

26. Pb AND Sr ISOTOPIC STUDIES OF IGNEOUS ROCKS CORED DURING LEG 31 OF THE DEEP SEA DRILLING PROJECT

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ABSTRACT

A number of igneous rock samples cored in the Philippine Sea during Leg 31 have been analyzed for Pb and Sr isotopic abundances and U-Th-Pb concentrations. Petrographic study of these samples indicates that all except the two-pyroxene andesite sample cored at Site 296 show evidence of alteration on the sea floor. The alteration products appear mainly to be montmorillonitic clays with minor calcite and K-feldspar. Mineralogically, the basalts from Sites 292 and 294 appear to have alkaline affinities, while the U/Pb and Th/U ratios of samples from Site 291 are more suggestive of ocean ridge basalts.

The Sr isotopic compositions of all Philippine Sea samples are very similar averaging 0.7037 ± 0.0002 . It is not clear what effect alteration may have had in producing these ratios. In terms of Pb isotopes, the Philippine Sea basalts have Pb^{206}/Pb^{204} and Pb^{207}/Pb^{204} ratios in the range of fresh ocean floor basalts. However, the Pb^{208}/Pb^{204} ratios of these samples are significantly higher than those of most known ocean floor basalts. The explanation for these high ratios favored in this report involves major increases in the Th/Pb and, to a lesser extent, the U/Pb ratios in the basalts during or shortly after the time of their formation.

The Pb isotopic composition and U-Th-Pb concentrations in the andesite from Site 296 are similar to andesites from west central Japan except for a somewhat higher initial Pb^{208}/Pb^{204} ratio. The cause for this high ratio is not obvious although it may involve an isotopically anomalous source region.

INTRODUCTION

Isotopic analyses of Pb and Sr in a number of igneous rock samples cored during Leg 31 are the main subject of this chapter. Analyses of U-Th-Pb concentrations in these samples are also presented. An important aspect of this work concerns the effects of sea-floor alteration on the Pb and Sr isotopic compositions of sea-floor basalts.

The extent of alteration of the analyzed samples is clearly evident in thin section. Petrographic descriptions of these samples are presented in the Appendix. Nearly all of the samples except Site 296, Core 55, Section 1 show some evidence of alteration, consisting most commonly of the replacement of olivine and/or interstitial glass with clays. The work of Bass et al. (1973) suggests that these clays are montmorillonites of various types including green, brown, and colorless varieties. Calcite accompanies the clays in some samples. Although the pyroxenes nearly always appear unaltered, olivine is evident only as relict outlines and plagioclase shows signs of incipient alteration in some samples.

All of the samples analyzed are basalts petrographically, except for Site 296, Core 55, Section 1, which is a two-pyroxene andesite. The basalts from Sites 292 and 294 have pyroxenes which are likely to be high in titanium as suggested by their purplish-brown color in plain light. This, in addition to the K-feldspar found in samples from Site 292, suggests that these basalts may have alkaline affinities. A program of major and trace element analyses of about 20 igneous rock samples from Leg 31 currently being conducted should provide definitive data concerning these suggestions. The andesite from Site 296 is indistinguishable from the two-pyroxene andesites common to most volcanic arcs around the Pacific basin. This suggests the ridge from which this sample was cored could be part of a now inactive volcanic arc.

ANALYTICAL METHODS

Samples were received as 10-cc mini cores. These cores were split so that a thin section could be made of one half while the other half was used for isotopic analysis. For the isotopic analysis, the samples were broken into fragments of approximately $\frac{1}{2}$ cm size and washed in warm twice distilled 2N HCl for 20 to 30 min. This was done mainly to eliminate any possible contaminating

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effects due to the original coring operation or the minicoring done at DSDP laboratories. After the acid wash, the samples were washed in three times distilled H₂O and pulverized in a boron carbide mortar.

The procedures used for sample dissolution and the separation of Pb, Sr, U, and Th were identical to those reported in Bass et al. (1973). Teflonware was used throughout to minimize the analytical blank. Blank concentrations for the whole chemical procedure were between 5-15 ng/g for Pb and 1-2 ng/g for Sr. The U and Th blanks measured in the Santa Barbara laboratory were an insignificant part of the concentrations reported here.

The precision of the analyses was normally within the following limits (1σ): $Pb^{208}/Pb^{204} = \pm 0.20\%$, $Pb^{207}/Pb^{204} = \pm 0.15\%$, $Pb^{206}/Pb^{204} = \pm 0.10\%$. All reported values have been normalized to absolute values through the NBS 981 standard. Shields' 1968 values for the NBS 981 standard were used in the normalization (Table 1).

PRESENTATION OF DATA

All of the isotopic and concentration data obtained in the present investigation are presented in Table 1. Consideration of these data will show that not only are there differences between samples from different sites in the Philippine Sea, but also differences between samples from the same site.

The samples from Site 291 appear to be the least altered in this section. The U, Th, and Pb concentrations of these samples and the Pb isotopic composition of Site 291, Core 5, Section 2 suggest close affinities with ocean ridge basalts (e.g., Church and Tatsumoto, in press). The Sr^{87}/Sr^{86} ratio of Site 291, Core 5, Section 2 is higher than average MORB ($Sr^{87}/Sr^{86} = 0.7027$; Hart, 1971 a, b). It is not clear whether this higher ratio is due to alteration effects or an originally higher initial Sr^{87}/Sr^{86} ratio. More work is needed on mineral separates of these samples to resolve this question.

The two samples from Site 292 are clearly distinct from the other samples in terms of U, Th, and Pb con-

centrations and Pb isotopic composition. The fact that Site 292, Core 46, Section 1 has a very high U^{238}/Pb^{204} ratio suggests it either gained U or lost Pb at some time in its history.

The consistency of the measured Sr^{87}/Sr^{86} ratios in the analyzed samples is remarkable in view of the range of alteration observed in these samples. Alteration must have some effect on these ratios although many factors can enter into the quantitative measure of this effect such as the extent of alteration, the nature of the fluid phase involved in the alteration process, the concentrations of Sr and Pb in the mineral phases, interstitial phases and the fluid phase, and the isotopic composition of Sr in all these phases. Suffice it to say that the Sr^{87}/Sr^{86} ratios in these samples vary only from 0.70350 to 0.70385 regardless of the degree of alteration.

The pyroxene andesite sample (Site 296, Core 55, Section 1) is distinct from the other samples in U-Th-Pb concentrations, Pb isotopic composition, as well as mineralogy. Again, the concentration and isotopic data are more like those of volcanic arc lavas (e.g., see Hedge and Knight, 1969, p. 17, Sample 204) than ocean basin lavas. The high Pb^{208}/Pb^{204} ratio coupled with a low Pb^{206}/Pb^{204} ratio is somewhat unusual. The unaltered nature of the sample suggests that the unusual Pb isotopic composition is not due to secondary alteration. Since two separate analyses of this sample yielded essentially the same isotopic ratios, the high Pb^{208}/Pb^{204} ratio also does not appear to be a result of contamination with the Pb^{208} spike used in the concentration analyses. Correction for radiogenic additions of Pb^{208} to the Pb in the sample assuming a 30 m.y. age (Ingle et al., 1973) does not significantly change the unusual character of this Pb. A more realistic possibility is that the source area for this lava was isotopically distinct. Tatsumoto (1966) has reported a Pb isotopic analysis for a trachyandesite from Iwo Jima which is also unusual compared to other western Pacific volcanic arc lava Pb isotopic compositions. Perhaps these unusual isotopic compositions are characteristic of the arc lavas erupted in this area. Further work may resolve this problematic situation.

TABLE 1
Isotopic and Concentration Data

Sample (Interval in cm)	Pb^{206}/Pb^{204a}	Pb^{207}/Pb^{204a}	Pb^{208}/Pb^{204a}	U^{238}/Pb^{204}	Sr^{87}/Sr^{86b}	Pb ^c	U ^c	Th ^c	U/Pb	Th/U	Age ^d
290-8, CC	—	—	—	—	0.70364	—	—	—	—	—	—
291-5-"A", 146-148	—	—	—	—	—	0.667	0.230	0.661	0.345	2.87	—
291-5-2, 115-117	18.437	15.452	38.463	14.21	0.70350	0.279	0.057	0.242	0.204	4.25	—
292-42-2, 68-70	—	—	—	—	0.70385	0.504	0.317	1.33	0.629	4.21	—
292-46-1, 120-122	19.206	15.566	39.265	73.50	0.70382	0.455	0.456	1.617	1.002	3.55	0
	18.826	15.548	38.818	—	—	—	—	—	—	—	37.5
294-7-1, 119-121	18.561	15.538	38.602	—	0.70375	2.600	—	—	—	—	—
296-55-1, 135-137	18.363	15.538	39.006	18.41	0.70382	6.692	1.79	6.86	0.267	3.83	0
	18.368	15.552	39.014	—	—	—	—	—	—	—	0
	18.262	15.540	38.878	—	—	—	—	—	—	—	30

^a Absolute ratios normalized to N.B.S. 981 values reported by Catanzaro (1968). Atom ratios.

^b Sr^{86}/Sr^{88} normalized to 0.1194; Eimer and Amend $SrCO_3$ standard = 0.70800; precision of Sr^{87}/Sr^{86} ratios is ± 0.0002 ($2\sigma_x$).

^c Concentrations in parts per million.

^d Age (m.y.) used to correct observed Pb isotope ratios to initial ratios (see McKee, this volume).

DISCUSSION

One of the fundamental questions that one might hope to clarify using isotopic analyses is whether the Philippine Sea basalts originated in an inter-arc basin environment or at an oceanic spreading center. Unfortunately, since only a very limited number of Pb and Sr isotopic analyses are available for inter-arc basin lavas, this question cannot be fully treated at the present time. Nonetheless, I will attempt in the following paragraphs to discuss the Philippine Sea data from this standpoint using the available data.

In terms of Sr isotopic compositions, the Philippine Sea basalts (average $\text{Sr}^{87}/\text{Sr}^{86} = 0.70371$) are clearly more radiogenic than the Mariana inter-arc basin basalts (average $\text{Sr}^{87}/\text{Sr}^{86} = 0.7028$; Hart et al., 1972). Whether this difference is due to the fact that the Philippine Sea basalts have been altered or whether their initial ratios were actually different is not clear. Corrections for the addition of radiogenic Sr^{87} to the Sr in the samples, assuming an average age of 35 m.y. and using preliminary Rb and Sr concentration analyses, cannot resolve this basic discrepancy. Hart (1971b) has reported $\text{Sr}^{87}/\text{Sr}^{86}$ ratios for apparently fresh basalts from the inter-arc basin behind the Tonga arc which overlap the range of the Philippine Sea basalts suggesting that inter-arc basin basalts may show as much variation in $\text{Sr}^{87}/\text{Sr}^{86}$ ratios as do ocean basin basalts (e.g., Peterman and Hedge, 1971).

The range in $\text{Pb}^{206}/\text{Pb}^{204}$ and $\text{Pb}^{207}/\text{Pb}^{204}$ observed in the Philippine Sea basalts is similar to the range observed by Church and Tatsumoto (in press) in fresh and slightly altered basalts from the eastern Pacific Ocean ridge systems (Figure 1). Compared to the Mariana inter-arc basin basalts analyzed by Meijer (1974), the Philippine Sea basalts are generally more radiogenic. Although sea-floor alteration and radioactive decay may have changed the Philippine Sea basalt Pb isotopic compositions, it is unlikely that these processes could convert the ratios observed in the Mariana inter-arc basin basalts ($\text{Pb}^{206}/\text{Pb}^{204} = 18.02 - 18.19$, $\text{Pb}^{207}/\text{Pb}^{204} = 15.38 - 15.49$, $\text{Pb}^{208}/\text{Pb}^{204} = 37.75 - 37.82$) to those observed in the Philippine Sea samples in a 30-40 m.y. period. As with the Sr isotopic data, inter-arc basin basalts may show a range of Pb isotopic compositions similar to that of oceanic basalts once a sufficient number of true inter-arc basin basalts have been analyzed.

The $\text{Pb}^{208}/\text{Pb}^{204}$ ratios of the Philippine Sea basalts are clearly higher than those reported by Church and Tatsumoto (in press) for eastern Pacific ridge basalts (Figure 1). Although these higher ratios could be the result of isotopic differences between the mantle source areas for these two suites of samples, a large part of the difference between them can be explained by sea-floor alteration. Alteration could change the original isotopic composition of the basalts in two ways: (1) by exchanging more radiogenic Pb with the original Pb in the basalt, and (2) by increasing the U/Pb and Th/Pb ratios of the basalts.

If an exchange mechanism were the sole cause of the high ratios, it would seem likely that the more radiogenic component would come either from seawater or

the overlying sediments. Analyses of the Pb isotopic compositions of several Philippine Sea sediment samples reported by Sun (1973) are plotted in Figure 1. From these data it is evident that although sediment Pb contamination may explain the high $\text{Pb}^{208}/\text{Pb}^{204}$ ratios of 291, Core 5, Section 2 and 294, Core 7, Section 1, it cannot explain the high ratio observed in 292, Core 46, Section 1. A similar argument can be made regarding seawater contamination using Mn-nodule Pb (Reynolds and Dasch, 1971) as an approximation of seawater Pb. The high $\text{Pb}^{206}/\text{Pb}^{204}$ and $\text{Pb}^{208}/\text{Pb}^{204}$ ratios reported by Bass et al (1973) for altered basalts from DSDP Leg 17 present similar problems.

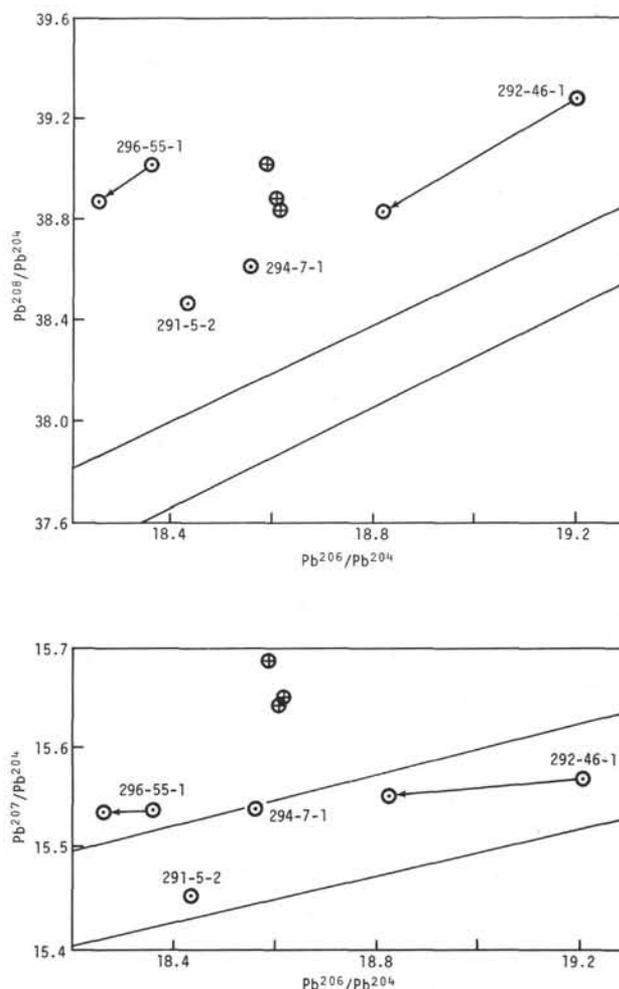


Figure 1. Plots of $\text{Pb}^{206}/\text{Pb}^{204}$ versus $\text{Pb}^{207}/\text{Pb}^{204}$ and $\text{Pb}^{206}/\text{Pb}^{204}$ versus $\text{Pb}^{208}/\text{Pb}^{204}$ for the Philippine Sea igneous rock samples (\bullet), Philippine Sea sediments ($+$) as reported by Sun (1973) and oceanic basalts from the eastern Pacific Ocean (fields enclosed by fine lines) reported by Church and Tatsumoto (in press). The lines with arrows point to the age corrected ratios assuming ages given in Table 1. Decay constants used in making the age corrections are as follows: $U^{235} = 0.98485 \times 10^{-9} \text{ yr}^{-1}$, $U^{238} = 0.15525 \times 10^{-9} \text{ yr}^{-1}$ and $Th^{232} = 0.49475 \times 10^{-9} \text{ yr}^{-1}$.

Increases in the Th/Pb and U/Pb ratios of the Philippine Sea basalts either during or shortly after their time of formation appear to be a more likely mechanism for producing the "high" observed Pb^{208}/Pb^{204} and Pb^{206}/Pb^{204} ratios. To illustrate, I have recalculated the initial Pb isotopic ratios that 292, Core 46, Section 1 would have had shortly after the time of its formation assuming that (1) it acquired its high Th/Pb and U/Pb ratios shortly after the time of its formation, and (2) it has remained a closed system isotopically since the time of alteration. McKee (this volume) has dated samples from Site 292 by the K-Ar method, and reported an average age of approximately 37.5 m.y. This value was used to recalculate the Pb isotopic ratios as given in Table 1 and plotted in Figure 1. The recalculation produces a shift in the Pb isotope ratios of Site 292, Core 46, Section 1, which causes them to lie closer to the trend of fresh oceanic basalts from the eastern Pacific Ocean (Church and Tatsumoto, in press). The fact that the recalculated ratios do not plot directly within the ocean floor basalt field could be due either to minor contamination with more radiogenic Pb, or to differences in the Pb isotopic composition of the source areas producing these different suites of lavas. Isotopic analyses of fresh mineral separates may help to resolve this question. Unfortunately, this still does not provide a definitive answer to the question posed initially concerning the environment in which these samples were formed. Hopefully, as more isotopic data become available, a more definitive answer to this question will become apparent.

CONCLUSIONS

1. Petrographic study of the samples discussed in this report and numerous additional samples indicates that nearly all of the igneous rock samples cored during DSDP Leg 31 are all altered to some degree. The most common alteration products appear to be montmorillonitic clays with minor amounts of calcite and possibly phillipsite. Pyroxenes and to a lesser extent plagioclases appear unaltered, while olivines and interstitial glass are usually completely replaced.

2. The mineralogy of basalts from Sites 292 and 294 is suggestive of alkali basalts, while samples from Site 291 have U/Pb and Th/Pb ratios similar to those of ocean ridge basalts. A two-pyroxene andesite from Site 296 looks very much like two-pyroxene andesites found in most circum-Pacific volcanic arcs. This suggests that the ridge from which this sample was cored was part of a now inactive volcanic arc.

3. The range in Sr^{87}/Sr^{86} ratios of all samples analyzed is very limited. The average value is 0.7037 ± 0.0002 . It is not clear to what extent alteration has effected these ratios.

4. The Pb^{206}/Pb^{204} and Pb^{207}/Pb^{204} ratios of three of the analyzed Philippine Sea basalts are within the range of eastern Pacific ridge basalts, while the Pb^{206}/Pb^{204} ratio of a basalt from Site 292 is higher than most ridge basalts. The Pb^{208}/Pb^{204} ratios of all the Philippine Sea basalts are higher than most ridge basalts. The explanation for these high ratios favored in the present report

involves a combination of an increase in the U/Pb and Th/Pb ratios of the basalts during or shortly after their time of formation causing relatively rapid increases in the Pb^{208}/Pb^{204} and Pb^{206}/Pb^{204} ratios, and either contamination with more radiogenic Pb or extraction from a distinct mantle source area.

5. The Pb isotopic composition of the andesite from Site 296 is unusual and suggests additional isotopic work in this area may prove to be worthwhile.

6. The question of whether the western Philippine Sea lithosphere was formed in an inter-arc basin environment or an ocean ridge environment cannot be resolved with the presently available isotopic and trace element data.

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APPENDIX
Sample Descriptions

Sample 290-8, CC: Megascopic—fine-medium grained greenish-gray basalt, nonvesicular, with randomly distributed limonitic specks and ubiquitous Fe-Ti oxides. No obvious sulfide phases present (probably altered to limonite). Microscopic—medium-grained basalt, intersertal texture with pyroxene partially enclosing plagioclase laths; plagioclase fresh, zoned, twinned, acicular, 0.2-0.5 mm grains, approximately 35%; clinopyroxene fresh, anhedral, stubby grains, 0.05-0.5 mm, 30%; altered interstitial areas mostly clays and iron oxides, 35%; no vesicles.

Sample 291-5 (center bit "A"), 146-148 cm: Megascopic—dense, grayish-black, very fine grained basalt with only minor alteration evident, no obvious sulfide phases present. Microscopic—basalt with quench texture, fine grained with occasional plagioclase microphenocrysts, 0.05-0.5 mm, fresh; quench clinopyroxene and plagioclase, fresh, approximately 20%; clinopyroxene also in small stubby crystals, 0.01-0.02 mm; most of sample is a brown felted mass of iron oxides and fresh(?) glass; some clay alteration evident in several cavities present, 1%-2%.

Sample 291-5-2, 115-117 cm: Megascopic—fine-grained greenish-gray basalt with orange specks, nonvesicular, no obvious sulfide phases present, friable. Microscopic—fine-medium grained basalt with intersertal texture; plagioclase fresh, acicular, twinned, 0.2-0.5 mm, 35%; clinopyroxene, fresh, stubby crystals, 0.05-0.5 mm, 30%; interstitial areas altered to clay and iron oxides.

Sample 292-42-2, 68-70 cm: Megascopic—medium-grained, dark greenish-gray vesicular basalt, vesicles approximately 15% lined with a drusy coating of greenish brown mineral (Mn-mineral?), some sulfides present, 1%-2%. Microscopic—medium-grained basalt, intergranular texture, possible olivine relicts (<1%) altered to clays; plagioclase phenocrysts An_{70} , 0.3 mm maximum, zoned mainly on outer rims (0.05 mm), incipiently altered to clays along cracks, 1%-2%;

groundmass clinopyroxene, 15%-20%, sector zoned, twinned, equigranular, mostly fresh, 0.02-0.1 mm, purplish tinge; groundmass plagioclase, An_{60-70} , $x = 0.05$ mm, minor alteration to clays, 45%-50%; interstitial areas altered to clays, approximately 30%; skeletal Fe-Ti oxides, 2-3%.

Sample 292-46-1, 120-122 cm: Megascopic—medium-grained vesicular greenish-gray basalt; vesicles 5%, 2-3 mm, contain white and greenish-brown crystals growing out from walls, some crystals platy others columnar; sulfide phases common. Microscopic—medium-grained basalt, intergranular texture; plagioclase phenocrysts, An_{70} , zoned along outer rims, 0.5 mm maximum, incipiently altered to clays, 2%-3%; groundmass pyroxene, fresh, stubby, 0.03-0.1 mm, purplish-brown color, 20%-25%; groundmass plagioclase, incipiently altered to clays, 35%-40%; skeletal Fe-Ti oxides, 4-5%, 0.1 mm; interstitial areas altered to K-feldspar and green, brown, and clear clays, 35%-40%.

Sample 294-7-1, 119-121 cm: Microscopic—fine-grained plagioclase-rich basalt; plagioclase incipiently altered to clays, $x = 0.05$ mm, 55%; quench clinopyroxene, fresh looking, purplish-brown color, 10%; acicular Fe-Ti oxides, 2%-3%; interstitial material altered to clay and calcite, calcite also fills several vesicles in slide.

Sample 296-55-1, 135-137 cm: Megascopic—medium-grained porphyritic, light gray, nonvesicular andesite, no obvious sulfides. Microscopic—porphyritic orthopyroxene-augite andesite, pilotaxitic texture; plagioclase phenocrysts 0.1-0.3 mm, commonly show sieve texture, complex zoning, inclusions of glass and minor pyroxene, commonly euhedral, fresh, 15%-20%; augite phenocrysts, 0.05-0.1 mm, commonly euhedral, contain many fine-grained inclusions especially along rims, occasionally twinned, 6%-8%; orthopyroxene microphenocrysts, euhedral, contain small inclusions, fresh, 5%-6%; possible olivine relicts, only mesh texture and outline preserved, 3-4 grains; Fe-Ti oxides 0.01-0.02 mm, 1%-2%; groundmass comprised of very fine grained fresh plagioclase laths, 30%; clinopyroxene, 6-7%; orthopyroxene, 2-3%; Fe-Ti oxides 1%-2%; and fresh light brown glass, 30%.