

Reports

Genetic Relations of Oceanic Basalts as Indicated by Lead Isotopes

Abstract. *The isotopic compositions of lead and the concentrations of lead, uranium, and thorium in samples of oceanic tholeiite and alkali suites are determined, and the genetic relations of the oceanic basalts are discussed. Lead of the oceanic tholeiites has a varying lead-206 : lead-204 ratio between 17.8 and 18.8, while leads of the alkali basalt suites from Easter Island and Guadalupe Island are very radiogenic with lead-206 : lead-204 ratios between 19.3 and 20.4. It is concluded that (i) the isotopic composition of lead in oceanic tholeiite suggests that the upper mantle source region of the tholeiite was differentiated from an original mantle material more than 1 billion years ago and that the upper mantle is not homogeneous at the present time, (ii) less than 20 million years was required for the crystal differentiation within the alkali suite from Easter Island, (iii) no crustal contamination was involved in the course of differentiation of rocks from Easter Island; however, some crustal contamination may have affected Guadalupe Island rocks, and (iv) alkali basalt may be produced from the tholeiite in the oceanic region by crystal differentiation. Alternatively the difference in the isotopic composition of lead in oceanic basalts may be produced by partial melting at different depths of a differentiated upper mantle.*

The genetic relations of tholeiite and alkali basalt have been debated. Powers (1) and Macdonald (2) have concluded that alkali basalt is a derivative from a tholeiitic parent; but, on the other hand, Kuno and his colleagues (3) and Yoder and Tilley (4) have suggested that two or more independent parental magmas are generated in the mantle by frac-

tional melting of peridotite under different conditions of pressure and temperature.

Recent petrochemical studies by Engel *et al.* (5) on the oceanic basalts suggest that the oceanic tholeiitic basalts are the principal, or only, parental magma generated in the upper mantle under the ocean. The tholeiitic basalts are charac-

terized by extremely low content of alkali metals, alkaline earths, phosphorus, titanium, zirconium, thorium, and uranium in comparison with the oceanic alkali basalts that cap submarine and island volcanos. Engel *et al.* suggest that the alkali-rich basalts are fractionated from oceanic tholeiites by processes of magmatic differentiation.

Tatsumoto *et al.* (6) reported that the amounts of thorium and uranium and the ratio of Th/U in the oceanic tholeiites are quite low when compared with these values in alkali basalts from oceanic localities. The Th/U ratio of the oceanic tholeiites averages about 1.8 and this value is only half of that commonly inferred for the crust and the upper mantle (7-9). Hence, the questions are: (i) What is the isotopic composition of lead in typical oceanic basalts, or does the isotopic composition of lead provide some suggestions regarding the genetic relations of the tholeiites and alkali basalts? (ii) Do the lead isotope studies confirm or invalidate the concept that the observed Th/U ratio is approximately that of the mantle source regions of the oceanic tholeiites?

The interrelations of the isotopic composition of lead and the content of uranium, thorium, and lead in volcanic rocks provide useful restrictions on the chemical differentiation system within the upper mantle and the crust since the isotopic composition of lead is the initial isotope ratios of lead plus the time-integrated result of the ratios U/Pb and Th/Pb in a given chemical system. It has been shown that U/Pb and Th/Pb ratios in volcanic rocks vary widely enough to produce significant isotopic differences (10). A number of analyses have recently been reported for the isotopic composition of lead in volcanic rocks (10-12). Except for (10), however, these papers have discussed only lead isotopes without knowing the U/Pb and Th/Pb ratios in the rocks; the interpretations are thus limited.

The isotopic composition of lead and the concentrations of lead, uranium, and thorium in oceanic basalts will be given in this report and their genetic implications will be discussed. Three tholeiitic basalt samples are from the Mid-Atlantic Ridge and three are from the East Pacific Rise. Two alkali basalt series are from Easter Island and Guadalupe Island. Petrochemical studies of these oceanic tholeiites and Guadalupe Island rocks have already been reported (5).

Lead in the alkali basalts was extracted by heating ($\sim 1100^{\circ}\text{C}$) a mix-

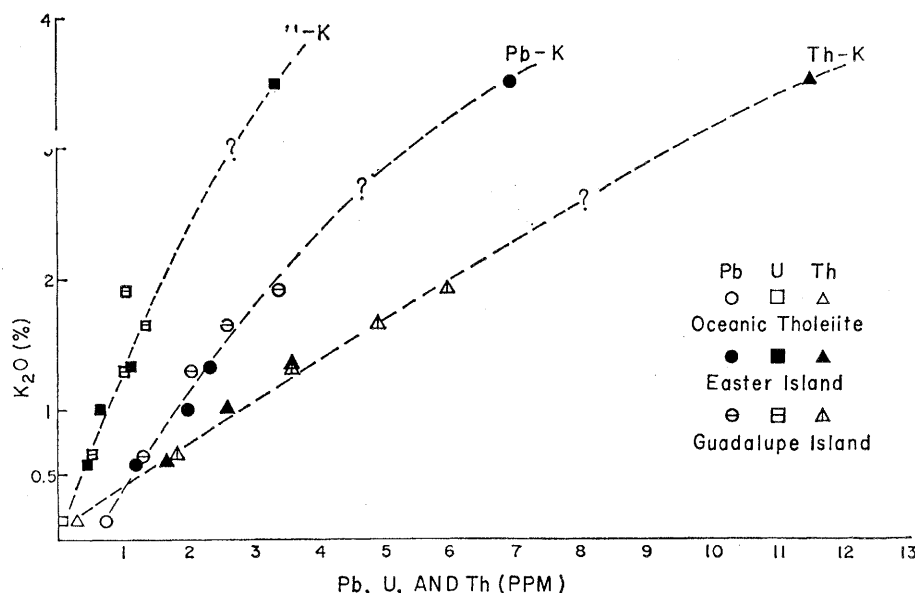


Fig. 1. Relations of concentrations between potassium and lead, uranium, and thorium.

ture of nine parts powdered sample and one part graphite powder under vacuum ($10^{-3} \sim 10^{-5}$ mm-Hg) (13). A modified procedure was used for tholeiite because of its low lead content. An uncrushed block sample (~ 40 g) was placed in a graphite crucible and heated to 1250°C in a vacuum. All samples melted at around 1100° to 1150°C , and more than 90 percent of the lead was usually recovered. The isotopic composition of the lead from AD3 was obtained by two methods: pyrochemical extraction and chemical decomposition. The results are compared in Table 1. Two additional comparisons of the two methods will be reported by Doe (14). The differences in isotopic composition were less than measurement errors, and no fractionation was observed during pyrochemical extraction. The volatilized lead was deposited on a cold finger and was purified further by barium coprecipitation (10) and a conventional dithizone extraction. Each sample included more than $15\text{ }\mu\text{g}$ of lead, while laboratory contamination amounted to only about $0.03\text{ }\mu\text{g}$ of lead per analysis. The concentrations of lead, uranium, and thorium were determined by isotope dilution (10). The mass spectrometry for the measurement of lead isotope ratios was the same as reported earlier (10). No mass discrimination factor is applied to the observed ratios. The reliability of the isotopic ratio determination may be estimated from the replicate measurements in Table 1 and from the CIT lead standard used by Tatsumoto (10).

The concentrations of lead, uranium, and thorium for oceanic tholeiites and members of the two alkali series are listed in Table 1 together with concentrations of Na_2O and K_2O . It should be emphasized that the concentrations of lead, uranium, and thorium in the oceanic tholeiites are about one-half to one-fourth those of the tholeiites from Hawaii and Japan (10), and that positive correlations exist between the amounts of potassium and lead, uranium, and thorium in the analyzed rock suites (Fig. 1) (15).

The observed $\text{U}^{238}/\text{Pb}^{204}$ ratios increase toward the rhyolite in the oceanic tholeiite and alkali series; however, observed $\text{Th}^{232}/\text{U}^{238}$ ratios have no progressive change. The $\text{Th}^{232}/\text{U}^{238}$ ratios are extremely low for the oceanic tholeiites (about 2) (6), as are the Japanese tholeiites (10), but they are high in the alkali series (about 4). The $\text{Th}^{232}/\text{U}^{238}$ ratio in labradorite-andesine

alkali basalt from Guadalupe Island is very high because of the low uranium content (15). Conceivably the irregularity of $\text{Th}^{232}/\text{U}^{238}$ might be reflected by the oxygen pressure and water content in magmas (5, 16), since uranium has two oxidation states (+4 and +6) while thorium has one (+4). The relation of $\text{U}^{238}/\text{Pb}^{204}$ and $\text{Th}^{232}/\text{U}^{238}$ ratios will be discussed further in conjunction with genetic relations of oceanic basalt.

The isotopic compositions of lead in the tholeiites and the alkali suites are given in Table 1. The observed lead isotope ratios for Easter Island rocks are similar to those of the alkali feldspar

[sample DWHD-47(F)] in volcanic sediments dredged from the East Pacific Rise (9), and the measured ratios of Ourito obsidian from Easter Island agree within experimental error with the value reported by Patterson and Duffield (17). The leads of all samples are further compared in Fig. 2, which is a combined diagram of $\text{Pb}^{206}/\text{Pb}^{204}$ versus $\text{Pb}^{207}/\text{Pb}^{204}$ (lower part) and $\text{Pb}^{206}/\text{Pb}^{204}$ versus $\text{Pb}^{208}/\text{Pb}^{204}$ (upper part). The primary growth curves (G) for a chemically closed system with present-day $\text{U}^{238}/\text{Pb}^{204}$ ratios (μ_0) of 8.6 and 8.8 since the earth formed (4.55 billion years ago) are shown in the lower part

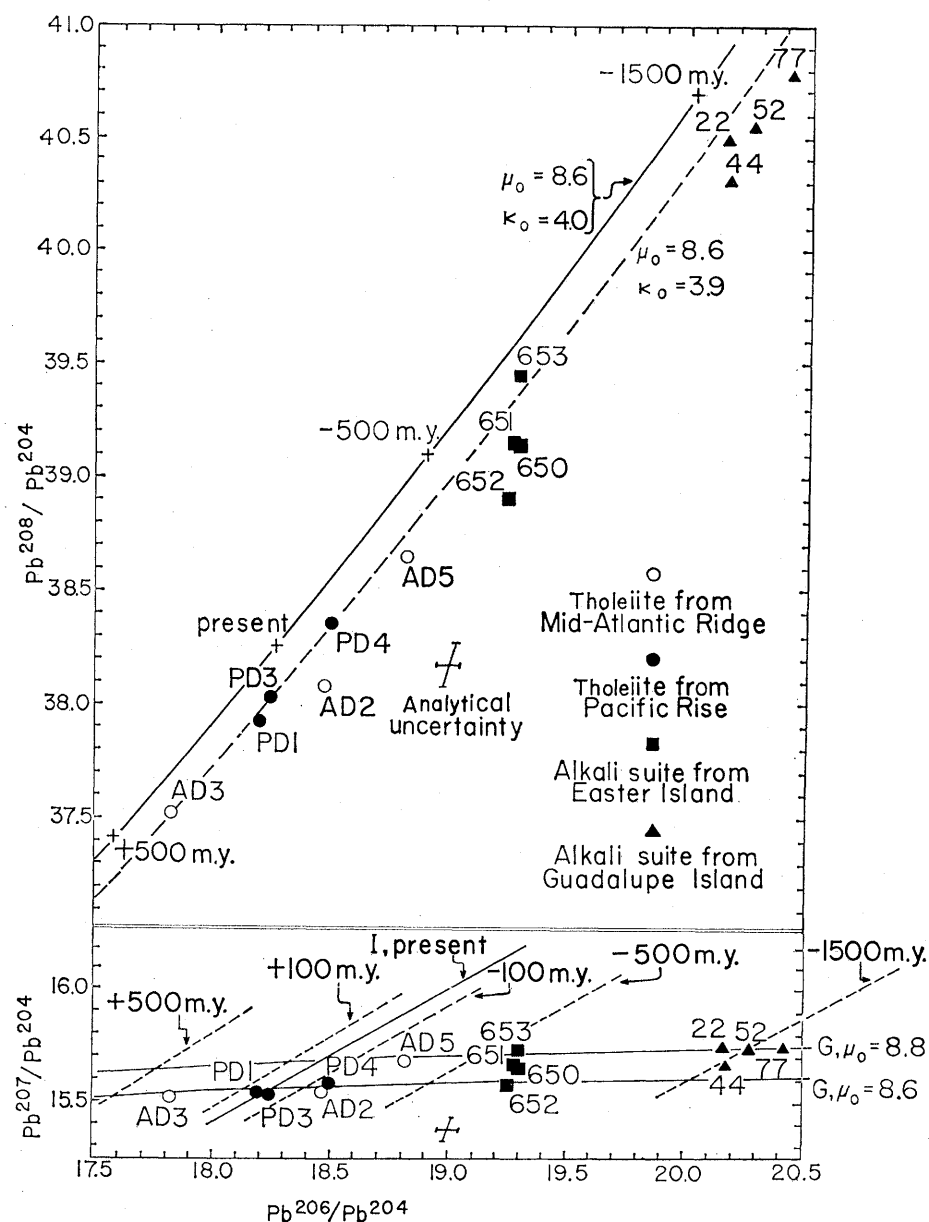


Fig. 2. Ratios of $\text{Pb}^{206}/\text{Pb}^{204}$ versus $\text{Pb}^{207}/\text{Pb}^{204}$ (lower part) and $\text{Pb}^{206}/\text{Pb}^{204}$ versus $\text{Pb}^{208}/\text{Pb}^{204}$ (upper part) in tholeiites and rocks from Easter and Guadalupe Islands. The primary growth curves having $\mu_0 = 8.6$ and 8.8 (lower part), $\mu_0 = 8.6$ and $\kappa_0 = 4.0$ and $\mu_0 = 8.6$ and $\kappa_0 = 3.9$ (upper part) are given by G. The primary isochrons (I) are also given for present, ± 100 million years, ± 500 million years, and -1500 million years.

Table 1. Concentrations of alkali, lead, uranium, and thorium and the isotopic composition of lead in volcanic rocks of Easter and Guadalupe Islands and in oceanic tholeiites.

Sample No.	Sample description	Concentration					Observed atomic ratio		Method†	Isotopic composition of lead (atomic ratio)						
		Percentage		Parts per million			U ²³⁸ /Pb ²⁰⁴	Th ²³² /U ²³⁸		Pb ²⁰⁶ /Pb ²⁰⁴	Pb ²⁰⁶ /Pb ²⁰⁷	Pb ²⁰⁶ /Pb ²⁰⁸	Pb ²⁰⁷ /Pb ²⁰⁴	Pb ²⁰⁸ /Pb ²⁰⁴		
		K ₂ O*	Na ₂ O*	Pb	U	Th										
<i>Easter Island rocks</i>																
PV 650	Sodic rhyolite glass (Cerro Ourito obsidian)	3.51	4.94	6.94	3.33	11.51	31.4	3.57	PP	19.306 19.31	1.2302 1.232	0.4923 0.493(17)	15.66	39.15		
PV 652	Andesine andesite	1.33	4.31	2.33	1.11	3.61	30.9	3.37	PP	19.253	1.2353	0.4944	15.58	38.94		
PV 653	Alkali basalt	1.00	3.26	1.98	0.63	2.58	21.1	4.19	PP	19.301	1.2271	0.4891	15.73	39.46		
PV 651	Tholeiite with alkali basalt affinity	0.58	3.30	1.20	0.44	1.67	23.9	3.93	PP	19.280	1.2302	0.4923	15.67	39.16		
<i>Guadalupe Island rocks</i>																
Gu 77	Labradorite-andesine alkali basalt									20.421 20.452	1.3016 1.2955	0.5012 0.5010				
Gu 52	Labradorite-andesine alkali basalt	1.93 1.65	4.80 4.08	3.39 2.58	1.05 1.36	7.16 4.91	21.0 35.6	6.82 3.73	PP PP	20.437 20.278	1.2986 1.2894	0.5011 0.5001	15.74 15.73	40.78 40.55		
Gu 22	Labradorite alkali basalt	1.30	4.00	2.04	1.00	3.60	33.1	3.72	PP	20.172	1.2801	0.4981	15.76	40.49		
Gu 44	Labradorite olivine basalt with alkali affinity	0.65	2.25	1.30	0.51	1.83	26.5	3.68	PP	20.178	1.2877	0.5006	15.67	40.31		
<i>Oceanic tholeiites‡</i>																
AD 2	Tholeiite	0.14	3.01	1.29	0.16	0.15	7.9	0.9	PB PP	18.470 18.471	1.1880 1.1891	0.4845 0.4854				
AD 3	Tholeiite	0.24	2.66	1.15	.10	.29	5.6	3.0	C PB	17.823 17.824	1.1459 1.1484	0.4740 0.4761	15.54	38.01		
AD 5	Tholeiite	.08	2.92	0.56	.09	.13	10.5	1.4	PB PB	17.824 18.815 18.817 18.816	1.1472 1.2017 1.1987 1.2002	0.4751 0.4866 0.4870 0.4868	15.54	37.52		
PD 1	Tholeiite	.06	2.18	.49	.05	.13	6.4	2.8	PB	18.193	1.1711	0.4797	15.54	37.93		
PD 3	Tholeiite	.24	2.91	.49	.09	.21	11.6	2.5	PB PB PB	18.211 18.229 18.280	1.1739 1.1740 1.1754	0.4798 0.4798 0.4793				
PD 4	Tholeiite	.11	2.75	.53	.07	.12	8.5	1.7	PB	18.240 18.497	1.1744 1.1870	0.4796 0.4825	15.53 15.58	38.03 38.34		
Average for oceanic tholeiites		0.15		0.75	0.09	0.17										

* The alkali contents are from A. E. J. Engel (private communication) for the Easter Island rocks, and (5) for Guadalupe Island rocks and oceanic tholeiites. † C, PB, and PP denote chemical decomposition by HF + HClO₄, pyrochemical extraction from a block, and pyrochemical extraction from powdered sample, respectively. ‡ The contents of uranium, thorium, and Th/U ratios in oceanic tholeiites are from (6).

of Fig. 2. The primary isochrons (*I*), in which lead evolved in chemically closed and coetaneous systems with initial U^{238}/Pb^{204} ratios, are shown for the present, ± 100 million years, ± 500 million years, and -1500 million years. In the upper part of Fig. 2, the primary growth curves (*G*) are shown for $\mu_0 = 8.6$ and $\kappa_0 = 4.0$ by a solid line, and for $\mu_0 = 8.6$ and $\kappa_0 = 3.9$ by a dashed line. These primary isochrons and the primary growth curves that are defined by meteoritic lead are shown as a convenient frame of reference (*I8*). The μ and κ are the conventional means of indicating the atomic ratios of U^{238}/Pb^{204} and Th^{232}/U^{238} today, as calculated from the lead isotopes. The μ_0 and κ_0 indicate the ratios in a chemically closed system since the earth formed. The actually observed atomic ratios of U^{238}/Pb^{204} and Th^{232}/U^{238} from the concentrations are always expressed as *observed* μ and *observed* κ .

The average of the leads of the oceanic tholeiites fits the $\mu_0 = 8.6$ growth curve. The primary isochron which fits the oceanic tholeiites spreads between ± 300 million years. The spread is most pronounced for Atlantic samples. *PD1* and *PD3* fit an apparent growth curve $\mu_0 = 8.55$ and are very close to the present isochron ("Geochron").

The apparent μ_0 of these tholeiites is lower than that of ordinary galena [$\mu_0 = 9.0 \sim 9.1$ (*I9*)], "crustal lead" [$\mu_0 = 8.95$, readjusted value (*I0*) from (*9*)], or $\mu_0 = 9.0$, calculated from marine lead (*8*)], Gough Island basalts [$\mu_0 = 8.8$ (*I1*)], and Japanese basalts [$\mu_0 = 8.75$ (*I0*)]. However, it is similar to the μ_0 calculated for Hawaiian rock lead (especially to that for the lead in alkali basalt) (*I0*). These relations are illustrated in Fig. 3 in which Pb^{206}/Pb^{207} is plotted against Pb^{206}/Pb^{204} . The lowest μ_0 which fits the oceanic tholeiite indicates that the leads are more "primitive" than the others; in other words, the leads from the galena, "the crust," and Gough Island and Japanese basalts evolved in systems with higher μ a long time ago, so that the apparent μ_0 increased along with mantle differentiation in a manner such as Patterson and Tatsumoto (*9*) have discussed. The

Fig. 3 (right). Ratios of Pb^{206}/Pb^{207} versus Pb^{206}/Pb^{204} for volcanic rocks. For comparison, the values in volcanic rocks from Hawaii, Japan, and Iwo-Jima (*I0*), Gough and Ascension Islands (*I1*), and the Oregon Coast (*25*), and in a feldspar from East Pacific Rise (*9*) are also plotted.

Table 2. Comparison between observed μ and κ and calculated μ_1 and κ_1 by two step model.*

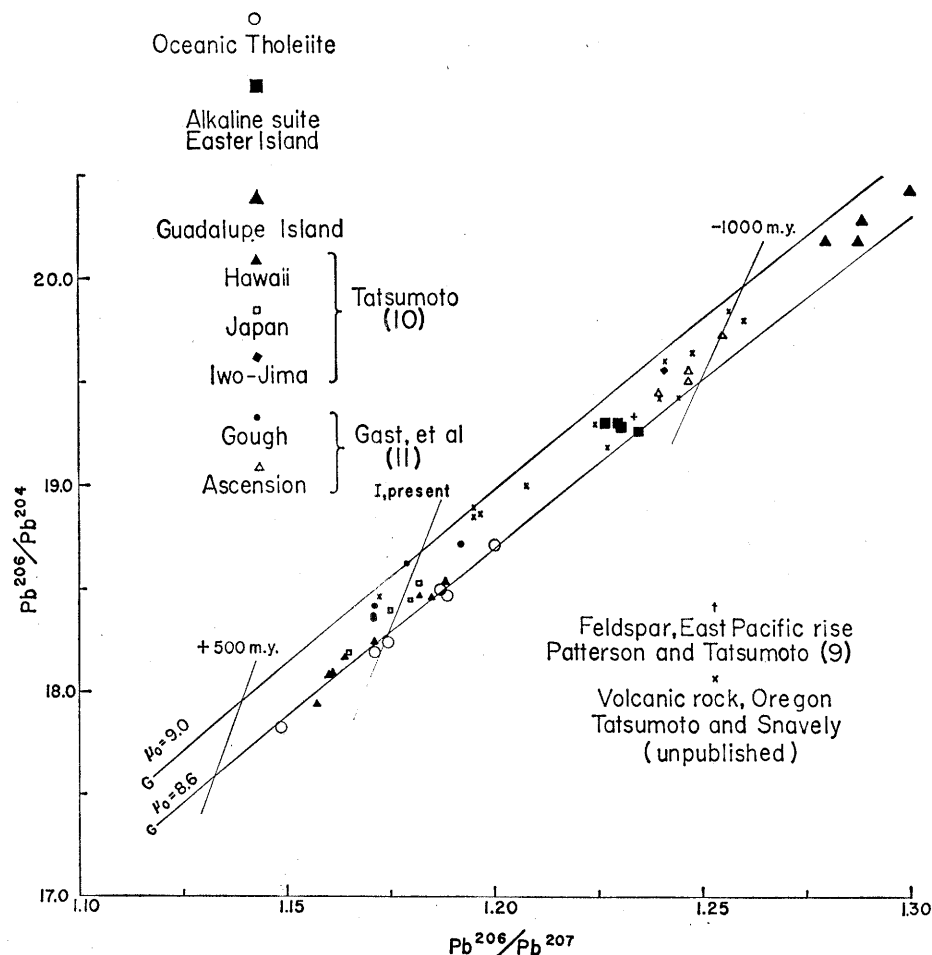
	AD3	PD4	AD5	PV651
<i>Observed</i>				
Pb^{206}/Pb^{204}	17.82	18.50	18.82	19.28
Pb^{208}/Pb^{204}	37.52	38.36	38.65	39.16
μ	5.6	8.5	10.5	23.9
κ	3.0	1.7	1.4	3.9
<i>Calculated</i>				
Starting $\mu_0 = 8.6$ and $\kappa_0 = 4.1^\dagger$				
Differentiation age;				
$t = 1.5$ billion years	μ_1 6.9	9.5	10.7	12.7
	κ_1 3.3	3.5	3.5	3.5
1.0 billion years	μ_1 5.9	10.0	11.9	14.7
	κ_1 2.7	3.3	3.3	3.3
0.5 billion years	μ_1 2.6	11.5	15.5	21.3
	κ_1 (—)	2.6	2.7	3.0
0.3 billion years	μ_1 0.6	13.6	20.3	30.1
	κ_1 (—)	2.0	2.31	2.7

* AD3, AD5, and PD4 were chosen to cover the range of isotopic composition of oceanic tholeiites; PV 651 is one example for alkali basalts. † The primary μ_0 and κ_0 in the source changed to μ_1 and κ_1 at t billion years ago. The necessary μ_1 and κ_1 are calculated to produce observed Pb^{206}/Pb^{204} and Pb^{208}/Pb^{204} ratios according to the following two equations:
Observed $Pb^{206}/Pb^{204} = 9.56 + \mu_0 (e^{\lambda_{238}t} - 1) + \mu_1 (e^{\lambda_{238}t} - 1)$
Observed $Pb^{208}/Pb^{204} = 29.71 + \mu_0 \kappa_0 (e^{\lambda_{232}t} - 1) + \mu_1 \kappa_1 (e^{\lambda_{232}t} - 1)$
 T (age of the earth) = 4.55 billion years.

primitiveness of lead in the oceanic tholeiite also is suggested in Fig. 4 in which Pb^{208}/Pb^{207} versus Pb^{208}/Pb^{204} are plotted. In Fig. 4, the leads of the oceanic tholeiite cluster near Hawaiian leads (*I0*) and have smaller apparent μ_0 and κ_0 than the others.

The lead in the alkali suite from Easter Island is radiogenic and shows a

Pb^{206} aberration from the model, and this fits the -750 million-year isochron without any spread. The Pb^{206}/Pb^{204} ratios lie within the range of 19.25 to 19.31, which is almost within the limits of experimental error, but the Pb^{208}/Pb^{204} ratios have a little variation (38.9 to 39.5). The apparent μ_0 of Easter Island lead is similar to that of



Ascension Island (11) and spreads between 8.6 and 8.8. The leads from Guadalupe Island are extremely radiogenic and show the model age of -1500 million years (Fig. 2). The Pb^{206}/Pb^{204} and Pb^{208}/Pb^{204} ratios of Guadalupe Island lead are the largest so far reported for

leads in volcanic rocks. The Pb^{206}/Pb^{204} and Pb^{208}/Pb^{204} ratios have a little variation (20.12 to 20.42 and 40.3 to 40.8, respectively).

It should be emphasized that the isotopic composition of lead in young volcanic rocks is somewhat variable

(Figs. 3 and 4) and suggests an isotopically nonuniform upper mantle (10, 11). This statement is true even for the source region (probably the upper mantle) of the most "primitive" oceanic tholeiite. These variations suggest that the source region of the tholeiite was differentiated from the original upper mantle material some time ago, as discussed previously for low Th/U and Sr^{87}/Sr^{86} ratios (6); therefore a chemically heterogeneous upper mantle exists. The upper mantle is an open system chemically; thus, the lead in basalts extruded from the upper mantle could not have developed in a single closed system in contrast to the idea that conformable ore lead came from an isotopically homogeneous mantle (10). The differentiation of the upper mantle by crystal differentiation, diffusion, convection, or other processes may have been accomplished over a prolonged period, as discussed by Patterson and Tatsumoto (9) and by Tatsumoto (10). For simplicity, the two-step model will be used for considering the relationship between the isotopic composition of lead and μ and κ .

In Table 2, the necessary μ and κ are calculated to generate the observed Pb^{206}/Pb^{204} and Pb^{208}/Pb^{204} ratios, assuming a two-step model in which the primary μ_0 and κ_0 changed to μ_1 and κ_1 by differentiation at t billion years ago. The observed μ and calculated μ_1 of the oceanic tholeiite agree if we consider that differentiation occurred about 1.0 to 1.5 billion years ago. The κ_1 of a differentiation age of 1.0 to 1.5 billion years for AD3 matches that of the observed κ but does not match for PD4 and AD5. The reason for the mismatch of PD4 and AD5 is not clear. It might be due to the partial melting of source material, to uncertainty about the age of the earth, or to the uncertainties of measurement of the Pb^{208}/Pb^{204} ratios in the present data and that of primordial lead. Or, the mismatch may be due to other causes such as inappropriateness of the two-step model. It is possible to reduce the κ_1 value to better agreement with the observed value if one uses the greatest Pb^{208}/Pb^{204} value for primordial lead within the uncertainties reported by Murthy and Patterson (20), which is 1.5 percent higher than their value of 29.71. The κ_1 value also can be reduced if a greater value such as 4.7 billion years instead of 4.55 billion years is used for the age of the earth, as suggested by Tilton and Steiger (21) and Wasserburg *et al.* (22).

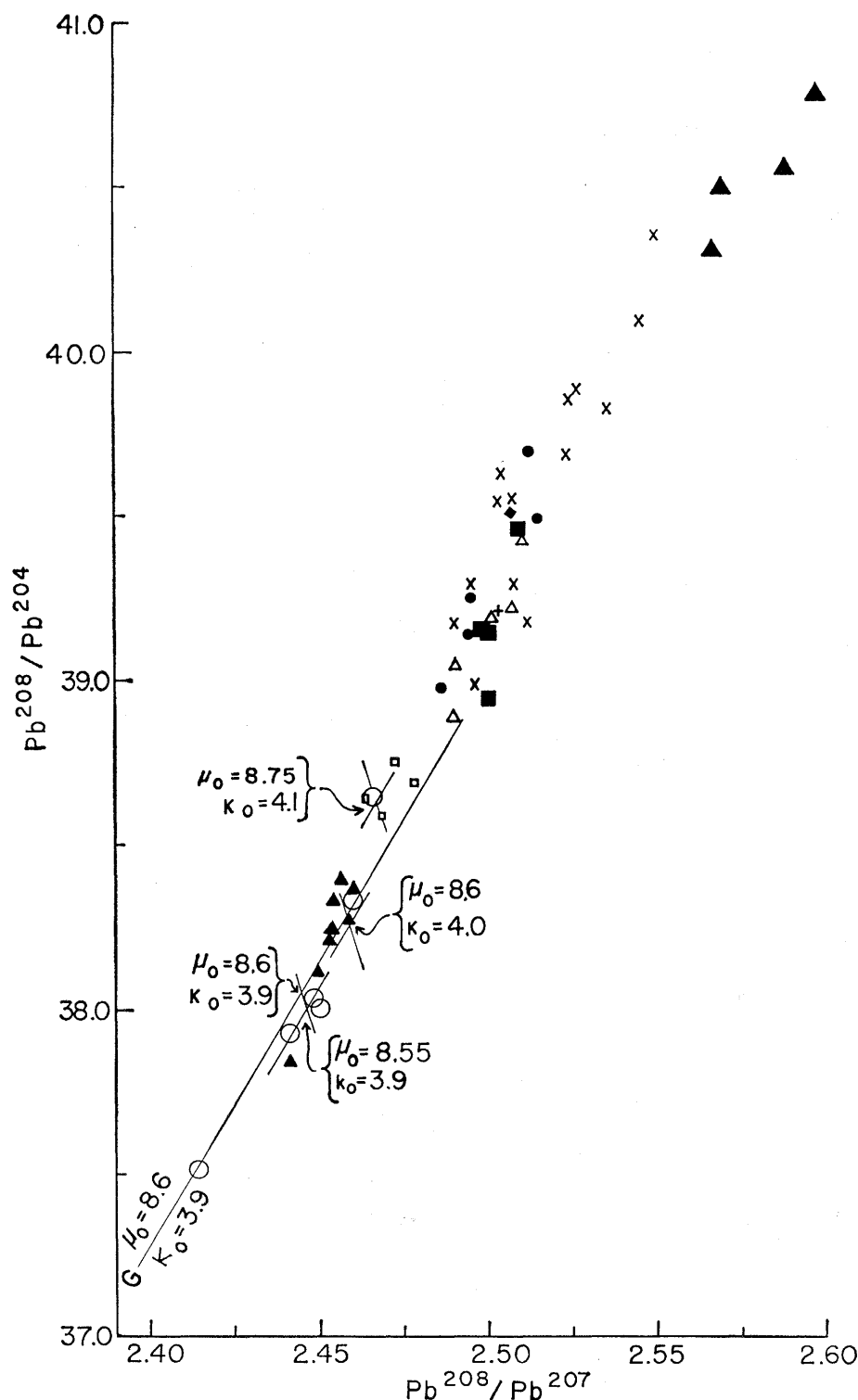


Fig. 4. Ratios of Pb^{208}/Pb^{207} versus Pb^{208}/Pb^{204} . The line labeled by G is a primary growth curve of $\mu_0 = 8.6$ and $\kappa_0 = 3.9$. Leads of the oceanic tholeiites and Hawaii are close to $\mu_0 = 8.6$ and $\kappa_0 = 3.9$ of a closed system. Lead of Japan and Gough Island have higher μ_0 's and κ_0 's as stated in the text. The symbols are as for Fig. 3.

Calculation of μ_1 for alkali suites of Easter and Guadalupe Islands by the two-step model suggests that the magma source region was differentiated about 0.3 to 0.5 billion years ago from the source of the Pacific tholeiites; however, the calculated κ_1 is much smaller than the observed. In this model the observed κ cannot be of a magma source because κ must have had a smaller value, similar to that of the tholeiite, in the past in order to match the observed $\text{Pb}^{208}/\text{Pb}^{204}$ ratios in the alkali series. This relation holds true even if κ_0 is as low as 3.9.

The $\text{Pb}^{206}/\text{Pb}^{204}$ versus observed $\text{U}^{238}/\text{Pb}^{204}$ (μ) and $\text{Pb}^{208}/\text{Pb}^{204}$ versus the observed $\text{Th}^{232}/\text{Pb}^{204}$ ($\mu \cdot k$) are plotted in Figs. 5 and 6, respectively. The significance of these plots is similar to that of isochron diagrams of $\text{Rb}^{87}/\text{Sr}^{86}$ versus $\text{Sr}^{87}/\text{Sr}^{86}$ which are commonly used in rubidium-strontium geochronology studies. However, for the interpretation of the present figures, it involves an important assumption. That is, $\text{U}^{238}/\text{Pb}^{204}$ or $\text{Th}^{232}/\text{Pb}^{204}$ ratios should be the same for basalt and its source if ages are to be determined.

Applying the two-step model, if the observed $\text{U}^{238}/\text{Pb}^{204}$ ratio is representative for the magma source, and if these rocks of the magma source were differentiated from a common parent material whose $\text{Pb}^{206}/\text{Pb}^{204}$ ratio was $\text{Pb}^{206}/\text{Pb}^{204}_0$ at the same time t years ago, then the present observed $\text{Pb}^{206}/\text{Pb}^{204}_p$ is:

$$\left(\frac{\text{Pb}^{206}}{\text{Pb}^{204}} \right)_p - \left(\frac{\text{Pb}^{206}}{\text{Pb}^{204}} \right)_0 = \frac{\text{U}^{238}}{\text{Pb}^{204}} (e^{\lambda_{238}t} - 1)$$

which is the equation of a straight line where t is calculated from the slope of the line, $\tan \theta$. Similarly, t can be calculated from the plots of $\text{Pb}^{208}/\text{Pb}^{204}$ versus $\text{Th}^{232}/\text{Pb}^{204}$ diagram.

From Fig. 5, it appears that the magma sources for oceanic tholeiite were differentiated from a parental upper mantle material at about 1.2 billion years ago, which is in good agreement with the calculated model age (23). The agreement of the two different calculations for an age of differentiation for uranium-derived lead suggests that the oceanic tholeiite was seemingly formed by nearly complete melting of the differentiated upper mantle, or that lead and uranium migrated together during partial melting and thorium remained behind. The latter case is preferred because (i) no isochron age for the tholeiite is obtained from Fig. 6 in

which $\text{Pb}^{208}/\text{Pb}^{204}$ is plotted against observed $\text{Th}^{232}/\text{Pb}^{204}$, and (ii) the points are located lower than the line of a model isochron age of 1190 million years in which μ and κ are unchanged from a primary μ_0 (8.6) and κ_0 (3.9).

In contrast to the tholeiites, the convergence line for the alkali suite on Easter Island in Fig. 5 is vertical to the abscissa and no age can be obtained. This means that: (i) If the alkali suite on Easter Island differentiated from tholeiite, the entire process, including the generation of the rhyolite (Ourito obsidian), took less than 20 million years (this time is within the limits of error of $\text{Pb}^{206}/\text{Pb}^{204}$ measurement), because if the differentiation took more than 20 million years, the measured $\text{Pb}^{206}/\text{Pb}^{204}$ ratio should vary more than 0.1 within the suite. (ii) There has been

little or no contamination or assimilation of oceanic crust or other sedimentary rocks by the magmas during differentiation in this suite; if there were crustal contamination, the convergence line should be either inclined, or declined, because of additions of more radiogenic or less radiogenic lead. The former case applies to the suite on Guadalupe Island. The variations of $\text{Pb}^{206}/\text{Pb}^{204}$ ratios in the suite beyond the experimental error suggest that assimilation of more radiogenic lead or, alternatively, longer time (about 50 million years) is involved for the differentiation of the alkali suite on Guadalupe Island.

Engel *et al.* (5) discovered the progressive changes in chemical composition between the tholeiite and the alkali series of volcanic rocks on the higher

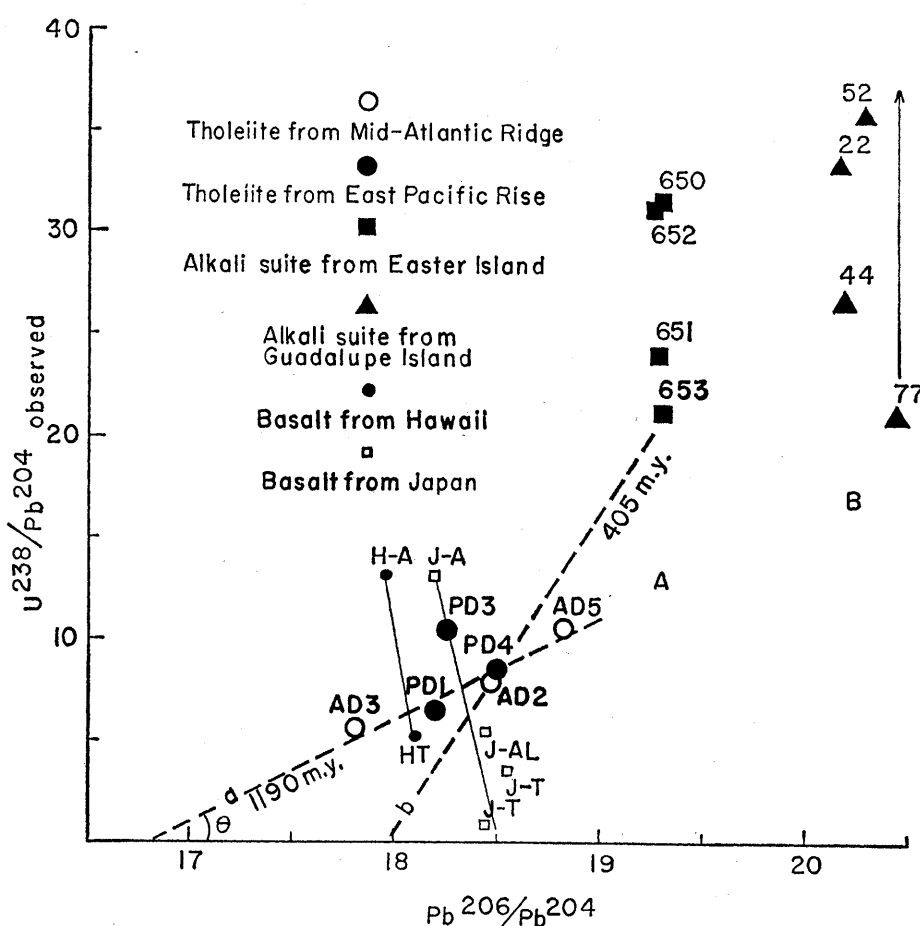


Fig. 5. Ratios of $\text{Pb}^{206}/\text{Pb}^{204}$ versus observed $\text{U}^{238}/\text{Pb}^{204}$. The convergence lines for the oceanic tholeiites and alkali suite from Easter Island show 1200 million years and 0 million years. However, PD3 does not fit on the isochron. It may be due to the sample heterogeneity (6). For the significance of the isochron age, refer to (23). Tholeiite near or from Easter and Guadalupe Islands, if any, should be at A and B respectively, if it is a parent of the alkali suite. If the alkali suite of Easter Island differentiated from a Pacific tholeiite (PD4), the differentiation age of Easter Island lead is about 400 million years. The convergence lines of Hawaiian rocks (HT-H-A) and Japanese rocks (J-T-J-AL-J-A) show negative ages. It may suggest that the sources are different for alkali basalt and tholeiite. The uranium content of Gu 77 is "irregularly" low. It should be higher than observed value in the source region as indicated by an arrow [refer to (15), Figs. 1 and 6].

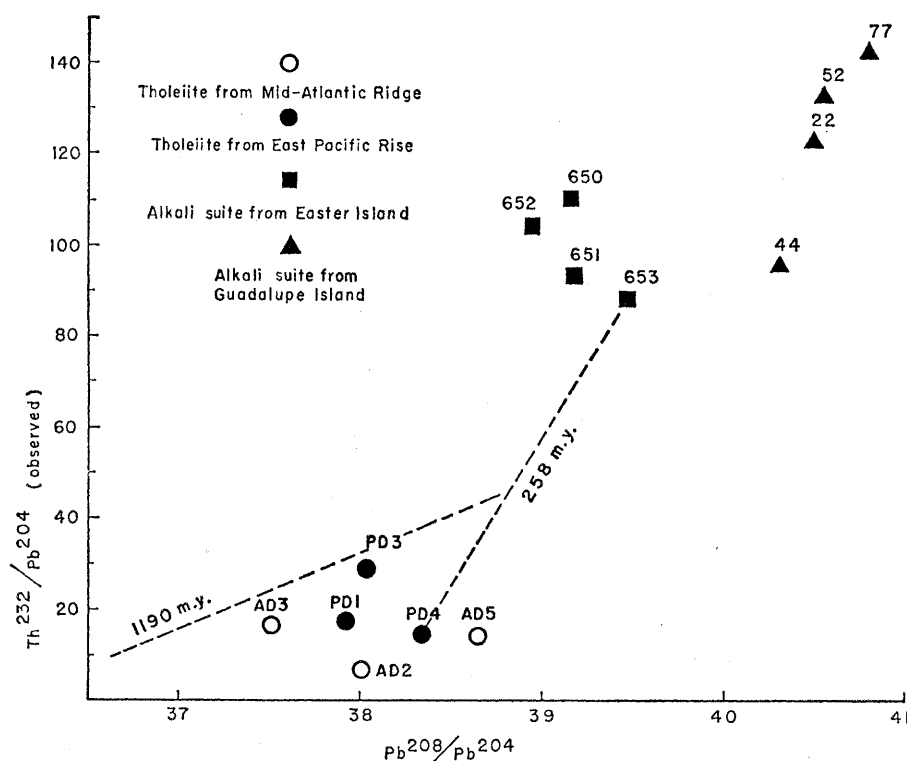


Fig. 6. Observed $\text{Th}^{232}/\text{Pb}^{204}$ versus $\text{Pb}^{208}/\text{Pb}^{204}$. The meaning of this figure is the same as Fig. 5; however, no convergence age can be obtained for the oceanic tholeiite. The isochron age for tholeiite to alkali basalt is about 260 million years, if the alkali basalts differentiated from a Pacific tholeiite.

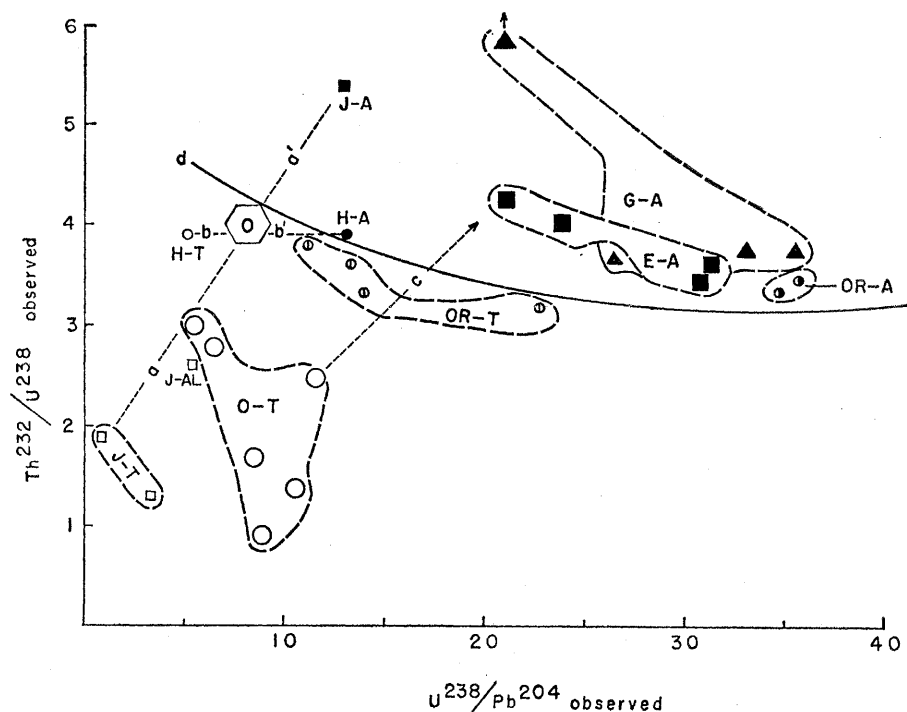


Fig. 7. Observed μ ($\text{U}^{238}/\text{Pb}^{204}$) versus observed κ ($\text{Th}^{232}/\text{U}^{238}$). O-T, E-A, and G-A are for oceanic tholeiites and alkali suites from Easter and Guadalupe Islands. J-T, J-AL, and J-A indicate tholeiite, high-alumina basalt and alkali basalt from Japan; H-T and H-A are for tholeiite and alkali basalt from Hawaii; OR-T and OR-A indicate tholeiites and alkali basalts from Oregon coast. Presumably, the source region of basalts of Japan and Hawaii differentiated from an original mantle material (O) along $a-a'$ and $b-b'$ lines; however, oceanic tholeiite was differentiated from O and evolved to Easter Island alkali suite along line c. All of the alkali basalts and their derivatives are above line d. The line was drawn arbitrarily.

volcanos of the oceans. They concluded that (i) the oceanic tholeiites are the only parental basaltic magma erupted from the mantle, and (ii) the alkali-rich basalts are derived from the tholeiites through gravitative differentiation in and below the higher volcanos in the oceans.

For the theory of "fractional" differentiation of alkali basalt from tholeiite, one of the main points of objection by those who consider different sources for tholeiite and alkali basalt has been the greater enrichment of alkali in alkali basalt compared with tholeiite if the parent is peridotite. Macdonald and Katsura (2) suggested that alkali enrichment can be accomplished by crystallization differentiation that is controlled by separation of pyroxene and also possibly by mechanisms such as volatile transfer (transfer of volatile compounds, carrying alkalies in solution) and thermodiffusion.

On the other hand, Yoder and Tilley (4) reported that there is a thermal barrier between tholeiite and alkali basalt for relatively high pressures, and that these two major basalt series may have evolved from a single parent at different depths. Additional minor variations in the magma may result from settling of crystals, oxidation or reduction, gas fluxing, contamination, and other processes.

The concept of a barrier between tholeiite and alkali basalt has been erased by recent even higher pressure phase-equilibrium studies (24), and the main mechanism to produce alkali basalt from tholeiite, if such occurs, could be a crystallization differentiation.

Several aspects of the isotopic and concentration data in this study suggest that alkali basalt may be a differentiate of parent tholeiite:

1) There is a progressive increase in contents of lead, uranium, and thorium from oceanic tholeiite to alkali basalt and other alkalic derivatives as would be expected in differentiation.

2) The source of the alkali series must have passed through a low κ stage similar to tholeiite as deduced from Fig. 2 where samples from Easter and Guadalupe Islands are below the growth curves involving $\text{Pb}^{208}/\text{Pb}^{204}$ ratios and as calculated in Table 2 for sample PV651.

3) The mechanism of transfer of volatile compounds probably is a subordinate process because enrichment in alkali and lead is accompanied by an

increase in refractory elements such as thorium. In fact, the observed $\text{Th}^{232}/\text{Pb}^{204}$ in the alkali series is generally too great to account for the observed $\text{Pb}^{208}/\text{Pb}^{204}$.

4) If significant assimilation of oceanic crust were important to the formation of the alkali series, variation of lead isotopic composition might be expected within the series such as was found for the series from the Guadalupe Islands. Since there was no isotopic effect in the alkali series of Easter Island, assimilation of oceanic crust was probably not important in producing that alkali series.

5) It is unlikely that tholeiite could be a differentiate from alkali basalt because the calculated age for differentiation of the source of tholeiite (about 1200 million years) is greater than a calculated age of differentiation of a Pacific tholeiite near Easter Island from the alkali series of Easter Island (< 400 million years).

6) If the alkali series is a differentiate from tholeiite, no isotopic effect should be observed. Probable tholeiite parents for Easter and Guadalupe alkali series are not available for analysis. Oceanic tholeiites range in lead isotopic composition, however, and only twice the observed range is necessary to yield tholeiites with lead isotopic composition similar to the most radiogenic alkali series rocks (Guadalupe). There may be tholeiites whose $\text{Pb}^{206}/\text{Pb}^{204}$ ratios are radiogenic (points A and B in Fig. 5). Even though good tholeiites of the appropriate isotopic composition are not known, a tholeiite with alkali basalt affinities from Easter Island is similar in lead isotopic composition to the alkali series rocks from that island. If tholeiites and alkali basalts are generated at different depths, a systematic difference in lead isotopic composition between the two rock types might be expected. Existing data do not completely rule out this alternative to differentiation.

The foregoing discussions of the genetic relation of tholeiite and alkali basalt may be limited to the oceanic tholeiite and the oceanic alkali basalt because, as discussed earlier, the source region of the oceanic tholeiite differentiated quite a long time ago and the upper mantle is not homogeneous. Therefore, isotopic variation is possible, especially in well-differentiated continental regions, by tapping out magma from different zones.

The relation between the observed μ

and κ in basalts is shown in Fig 7. The values of basalt from Hawaii and Japan (10) and the Oregon Coast (25) are also shown for comparison. From the many lead isotope studies, the original material in the mantle is thought to be $\mu_0 \approx 8.6$ and $\kappa_0 \approx 4$ (indicated by O in Fig. 7). The Japanese tholeiite and high alumina basalt have low observed μ 's and κ 's, but Japanese alkali basalt has high observed μ 's and κ 's. It has been stated (10) that Japanese alkali basalt must pass through a low μ and κ stage. The situation is similar for Easter Island and Guadalupe Island alkali suites, that is, they have high observed μ and κ , but their lead isotopic composition shows that they had a low κ in the past. Therefore, the following interpretation, rather than splitting into two sources (like Japanese tholeiite and alkali basalt) from an original upper mantle material, is more likely for oceanic basalts. That is, starting from O in Fig. 7, the μ and κ in the source region of oceanic basalts were once low like oceanic tholeiite, then the μ and κ were increased by further differentiation.

The alternative mechanism of generation of the two basalt series from different depths could result in a variety in the isotopic composition of lead in young basalts because of different kinds of partial melting under different conditions of pressure and temperature. Laboratory experiments show that lead is very mobile and can easily be extracted from the basalt by heating. Uranium also may be movable in magma if it makes a volatile compound like uranium halide. Thorium, however, is more refractory than lead and uranium. In the previous section it was found that the calculated μ of oceanic basalt matches well the observed μ (Table 2), which suggests that lead and uranium were volatilized from the bulk of the source material and transferred to the melted portion, but thorium tended to remain in the unmelted portion so that the agreement between the calculated κ and the observed κ is poor. There may be variety in isotopic composition of lead and Th/U ratio by partial melting of different mineral components of a single mantle peridotite; however, I prefer the heterogeneous upper mantle model because the mantle is no longer homogeneous after one partial melting event (10).

M. TATSUMOTO

U.S. Geological Survey,
Denver, Colorado

References and Notes

1. H. A. Powers, *Amer. J. Sci. Ser. 5* **30**, 57 (1935); *Geochim. Cosmochim. Acta* **7**, 994 (1955).
2. G. A. Macdonald, *Bull. Geol. Soc. Amer.* **59**, 1009 (1949); — and T. Katsura, *Pacific Sci.* **15**, 358 (1961).
3. H. Kuno, *Bull. Volcanol.* **20**, 37 (1959); I. Kushiro and H. Kuno, *J. Petrol.* **4**, 75 (1963).
4. H. S. Yoder, Jr., and C. E. Tilley, *J. Petrol.* **3**, 342 (1962).
5. A. E. J. Engel and C. G. Engel, *Science* **144**, 1330 (1964); **146**, 477 (1964); C. G. Engel and R. L. Fisher, *ibid.* **150**, 605 (1965); A. E. J. Engel, C. G. Engel, R. G. Havens, *Bull. Geol. Soc. Amer.* **76**, 719 (1965).
6. M. Tatsumoto, C. E. Hedge, A. E. J. Engel, *Science* **150**, 886 (1965).
7. R. D. Russell and R. M. Farquhar, *Geochim. Cosmochim. Acta* **19**, 41 (1960).
8. T. J. Chow and C. C. Patterson, *ibid.* **26**, 263 (1962).
9. C. C. Patterson and M. Tatsumoto, *ibid.* **28**, 1 (1964).
10. M. Tatsumoto, *J. Geophys. Res.* **71**, 1721 (1966).
11. P. W. Gast, G. R. Tilton, C. E. Hedge, *Science* **145**, 1181 (1964).
12. A. Masuda, *Geochim. Cosmochim. Acta* **28**, 291 (1964); C. C. Patterson, *Recent Researches in the Fields of Hydrosphere, Atmosphere, and Nuclear Geochemistry* (Maruzen, Tokyo, 1964), p. 257.
13. A. Masuda, *J. Earth Sci. Nagoya Univ.* **10**, 117 (1962).
14. B. R. Doe, *J. Petrol.*, in press.
15. One exception is observed; the uranium content in the sample of labradorite-andesine alkali basalt from Guadalupe Island deviates from the relation curve. The uranium content listed in Table 1 is an average of five replicate analyses (1.05 ± 0.02 ppm) using two different decomposition techniques; $\text{HF} + \text{HClO}_4$ and Na_2CO_3 fusion.
16. E. F. Osborn, *Amer. Mineralogist* **47**, 211 (1962).
17. C. C. Patterson and B. Duffield, *Geochim. Cosmochim. Acta* **27**, 1180 (1963).
18. For detailed explanation of the definition of the primary isochron and the primary growth curve, refer, for example, to A. Holmes, *Nature* **157**, 680 (1946); F. Houtermans, *Naturwiss.* **33**, 185 (1946); C. C. Patterson, *Geochim. Cosmochim. Acta* **10**, 230 (1956); R. D. Russell and R. M. Farquhar, *Lead Isotopes in Geology* (Interscience, New York, 1960).
19. R. G. Ostic, R. D. Russell, P. H. Reynolds, *Nature* **199**, 1150 (1963); R. G. Ostic, "Isotopic investigation of conformable lead deposits" thesis, University of British Columbia, 1963; B. R. Doe, *J. Geophys. Res.* **67**, 2895 (1962), the μ_0 value is readjusted to the Re filament value.
20. V. R. Murthy and C. C. Patterson, *J. Geophys. Res.* **67**, 1161 (1962).
21. G. R. Tilton and R. H. Steiger, *Science* **150**, 1805 (1965).
22. G. J. Wasserburg, D. S. Burnett, C. Frondel, *ibid.*, p. 1814.
23. The convergence line which was drawn to all oceanic tholeiites might be inappropriate because the differentiations of the source regions of the oceanic tholeiites under the Mid-Atlantic ridge and the East Pacific rise conceivably occurred at different times. The model age calculations from lead isotopes, as well as the Rb/Sr and $\text{Sr}^{87}/\text{Sr}^{86}$ relations (6), however, suggest similar differential age for the source regions under both the ridge and rise.
24. *Carnegie Inst. Year Book* **63** (1965).
25. M. Tatsumoto and P. D. Snively, Jr., in preparation (abstract; M. Tatsumoto, *Trans. Amer. Geophys. Union* **46**, 165 (1965)).
26. I am grateful for the many valuable comments and the samples supplied by A. E. J. Engel, University of California. I wish to thank my many colleagues in the U.S. Geological Survey, especially R. G. Coleman, B. R. Doe, W. B. Hamilton, and C. E. Hedge for their helpful discussions. E. E. Wilson developed a special instrument for the furnaces to increase temperature automatically. R. J. Knight gave valuable help in chemistry on the samples from Guadalupe Island and M. Delevaux maintained mass spectrometers.
27. Publication authorized by the director, U.S. Geological Survey.

27 June 1966