11. STRONTIUM ISOTOPE GEOCHEMISTRY OF LEG 113 INTERSTITIAL WATERS AND CARBONATES¹

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ABSTRACT

The concentration of dissolved Sr and the distribution of 87 Sr/ 86 Sr isotope ratios in Leg 113 interstitial waters may be interpreted in terms of mixing of Sr from four different reservoirs: indigenous seawater, marine carbonate minerals, and basaltic and siliceous detrital material. The input to the pore water from these reservoirs is determined by the reactivity of the reservoir rather than its size. The presence of strontium derived from siliceous detrital material is unequivocally demonstrated in the pore waters of the hemipelagic deposits, and is also significant in the calcareous Maud Rise sediments due to the unusually low degree of carbonate recrystallization. Also, alteration of basic volcanic material is important at several sites.

INTRODUCTION

In marine sediments the bulk of the elements is held in the solid phases while the pore water serves as a conduit between reacting solid phases and their products. Thus, pore waters are very sensitive indicators of incipient diagenetic reactions. Variations in the chemical composition of interstitial waters have proven to be useful for the delineation of elemental budgets (e.g., Manheim, 1976; Gieskes and Lawrence, 1976; Baker, 1986). However, because several different mechanisms may leave essentially the same imprint on the concentration profiles of dissolved constituents, major element chemistry on its own is not always sufficient to fingerprint reactions. For example, the frequently observed depletion of pore water magnesium and concomitant enrichment of calcium (Gieskes, 1973; McDuff, 1981; Baker, 1986) have been attributed to dolomitization (Manheim, 1976), reversed weathering (Wollast, 1974), and to alteration of volcanic matter dispersed in the sediments (Perry et al., 1976) or associated with the volcanic basement (McDuff, 1981; Baker, 1986). Because the amounts of authigenic phases formed are generally too small to be quantified reliably, only in special situations will petrographic examinations be diagnostic.

⁸⁷Sr/⁸⁶Sr isotope ratio determinations of Sr dissolved in the pore water add an extra dimension to such studies. For example Hawkesworth and Elderfield (1976) were able to identify alteration of volcanically derived material and recrystallization of carbonates as the controlling mechanisms, rather than pore water/basement interactions. Subsequently, several similar studies summarized by Elderfield and Gieskes (1982) have shown that ⁸⁷Sr/⁸⁶Sr isotope ratios are sensitive indicators of diagenetic reactions.

Other applications of pore water/rock strontium isotope geochemical studies include studies of seawater-oceanic crust interactions (Spooner, 1976; Palmer and Elderfield, 1985) and determination of the rate of recrystallization of carbonates in pelagic carbonate deposits (Richter and Depaolo, 1987).

The main aims of the present work are to establish the distribution of ⁸⁷Sr/⁸⁶Sr isotope ratio in pore water and carbonates recovered during Leg 113, and to delineate the controlling mechanisms.

ANALYTICAL METHODS

The interstitial water were extracted on board using a stainless steel hydraulic press and the procedures described by Manheim and Sayles (1974). The concentration dissolved strontium was determined by atomic absorption spectrophotometry (AAS) after adjusting the samples to 4000 ppm La³⁺ with LaCl₃ (Gieskes, 1973). For ⁸⁷Sr/⁸⁶Sr isotope analysis, Sr was concentrated from pore water samples using miniature ion-exchange columns. The strontium concentrates were analyzed on a Finnigan MAT 261 machine (Institute of Energy Technology) in the static multicollector mode. ⁸⁷Sr/⁸⁶Sr ratios were normalized to 86Sr/88Sr = 0.1194. Measurements of the NBS 987 standard during the period of analysis averaged 0.710251 ($2\sigma = 0.00003$).

A 1-hr treatment of bulk sediments with cold 0.1N HCl was used to liberate adsorbed strontium, strontium in exchange positions, and strontium held in carbonates. The 87 Sr/ 86 Sr ratio of the leached strontium was subsequently determined as described above. The concentration of calcium, magnesium, and strontium in the acid extract were determined by AAS as described above and back-calculated to percent (Ca) and ppm (Mg and Sr) of bulk sample (only for Sites 689 and 690).

The δ^{18} O values of the pore waters were determined using the CO₂ equilibration method. Isotope analyses were performed on a Finnigan MAT 251 mass spectrometer. The results are reproducible to ± 0.2 % (2 sigma).

SITE DESCRIPTION

Site 689 (Maud Rise)

The site is located near the crest of Maud Rise (Fig. 1) at a water depth of 2080 m. Whole round samples for squeezing were taken from cores collected from Hole 689B drilled to 297.3 mbsf, terminated approximately 40 m above basement. The recovered sediments are pelagic, biogenic in origin, and range in age from Campanian/Maestrichtian to Pleistocene. The sequence has been divided into three lithostratigraphic units based upon compositional differences and diagenetic maturity.

Unit I (0-31.0 mbsf; late Miocene to Pleistocene) consists predominantly of diatom ooze with varying amounts of other biosiliceous components.

Unit II (31.0-149.1 mbsf; late Eocene to late Miocene) consists of a mixture of biosiliceous and calcareous oozes. The calcareous component (mainly nannofossils) increases downward.

Unit III (149.1–297.3 mbsf; Campanian? to late Eocene) consists of nannofossil ooze and chalk.

Zeolites, chert, and altered volcanic ash-bearing layers are minor components. The clay size fraction (Barker, Kennett, et al., 1988) is dominated by smectite and illite down to 110 mbsf and by smectite below 110 mbsf.

¹ Barker, P. F., Kennett, J. P., et al., 1990. Proc. ODP, Sci. Results, 113: College Station, TX (Ocean Drilling Program).

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Figure 1. ODP Leg 113 site locations.

Site 690 (Maud Rise)

This site is situated on the southwest flank of Maud Rise in 2914 m of water. Whole round samples for squeezing were taken from cores from Hole 690B drilled to 213.4 mbsf and from Hole 690C, terminated in basaltic rock at 321.2 mbsf. The sediments range in age from late Cretaceous to Pleistocene. The sequence is mainly biogenic, but terrigenous and volcanic detritus become a significant component around 140 mbsf. Thin, rare, chert layers were recovered. The sequence has been divided into six lithostratigraphic units.

Unit I (0-24.4 mbsf; late Miocene to Pleistocene) consists of 2.1 m of foraminiferal ooze overlying diatom ooze with varying amounts of other biosiliceous components.

Unit II (24.4-92.9 mbsf; early Oligocene to late Miocene) consists of pure and mixed biogenic siliceous and calcareous oozes.

Unit III (92.9-137.8 mbsf; late Paleocene to early Oligocene) consists of foraminiferal oozes.

Unit IV (137.8-281.1 mbsf; late Maestrichtian to late Paleocene) consists of nannofossil oozes and chalks with a terrigenous component of 10%-15%.

Unit V (281.1-317.0 mbsf; late Campanian(?) to late Maestrichtian) consists predominantly of terrigenous material (including volcanic glass) and contains calcareous ooze and chalk.

Unit VI (317.0-321.1 mbsf) consists of 1.71 m of amygdaloidal pyroxene olivine basalts.

The clay size fraction (Barker, Kennett, et al., 1988) is dominated by smectite except in the upper 90 mbsf where illite is equal to or more common. Chlorite + kaolinite is present down to 213 mbsf. Zeolites are present at 230–260 mbsf.

Site 693 (Dronning Maud Land margin)

Site 693 lies on a mid-slope bench of the Weddell Sea margin of East Antarctica in 2359 m of water. Whole round samples for squeezing were taken from cores from Hole 693A drilled to 474.3 mbsf and from Hole 693B drilled to 403.1 mbsf. The seismic records indicate the presence of at least another 1000 m of sediments below our maximum penetration. The sediments range in age from lower Cretaceous to Pleistocene. The sequence has been divided into seven lithostratigraphic units. Unit I (0-12.2 mbsf; Pleistocene) consists of foraminiferbearing clayey mud.

Unit II (12.2-31.4 mbsf; late Pliocene to Pleistocene) consists of clayey mud.

Unit III (31.4-325.8 mbsf; late Oligocene to late Pliocene) consists of diatom mud and diatom-bearing silty to clayey mud.

Unit IV (325.8-345.1 mbsf; late Oligocene) consists of alternating diatomaceous mud and ooze and minor muddy nannofossil ooze and nannofossil-bearing clayey mud.

Unit V (345.1-397.8 mbsf; Oligocene) consists predominantly of diatomaceous mud and silt.

Unit VI (397.8-409.0 mbsf; Cretaceous-?Albian to Santonian) consists of radiolarian diatomite.

Unit VII (409.0-483.9 mbsf; early Cretaceous-Albian) consists of terrigenous claystones and mudstones with organic rich beds.

The clay size fraction (Barker, Kennett, et al., 1988) is dominated by illite down to 403.1 mbsf and by smectite from 403.1 mbsf to 483.9 mbsf. Kaolinite and chlorite are present in most samples above 403.1 mbsf. Volcanic glass and opaques are present throughout as minor components.

Site 694 (Weddell abyssal plain)

Site 694 is situated on the northern part of the Weddell abyssal plain in 4653 m of water. Whole round samples for squeezing were collected from cores from Hole 694B drilled to 179.2 mbsf and from Hole 694C terminated at 391.3 mbsf. The seismic records indicate the presence of more than 1000 m of sediment below our total depth. The recovered sediments range in age from middle Miocene to Pleistocene and are mostly terrigenous with a minor biosilicious component. Four lithostratigraphic units were defined.

Unit I (0-21.1 mbsf; early Pliocene to Pleistocene) consists of terrigenous clay and clayey mud with minor silt and diatombearing clayey mud.

Unit II (21.1-111.5 mbsf; early Pliocene?) consists predominantly of well- to moderately-well-sorted lithic and quartz sands.

Unit III (111.5-304.3 mbsf; middle Miocene? to early Pliocene?) consists of hemipelagic sediments and turbidites, graded silt sequences with some diatoms and diatom-bearing silty and clayey muds with interbedded silts and sandy muds. Unit IV (304.3-391.3 mbsf; middle Miocene) consists of diatom-bearing and diatomaceous claystones, with silt near the base.

The clay size fraction (Barker, Kennett, et al., 1988) is dominated by illite and chlorite. The proportion of chlorite increases downward while the abundance of kaolinite and smectite generally decrease toward the base of the sequence. Opaques are abundant.

Site 695 (South Orkney microcontinent)

Site 695 is located on the southeastern edge of the South Orkney microcontinent in 1305 m of water. Whole round samples for squeezing were sampled from cores recovered from Hole 695A drilled to 341.1 mbsf. The seismic records indicate the presence of at least another 1500 m of sediment below. The sediments are biosiliceous hemipelagic deposits and range in age from Miocene/Pliocene to Pleistocene. Three lithostratigraphic units were defined.

Unit I (0-190.0 mbsf; Pleistocene to early Pliocene) consists of diatom-bearing silty and clayey muds, muddy diatom ooze, diatom ooze, and diatom silty muds.

Unit II (190.0-306.9 mbsf; early Pliocene) consists of diatom-bearing silty and clayey muds.

Unit III (306.9-341.1 mbsf; early Pliocene to late Miocene) consists of silty mud with 0%-10% diatoms.

The clay size fraction (Barker, Kennett, et al., 1988) is dominated by illite and chlorite, the proportion of kaolinite decreases downhole, while the relative abundance of smectite varies randomly. Volcanic glass and rock fragments are observed throughout.

Site 696 (South Orkney microcontinent)

Site 696 is situated on the southeastern margin of the South Orkney microcontinent, under 650 m of water. Whole round samples for squeezing were taken from cores from Hole 696A drilled to 106.0 mbsf and from Hole 696B terminated at 645.6 mbsf. The seismic records indicate total sediment thickness of at least 1200 m. The sediments are terrigenous, hemipelagic, and pelagic and range in age from middle/upper Eocene to Pleistocene. The sequence has been divided into seven lithostratigraphic units.

Unit I (0-64.2 mbsf; Pleistocene to early late Pliocene) consists primarily of diatomaceous muds and oozes.

Unit II (64.2-124.8 mbsf; early late Pliocene to early Pliocene) consists mainly of diatom-bearing silty and clayey mud. Unit III (124.8–211.8 mbsf; early Pliocene to late Miocene) consists predominantly of silty and clayey mud, with less important diatom-bearing clayey mud.

Unit IV (211.8-260.1 mbsf; late Miocene) consists of diatom ooze and muddy diatom ooze.

Unit V (260.1-269.7 mbsf; late Miocene) consists of coarsegrained sand.

Unit VI (269.7-529.8 mbsf; late Miocene to middle Miocene) consists of diatom ooze, mud-bearing diatom ooze, diatomite, and mud-bearing diatomite.

Unit VII (529.8-645.6 mbsf; middle? to late Eocene) consists of sandy mudstone, claystone, clayey mudstone, silty mudstone, and barren glauconitic silty mudstone.

The clay size fraction (Barker, Kennett, et al., 1988) is dominated by chlorite and illite down to 500 mbsf and by smectite from 500 mbsf to 645.6 mbsf. Volcanic ash is present as layers throughout most of the sequence and is particularly abundant between 269.7 and 529.8 mbsf; finely dispersed glass constitutes 4%-8% of the sediment (smear slide estimates).

Site 697 (South Orkney microcontinent-Jane Basin)

Site 697 is located in Jane Basin under 3484 m of water. Whole round samples for squeezing were taken from cores from Hole 697A drilled to 20.9 mbsf and from Hole 697B terminated at 322.9 mbsf. The seismic records indicate a total sediment thickness of at least 800 m. The recovered sediments are hemipelagic with a minor biosilicious component, and range in age from Pliocene to Pleistocene. Two lithostratigraphic units were defined.

Unit I (0-293.0 mbsf; Pliocene to Pleistocene) consists of silty mud, diatom-bearing silty mud, clayey mud, and diatom-bearing clayey mud.

Unit II (293.0-322.9 mbsf; early Pliocene) consists of silty and clayey mud.

The clay size fraction (Barker, Kennett, et al., 1988) is dominated by illite and chlorite; kaolinite and smectite are present at most depths. Volcanic ash is abundant.

RESULTS

The strontium isotope data are presented in Tables 1–7 and compared to the seawater ⁸⁷Sr/⁸⁶Sr-time curve (data from De-Paolo and Ingram, 1985; Palmer and Elderfield, 1985; DePaolo, 1986; Koepnick et al., 1985; Hess et al., 1986) on Figures 2–5. The shaded areas represent the spread of data used to draw the time curve. For the calcareous sequences (Sites 689 and 690) we

Table 1. Interstitial water concentration data and concentration of elements in bulk sediment (extracted with 0.1N HCl), Site 689.

Core, section, interval (cm)	Depth (mbsf)	Age (Ma)	Sr ²⁺ (μm)	⁸⁷ Sr/ ⁸⁶ Sr (I.W) ^a	δ ¹⁸ O (‰) ^b	⁸⁷ Sr/ ⁸⁶ Sr (A.E.) ^c	Ca (ppt) ^c	Sr (ppm) ^c	Mg (ppt) ^c
113-689B-									
1H-3, 78-85	3.78	3.8				0.70946	0	12	0.9
1H-3, 145-150	4.45	4.1	101	0.70902	0.2	0.70926	2	19	1.2
2H-2, 66-70	7.46	4.5	104	0.70905	0.7	0.70937	1	16	0.5
3H-4, 120-125	20.50	6.0	111	0.70885	-0.1	0.70905	0	15	1.1
6H-4, 120-125	49.00	12.0	128	0.70873	-1.2	0.70909	2	34	1.7
9H-4, 120-125	77.80	23.0	147	0.70857	-1.7	0.70811	309	989	1.2
12H-4, 115-125	106.65	32.0	156	0.70852	-1.2	0.70797	348	1015	1.1
15H-4, 115-125	135.55	38.0	147	0.70834	-1.5	0.70775	351	1459	1.3
18H-4, 115-125	164.45	44.5	194	0.70822	-1.4	0.70778	309	1171	1.2
22X-4, 115-125	203.15	55.8	200	0.70812	-0.9	0.70780	349	783	1.3
25X-4, 115-125	232.25	66.4	209	0.70807	-0.9	0.70775	352	887	1.3
28X-3, 115-125	259.72	70.0	219	0.70808	-0.4	0.70783	359	1102	1.6

^a Interstitial water.

^b Relative to SMOW.

^c Released by extraction with 0.1N HCl (ppt = parts per thousand, ppm = parts per million).

Table 2. Interstitial water concentration data and concentration of elements in bulk sediment (extracted with 0.1N HCl), Site 690.

Core, section, interval (cm)	Depth (mbsf)	Age (Ma)	Sr ²⁺ (μm)	⁸⁷ Sr/ ⁸⁶ Sr (I.W) ^a	δ ¹⁸ O (‰) ^b	⁸⁷ Sr/ ⁸⁶ Sr (A.E.) ^c	Ca (ppt) ^c	Sr (ppm) ^c	Mg (ppt) ^c
113-690B-									
2H-3, 145-150	6.55	5.0	99	0.70953	-0.5	0.70940	1	18	1.3
3H-4, 120-125	17.40	6.5	104	0.70911	-0.5	0.70942	1	19	1.3
6H-4, 120-125	46.50	17.5	109	0.70887	-1.1	0.70864	135	627	1.7
9H-4, 120-125	75.50	28.0	138	0.70880	-1.4	0.70806	320	994	1.7
12H-5, 120-125	106.00	35.0	127	0.70859	-2.1	0.70774	376	1191	1.4
16H-4, 120-125	143.50	44.0	147	0.70842	-1.6	0.70779	310	839	1.6
19H-4, 115-125	172.55	49.0	146	0.70836	-2.1	0.70781	336	894	1.1
22H-3, 120-125	189.40	53.0	146	0.70831	-2.3	0.70779	341	995	1.4
25H-4, 115-125	209.85	58.0	155	0.70828	-2.5	0.70781	366	1092	0.9
113-690C-									
11X-3, 115-125	208.35		127	0.70832	-1.5	0.70783	294	834	1.7
14X-4, 115-125	238.85	65.0	124	0.70823	-1.5	0.70792	373	1000	1.2
17X-2, 115-125	264.45	69.0	160	0.70819	-1.5	0.70787	279	960	1.4
20X-4, 115-125	296.45	72.0	159	0.70828	-1.7	0.70788	364	962	5.5

^a Interstitial water. ^b Relative to SMOW.

^c Released by extraction with 0.1N HCl (ppt = parts per thousand, ppm = parts per million).

Table 3. Interstitial water concentration data, Site 693.

Core, section, interval (cm)	Depth (mbsf)	Age (Ma)	Sr ²⁺ (μm)	⁸⁷ Sr/ ⁸⁶ Sr (I.W) ^a	δ ¹⁸ O (‰) ^b
113-693A-			7		
2R-4, 145-150	8.45	0.5	76	0.70925	-0.5
4R-3, 120-125	26.10	2.3	85	0.70911	-0.7
6R-4, 120-125	46.80	3.0	94	0.70898	-1.1
9R-4, 120-125	75.80	4.0	94	0.70887	-0.7
12R-4, 120-125	104.60	4.5	85	0.70886	-1.5
18R-3, 120-125	161.10	5.1	103	0.70851	-2.3
25R-2, 120-125	227.30	7.0	124	0.70835	-2.8
113-693B-					
2X-3, 145-150	238.25	8.0	135	0.70847	-3.2
113-693A-					
28R-3, 120-125	257.80	8.5	127	0.70840	-1.1
113-693B-					
6X-4, 145-150	273.75	17.0	136	0.70810	-1.3
9X-3, 120-125	301.00	21.0	134	0.70809	-2.9
12X-3, 120-125	330.00	25.0	154	0.70784	-2.6
15X-2, 140-150	357.60	32.5	138	0.70778	-1.2
40R-2, 138-142	371.78	32.6	147	0.70773	-1.8
17X-1, 145-150	375.45	32.7	173	0.70773	-3.8
19X-2, 140-150	396.30	33.0	166	0.70770	- 3.1
113-693A-					
48R-2, 120-125	448.10	40.0	168		

^{a 87}Sr/⁸⁶Sr ratio of interstitial water strontium.

^b Relative to SMOW.

also present the isotope composition of the acid extracted strontium. Because the ⁸⁷Sr/⁸⁶Sr ratios of the acid extracts from the other sites turned out to be of little value, we only carried out a limited number of isotope ratio determinations. These are listed separately in Table 8.

Present-day seawater has an ⁸⁷Sr/⁸⁶Sr of 0.70920, and most of the pore waters have a value close to this in the shallowest sediment layers.

Examination of the δ^{18} O values in Tables 1-7 reveals that considerable scatter is present. In some instances (e.g., Cores

Table 4. Interstitial water concentration data, Site 694.

Core, section, interval (cm)	Depth (mbsf)	Age (Ma)	Sr^{2+} (μ m)	⁸⁷ Sr/ ⁸⁶ Sr (I.W) ^a	δ ¹⁸ Ο (‰) ^b
113-694B-	<u></u>	<u> </u>		<u> 50 6 </u>	10.12
1H-2, 145-150	2.95	0.8	83	0,70914	-0.3
3H-2, 120-125	17.79	4.5	92	0.70919	-0.8
22X-1, 145-150	151.85	5.4	93	0.71063	-0.6
24X-2, 120-125	172.30	6.8	93	0.70967	-1.1
13-694C-					
5X-1, 145-150	210.35	8.5	102	0.70998	-1.8
14X-4, 115-125	300.35	12.6	102	0.71017	-2.6
19X-2, 145-150	345.95	15.0	111	0.71025	-1.1
22X-2, 115-125	374.65		115	0.70983	- 2.7

^{a 87}Sr/⁸⁶Sr ratio of interstitial water strontium.

^b Relative to SMOW.

Table 5. Interstitial water concentration data, Site 695.

Core, section, interval (cm)	Depth (mbsf)	Age (Ma)	Sr ²⁺ (μm)	⁸⁷ Sr/ ⁸⁶ Sr (I.W) ^a	δ ¹⁸ O (‰) ^b
113-695A-					
1H-3, 145-150	4.45	1.4	93	0.70919	-1.2
3H-4, 140-145	18.10	2.8	89	0.70912	0.0
6H-3, 120-125	45.40	3.3	90	0.70910	-1.2
9H-2, 120-125	72.70	3.4	103	0.70897	-0.9
12H-4, 120-125	94.90	3.6	99	0.70890	-1.5
15H-3, 120-125	122.40	3.8	110	0.70886	-1.3
19X-4, 120-125	153.00	4.0	115	0.70876	-1.4
22X-3, 120-125	180.70	4.1	118	0.70857	-1.8
25X-3, 120-125	200.10	14.2	120	0.70874	-2.1
29X-2, 115-125	237.35	4.4	124	0.70879	-2.1
32X-4, 115-125	259.75	4.5	125	0.70880	-1.3
36X-2, 115-125	295.45	4.8	131	0.70879	-1.8
39X-3, 115-125	316.15	5.1	127	0.70887	-2.2
41X-3, 140-150	335.80	5.3	134	0.70877	- 2.3

^{a 87}Sr/⁸⁶Sr ratio of interstitial water strontium. ^b Relative to SMOW.

Table 6. Interstitial water concentration data, Site 696.

Core, section, interval (cm)	Depth (mbsf)	Age (Ma)	Sr ²⁺ (μm)	⁸⁷ Sr/ ⁸⁶ Sr (I.W) ^a	δ ¹⁸ O (‰) ^b
113-696A-					
2H-3, 120-125	6.70	0.6	112	0.70897	-0.4
5H-3, 120-125	35.40	3.6	125	0.70843	-2.2
8H-4, 120-125	65.70	4.3	123		-2.7
113-696B-					
3R-3, 120-125	90.40	4.4	123		-2.0
6R-3, 120-125	119.40	4.7	136	0.70764	-3.5
20R-2, 120-125	233.80	7.0	171	0.70681	- 3.5
26R-2, 120-125	291.70	8.4	172		-2.4
34R-1, 120-125	367.50	10.0	162	0.70722	-2.9
53R-4, 115-125	554.55	20.0	168	0.70766	- 3.9
59R-3, 115-125	611.05	31.0	160	0.70768	-4.1
62R-5, 115-125	641.96	41.0	167	0.70748	-4.0

a ⁸⁷Sr/⁸⁶Sr ratio of interstitial water strontium.

^b Relative to SMOW.

113-696B-3R-3, 113-696B-26R-2, and 113-696B-34R-1) anomalous δ^{18} O values are associated with anomalous sulfate concentrations, thus some contamination by seawater may have taken place. However, in most cases the scatter in the oxygen isotope data is not related to fluctuations in the concentration of any of the other dissolved constituents. The fluctuations are much larger than what one would expect from the reproducibility of the method (\pm 0.2‰) and are probably partly due to analytical difficulties. We believe the data represent the general trends, but do not trust them sufficiently to go into any detailed discussion.

Table 7. Interstitial water concentration data, Site 697.

Core, section, interval (cm)	Depth (mbsf)	Age (Ma)	Sr ²⁺ (μm)	⁸⁷ Sr/ ⁸⁶ Sr (I.W) ^a	δ ¹⁸ Ο (‰) ^b
113-697A-					
1H-4, 145-150	5.95	0.1	89	0.70915	-1.2
113-697B-					
1H-1, 145-150	19.45	0.4	89	0.70923	-1.4
3H-4, 120-125	42.80	0.9	97		-1.0
4H-4, 115-125	52.45	1.2	95	0.70950	-1.6
5H-3, 115-125	60.65	1.4	89	0.70914	-1.5
6H-2, 115-125	68.85	1.6	90	0.70913	-1.0
7H-2, 140-150	78.90	1.9	93	0.70912	- 2.5
8H-3, 140-150	90.10	2.2	93		- 3.3
9H-3, 115-125	99.55	2.3	93		-1.4
13X-3, 115-125	132.75	3.0	102	0.70911	-0.6
16X-4, 115-125	163.15	3.4	106	0.70911	
19X-3, 115-125	190.75	3.7	100	0.70911	-0.9
22X-2, 115-125	218.15	3.9	103	0.70911	-1.6
26X-2, 115-125	256.85	4.1	101	0.70925	-1.8
29X-2, 115-125	285.90	4.3	79	0.70926	
31X-2, 140-150	305.50	4.4	111	0.70936	-2.2

^{a 87}Sr/⁸⁶Sr ratio of interstitial water strontium.

^b Relative to SMOW.

Sites 689 and 690

The sediments deposited in the interval 72-14 Ma at these sites contain more than 70% carbonate (average 84%) Here the acid extracts give an average concentration of strontium in carbonate of 1200 ppm (\pm 190 ppm) and the discrepancy between



Figure 2. 87 Sr/ 86 Sr-ratios of pore water strontium and acid leached strontium vs. sediment age at Sites 689 and 690, data from Tables 1 and 2. The shaded area represents the spread of data used to draw the time curve (solid line).



Figure 3. 87 Sr/ 86 Sr-ratios of pore water strontium vs. sediment age at Sites 693 and 696, data from Tables 3 and 6. The shaded area represents the spread of data used to draw the time curve (solid line).





Figure 4. 87 Sr/ 86 Sr-ratios of pore water strontium vs. sediment age at Sites 694 and 697, data from Tables 4 and 7. The shaded area represents the spread of data used to draw the time curve (solid line).

Figure 5. 87 Sr/ 86 Sr-ratios of pore water strontium vs. sediment age at Site 695, data from Table 5. The shaded area represents the spread of data used to draw the time curve (solid line).

Table. 8. ⁸⁷Sr/⁸⁶Sr data on strontium extracted from bulk sediment with 0.1N HCl, Sites 693 and 696.

Core, section, interval (cm)	Depth (mbsf)	Age (Ma)	⁸⁷ Sr/ ⁸⁶ Si
113-693A-			
2R-4, 145-150	8.45	0.8	0.71009
6R-4, 120-125	46.80	3.0	0.71198
12R-4, 120-125	104.60	4.5	0.71114
25R-2, 120-125	227.30	7.0	0.71137
28R-3, 120-125	257.80	8.5	0.71111
113-693B-			
9X-3, 120-125	301.00	21.0	0.71102
15X-2, 140-150	357.60	32.5	0.71147
19X-2, 140-150	396.30	33.0	0.71102
48R-2, 120-125	448.10	40.0	0.71238
113-696A-			
2H-3, 120-125	6.70	0.6	0.70939
8H-4, 120-125	65.70	4.3	0.70929

the isotope composition of the acid liberated Sr and the seawater curve (Fig. 2) is less than the uncertainty in the assigned ages (Barker, Kennett, et al., 1988) In the diatomaceous shallower sections above 50 mbsf the 87Sr/86Sr of the acid liberated strontium varies erratically between 0.7086 and 0.7094. The concentration of dissolved strontium increases from that of seawater (0.090 mmol/L) in the shallower samples to 0.22 and 0.16 mmol/L in the deeper samples at Sites 689 and 690 respectively (Tables 1 and 2) At Site 689 the 87Sr/86Sr isotope ratio of dissolved strontium decreases smoothly from 0.70902 at 2.9 mbsf (3.0 Ma) to 0.70808 at 259.7 mbsf (70 Ma). With two exceptions, at 20.5 mbsf (6.0 Ma) and 49 mbsf (~12 Ma), the data plot at or above the seawater curve (Fig. 2). At Site 690 the Sr87/Sr86 isotope ratio of pore water strontium decreases smoothly from 0.70911 at 17.4 mbsf (6.5 Ma) to 0.70819 at 264.5 mbsf (69 Ma). The pore water strontium in the shallower sample (6.6 mbsf, 5.0 Ma) exhibits a ⁸⁷Sr/86Sr ratio of 0.70953. All data plot above the seawater curve (Fig. 2).

At Sites 693, 694, 695, 696, and 697 the sediments consist predominantly of hemipelagic deposits, very low in calcium carbonate (generally less than 0.2%).

Site 693

The ${}^{87}\text{Sr}/{}^{86}\text{Sr}$ isotope ratio of dissolved strontium (Fig. 3) decreases smoothly from 0.70925 at 8.5 mbsf (0.8 Ma) to 0.70770 at 396.3 mbsf (33 Ma) Except for the two shallowest samples all waters are less radiogenic than contemporaneous seawater. The concentration of dissolved strontium increases from 0.076 mmol/L in the shallower sample to 0.169 mmol/L at 448 mbsf (40 Ma). The ${}^{87}\text{Sr}/{}^{86}\text{Sr}$ of the acid liberated strontium ranges between 0.7101 and 0.7124.

Site 694

The 87 Sr/ 86 Sr of pore water strontium ranges between 0.70914 and 0.71063. The two shallowest samples plot close to the seawater curve (Fig. 4) Below 172 mbsf (6.8 Ma) 87 Sr/ 86 Sr increases smoothly and reaches 0.71025 at 346 mbsf (15 Ma) before decreasing abruptly to 0.70983 at 374.7 mbsf (sediments at this depth could not be dated) The highest value (0.71063) is observed at 151.9 mbsf (5.4 Ma) The concentration of dissolved strontium increases by about 20% from the surface to the deepest sample (Table 4).

Site 695

The pore water ${}^{87}\text{Sr}/{}^{86}\text{Sr}$ varies between 0.70919 and 0.70857. Above 45.4 mbsf (3.3 Ma) the pore water strontium isotope ratio plots close to the seawater curve (Fig. 5) Below this, ${}^{87}\text{Sr}/{}^{86}\text{Sr}$ falls rapidly to a minimum value of 0.70857 at 180.7 mbsf (4.1 Ma) below which considerable scatter is observed. The concentration of dissolved strontium increases from 0.093 mmol/L in the shallower sample to 0.135 mmol/L at 335.8 mbsf (5.3 Ma).

Site 696

The ${}^{87}\text{Sr}/{}^{86}\text{Sr}$ ratio of pore water strontium decreases from 0.70897 at 6.7 mbsf (0.6 Ma) to a pronounced minimum of 0.70681 at 233.8 mbsf (7.0 Ma, Fig. 3); all of the data plot below the seawater curve. The concentration of strontium increases from 0.113 mmol/L in the shallower sample to a plateau of about 0.170 mmol/L below 233.8 mbsf (7.0 Ma, Table 6).

Site 697

The pore water 87 Sr/ 86 Sr plots on or above the seawater curve. A maximum value of 0.70905 is observed at 52.5 mbsf (1.2 Ma) and the 87 Sr/ 86 Sr of the three deeper samples are also significantly higher than contemporaneous seawater (Fig. 4) The concentration of pore water strontium increases from 0.089 mmol/L at 5.95 mbsf (0.1 Ma) to 0.112 mmol/L at 305.5 mbsf (4.4 Ma, Table 7).

DISCUSSION

The Leg 113 pore water data agree with previous work which has shown that deep sea pore waters are usually enriched in strontium relative to seawater. In the present case this may be the result of addition of strontium from various possible reservoirs: carbonates and basic and siliceous detrital material. The Leg 113 interstitial waters may be grouped into three categories according to which of these components dominate the pore water strontium.

Sites 689 and 690

At both sites there are several reservoirs of strontium. In pelagic carbonate-rich deposits strontium derived from carbonates in the host rock is generally thought to be the main control on pore water 87Sr/86Sr and strontium concentration (Baker, 1986; Baker et al., 1982; Richter and Depaolo, 1987) Interaction with basaltic basement has also been invoked (Spooner, 1976), Hawkesworth and Elderfield (1976) demonstrated that strontium released during alteration of volcanic matter may also give a strong ⁸⁷Sr/⁸⁶Sr signal. At Site 690 there is a significant noncarbonate component consisting of eolian material below 137 mbsf (42 Ma) most probably derived from East Antarctica. A similar component may be present at Site 689 in smaller abundance. At Site 690 two thin virtually unaltered (Schandl and Wicks, this volume) basalt flows (possibly basement) were drilled, but the composition of the underlying basement is not known. At Site 690, the sediment sequence below 275 mbsf (70 Ma) consists predominantly of terrigenous material and contain 5%-20% carbonate.

The high ⁸⁷Sr/⁸⁶Sr of the pore water strontium is in marked contrast to what has been observed in other pelagic carbonaterich sequences (Richter and Depaolo, 1987; Elderfield et al., 1982) Both groups found the isotope composition of dissolved strontium to be dominated by strontium released during recrystallization of carbonates in the host rocks, thus giving pore water ⁸⁷Sr/⁸⁶Sr values similar to those of the corresponding carbonates. At Sites 689 and 690, however, the pore water strontium is systematically more radiogenic than the corresponding carbonates (acid extracts).

At the carbonate-dominated Lord Howe Rise (Richter and Depaolo, 1987) and the Ontong Java Plateau (Elderfield et al., 1982) locations, the pore waters rapidly reach stable levels of 0.7 and 0.8 mmol/L strontium respectively, whereas at Sites 689 and 690 the concentrations of dissolved strontium increase almost linearly downward from the seafloor to maximum values of 0.22 and 0.16 mmol/L respectively. This difference in strontium distribution is not related to differences in the Sr/Ca ratio of the carbonates (0.0029 to 0.0042 at Maud Rise, 0.0005 to 0.0015 at Ontong Java Plateau), or to different Mg/Ca ratios (Table 1 and 2). The Maud Rise, Lord Howe Rise, and Ontong Java Plateau sediments consist predominantly of nannofossil oozes and chalks. The difference in rate of deposition is not sufficiently large to account for the observed features, thus the comparatively low concentrations of pore water strontium in Maud Rise sediments must be due to low rates of recrystallization of the carbonates.

Based on experimental studies, Baker et al. (1980) found that the most important factors governing the rate of recrystallization of natural carbonates are temperature, effective stress and the amount of noncarbonate material. At the Lord Howe Rise and the Ontong Java Plateau, the pore waters attain their high contents of strontium within the upper 100-200 m, thus the differences in rate of recrystallization do not seem to be directly related to differences in effective stress. Baker et al. (1980) found that, qualitatively, the rate of recrystallization was inversely related to the amount of silica in solution at the end of the experiments, and attributed the retarding effect of noncarbonate material to surface chemical reactions involving the formation of siliceous surface complexes. In the Maud Rise sediments, the noncarbonate fraction is on the average 10% (absolute) greater than at Lord Howe Rise and Ontong Java Plateau. The concentration of dissolved silica, however, is actually lower than in Ontong Java Plateau pore waters. It seems that the question can only be resolved by detailed surface chemical methods. At this stage we can only suggest that the comparatively low strontium concentrations are due to retarded rate of recrystallization, and that the rate terms used by Baker et al. (1982) and Richter and Depaolo (1987) to describe rate of recrystallization of carbonates are not valid for Sites 689 and 690.

The low degree of carbonate recrystallization at Sites 689 and 690 afford excellent opportunity to assess other influences on pore water composition. Evidently strontium with a higher ⁸⁷Sr/⁸⁶Sr isotope ratio than contemporaneous seawater must have been supplied to these pore waters. This strontium could have been derived from the seawater above or from the basement below or been generated within the sediment column. Based on a compilation of data from the DSDP program, Elderfield and Gieskes (1982) stated that no site yet studied shows unequivocal evidence of a ⁸⁷Sr/86Sr isotope signal reflecting diagenesis of terrigenous detritus. They attributed similar ⁸⁷Sr/⁸⁶Sr profiles to diffusive communication with the overlying seawater. However, at Sites 689 and 690, despite the smoothly decreasing 87Sr/86Sr isotope ratio with increasing depth, the increase in total strontium is more than sufficient to cause the concentration of ⁸⁷Sr to increase with depth. Thus there will be a net upward flux of both ⁸⁷Sr and ⁸⁶Sr. Although the isotope ratio will be modified by diffusion, there is little doubt that there is a source of Sr with a relatively radiogenic isotope signature, situated either in the recovered sediments or below. Although the immediately underlying rocks are of basaltic composition the nature of ultimate basement is not known, and the possibility that the strontium is diffusing upward from a siliceous basement cannot be excluded.

At Site 690 the high ⁸⁷Sr/⁸⁶Sr strontium may possibly come from the underlying terrigenous section. By using the tracer diffusion coefficient for strontium given by Li and Gregory (1974) the formation factor for nannofossil oozes (Berner, 1980), and a porosity of 0.5 (chosen as an average for the whole sequence) the distance of diffusion during 70 m.y. is more than 700 m. Thus, there is no doubt that if there is a supply of strontium from below, then diffusion will be rapid enough for the signal to be detected throughout the 300-m-thick sediment column. However, the fact that below 90 mbsf (about 30 Ma), the ⁸⁷Sr/⁸⁶Sr values run almost parallel to the seawater curve (Fig. 2) suggests that the system is buffered by strontium derived from carbonates in the host rocks. By assuming that the concentration of seawater strontium has remained constant over the last 70 m.y. and that the pore waters were deposited in isotopic equilibrium with ocean water and marine carbonate, and by ignoring diffusion, the relative amount of strontium released from carbonates and terrigenous material may be estimated.

The most likely source of the terrigenous material is Queen Maud Land. Igneous rocks from this area typically have ⁸⁷Sr/ ⁸⁶Sr ratios of 0.728 (Barton and Copperthwaite, 1983).

In this study we recognize four reservoirs of strontium: indigenous seawater, marine carbonate minerals, and basaltic and siliceous detrital material. Because for each site we are able to eliminate one of these sources (based on geochemical considerations) and because we assume that the original pore water was deposited in isotopic equilibrium with seawater, we are left with only two variables. By making educated guesses of the ⁸⁷Sr/⁸⁶Sr ratios of the strontium in these reservoirs we are able to estimate the relative amount of strontium from each of them by manipulating equations 1 and 2.

$$Sr_{to} = Sr_w + Sr_1 + Sr_2 \tag{1}$$

$$\mathbf{R} = (\Sigma \mathbf{Sr}_1 \cdot \mathbf{R}_i / \mathbf{K} + \mathbf{R}_i) / (\Sigma \mathbf{Sr}_i / \mathbf{K} + \mathbf{R}_i)$$
(2)

Where Sr_{to} is the observed total number of moles of strontium per unit volume, Sr_w , Sr_1 , and Sr_2 are the number of moles derived from indigeneous seawater and reservoir 1 and 2, respectively. R_0 and R_i are the observed ${}^{87}Sr/{}^{86}Sr$ isotope ratio and the isotope ratio of reservoir i, respectively. The summations are carried out over all end members. K is a constant given by equation 3 that relates the total number of moles of strontium (Sr_{ti}) to the ${}^{87}Sr/8^6Sr$ ratio (R_i) and number of moles of ${}^{86}Sr$.

$$Sr_{ti} = (K + R_i) \cdot {}^{86}Sr_i \qquad (3)$$

The figures used to estimate K are those recommended by the Subcommission on Geochronology of the International Union of Geological Sciences and given by Faure (1986).

Because we are mainly interested in the relative amount of strontium derived from each source we are not considering diffusion. Implicitly we assume that all reactions are taking place simultaneously.

The profiles in Figure 6 are determined by the means of equation 1 and 2 and show the contribution to pore water Sr of material derived from carbonate and siliceous sources. The profiles reveal two important aspects. First, that instead of showing a systematic increase with increasing depth as one would expect if strontium with a high 87Sr/86Sr isotope ratio was supplied from below, the amount of strontium derived from terrigenous material is almost constant throughout. Secondly, assuming the choice of ⁸⁷Sr/⁸⁶Sr isotope ratio of the terrigenous strontium to be 0.728, it takes only around 3% of this strontium to produce the high 87Sr/86Sr ratios in the pore waters. It appears that there is an internal source of strontium with a high ⁸⁷Sr/⁸⁶Sr isotope ratio. Most likely this strontium is derived from the eolian terrigenous material within the sediments. The fact that one estimates about the same contribution of terrigenous strontium at the two sites, despite the much lower abundance of terrigenous material at Site 689, indicates that the availability of radiogenic



Figure 6. Amount of pore water strontium derived from terrigenous and carbonate sources at Sites 689 and 690, estimated by the means of equation 1 and 2 and by assuming a ⁸⁷Sr/⁸⁶Sr ratio for the terrigenous strontium of 0.728.

strontium is not directly related to the amount of bulk eolian material.

Sites 693, 695, and 696

At Sites 693, 695, and 696 the strontium isotope signature of the pore waters plot at or below the seawater curve (Figs. 3 and 5). Low 87 Sr/ 86 Sr isotope ratios in pore waters have previously been attributed to interaction with basaltic volcanic material (Hawkesworth and Elderfield, 1976; Elderfield and Gieskes; 1982).

Egeberg et al. (this volume) show that most of the variations in major element chemistry of Leg 113 interstitial waters may be interpreted in terms of reactions involving alteration of volcanic detritus dispersed in the sediments. The irregular Sr isotope variations in pore waters at Sites 695 and 696 confirm that there are local sources of strontium involved. The low 87Sr/86Sr ratios of dissolved strontium suggest that much of the strontium added to the pore waters to account for the downward increase in strontium content is derived from volcanic material. This is further substantiated by the significant correlation between the δ^{18} O signatures of the pore waters and the 87 Sr/ 86 Sr ratios of dissolved strontium (Fig. 7) At these sites the concentration of carbonate is very low (less than 0.1%, Barker, Kennett, et al., 1988) and the contribution of carbonate strontium to the pore water may be neglected. Thus the pore water strontium may be considered as a mixture of indigenous seawater, strontium derived from altered basic volcanic material, and siliceous terrigenous material. The relative contribution of strontium from these sources may be estimated by assigning each an appropriate 87Sr/86Sr ratio. The most conservative estimate of the amount of strontium derived from basic volcanics is obtained assigning them the lowest reasonable 87Sr/86Sr ratio for local volcanic matter (0.703) For strontium derived from terrigenous material we may use the same ⁸⁷Sr/⁸⁶Sr ratio as above (0.728). Figure 8 shows the amount of strontium the pore water in each unit of pore water could have acquired from the two sources (assuming constant porosity of 0.5) The amount of strontium supplied from terrigenous material is about the same at all sites (0-5 μ mol), whereas the



Figure 7. ⁸⁷Sr/⁸⁶Sr-ratios of pore water strontium vs. oxygen isotope distribution at Sites 693, 695, and 696, data from Tables 3, 5, and 6.



Figure 8. Amount of pore water strontium derived from terrigenous and basic volcanic sources at Sites 693, 695, and 696, estimated by the means of equation 1 and 2 and by assuming a ${}^{87}\mathrm{Sr}/{}^{86}\mathrm{Sr}$ ratio of terrigenous strontium of 0.728 for the terrigenous strontium and 0.703 for the volcanogenic strontium.

amount derived from basic volcanic matter is considerably higher at Site 696 compared to the other sites. The pronounced drop in the ⁸⁷Sr/⁸⁶Sr ratio at Sites 695 and 696 (at 180.7 mbsf (4.1 Ma) and 233.8 mbsf (7.0 Ma) respectively) is due to high concentrations of volcanically derived strontium relative to that derived from terrigenous material.

An independent test of the assumptions involved in the calculations may be obtained by comparing the relation between the δ^{18} O signature of the pore waters and the estimated amount of volcanically derived strontium with that predicted by alteration of volcanic material as proposed by Egeberg et al. (this volume) The curve in Figure 9 is constructed using an average concentra-



Figure 9. Oxygen isotope distribution (data from Tables 3, 5, and 6) vs. amount of strontium derived from alteration of basic volcanic material (estimated by the means of equation 1 and 2) The solid line is established by the means of equation 5 and 6 in Egeberg et al. (this volume) and by assuming a concentration of strontium in volcanic glass of 100 ppm.

tion of strontium in basaltic material of 100 ppm (Spooner, 1976) Inspection of Figure 9 reveals a good simulation of the overall trend. Much of the scatter may be attributed to compositional variations in the reacting material.

Sites 694 and 697

The pore water strontium at Site 694 probably has the most radiogenic ⁸⁷Sr/⁸⁶Sr values yet observed at DSDP or ODP sites. The average ⁸⁷Sr/⁸⁶Sr ratio of dissolved strontium in Site 694 pore water is 0.70986. Ratios (⁸⁷Sr/⁸⁶Sr) greater than that of present-day seawater (0.70920) are rare in DSDP/ODP pore waters.

As with the other Leg 113 sites, the variations in major element chemistry at Sites 694 and 697 may be interpreted in terms of alteration of volcanic debris dispersed in the sediments (Egeberg et al., this volume) Evidently the effect of volcanically derived strontium is completely masked by strontium derived from terrigenous material. At Site 697 unusually high 87Sr/87Sr isotope ratios are confined to the deeper samples.

The poor core recovery at Site 694 was attributed to the presence of coarse sands, and large parts of the recovered sequence consist of turbidites with abundant lithic fragments. Site 694 may be the first DSDP/ODP site where the presence of strontium released from terrigenous material is unequivocal in the pore water.

At Site 694 the content of sedimentary carbonate is so low (less than 0.1%) that the only significant sources of strontium in the pore waters are indigenous seawater, terrigenous material, and altered basic volcanic detritus. The profiles in Figure 10 are determined using the same input parameters as at Sites 693, 695, and 696. Comparison with Figure 8 shows that the high ⁸⁷Sr/⁸⁶Sr isotope ratios at Site 694 are not caused by excessively large amounts of terrigeneously derived strontium, but rather by the comparatively low amounts of volcanically derived strontium. Thus the radiogenic input from the terrigenous material is not fully compensated by a nonradiogenic input from basic volcanic material. Similar calculations may be carried out for Site 697.



Figure 10. Amount of dissolved strontium derived from terrigenous and basic volcanic sources at Site 694, estimated by the means of equation 1 and 2 and by assuming a ${}^{87}\text{Sr}{}^{86}\text{Sr}$ ratio of terrigenous strontium of 0.728 and 0.703 for the volcanogenic strontium.

CONCLUSIONS

The strontium concentration and the distribution of ⁸⁷Sr/ ⁸⁶Sr ratios in Leg 113 interstitial waters may be interpreted in terms of mixing of strontium from four reservoirs, indigenous seawater, carbonates, altered basic volcanics, and siliceous terrigenous detrital material. The 87Sr/86Sr ratio of the pore water is determined by the magnitude of the end member isotope ratio and its reactivity, rather than by the size of the reservoir. Thus, at the carbonate-rich sites (689 and 690) the amount of strontium held in carbonates is more than 50 times the size of that held in the terrigenous material, yet the 87Sr/86Sr ratio of the interstitial waters is dominated by strontium released from terrigenous material. This indicates an almost complete lack of recrystallization of carbonates. At the hemipelagic sites the relative size of the terrigenous and basic volcanic strontium reservoir is roughly 20:1, yet at Sites 693, 695, and 696, the strontium isotope composition of dissolved strontium is dominated by strontium from the reactive volcanic reservoir. Only at Site 694 and in the deepest samples from Site 697 does strontium from siliceous terrigenous material dominate. This is the result of a low contribution from the other reservoirs due to low concentrations of carbonate and limited alteration of basic volcanic material.

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