

Reports

Isotopic Composition of Strontium in Volcanic Rocks from Oahu

Abstract. Analysis of several well-documented specimens from each of the three volcanic series on Oahu gives the following mean ratios of Sr^{87} to Sr^{86} : the Waianae series, 0.7030 ± 0.00010 ($\bar{\sigma}$); the Koolau series, 0.70385 ± 0.00009 ($\bar{\sigma}$); and the Honolulu series, 0.7029 ± 0.00006 ($\bar{\sigma}$). The mean ratio of Sr^{87} to Sr^{86} of the Koolau series specimens is significantly higher than the means of the other two series. With one exception, significant differences in Sr^{87}/Sr^{86} within a series were not found, even though some large compositional differences existed.

Several authors have recently reviewed the application of variations in the abundance of Sr^{87} to problems of geochemistry and igneous petrology (1). Strontium-87 is the daughter of Rb^{87} , and the present Sr^{87}/Sr^{86} ratio in an igneous rock reflects (i) the initial, inherited Sr^{87}/Sr^{86} ratio of its parent material, (ii) radiogenic Sr^{87} added to the parent material, and (iii) radiogenic Sr^{87} added to the rock itself after formation from its parent material. The Sr^{87}/Sr^{86} ratios of volcanic rocks thus provide information concerning the Rb/Sr ratios of their source materials. In addition, since the isotopes of strontium apparently do not undergo appreciable natural fractionation, several rocks derived from the same parent magma and uncontaminated with foreign material with a different Sr^{87} abundance should have had identical Sr^{87}/Sr^{86} ratios at the time they crystallized. Rocks with significantly different initial Sr^{87}/Sr^{86} ratios either are not comagmatic, or are comagmatic and contaminated.

Much of the previous work in the application of Sr^{87} as a natural tracer has involved oceanic volcanic rocks, primarily because in oceanic environments the possibility of contamination of magmas with crustal rocks enriched in radiogenic Sr^{87} is greatly reduced, although perhaps not eliminated. The Sr^{87}/Sr^{86} ratios found in oceanic volcanic rocks have been very uniform, usually varying between 0.702 and 0.705. Gast *et al.* (2) found both intra-island (within a single island) and inter-island (between a group of islands)

variations in the Sr^{87}/Sr^{86} ratios of rocks from Ascension and Gough islands in the Atlantic. The intra-island variations in both cases were correlated with rock composition, an effect that has not been observed for any of the other individual islands that have been studied.

A number of Sr isotope analyses of Hawaiian volcanic rocks have been reported (1, 3–5). Lessing and Catanzaro (3) attributed differences in the Sr^{87}/Sr^{86} ratios of Hawaiian basalts and trachytes to assimilation of sediment by Hawaiian lavas. Powell *et al.* (5) found that four specimens with low SiO_2 contents from the Honolulu series on Oahu had significantly lower Sr^{87}/Sr^{86} ratios than four specimens with higher SiO_2 contents from Hawaii. They were unable to determine whether this difference was related to the different chemical compositions or to the different geographic locations of the two groups of samples.

Our study was undertaken to extend the previous work on Hawaiian rocks, to try to remove the ambiguity resulting from the work of Powell *et al.* (5), and to try to gain information about the genetic relations between the melilitite- and nepheline-bearing lavas and the abundant tholeiitic basalts on Oahu.

The general geology of Oahu has been described by Stearns and Vaksik, and Stearns (6). Three different volcanic series are clearly distinguishable. The oldest, the Waianae series, is composed of tholeiitic basalts that range in age from 3.5 to 2.75 million years (7). The Koolau series also is tholeiitic;

specimens dated by McDougall (7) range in age from 2.55 to 2.15 million years. The strongly undersaturated lavas of the Honolulu series were erupted locally in the southeastern part of the Koolau Range during late Pleistocene and Recent time.

We have analyzed a number of well-documented specimens from each of the three volcanic series on Oahu. Twenty-one of the 25 volcanic rock specimens studied had been chemically analyzed and in some cases examined petrographically in earlier work. The other four specimens came from outcrops for which detailed descriptions and chemical analyses are available. All volcanic rock specimens were analyzed for Sr^{87}/Sr^{86} at least twice; in general, these replicate runs were performed on the same strontium separate. The specimens were not analyzed in groups according to volcanic series. Analyses of specimens from the Koolau series were interspersed with those of specimens from the other series. Age corrections to the measured Sr^{87}/Sr^{86} ratios were not required.

All isotope ratio measurements were made within a period of 6 weeks on a mass spectrometer with a 30-cm radius and a 90° sector. A double-filament source, rapid switching of the magnet current, an expanded scale recorder, and a Faraday cup collector were used. No rubidium analyses had been made on the instrument used, and no corrections were made to our raw Sr^{87}/Sr^{86} ratios other than to normalize them to $Sr^{86}/Sr^{88} = 0.1194$. Pressures in both the source and analyzer regions were usually less than 5×10^{-8} mm-Hg. Eight analyses of the Eimer and Amend interlaboratory strontium standard gave $Sr^{87}/Sr^{86} = 0.7078 \pm 0.00035$ (σ). The precision of a single analysis, calculated from 23 duplicate analyses and one triplicate analysis, is ± 0.0003 (σ). The precision of the means of the duplicate analyses is therefore approximately ± 0.0002 ($\bar{\sigma} = \sigma/N^{1/2}$), or about ± 0.0004 at the 95 percent confidence level.

It is clear from Fig. 1 that significant differences exist between the Sr^{87}/Sr^{86} ratios of the Waianae and Honolulu series and those of the Koolau series. The mean Sr^{87}/Sr^{86} ratios and standard deviations of the mean for the three are: the Waianae series, 0.7030 ± 0.00010 ; the Koolau series, 0.70385 ± 0.00009 ; and the Honolulu series, 0.7029 ± 0.00006 . Standard *t*-tests can be applied to test the significance of the differences between the means of the

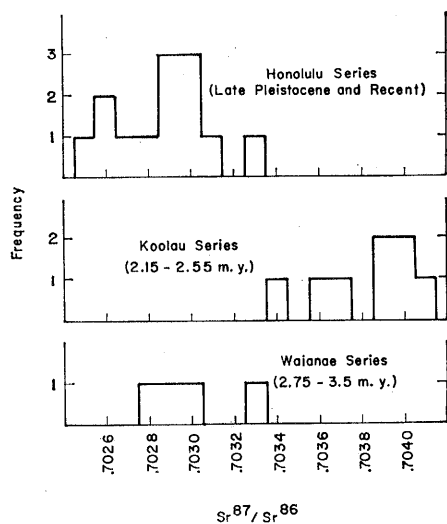


Fig. 1. Ratios of Sr^{87} to Sr^{86} of rocks from the three volcanic series on Oahu. Ages (million years) from McDougall (7).

three pairs of series. These tests give the following results: Honolulu compared to Koolau series, $t = 10.3$, degrees of freedom (df) = 19, difference significant at confidence levels exceeding 99 percent; Waianae compared to Koolau, $t = 6.38$, $df = 10$, difference significant at confidence levels exceeding 99 percent; and Honolulu compared to Waianae, $t = 1.25$, $df = 15$, difference not significant at confidence level of 80 percent. Specimen 9991 was analyzed six times with two dissolutions, and gave $\text{Sr}^{87}/\text{Sr}^{86} = 0.7034 \pm 0.0001$ ($\bar{\sigma}$). From the results of the individual $\text{Sr}^{87}/\text{Sr}^{86}$ analyses, the difference between the mean $\text{Sr}^{87}/\text{Sr}^{86}$ ratio of this specimen and the mean for the other Koolau series specimens is significant at confidence levels greater than 99 percent ($t = 3.1$, $df = 22$). The results for sample 9991 were nevertheless included in calculating the mean for the Koolau series in order to avoid a possible bias in the results. The reason for the lower $\text{Sr}^{87}/\text{Sr}^{86}$ ratio of this specimen is unknown.

These results show that intra-island variations in $\text{Sr}^{87}/\text{Sr}^{86}$ ratio, as well as the inter-island variations found by Powell *et al.* (5), occur in the Hawaiian Islands. Furthermore, on Oahu the isotopic variations occur between different volcanic series. With the exception already noted, significant differences in $\text{Sr}^{87}/\text{Sr}^{86}$ within a series were not found. In both the Koolau and Waianae series, alkalic basalts have the same Sr^{87} abundance as tholeiitic basalts. Within the Honolulu series, a basanite with 45 percent silica has the same Sr^{87}

abundance as melilite-nepheline basalts with 36 to 37 percent silica. Within the Waianae series, the Mauna Kuwale rhyodacite, with 67 percent silica, has the same low Sr^{87} abundance as the other members, including C-47, an alkalic basalt with 46 percent silica. These results are consistent with the belief that the rocks within each series are comagmatic.

Our data show no correlation between Sr^{87} abundance, time of eruption, and general chemical composition of a series. The oldest and youngest series, the Waianae and Honolulu, are tholeiitic and strongly undersaturated, respectively, and they have identical and relatively low $\text{Sr}^{87}/\text{Sr}^{86}$ ratios. The Koolau series, of intermediate age and of tholeiitic composition, has a distinctly higher Sr^{87} abundance than the other two.

These results suggest that caution must be used in interpreting intra-island variations in Sr^{87} abundance. Unless adequate stratigraphic control is available, and unless a number of specimens are analyzed, it may not be possible to determine whether such isotopic variations reflect the presence of more than one volcanic series, or whether they are related to variations in chemical composition within a single stratigraphic unit.

The possible causes of intra-island variations in the abundance of radiogenic isotopes have been discussed by Gast *et al.* (2), Powell *et al.* (5), and Tatsumoto (8). Lessing and Catanzaro (3) found an inverse relation between $\text{Sr}^{87}/\text{Sr}^{86}$ and K/Rb for Hawaiian rocks, and concluded that Hawaiian lavas are contaminated with marine argillaceous sediments. Taubeneck (9) states that the geochemical evidence indicates that K/Rb variations correlate with degree of differentiation and need not be caused by contamination. Gast *et al.* (2) and Tatsumoto (8) presented evidence that strongly suggests that intra-island variations in radiogenic isotope abundances are not caused by contamination.

On Oahu, contrary to the situation on the other islands for which strontium isotope data are available, the latest rocks are lower in SiO_2 content, and as low or lower in Sr^{87} abundance than the predominant basaltic types. Only contamination with a low-silica or nonsilicate material could produce the first effect. In a mid-oceanic environment, the only materials that fit these requirements are coral and *Glo-*

bigerina ooze. Daly (10) proposed that the alkalic and melilite-bearing rocks of the Hawaiian Islands were formed by "... reaction of the primary basaltic melt with sedimentary material rich in calcium carbonate," and he suggested that this sedimentary material was *Globigerina* ooze. Winchell (11) showed that the geologic evidence is strongly against this hypothesis for the Honolulu series.

In Table 1 we report $\text{Sr}^{87}/\text{Sr}^{86} = 0.7088$ for a coral and shell rock from Diamond Head. Ocean water has a nearly constant $\text{Sr}^{87}/\text{Sr}^{86}$ ratio of about 0.709 (12). The work of Faure *et al.* (13) with freshwater shells suggests that tests of *Globigerina* formed in sea water would likewise have a $\text{Sr}^{87}/\text{Sr}^{86}$ ratio of about 0.709. Addition of any of the low-silica materials available in mid-ocean, including ocean water, to a basalt magma would raise its $\text{Sr}^{87}/\text{Sr}^{86}$ ratio, not lower it. The Sr isotope data therefore indicate that the Honolulu series was not derived from the Waianae or Koolau series parent magma by carbonate syntexis. It is still possible that some other parent magma with a lower Sr^{87} abundance, such as the oceanic tholeiites described by Tatsumoto *et al.* (14), produced the Honolulu series by assimilation of carbonate material. Tatsumoto *et al.* (14) give 115 parts per million and 0.702 as the average Sr content and $\text{Sr}^{87}/\text{Sr}^{86}$ ratio of oceanic tholeiites. Turekian and Wedepohl (15) give 2000 parts per million as the average Sr content of deep-sea carbonates; the $\text{Sr}^{87}/\text{Sr}^{86}$ ratio of such carbonates can be assumed to be approximately 0.709. It would appear that addition of an amount of carbonate sediment to an oceanic tholeiite magma sufficient to cause the formation of significant amounts of strongly undersaturated magma would raise the $\text{Sr}^{87}/\text{Sr}^{86}$ ratio of the derivative magma to a value higher than that found for the Honolulu series. For example, a mixture of 90 percent oceanic tholeiite and 10 percent carbonate sediment would have a $\text{Sr}^{87}/\text{Sr}^{86}$ ratio of about 0.707.

The Sr isotope data therefore quantitatively confirm the conclusions of Winchell (11) that the strongly undersaturated rocks of the Honolulu series have not been derived from the Koolau series parent magma by carbonate syntexis or by igneous differentiation. The isotopic data are consistent with the hypothesis that the Waianae and Honolulu series are comagmatic, but on

geological grounds this seems to be very unlikely. If the melilite- and nepheline-bearing rocks on Oahu are derived independently of alkalic and tholeiitic basalt magma, there may be good reason to believe that the melilite basalts of kimberlite provinces, and the melilitic rocks of the carbonatite suite, have likewise arisen independently of normal basalt magma.

The second possible explanation of intra-island variations in the abundance of radiogenic isotopes (1, 2, 5, 8) is that they are due to chemical, and resulting isotopic, heterogeneities in the upper-mantle parent materials of the rocks. Yoder and Tilley (16) and Kushiro and Kuno (17) have suggested that the more alkalic magmas are derived from greater depths than tholeiitic magmas. Powell *et al.* (5) postulated an overall decrease with depth of the Rb/Sr and Sr^{87}/Sr^{86} ratios of the upper mantle. If both hypotheses are correct, alkalic magmas would consistently show lower Sr^{87} abundances than tholeiitic ones. Our results show no such simple correlation between chemical composition and Sr^{87} abundance.

It is possible that isotopic heterogeneities on the scale of individual mineral grains occur in the upper mantle and that under different conditions different proportions of these minerals melt, giving rise to magmas with different Sr^{87}/Sr^{86} ratios. This effect might or might not be combined with a net decrease of Rb/Sr with depth. Our results are compatible with this general hypothesis, but again indicate that the actual magma-producing mechanisms and their time-depth relationships are complicated.

A third hypothesis to explain the observed intra-island variations in Sr^{87}/Sr^{86} is that Oahu and the other islands of the Hawaiian chain have drifted across regions of the mantle having different Rb/Sr and Sr^{87}/Sr^{86} ratios. That such lateral inhomogeneities in radiogenic isotope abundances do occur in the ocean basins is shown by the work of Gast *et al.* (2) and Tatsumoto (8). According to the K-Ar age determinations of McDougall (7), the Waianae series was extruded over a period of about 0.75 million years, and the Koolau series over a period of about 0.40 million years. The difference in age between the youngest dated Waianae specimen and the oldest dated Koolau specimen is about 0.20 million years. The drift hypothesis requires that for about 0.75 million years the magma

sources of the Waianae series maintained an approximately constant Sr^{87} abundance, then within 0.20 million years a region with a different abundance of Sr^{87} was reached and the Koolau series extruded. This demands either relatively rapid changes in the drift velocity or relatively rapid lateral changes in the Rb/Sr and Sr^{87}/Sr^{86} ratios of the upper mantle, both of which are possible. Perhaps it is fair to state that intra-island variations in

radiogenic isotope abundances are consistent with the hypothesis of spreading sea floors and drifting islands, but that such variations might occur if the sea floors were static.

In summary, the intra-island variations in Sr^{87} abundance found for Oahu show that all the rocks there were not derived from a common parent magma, and that they were not formed by limestone syntaxis. These variations are believed to be caused by chemical,

Table 1. Isotopic composition of strontium in rocks from the three volcanic series on Oahu. With the exception of specimens B-975, R13, R16, R131, and TK-1501, chips of the original specimens described in the references were analyzed. Specimen TK-1501 was collected from the original locality by Macdonald. The sample numbers used in the references have been retained where possible. Specimen 9962 and all of those from the Koolau series have been renamed by us using the system of Macdonald and Katsura (19). Unless otherwise indicated, the Sr^{87}/Sr^{86} ratios listed are the means of duplicate analyses; their precision is about ± 0.0002 ($\bar{\sigma}$). The Rb/Sr ratios were measured by x-ray fluorescence; their precision is about 15 percent. Where no Rb/Sr ratios are given, Rb is less than 11 parts per million.

Sample No. and description	Location	$\frac{Sr^{87}}{Sr^{86}}$ *	$\frac{Sr^{86}}{Sr^{88}}$ (% by wt)	SiO ₂	$\frac{Rb}{Sr}$	Ref.
<i>Honolulu series</i>						
10399, Melilite-nepheline basalt	Kaau lava flow	0.7031	0.1194	36.7	0.0088	(20)
9960, Melilite-nepheline basalt	Kalihi flow	0.7029	0.1197	36.8	0.0058	(20)
10401, Melilite-nepheline basalt	Mokapu peninsula	0.7030	0.1198	37.2		(20)
88126:85-0, Melilite-nepheline basalt	Moilili quarry	0.70285	0.1197	36.3	0.023	(21)
B-975, Melilite-nepheline basalt	Moilili quarry	0.7026	0.1195		0.019	(22)
R16, Melilite-nepheline basalt	Moilili quarry	0.70295	0.1195		0.023	(23)
10400, Nepheline basalt	Training School flow	0.70305	0.1195	37.1	0.016	(20)
9961, Nepheline basalt	Lower Nuuanu flow	0.7025†	0.1195	38.6	0.021	(20)
R13, Nepheline basalt	Upper Nuuanu flow	0.7027	0.1195		0.025	(23)
9982, Nepheline basanite	Black Point	0.70305	0.1198	42.9	0.036	(20)
10402, Nepheline basanite	Kalama Crater	0.7033	0.1193	43.9	0.040	(20)
C-32, Basanitoid	Kaimuki Shield	0.7029	0.1190	43.1	0.033	(19)
9962, Basanite	Koko Head	0.7026	0.1196	45.1	0.021	(20)
<i>Koolau series</i>						
11320, Alkalic basalt	Boulder in Monanalu Valley	0.70395	0.1191	48.3		(24)
10396, Tholeiitic basalt	Waiahole Valley	0.7040	0.1192	48.7		(24)
9948, Tholeiitic olivine basalt	Makapuu Point	0.70405	0.1189	49.6		(24)
9991, Tholeiitic basalt	Waiakeakua branch of Manoa Stream	0.7034‡	0.1192	50.1		(24)
9980, Tholeiitic basalt	Oahu shore opposite Wananapaoa Islet	0.70365‡	0.1192	51.0	0.025	(24)
10404, Tholeiitic basalt (Kailua member)	N. of Kailua junction	0.7039	0.1191	51.2		(24)
10403, Tholeiitic basalt	S. of Makapuu Head	0.70415	0.1193	51.9		(24)
R131, Quartz dolerite	Palolo quarry	0.70375	0.1191	52.3		(25)
<i>Waianae series</i>						
C-16, Tholeiite	Lower member, Nanakuli Valley	0.7029	0.1193	49.5	0.027	(19)
C-47, Alkalic basalt	Middle member, Nanakuli Valley	0.70305	0.1193	45.6	0.019	(26)
C-170, Hawaiite	Upper member	0.7033	0.1193	46.4	0.025	(27)
TK-1501, Rhyodacite	Upper member (?), Mauna Kuwale	0.70285	0.1193	66.8	0.36	(19)
<i>Shell rock and standard</i>						
88126:1-0, Coral and shell rock	Sea level, south base of Diamond Head	0.7088§	0.1197			(21)
Strontium standard		0.70776	0.1192			

* Normalized to $Sr^{86}/Sr^{88} = 0.1194$. † Mean of three analyses. ‡ Mean of six analyses, two dissolutions. § Single analysis. || Eimer and Amend (lot 492327), mean of eight determinations.

and resulting isotopic, heterogeneities in the upper mantle source material of the rocks on Oahu; the detailed nature of such heterogeneities and of the magma-producing process remain unknown. As Gast (18) has pointed out, the isotopic data indicate that at least some chemical differences among spatially related volcanic rocks may reflect differences in the chemistry of their source materials, rather than the effects of contamination or igneous differentiation.

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Some Doubts about the Earth's Dust Cloud

Abstract. Considerable doubt is cast on the validity of past satellite measurements of micrometeoroid fluxes in which piezoelectric microphones have been used as detectors. Data have been obtained from satellite and laboratory experiments which show that the microphone crystals emit noise when subject to slowly varying temperatures. The rate of noise is consistent with past flight data which have previously been interpreted on the basis of micrometeoroid impacts. These measurements have given rise to the theory that the earth is surrounded by a cloud of dust, although no satisfactory mechanism has yet been found to explain this apparent phenomenon. On the basis of the results reported here, it now appears that whether or not a concentration of dust exists in the vicinity of the earth, the data from satellite microphone measurements should not be used to support such a hypothesis.

Over the last 7 years there has been a considerable volume of literature concerning the apparent existence of a concentration of dust in the vicinity of the earth. Evidence for this concentration consists primarily of the direct measurements reported by Dubin and McCracken (1) and Alexander *et al.* (2). Whipple (3) and Singer (4) have examined various possible sources of this dust, but no satisfactory mechanism has yet been found to account for the apparent increase in the flux of particles of masses between 10^{-7} and 10^{-13} g over fluxes deduced from photometric measurements of zodiacal

light. Moroz (5) has presented arguments to show that debris splashed from the moon by meteoric impact cannot be a sufficient source of these dust particles. Many attempts have been made to explain the enhancement by the gravitational attraction of the earth. Dole (6), and also Hale and Wright (7), are among those who have studied this, but as Southworth (8) has pointed out, gravitational attraction can only increase the flux near the earth if the particles in heliocentric orbits have very small velocities relative to the earth. This implies an orbital distribution for these particles which is

not realistic. The data of Hawkins (9) from ratio meteor studies show no evidence for such a confined distribution of orbits.

The measurements themselves seem to rule out the possibility that the dust particles are in long-term closed orbits around the earth. Rapid temporal variations of particle flux have been observed on nearly every satellite and these variations could not possibly exist if these particles were in geocentric orbits with periods of many days. The results of direct measurements of dust particles near the earth are difficult to explain theoretically, and this has prompted Singer (10) to announce his disbelief in the existence of the earth's dust cloud. Experimental evidence has now accumulated to support this view by throwing grave doubts on the validity of the direct measurements. The data presented below suggest that the microphone measurements consist largely, if not completely, of noise generated by the experiments themselves under changing temperature conditions. Thus the prime evidence for the existence of the earth's dust cloud is negated, and there is no longer any need for a theoretical explanation of such a large apparent enhancement of particle flux.

Direct evidence of the noise generated by microphone experiments has been obtained both in interplanetary space and in the laboratory. The former evidence was obtained from the flight of the OGO II interplanetary dust particle experiment. The basic sensor for this experiment has been previously described in connection with an earlier experiment on the OGO I satellite (11), and only a brief description need be given here. A micrometeoroid is detected through its impact on a thin film capacitor plate after it has passed through two very thin (1500-Å) films. A lead zirconate microphone crystal is bonded to the back of each glass capacitor plate to provide a measure of the impulse imparted to the plate by the impact of the particle. Figure 1 shows the basic sensor.

One hundred hours of data from the OGO II experiment have been analyzed to date. Two major conclusions to be drawn from these data are: first, there have been no detectable signals from the sensors that could have been caused by micrometeoroid impact, and second, the microphone systems have been emitting noise. It is this