

# Partition geochemistry of sediments from DSDP 424 in the Galapagos Hydrothermal Mounds Field

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**ABSTRACT.** Sediments from DSDP 424, a core taken in a hydrothermal mound in the Galapagos Mounds Field, comprise biogenic oozes, siliceous ash-rich layers and two varieties of hydrothermal precipitates, an Fe-rich clay (nontronite), and Mn oxides. Compositionally, these sediments differ considerably from one another and vary in proportion down the core to basement. Geochemical partition studies on the sediments have indicated a variable fractionation of the elements that they contain between clays, carbonate, and Mn oxides with the hydrothermal phases being strongly depleted in trace elements. The hydrothermal sediments are thought to have been formed as a result of the fractional precipitation of Fe silicates and Mn oxides from ascending hydrothermal solutions which are depleted in trace elements due to subsurface precipitation of sulphides, and have had insufficient time to scavenge trace metals from sea water due to their rapid precipitation

THE Galapagos Spreading Center (fig. 1) forms part of the global mid-ocean ridge system. It is spreading at a half rate of 35 mm/yr (Klitgord and Mudie, 1974) and heat flow and bottom-water temperature measurements show an increase of these parameters in this area relative to that surrounding (Sclater *et al.*, 1974; Detrick *et al.*, 1974) indicating that it is hydrothermally active.

Hydrothermal mounds are located within an area of high heat flow, between 18 and 25 km from the spreading axis. Their height varies from less than a metre to over 20 m, the small mounds having gentle slopes while the larger ones have steeper slopes. Within the mounds, black Mn-oxide crusts have been observed overlying orange Fe oxides, which in turn overlie green nontronite (Corliss *et al.*, 1978). The chemistry of these deposits showed that Fe and Mn were strongly fractionated. Fe was found almost exclusively in the nontronite or Fe

oxides while Mn formed a separate phase consisting of todorokite and birnessite (Corliss *et al.*, 1978).

In the present study, some hydrothermal deposits drilled at DSDP 424 in a sea-floor mound field 22 km south of the Galapagos Spreading Center have been investigated as follows: (1) The bulk chemical composition of the deposits has been determined, and the profiles of the vertical distribution of their metals studied. (2) The phases present in the sediments have been separated chemically and the proportional distribution of metals in each determined. (3) An attempt has been made to deduce the process of formation of the deposits.

## *Lithology—stratigraphy*

The general lithology of the sediments drilled at DSDP 424 has been summarized by Hekinian *et al.* (1978). The following material was recovered: hydrothermal deposits, foraminifer-nanofossil ooze, siliceous-nanofossil ooze, and siliceous-ash-rich layers. The hydrothermal deposits can be divided into two classes: (a) green smectite of nontronitic composition, and (b) Mn oxides.

The top of the core down to 13.8 m consists of hydrothermal sediment, made up essentially of green nontronitic mud intermixed with Mn oxide. At 13.8–14.2 m, a thin layer of foraminifer-nanofossil ooze occurs, in which Mn oxides are present. Hydrothermal sediments similar to those found in the upper part of the sediment sequence occur in the core at 14.2–15.1 m and 15.5–16.2 m, with foraminifer-nanofossil ooze between them. Between 16.2 and 19.0 m there is a layer of carbonate sediments which is followed by another horizon of green nontronitic mud (19.0–19.5 m). From 19.5 m down to the bottom of the sediment column the core is composed of foraminifer-nanofossil ooze.

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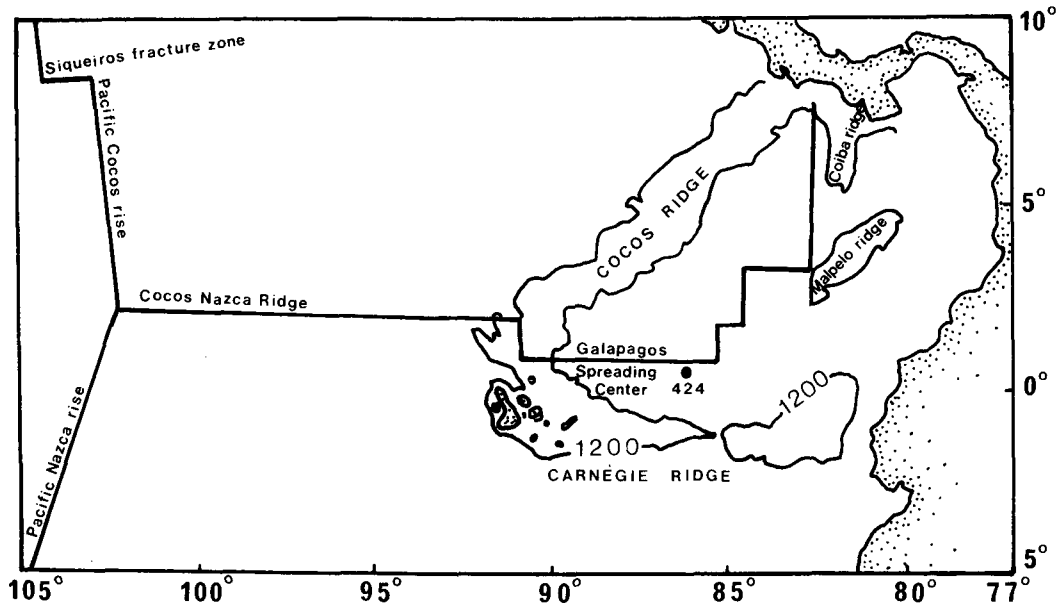


FIG. 1. Location of the Galapagos Spreading Centre.

From the presence of radiolaria, which are abundant and well preserved in a large part of the core, the age of the basal sediments has been determined to be not greater than 1.2 Ma.

#### *Chemistry of the deposits*

**Bulk chemical composition.** Chemical analyses of nineteen samples from various depths of DSDP 424 were carried out by Atomic Absorption Spectrophotometry for Mn, Fe, Ni, Co, Pb, Zn, Ca, Mg, and Al. The results are given on a carbonate-free basis in Table I. The attack used was a total HF-HClO<sub>4</sub> dissolution leaving no residue. The accuracy of the determinations was checked by analysis of international standards, and the precision of the determinations were better than  $\pm 10\%$ . The Atomic Absorption instrument used was a Perkin Elmer 403, and the readings it gave were corrected for Ca interference in high-carbonate samples. Silica was determined separately.

The chemical composition of the deposits shows that certain sediment horizons occur which are markedly ferruginous, whereas in other layers the concentration of Fe is lower and in some cases is similar to that in normal pelagic clays. Fe is depleted in the Mn crusts at the top of the sediment sequence while its concentration in the underlying green hydrothermal sediments ranges between 21 and 22.5%. Mn is depleted in these latter sediments.

From 19.5 m down to the basement, where carbonate sediments occur, the concentration of Fe tends to decrease and at the basement it reaches values which are comparable with those of normal pelagic sediments. It is observed that at the top of each of the ferruginous horizons a thin Mn-rich layer occurs, whereas at greater depths the whole of each ferruginous horizon is exceedingly depleted in Mn.

The trace-element data for the highly ferruginous and manganiferous horizons show that these sediments are extremely depleted in trace metals such as Ni, Co, Zn, and Cu, when compared with the average composition of Pacific surface pelagic clays and especially Mn nodules and mid-ocean ridge metalliferous sediments at large (Cronan, 1969, 1976a). In the upper Fe-rich clays (0–13.8 m) the concentrations of these metals do not show any significant variation with increasing depth. In contrast, there is a tendency for their concentration to increase with depth in the carbonate sediments (fig. 2). The maximum concentrations of Ni, Co, and Zn occur at 17.4 m (346, 60, 418 ppm, respectively), while Cu shows its highest concentration at 30.6 m (324 ppm). In both cases these depths fall within the foraminifer-nanofossil ooze layers. The vertical variations of Ni, Co, Cu, and Zn are, in large part, positively correlated with each other.

In contrast to the other trace metals, Pb does not show any depletion in the Fe-rich sediments,

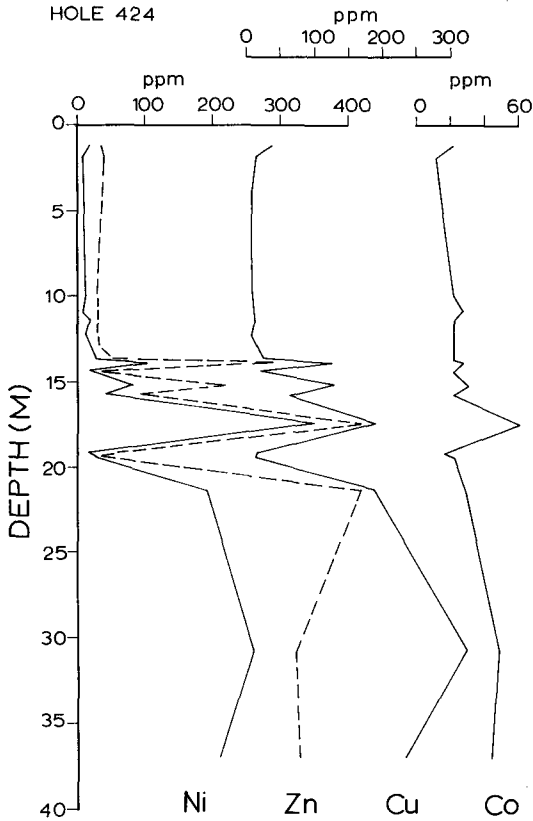


FIG. 2. Variations of Ni, Zn, Cu, and Co with depth in DSDP 424.

its concentrations being similar to the average concentration of Pb in normal pelagic clays. Furthermore, the vertical variations of Pb show a different pattern from that produced by the vertical distributions of Ni, Co, Cu, and Zn. A decrease in the concentration of Pb with increasing depth within the upper ferruginous horizon occurs which continues into the carbonate sediments below. The two peaks in the concentration of Pb which have been found at depths of 1.20 and 19.10 m are associated with the Mn-rich layers occurring at the tops of the ferruginous horizons.

The Fe- and Mn-rich sediments are characterized by a very low Al content, while in the other sediments the concentration of Al varies with that of  $\text{CaCO}_3$ . The concentration of  $\text{SiO}_2$  ranges between 39.03 and 47.32% on a CFB. There is a tendency for its concentration to decrease with increasing depth in DSDP 424. A positive linear correlation (fig. 3) between  $\text{SiO}_2$  and Fe concentrations was found.

The average chemical composition of the Galapa-

gos green hydrothermal clay material shows similarities with those of clay-rich sediments described from the Gulf of Aden (Cann *et al.*, 1977), and from transform fault A in the FAMOUS area (Hoffert *et al.*, 1978).

**Partition analysis.** The use of partition analysis has become an important tool in the elucidation of the distribution of elements between various coexisting phases of marine sediments which are too fine-grained for it to be possible to physically separate their individual constituents (Goldberg and Arrhenius, 1958; Chester and Hughes, 1967; Cronan, 1976a). In the present investigation, partition analysis has been carried out on ten samples in order to investigate the distribution of Ca, Mn, Fe, Ni, Co, Pb, Zn, Cu, and Al between their constituent phases. The method used was a modification of that of Chester and Hughes (1967), described in detail by Cronan (1976a). The samples were leached first with acetic acid to remove carbonate phases and adsorbed ions, secondly with hydroxylamine HCl to remove reducible Mn and ferromanganese oxides, thirdly with HCl to remove iron oxides and attack silicates, and finally with HF,  $\text{HClO}_4$  to take up the residue resistant to the previous attacks. The partition data are summarized in Table II, where the sediments have been divided into Fe-rich clays, carbonates, and Mn-rich sediments. The relatively pure manganese oxide crust from the top of the core was not subjected to partition analysis because insufficient material was available.

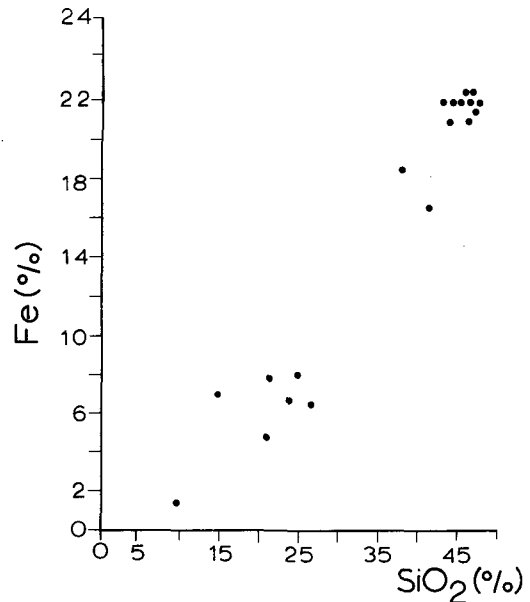


FIG. 3. Fe against  $\text{SiO}_2$  in samples analysed.

Table I: Chemical composition of sediments from the Galapagos spreading center (Leg 54, Hole 424), expressed on a carbonate-free basis.

Sample Number	Depth m	Ca %	CaCO <sub>3</sub> %	Mn %	Fe %	Ni ppm	Co ppm	Pb ppm	Zn ppm	Cu ppm	Mg %	Al %	SiO <sub>2</sub> %	Fe/Mn	
1	6	Top	1.02	0.01	33.00	7.2	81	16	48	126	39	1.65	0.15	13.84	0.22
2	15	1.20	0.33	0.01	3.80	22.0	17	22	150	35	14	2.20	0.09	42.72	5.79
3	39	1.94	0.17	0.01	0.35	22.5	7	12	80	39	8	2.30	0.06	45.99	64.2
4	48	9.87	0.18	0.01	0.078	22.0	12	22	95	28	9	2.50	0.03	45.22	282
5	51	10.97	0.19	0.01	0.077	22.0	7	27	80	29	11	2.65	0.05	47.32	285
6	38	11.38	0.15	0.01	0.054	22.0	17	22	50	30	12	2.70	0.07	46.56	407
7	2	12.21	0.17	0.01	0.063	22.5	12	22	50	27	9	2.47	0.05	46.32	357
8	25	12.91	0.17	0.01	0.067	22.0	17	22	50	33	12	2.40	0.07	45.06	328
9	28	13.72	0.21	0.01	0.090	21.0	27	22	50	54	24	2.70	0.43	47.00	233
10	19	13.91	17.25	40.98	0.314	13.1	101	27	35	289	125	2.77	2.19	36.19	41.82
11	18	14.40	0.22	0.01	0.105	22.0	17	22	50	38	20	2.50	0.17	44.67	209
12	1	15.22	15.50	36.53	0.300	12.6	82	30	46	218	129	3.17	2.14	39.28	42.0
13	32	15.75	4.02	7.34	0.108	17.8	40	22	45	94	63	2.89	1.16	44.57	164
14	47	17.41	18.50	44.16	0.457	12.0	346	60	41	418	189	3.72	4.30	42.65	26.2
15	12	19.10	0.52	0.01	10.75	18.4	16	17	294	57	13	2.15	0.05	38.08	1.72
16	54	19.36	0.29	0.01	0.080	21.0	27	22	49	35	14	2.75	0.21	43.95	262
17	35	21.30	20.75	49.89	0.409	9.4	190	29	37	418	189	7.65	4.19	41.35	23.1
18	7	30.69	11.50	26.35	1.603	8.9	261	49	71	323	324	3.10	3.74	35.66	5.59
19	14	36.90	30.75	75.32	2.068	5.1	210	44	75	329	233	3.34	5.27	39.03	2.50
Average of nine ferruginous samples			0.19	0.01	0.107	21.9	16	22	62	35	13	2.55	0.13	45.79	

Table II Summary of partition analyses. The results are expressed as percentages of bulk composition.

		Ca	Mn	Fe	Ni	Co	Pb	Zn	Cu	Al
Fe-rich clays	A	51.62	31.06	1.19	5.88	8.33	1.01	3.57	8.88	13.54
	B	32.58	29.72	2.39	51.64	7.58	65.80	8.81	39.39	6.38
	C	0.98	33.52	95.08	34.64	15.15	4.46	43.78	15.27	48.48
	D	14.83	5.69	0.21	7.84	68.93	28.74	43.85	36.44	31.61
	E	0.75	0.61	21.07	20.00	21.00	69.00	67.00	21.00	0.27
Carbonate sediments	A	95.65	41.93	1.42	13.61	12.50	16.82	5.44	18.33	2.81
	B	1.00	5.00	2.11	22.75	15.00	4.76	6.31	21.12	2.65
	C	0.00	49.23	80.97	62.30	72.50	59.37	68.0	33.71	57.88
	D	3.35	3.85	14.78	1.35	0.00	19.05	20.17	26.86	36.67
	E	17.87	0.22	7.22	126.00	25.00	22.00	202.00	89.00	1.84
Mn-rich sediments	A	65.39	2.23	0.76	0.00	0.00	0.00	1.76	7.69	20.00
	B	19.23	89.49	2.76	81.25	11.76	94.90	8.77	46.15	0.00
	C	1.92	4.93	93.01	0.00	11.76	0.00	21.05	0.00	40.00
	D	13.46	3.35	3.47	18.75	76.48	5.10	68.42	46.15	40.00
	E	0.52	10.75	18.45	16.00	17.00	294.00	57.00	13.00	0.05

A: Acetic acid leach (HAc).

B: Acid-reducing agent leach (i.e. hydroxylamine hydrochloride).

C: HCl leach.

D: HCl-insoluble residue.

E: Bulk composition (Ca, Mn, Fe and Al in wt. %, remainder in ppm).

When considered over all, the partition data indicate that there are considerable differences in the partition of elements between the Fe-rich clays, the carbonate sediments, and the Mn-rich sediments (Table II).

In the Fe-rich clays most of the Fe and Al are associated with the HCl-soluble fraction in the clay minerals, Fe in the nontronite and Al probably in a non-authigenic aluminosilicate, but Al is very low over all. The majority of the Ni and Pb are located in the minor hydroxylamine HCl-soluble fraction. Cobalt is more concentrated in the HCl-insoluble residue, while Zn is equally distributed between the HCl-soluble fraction and the HCl-insoluble residue. Other elements show more even distributions. The presence of significant amounts of Co, Pb, Zn, and Al in the HCl-insoluble residue of the Fe-rich clays would suggest the incorporation of some basaltic detrital material in these sediments. Since there is little or no carbonate material in these sediments, the high proportion of Mn associated with the acetic acid-soluble fraction may reflect its removal from adsorption sites on the surface of the Fe-rich clays, and hydroxylamine-HCl-soluble Mn may be present in physically admixed oxides.

Except for Ca, the majority of the elements (Mn, Fe, Ni, Co, Pb, Zn, Cu, and Al) in the carbonate sediments are associated with the HCl-soluble fraction. This may reflect the processes of normal pelagic sedimentation, where these elements are incorporated into the non-authigenic clay fraction of the sediments. However, the significant concentrations of Mn, Pb, and Cu in the acetic-acid-soluble fraction points to the removal of these elements from biogenic calcium carbonate or from coatings on carbonate material.

The Mn-rich sediments show partition patterns different from those of Fe-rich clays and the carbonate sediments, but are more similar to those observed for the Fe-rich clays than for the carbonates. This is most likely to be due to the presence of Fe-rich clay material admixed in these sediments. In the Mn-rich sediments, Mn, Ni, and Pb are associated with the acid-reducible Mn oxide fraction, while Co and Zn are predominately in the HCl-insoluble detrital residue. Copper is equally distributed between the acid-reducible fraction and the HCl-insoluble residue. The minor aluminum is equally distributed between the HCl-soluble and the HCl-insoluble fractions, with some in the acetic acid-soluble fraction. The high percentage of Mn associated with the hydroxylamine HCl-soluble fraction indicates the presence of a large proportion of reducible Mn oxides in the sediments.

### Discussion

The observations described in this paper support discussion of a number of problems. What are the reasons for the strikingly different compositions of the different hydrothermal constituents of the sediments, and why are they all so low in trace elements? Are the deposits primary precipitates, or are they diagenetic in origin? Do the data indicate episodic or continuous accumulation of hydrothermal precipitates in the Galapagos Mounds Area? Where do they fit into the global mid-ocean ridge hydrothermal fractionation sequence (Cronan, 1976b, 1980), parts of which are being found in more and more locations on the sea floor.

The fact that the Mn-crusts present at the top of the sediments contain very little Fe, while the underlying Fe-rich clays are depleted in Mn, indicates that a fractionation between Fe and Mn occurs in the sediments. The experimental conditions for the direct precipitation of nontronite from solutions containing Fe and Si require a reducing environment (Harder, 1976). Therefore during the precipitation of the nontronite, Mn remains in solution and precipitates later above the Fe silicates as Mn oxide when the redox potential rises, although some may escape altogether by dispersion through sea water.

That the Fe/Mn ratios vary vertically down the core, indicates that hydrothermal solutions have been discharged periodically in the mounds area rather than during one continuous process. The layers with high Fe/Mn ratios represent sediments deposited during the early phases of discharge of hydrothermal solutions whereas those having lower Fe/Mn ratios are sediments which have been formed from the residual solutions of the hydrothermal activity under more oxidizing conditions. This is consistent with observations on the Santorini hydrothermal deposits made by Smith and Cronan (1975), where the sediments from close to the fumarolic outlets are characterized by high Fe/Mn ratios in contrast to those away from the fumarolic outlets where the Fe/Mn ratios are low. Fluctuations in the Fe/Mn ratios are also reported from sediments near Stromboli (Bonatti *et al.*, 1972), which suggest a similar process of sedimentation. Likewise, there is a marked fractionation of Mn from Fe in the Red Sea hydrothermal deposits, where a pattern including Mn- and Fe-rich geochemical zones was found around the Atlantis 11 Deep (Bignell *et al.*, 1976). Thus, there is evidence to conclude that the geochemical conditions which occurred during the precipitation of the Galapagos deposits were similar to those occurring in the Red Sea and the Santorini hydrothermal areas. The main difference is that in the Galapagos deposits

the results of this process are exhibited vertically, whereas in the Santorini and the Red Sea sediments they are exhibited horizontally.

It is notable that the concentrations of Ni, Co, Zn, and Cu are extremely low in the Galapagos hydrothermal sediments, particularly in the ferruginous horizons. Two possible explanations are envisaged for the depletion of these metals. (1) The hydrothermal solutions which are responsible for their formation are depleted in these metals. (2) The rapid precipitation of the major hydrothermal phases does not allow significant scavenging of the metals from sea water. Cronan (1976*b*) suggested that circulating sea water within the fractures and fissures of the upper part of the oceanic crust could mix with hydrothermal solutions and cause precipitation of metal sulphides and possibly other phases below the sea floor. This would lead to a depletion of sulphide-forming elements, such as most of the trace elements determined in this work, in the hydrothermal solutions at discharge.

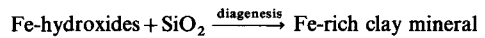
Although sub-sea-floor precipitation processes can explain why the Galapagos hydrothermal sediments are not enriched in trace metals, they are not able to explain why the concentrations of trace metals in these sediments are lower than the average concentrations which occur in normal pelagic sediments. It is known that 'scavenging' of metals from sea water by suspended material and transfer of them to the sea floor is an important process in the incorporation of trace metals into deep-sea sediments (Goldberg, 1954), and is a function of a number of factors. The factor which is probably most important in explaining the extremely low concentrations of trace metals in the Galapagos hydrothermal sediments is their rate of deposition. In view of the young age of the deposits, their rate of deposition must have been very rapid and the time of their residence in the sea water was so short that scavenging of metals would have been very limited.

Therefore, it is concluded that a combination of two factors determines the low trace-element content of the hydrothermal sediments; the rapid deposition of the hydrothermal precipitates together with the depletion of the hydrothermal solutions in trace metals as a result of sub-sea-floor precipitation processes.

An attempt has been made in the present study to investigate the form in which Si is present in the Galapagos sediments, and to determine the possible form in which it was precipitated from the hydrothermal solutions. The selective chemical analyses indicate that Fe is associated chiefly with the HCl-soluble fraction, which, along with the fact that there is a positive linear correlation (fig. 3) between Fe and the total SiO<sub>2</sub> leads to the conclu-

sion that the majority of Fe and Si in the sediments examined occur in the form of nontronite. This could have been formed either by direct precipitation from the hydrothermal solutions or from their reaction with silica-bearing sediments, or diagenetically from the reaction between Fe-hydroxides and SiO<sub>2</sub> after their precipitation from the hydrothermal solutions as two separate phases.

To examine the possibility of diagenetic formation of an Fe-rich clay mineral, after the precipitation of Fe and Si from the hydrothermal solutions, the following reaction should be considered.



If this reaction takes place in the Galapagos sediments, then the proportion of Fe present in the sediments associated with the Fe-rich clays should increase with time. This can be tested by examining variations down the core in the distribution of Fe between the fraction soluble in HCl and the other fractions, because the Fe which is present in the form of Fe silicate in the clay minerals will be dissolved in HCl. There is no tendency for the proportion of the Fe associated with the HCl-soluble fraction to increase with increasing depth in the core and thus it is considered that no transformation of one Fe phase to another occurs in the present sediments. It is, therefore, reasonable to conclude that the Fe-rich nontronitic clays found in the Galapagos sediments either precipitate directly from the hydrothermal solutions, or form by reaction of the hydrothermal solution with, and replacement of, the indigenous sediments of the area which are SiO<sub>2</sub>-bearing biogenic oozes.

In comparison with other hydrothermal deposits on the sea floor, those from the Galapagos Mounds Area described here can be classified as sharply fractionated deposits. In the global mid-ocean ridge hydrothermal fractionation sequence (Cronan, 1980) they would form after the precipitation of sulphides, which, as mentioned, probably occur within the oceanic crust below them, and before the precipitation of widely dispersed Fe-Mn oxides, which probably occur on the sea floor away from the immediate vicinity of the Mounds area. Their sharply fractionated nature is probably due to rapid cooling, mixing, and changes in Eh and other properties of the hydrothermal solutions on discharge.

### Conclusions

Hydrothermal sediments from DSDP 424 can be divided into an Fe-rich smectite (nontronite) phase and a Mn oxide phase. Both hydrothermal phases are low in trace elements, thought to be due to (i) the subsurface precipitation of the elements

concerned in sulphides and (ii) the rapid precipitation of the hydrothermal phases precluding the scavenging of trace metals from sea water.

Down-core variations in the proportion of the different hydrothermal phases present, reflected in part by variation in the Fe/Mn ratio, demonstrate that selective fractionation of Mn phases from Fe phases has taken place at discrete times in the past and that the hydrothermal precipitation has not been a continuous process.

The partition of elements between the different phases is as follows: Fe is principally present in the HCl-soluble Fe-rich clay fraction of all the sediments, Ca is largely present in biogenic debris, and Al as non-authigenic aluminosilicate detritus. The remaining elements are variably partitioned between these phases and Mn additionally occurs in an authigenic Mn-oxide phase in the Mn-rich sediments.

In the global mid-ocean ridge hydrothermal fractionation sequence of Cronan (1980), the deposits are thought to lie between early formed sulphides and later formed widely dispersed ferromanganese oxides.

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