the low heat-flow area ($o^{34}S$ = −41 to −49‰) are similar to those from the mounds regions. It must be stressed that accumulation of light sulfur (^{32}S) in sulfides is also observed in the deepest sediments investigated (up to 110 m sub-bottom depth in Hole 510). This peculiarity in the isotope composition of sulfide sulfur is assumed to be common in pelagic sediments in the area investigated. This is confirmed by the results of previous investigations (Migdisov et al., 1978) of the upper 4-meter-thick sediment layer of the SE Pacific (3°S–4°N; 86°W–101°W), including the area under discussion (Figs. 6 A,B). The values of $\delta^{34}S$ of pyrite dispersed in the sediments are very homogeneous and characterized by high ^{32}S accumulation. The average values of $\delta^{34}S$ and S_{pyr} concentrations are considered to be background levels for pyrite sulfur concentrations and isotope compositions in sediments of this whole area. One should note the close similarities between data obtained for the mounds and low heat-flow areas and background values. All the data presented support the thesis (Migdisov, Girin, et al., 1979) that high accumulation of ^{32}S in sulfides during the process of sulfate reduction (providing the process is taking place) is obtained in pelagic sediments at deep water depths and rather slow rates of accumulation of sediments. The common occurrence of the peculiarity is emphasized by Goldhaber and Kaplan's (1974) data on $\delta^{34}S$ values of pyrite sulfur from pelagic sediments in the Pacific ($\delta^{34}S$ up to −48‰) and the results of our investigation (Migdisov, Girin, et al., 1979) of pelagic sediments from the Indian Ocean ($\delta^{34}S$ = −46 to −48‰).

It is known (Kaplan and Rittenberg, 1964; Goldhaber and Kaplan, 1974) that accumulation of the light sulfur isotope results from the process of bacterial sulfate reduction, which is controlled by the rate of S^{-2} generation by cell. In turn, the rate of S^{-2} generation depends on sulfate concentration in the water, organic carbon content, and other nutrients present in the bacteria's environment.

For the sediments under discussion, the C_{org} content is found to be, on average, 0.4 to 0.6%. Similar concentrations are characteristic of the whole area's upper layer of sedimentary cover (Migdisov et al., 1978). Only the more deeply buried sediments from Hole 510 differ in elevated amounts of organic carbon (average C_{org} = 1.0%). Hence, all but Hole 510 sediments are assumed to contain moderate amounts of organic carbon. Most likely the high rate of sulfide generation cannot be provided by these concentrations.

Sulfate content in pore waters from the upper sediments and its sulfur-isotope composition is observed (Migdisov et al., 1978) to be similar to that of seawater ($S_{SO_4}{}^{2-}$ = 0.094%–0.104%; $\delta^{34}S$ = +19.2%–+21.0%). The data for pore waters from high heat-flow areas are close to the values ($S_{SO_4}{}^{2-}$ = 0.092%–0.112%; $\delta^{34}S$ = +18.3%–+19.7‰). Pore-water analysis (see Bender, this volume) showed that sulfate reduction occurs only in the upper horizons of the sediments not depleted in sulfate. The stability of the sulfate concentrations in pore water also suggested that the process of bacterial sulfate-reduction is restricted by the upper horizons of the sedimentary layer. These conditions are in accordance with a high accumulation of ^{32}S in sulfides.

In sediments from Holes 508, where direct evidences of H_2S generation at depth of the sedimentary sections were obtained, this process affects only slightly both sulfide formation and the isotope composition of sulfide sulfur. For example, in sediments from Hole 510, where the generation of H_2S was found to be maximum, the sulfide is characterized by the same isotope composition as in the upper layers of pelagic sediments. Bacterial reduction of sulfate in a closed or partly closed system (Goldhaber and Kaplan, 1974) results in a decrease in the sulfate-sulfur content and in accumulation of the heavy-sulfur isotope. In fact, we observed the peculiarities for Hole 510 pore waters ($\delta^{34}S$ up to +25.4‰). But if the process occurs in a closed system, isotopically heavier sulfides (compared with the upper horizons) will be formed. The absence of isotopically heavy sulfide sulfur in the deep layers of the hole section is probably

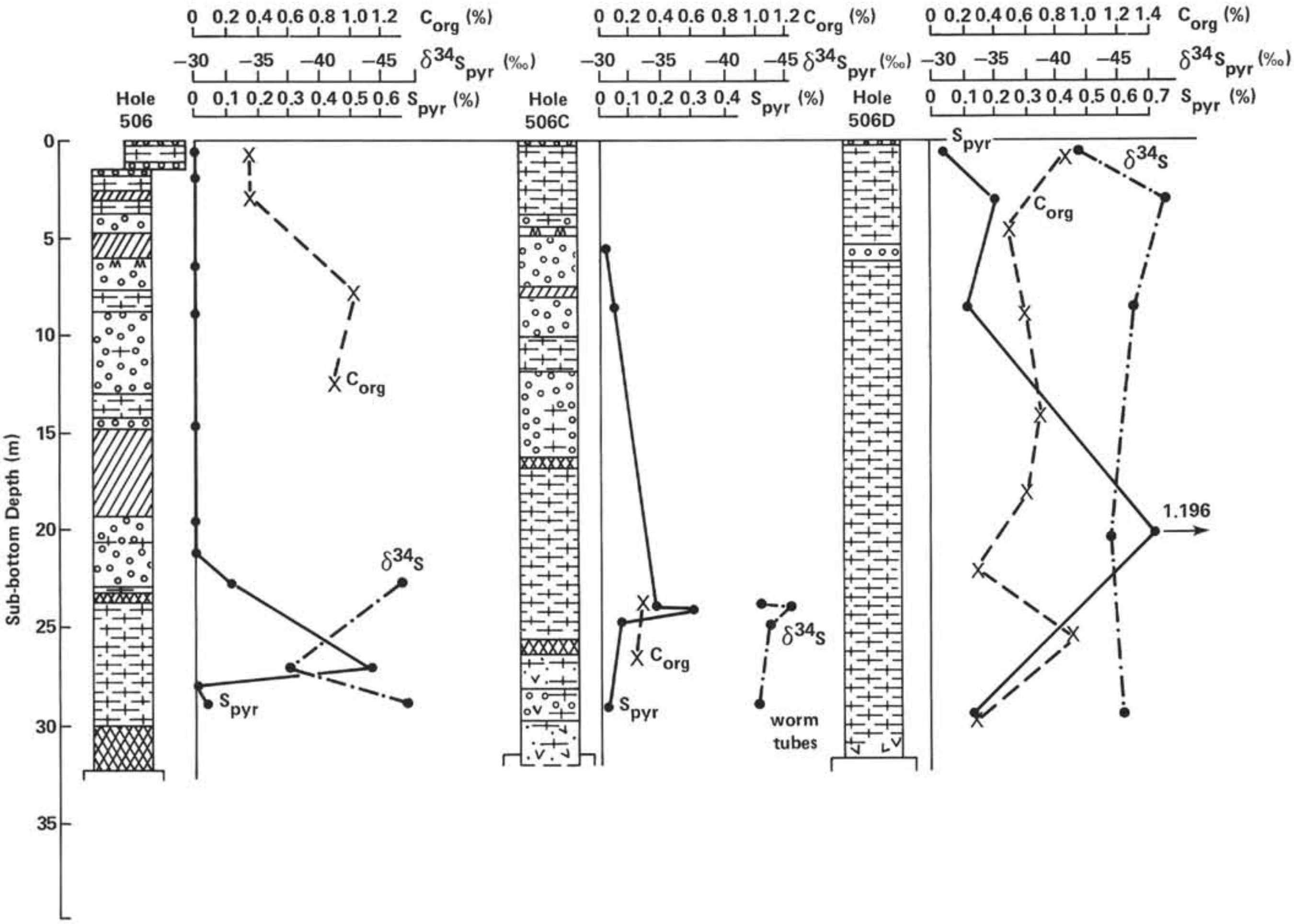

Figure 2. Concentrations of pyrite sulfur and organic carbon, plus $\delta^{34}S$ of pyrite sulfur, in sediments from Site 506.

related to a slight degree of sulfide pyritization. It has been mentioned repeatedly that the intensity of the process is limited by the ferrous iron content in sediments (Berner, 1964; Volkov, 1961; Strakhov, 1960; Goldhaber and Kaplan, 1974). It is probable that as a result of a low content of ferrous iron in sediments (FeO = 0.5%–0.6% in Hole 510 sediments and 0.4–1.6% in oozes from Hole 508) the H_2S generated within the depth of sedimentary layer has not had opportunity to form significant amounts of pyrite and too slight additions of newly formed pyrite are observed.

The only observation of fixation of isotopically heavy sulfide sulfur is in the lowest part of the sedimentary section of Hole 508. This contradicts the evidence on lower-sediment sulfate reduction compared with Hole 510 sediments, with decreasing NH_3 in pore waters with depth (Bender, this volume), and with the absence of any enrichment of pore-water sulfate in heavy-sulfur isotopes. It is thought that this lower-sediment sulfide enrichment is not related to common sulfate reduction processes in a closed system. In fact, the lower horizon of the section from Hole 508 is a layer of recrystallized, lithified carbonate lacking in siliceous tests. Well-crystallized but sparse crystals of pyrite were observed here, in contrast to the common framboidal form of pyrite in other horizons of pelagic sediments. In analytical determinations of pyrite sulfur, only traces or negligible amounts were observed. Dissolution, subtraction, and partial recrystallization of pyrite in the lowest layers of Hole 508 are thought to be associated with silica output and lithification of carbonate ooze (see Hole 508 core description) in this horizon. Mobilization of ferrous iron in this process (including dissolved pyrite), with the addition of generated and isotopically heavier H_2S, are seen to lead to formation of pyrite with a higher content of $\delta^{34}S$ above the partially lithified layer.

In general, it can be concluded that sulfide-sulfur minerals form mainly in the upper horizons of sediments in the area investigated. It is probable that a sufficient content of sulfate and limited concentration of organic carbon in sediments (and perhaps great ocean depths) led to a small rate of S^{2-} generation, and results in high ^{32}S accumulation in pelagic oozes.

Examples of highly dispersed sulfide-sulfur content (> 1%) and even occurrences of pyrite concentration in mounds-area pelagic sediments are not common. Their occurrence suggests bioturbation, implying both the presence of sulfate and a local enrichment of organic carbon. Meanwhile, the sulfur-isotope composition of the concentrated form of pyrite is identical to that dispersed in sediments. This implies that pyrite from the sediments is being incorporated into worm tubes.

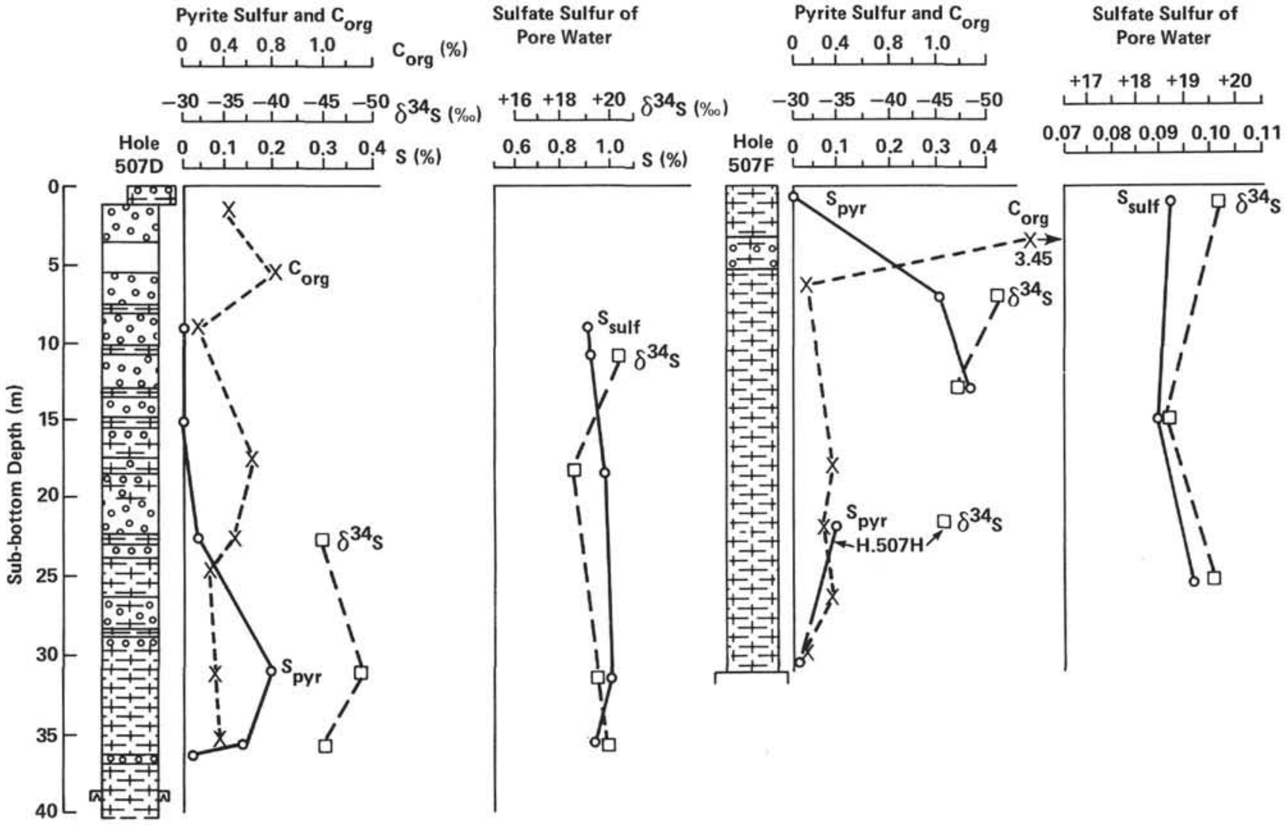

Figure 3. Concentrations of sulfide and sulfate sulfur and organic carbon, plus $\delta^{34}S$ of pyrite and sulfate sulfur values, in Site 507 sediments and pore waters.

The most remarkable fluctuation of sulfide-sulfur content observed in mounds pelagic ooze is presumably connected with the influence of hydrothermal solutions. The effect of these solutions percolating upward through the sedimentary cover on siliceous-carbonate oozes resulted in dissolution of silica and carbonate and formation of iron-rich smectites and manganese-oxide crust; the chemical reactions of this process have been proposed by Honnorez et al. (1981). One peculiarity of the process is the oxidized form of iron in the main resulting product—that is, in smectite. Hydrothermal solutions, in a reduced state, must be oxidized by oxygen (or MnO_2, NH_3, etc.) during the smectite formation. This suggests that oxidation must occur during the interaction between solutions and sediments and that reduced components in both must be oxidized. Thus, oxidation of organic carbon, and oxidation and dissolution of pyrite should be observed. This leads also to redistribution and inhomogeneity in the distribution of reduced minerals in mound oozes. Traces of pyrite, sometimes occurring in slightly reduced lower smectites (especially in mixed ones) confirm the relationship between pyrite content (or its preservation) and redox conditions in an interacting solution-ooze system.

It is probable that intensive circulation in the mounds will prevent us from tracing the oxidation of sulfides in sediments. Unusually low values of $\delta^{34}S$ of sulfate sulfur in mounds pore water ($\delta^{34}S$ to 18.2‰) may reflect this process. Nevertheless, the data suggest that the solutions are lacking in hydrothermal sulfur, or that its concentration is very small, assuming that the process of hydrothermal sediment formation is proceeding at the present time.

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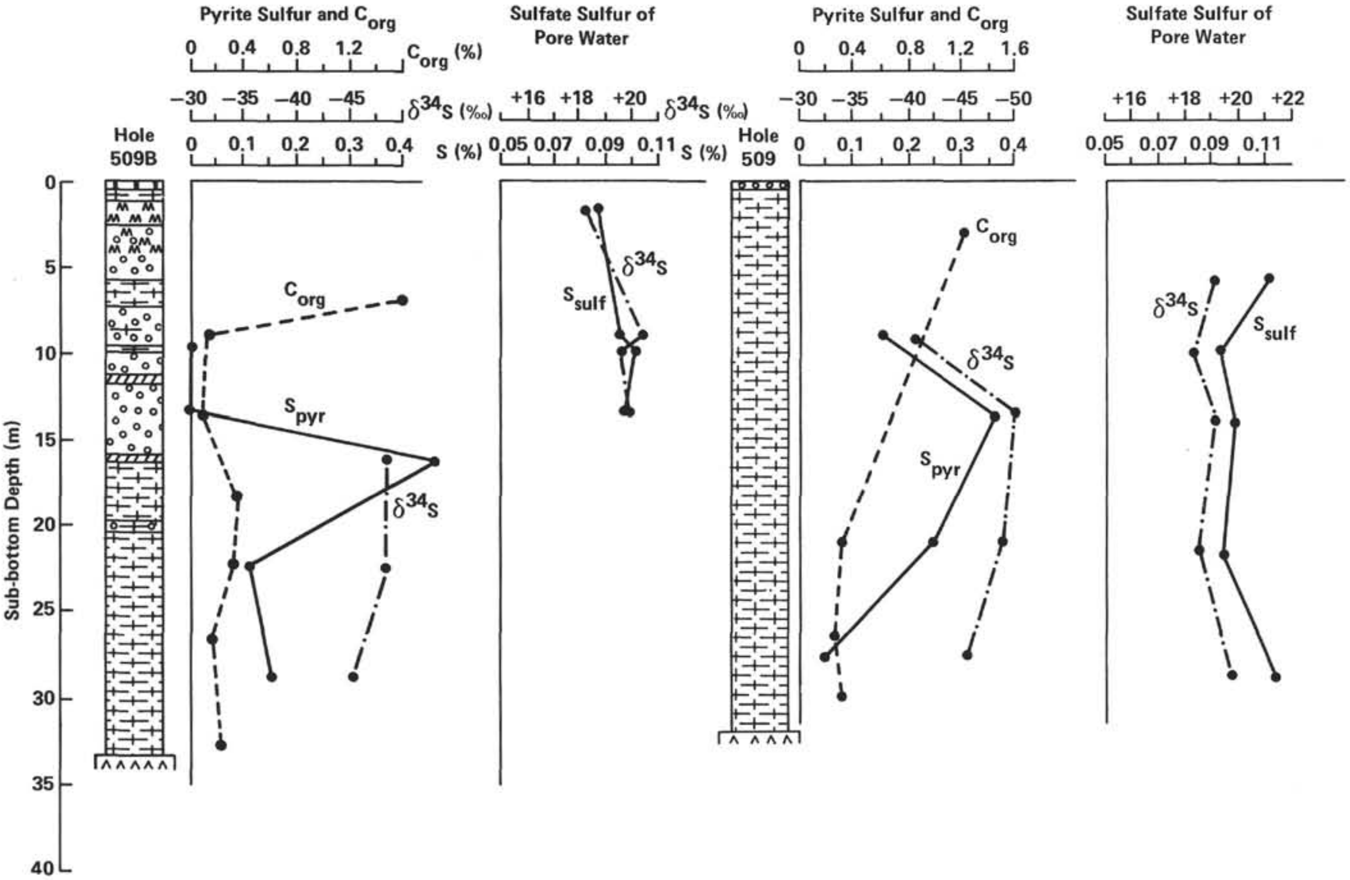

Figure 4. Concentrations of sulfide and sulfate sulfur and organic carbon, plus $\delta^{34}S$ of pyrite and sulfate sulfur values, in Site 509 sediments and pore waters.

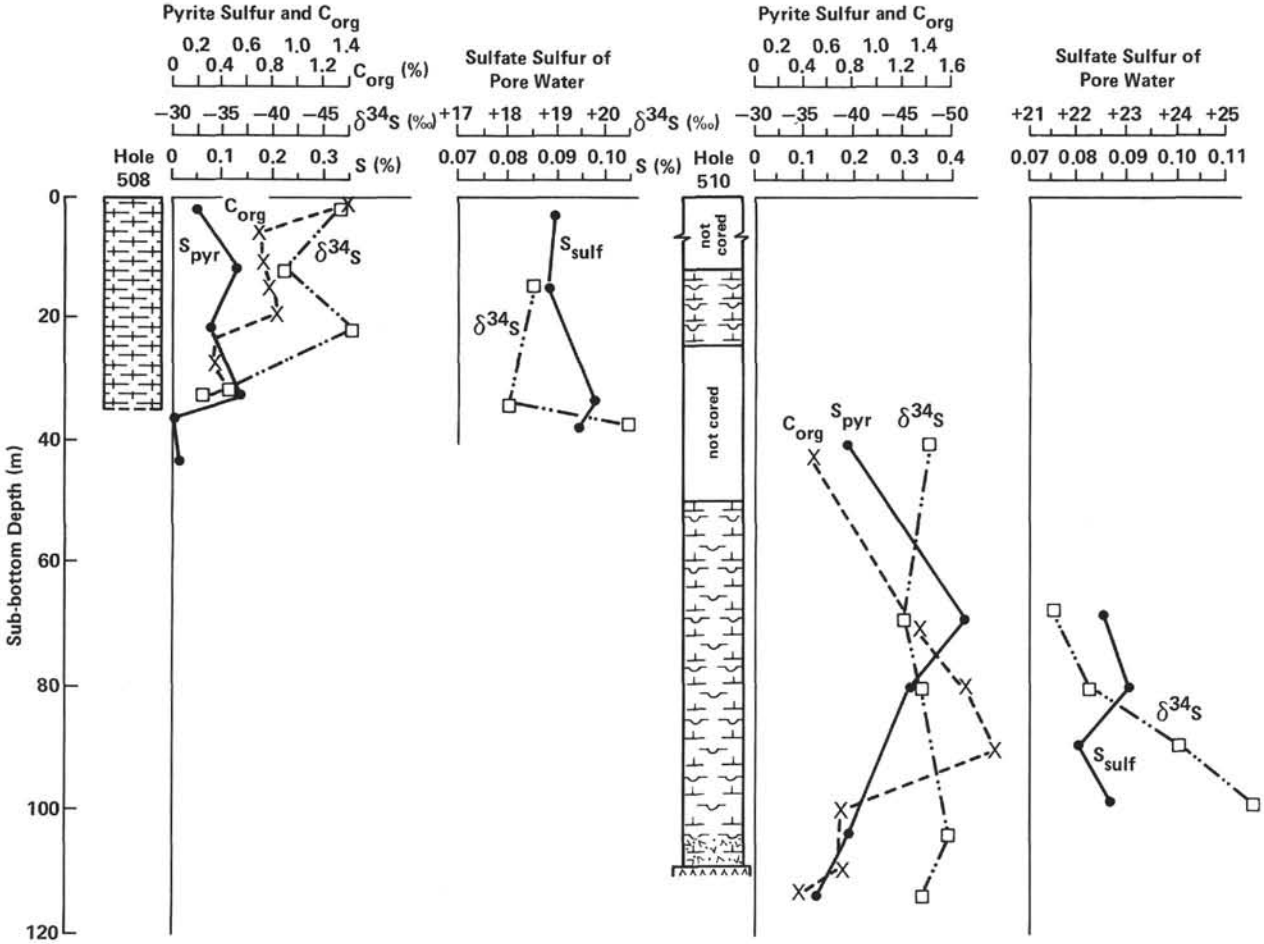

Figure 5. Concentrations of sulfide and sulfate sulfur and organic carbon, plus $\delta^{34}S$ of pyrite and sulfate sulfur values, in Holes 508 and 510 sediments and pore waters.

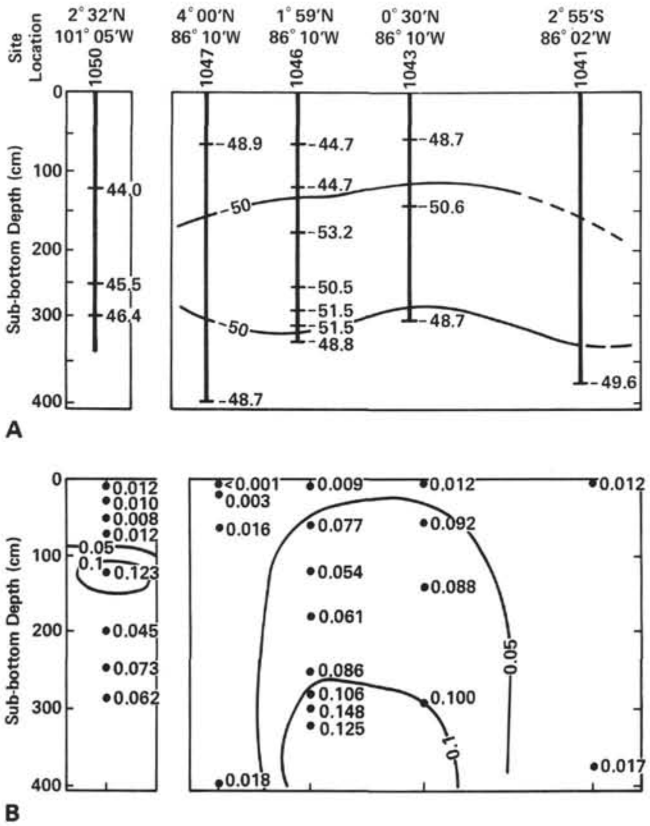

Figure 6. A. $\delta^{34}S$(‰) of the sulfide sulfur values in the upper layer of sediments from the southeast Pacific (from Migdisov et al., 1978). B. Pyrite sulfur concentrations (%).