tistical miracle, we simply do not know enough about the physical, chemical, and biological evolution of planets to assess with any degree of accuracy the sets of circumstances under which intelligent life forms may or may not emerge. If planetary systems are indeed extremely abundant one might conclude with equal conviction that man is not alone-that his equivalents may occupy hundreds or even thousands of bodies within our galaxy. Listening for evidence of the existence of such forms may indeed prove to be in the long run a profitable and exciting pursuit. HARRISON BROWN

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Isotopic Composition of Lead and Strontium from Ascension and Gough Islands

Abstract. Isotopic composition of lead and strontium has been determined in a series of rock samples from two islands on the Mid-Atlantic Ridge. Both interand intra-island variations exist in the abundance of radiogenic isotopes of both elements. Lead from basalt of Ascension Island has a Pb²⁰⁶-Pb²⁰⁴ ratio of 19.5, while the corresponding ratio at Gough Island is only 18.4. The Pb²⁰⁸-Pb²⁰⁴ ratios from the two islands do not differ. Conversely, strontium from basalt of Ascension Island is less radiogenic than that from Gough Island basalts. The trachytes of both islands have lead and strontium that is more radiogenic than that found in the basalts. The inter-island differences indicate the existence of regional variations in the uranium-lead and rubidium-strontium ratios of the upper mantle source of these rocks and show that isotope compositions are a means for investigating chemical heterogeneities in the mantle.

The isotopic composition of strontium in rocks derived from the mantle, for example, oceanic volcanic rocks, is determined by the relative abundances of rubidium and strontium during geologic time in the parent materials that produce the rock magmas. Gast (1) used this principle to place certain restrictions on the composition of the upper mantle of the earth. He showed that basaltic rocks contain strontium with much lower Sr⁸⁷-Sr⁸⁶ ratios than would be expected if the outer mantle had the same chemical composition as chondritic meteorites, an assumption that has been widely used in various calculations. Another use of isotopic data for strontium in basic rocks is based on knowledge of the amount of variation in isotopic composition from one rock

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to another; this information can be used to infer the amount and extent of regional variations in the ratio of rubidium to strontium in the upper mantle. Data obtained thus far indicate that the Sr⁸⁷-Sr⁸⁶ ratio of strontium from basic rocks is quite uniform (1-3). The isotopic composition of strontium in surface rocks is highly variable, depending on the age and rubidium-strontium ratio of the rock and on the isotopic composition of strontium initially inherited by the rock. If basic rocks contain strontium of narrowly defined isotopic composition, a potentially useful tracer is provided that can delineate rocks of deep-seated origin from those containing large admixtures of crustal materials. Several authors have discussed the use of strontium in this manner to study the origin of granitic rocks (1, 3, 4).

Arguments similar to those used for strontium can also be applied to the uranium-lead and thorium-lead systems. The former system is particularly valuable since two different isotopes of uranium, U^{285} and U^{288} , with greatly different half-lives decay to form two isotopes of lead, Pb²⁰⁷ and Pb²⁰⁶. There are relatively few data in the literature on the isotopic composition of lead in rocks. We have begun to investigate the isotopic composition of lead and strontium in a series of rocks from two typical islands on the Mid-Atlantic Ridge, Ascension Island and Gough Island, to establish the magnitude of both inter- and intra-island variations in the isotopic composition of these elements. An oceanic rather than a continental environment was chosen for these experiments to avoid possible contamination by radiogenic lead and strontium from the granitic rocks associated with the crust in continental areas. Rather detailed petrologic studies of both islands have been made by previous investigators (5, 6).

Isotopic compositions of lead and strontium, together with brief descriptions of individual samples used in this study, are given in Table 1. Analyses labeled U.S. Geological Survey were made on the mass spectrometer, having a 12-inch (30-cm) radius of curvature, that was used by Hedge and Walthall (3). Those labeled University of Minnesota were made on a mass spectrometer having a 6-inch (15-cm) radius of curvature, as described by Gast (7). Analyses labeled Department of Terrestrial Magnetism were made on a mass spectrometer [9-inch (22.5-cm) radius of curvature] with an electron multiplier ion detector. The lead analyses were interspersed with analyses of the reference sample from California Institute of Technology described by Chow and McKinney (8). Corrections for electron multiplier discrimination were determined from results on the reference sample. The reliability of the ratio determinations can be estimated from the duplicate measurements in Table 1. The duplicate data in this table represent chemical processing of two separate rock samples. The lead concentrations varied from 2 to 8 parts per million, except that G-19D contained about 20 parts per million. Fivegram samples were generally used; the reagent blanks were about 0.1 μg for this amount of sample. Hedge and Walthall (3) give additional data bearing

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on the reliability of the strontium ratio measurements. The long-term precision at the 95-percent confidence level for the normalized ratio of $Sr^{s\tau}$ to Sr^{s0} is ± 0.001 . Replicate determinations made over a short term have about half this uncertainty.

Lead isotope compositions for samples from both islands are shown in Fig. 1, in which the isotope ratios of lead evolving in chemically closed and coetaneous systems with varying uranium-lead ratios define straight lines called isochrons (9). In Fig. 2 the Pb²⁰⁶-Pb²⁰⁴ ratios are plotted against the Pb²⁰⁶-Pb²⁰⁷ ratios; in this diagram the isochrons approximate straight lines. The isochron defined by meteoritic lead, called the geochron (10), is shown in both Figs. 1 and 2 as the 0-million year isochron. The growth curve for a chemically closed system having a present-day U²³⁸-Pb²⁰⁴ ratio of 8.9 and an age of 4.55 billion years is also shown in both figures; it is similar to the curve derived by Ostic et al. (11) from selected samples of galena. Both the isochrons and the growth curves are shown primarily as a convenient frame of reference. Four of the samples from Gough Island fall very close to the geochron and are nearly consistent with an evolution in a chemically closed system 4.5 billion years old. The U²³⁸-Pb²⁰⁴ ratios inferred from these isotope compositions bracket the U²³⁸-Pb²⁰⁴ ratio derived from the selected galenas. Both the trachytic rocks studied seem to have somewhat more radiogenic lead than the basalt and trachyandesite. Sample G-3, in particular, is distinguishable from the other samples in that it has an excess of Pb²⁰⁶. The difference between the mean Pb²⁰⁶-Pb²⁰⁴ ratio of samples G-15 and G-95, and the mean of two determinations on G-3, cannot be ascribed to experimental errors. The uranium and lead contents of G-3 are 1.6 and 8.4 parts per million, respectively, giving a U238-Pb204 ratio of 14. This leads to a rate of change of 0.002 per million years in the ratio of Pb²⁰⁶ to Pb²⁰⁴ due to the decay of U²³⁸. Thus, even if the U²³⁸-Pb²⁰⁴ ratios of the other samples were zero, it would require 140 million years to generate the difference of 0.3 observed in the Pb²⁰⁶-Pb²⁰⁴. A preliminary K-Ar age determination on the trachyte G-19D suggests that the age of this rock is less than 2 million years. We thus conclude that the difference in the Pb²⁰⁶-Pb²⁰⁴ existed at the time when these rocks were emplaced. Therefore, the different rock types could not all be derived from an istotopically homogeneous magma.

The lead from the Ascension rocks is much more radiogenic than the Gough lead. The Pb²⁰⁶-Pb²⁰⁴ ratios for

Table 1. Isotopic compositions of lead and strontium in rocks from Gough Island and Ascension Island. The name of each rock is followed by the sample number assigned by LeMaitre (6) or by Daly (5). Sample descriptions for rocks from Ascension Island are taken from unpublished descriptions by Daly. The strontium-rubidium ratio for sample G-19D was determined by the isotope dilution method; all other strontium-rubidium ratios were determined by x-ray fluorescence.

Sample description and No.	Pb ²⁰⁶	Pb ²⁰⁶	Pb ²⁰⁶	Sr ⁸⁷ *	Sr
	Pb ²⁰⁴	Pb ²⁰⁷	Pb ²⁰⁸	Sr ⁸⁶	Rb
	Gough i	Island			
Olivine-poor basalt, G-132	18.36 ₆ †	1.1 7 1 ₃	0.4711	0.7045† .7038‡	
Porphyritic trachyandesite, G-15	18.37_{5}^{\dagger}	1.170_8	.4694	.7050†	13
Porphyritic trachybasalt, G-95	18.43_{3}^{\dagger}	1.171_{2}	.4695	.7043†	17
Sodalite, aegerine-augite	18.63 ₆ †	1.179_{6}	.4699	.7094†	0.0155
trachyte, G-19D	18.63_7^{\dagger}	1.178_{6}	.4688		0.5
Aegerine-augite trachyte, G-3	18.73_{8}^{\dagger}	1.191_{0}	.4738	.7050†	0.5
	18.71 ₄†	1.193 ₂	.4744		0.5
Aegerine-augite trachyte, G-18				.7069†	.05
	Ascension	ı İsland			
Olivine basalt (Travellers Hill) 2765	19.43 §	1.240	0.4958	0.7025† .7028‡	15
Olivine-poor basalt, (Portland Point) 2740	19.55 †	1.247	.5008	.7025† .7028‡	
Obsidian bomb 2775	19.50 ₀ †	1.247_{1}	.4973		
	19.51,†	1.246_{0}	.4975		
Aegerine-riebeckite trachyte	10 72 8	1 255	5000	7073+	0.25
Trachyandesite (S.E. Head) 2864	19.72 8	1.235	.5000	.7025†	0.25
Trachyte (S.E. Head) 2863				.70451	
	Stand	ards			
Lead (California Institute	16.72 †	1.0718	0.4556		
of Technology)	16.75 †	1.0703	.4547		

* Normalized to $Sr^{s6}/Sr^{s8} = 0.11940$. † U.S. Geological Survey. ‡ University of Minnesota. § Department of Terrestrial Magnetism.

all these samples are drastically displaced from the geochron; the isotope ratios yield negative model ages exceeding -600 million years. The variation within the set is somewhat less than that observed for the rocks from Gough Island; however, the highest Pb²⁰⁰-Pb⁵⁰⁴ ratio is again associated with a trachyte. Such leads have been designated anomalous leads in previous discussions. They are relatively common in continental regions: leads similar to those observed at Ascension Island have been reported from Mount Vesuvius (12) and New Mexico (13).

It is important to note that the abundance of Pb²⁰⁶ relative to Pb²⁰⁴ differs between the two islands much more than the abundance of Pb207 relative to Pb²⁰⁴. This requires that the lead in the samples from Ascension Island has evolved in a system in which the uranium-lead ratio was increased over that existing in the Gough Island source within approximately the past 1.5 billion years, after which time the abundance of U²²⁵ was too low to yield much additional Pb207. A secondary isochron for a billion-year-old subsystem is shown in Fig. 1. Figure 3 shows that the range of the Pb²⁰⁸-Pb²⁰⁴ ratios is about the same on each island. In order to generate the observed anomalies, the U²³⁸-Pb²⁰⁴ and U²³⁸-Th²³² ratios in this subsystem would have to be 15 and 0.5 today; that is, approximately 1.8 times the mean values derived for the source of normal leads such as the samples from Gough Island or the comformable leads of Ostic et al. (11).

Strontium isotope ratios for samples from both islands are shown plotted against a common variation index in Fig. 4. The initial ratios for two samples from Gough Island, G-19D and G-18, are somewhat uncertain due to the high rubidium-strontium ratios, and uncertainty in the age of these rocks. The magnitude of this uncertainty is indicated by the size of the correction for an age of 2 million years shown in Fig. 4. If the age of G-19D is in fact substantially less than 2 million years, the initial ratio of this rock is higher than that of the more basic rocks. For the rocks from Ascension Island, two trachyte samples (2838 and 2716) yield strontium isotope compositions which are definitely more radiogenic than those from the more basic rocks from this island. Thus, in at least one of the islands on the Mid-Atlantic Ridge the strontium, like the lead, is more radiogenic in the trachytes than in the more basic rocks. Lessing and

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Catanzaro (14) have observed a similar, somewhat smaller, difference for the basalts and trachytes of Hawaii. Figure 4 also shows that strontium in the more basic rocks of the two islands on the Mid-Atlantic Ridge is systematically and significantly different. The $Sr^{sr}-Sr^{sa}$ ratio for the rocks from Gough Island is higher by 2 parts in 700 than that for the rocks from Ascension Island.

Thus, a comparison of the results for lead and strontium indicates that (i) the intra-island variations are in accord with the usual geochemical behaviors of uranium, thorium, lead, rubidium, and strontium—that is, sources enriched in uranium and thorium with respect to lead are also enriched in rubidium with respect to strontium; and (ii) this situation is reversed for the inter-island variations where high Pb^{200} - Pb^{204} ratios are associated with low Sr^{87} - Sr^{86} ratios.

The variations could have arisen in two main ways: (i) by admixture of surficial or crustal lead and strontium with the lead and strontium in the primary magma, or (ii) from chemical, and ensuing isotopic, heterogeneities in the source of the magma, combined with magma-forming mechanisms which tended to preserve these heterogeneities. Several possible mechanisms might explain the data.

The magma-contamination hypothesis would be quite acceptable for a magma which has traversed tens of kilometers of continental crust, since highly radiogenic lead and strontium are relatively abundant in many crustal rocks. However, no abundant source of radiogenic lead and strontium is available in oceanic regions. The isotopic composition of lead in pelagic sediments has been investigated by Chow and Patterson (15). Their data for the Atlantic Ocean is included in Fig. 2. This graphical comparison indicates that neither the inter- nor intra-island variations in the lead isotope ratios are easily explained by mixing pelagic with magmatic lead. Only the difference between G-3 and the other samples from Gough Island can be readily explained by admixing some pelagic lead with magmatic lead. This possibility is eliminated by comparing the Pb208-Pb204 ratios for trachytes G-19D and 2716 with those determined in pelagic lead samples. The Pb208-Pb204 ratio for lead from both of the trachyte samples is higher than any of those determined for sediments or manganese nodules of the Atlantic Ocean (15). Thus, the lead isotope ratios are not consistent 11 SEPTEMBER 1964



Fig. 1. Isotope ratios for lead from Gough Island and Ascension Island compared with the meteorite isochron (0 m.y.) and the growth curve (G) for lead from conformable bodies of ore of Ostic *et al.* (11). I' is a secondary isochron representing the expected isotopic composition of leads from a 1 billion-year-old subsystem.

with the magma contamination hypothesis.

Lessing and Catanzaro (14) have concluded that the differences in the Sr^{s7}-Sr^{s6} ratio in the Hawaiian trachytes and basalts result from assimilation of sedimentary material by the trachytic magma. If the strontium isotope composition is considered independently of other factors, this explanation is equally valid for the differences found in this study. An Sr^{s7} - Sr^{s6} ratio of 0.720 \pm .003 has been determined (16) for a single sample of carbonate-free red clay from the Atlantic Ocean (27°17' N, 62°07'W). If strontium of this



Fig. 2. Lead from Gough Island and Ascension Island compared with the lead in pelagic sediments determined by Chow and Patterson (15). G, The growth curve for lead from conformable bodies of ore; NA, samples of pelagic lead from the North Atlantic; SA, pelagic lead from the South Atlantic.



Fig. 3. Diagram showing intra-island variations in the abundance of Pb^{208} and the absence of inter-island variations in abundance of Pb^{208} . G, The growth curve for lead from conformable bodies of ore.

isotope composition is mixed with magmatic strontium (Sr^{s7} - Sr^{s6} ratio, 0.703), in order to derive strontium isotopically similar to that in sample 2716, onefourth of the strontium in this sample must be of pelagic origin. However, because of the low strontium content of this rock (20 ppm) and because of the relatively high abundance of strontium in pelagic sediments (200 to 400 ppm), the proportion by weight of sediment to magma required to change the isotope composition is less than one part of sediment to 25 parts of original



Fig. 4. Ratios of Sr^{s7} to Sr^{s0} plotted against ($\frac{1}{3}SiO_2 + K_2O$) – (CaO + MgO). Samples 2765, 2740, and 2716 have not been chemically analyzed; their positions along the abscissa are therefore only approximate. The arrows indicate the position of the initial Sr^{s7} - Sr^{s6} ratio for samples G-19D and G-18 if they were 2 million years old. All other ratios will not be significantly changed in 2 million years.

magma. It is difficult to exclude this amount of contamination on chemical or mineralogic grounds. Because of the much higher strontium content of the more basic rocks, the amount of sediment which must be added to increase the Sr⁸⁷-Sr⁸⁶ ratio is very substantial. An increase of 0.002 in the Sr⁸⁷-Sr⁸⁶ ratio requires from 10 to 20 percent of sediment by weight. This amount of contamination is inconsistent with the rather uniform chemical composition and mineralogy of the rocks. Thus, the inter-island variations in the Sr⁸⁷-Sr⁸⁶ ratio probably cannot be explained by mixing pelagic sediments with a primary magma, even if the lead isotope compositions are neglected.

We have assumed that no older crystalline rocks exist in the vicinity of either Ascension Island or Gough Island. Yet, in his description of the geology of Ascension Island (5) Daly describes inclusions in some of the flows which he supposes are derived from an older terrane of granitic rocks. Based on more careful observations, Tilley (17) concludes that these inclusions are the plutonic equivalent of some of the more acidic extrusive rocks. If Daly's hypothesis regarding the origin of these inclusions is correct, particularly if this older terrane is several hundred million years old, there exists a source of both radiogenic strontium and lead intimately associated with the volcanic rocks of Ascension Island. However, even with this source of radiogenic strontium and lead it is difficult to devise an explanation of the observed isotope compositions. In particular, it is hard to see how the isotope composition of lead in the basalts (samples 2765 and 2745) can be affected without also changing the chemical composition and the strontium isotope composition.

Thus, it is possible that mixing of surface material with a parent magma may explain some of the observed isotopic variations, particularly the high Sr^{s7} - Sr^{s0} abundance in the trachytes. But the inter-island variations and most of the differences in lead isotope abundance do not fit this mechanism.

Chemical and isotopic heterogeneities within the mantle can lead to the observed isotope ratios in several ways. First, the small intra-island variations in both lead and strontium isotope compositions may result from local heterogeneities in the mantle, perhaps even on the scale of individual mineral grains. If the more silicic magmas are

an early melting fraction of the mantle which is formed primarily from mineral phases having high rubidium-strontium and uranium plus thorium-lead ratios, and if these magmas can be brought to the surface without complete mixing with later derived magmas, the observed isotope differences would result. Second, the inter-island differences may reflect regional variations in the rubidium-strontium and uranium-lead ratios in the mantle. We have already shown that a general increase in the uraniumlead ratio for the last billion years or so in the region of the source of the volcanic rocks on Ascension Island could explain the different Pb206 abundance in the two islands. This mechanism requires that the increase in the uranium-lead ratio was not accompanied by a corresponding increase in the thorium-lead ratio. The data for strontium show that the postulated enrichment of uranium with respect to lead would have taken place without enrichment of rubidium with respect to strontium. If correct, this explanation indicates that fractionation of these elements has occurred in the outer mantle in ways that do not follow those commonly observed in magmatic differentiation of plutonic rocks. This is probably more an indication of our ignorance of chemical fractionation processes in sub-crustal regions than an argument against the existence of chemically differentiated regions in the mantle. The work of Shavrova (18) and Gottfried et al. (19) on the thorium and uranium content of a series of circumpacific volcanic rocks gives some support to the high uraniumthorium ratios inferred for the source of the magmas on Ascension Island. Their work shows, first, that the uranium-thorium ratios of 1 : 2 are not uncommon in volcanic rocks and, second, that this ratio does not change with the composition of the rock. The second observation suggests that the observed uranium-thorium ratio is related to that in the source of the rock rather than to a magmatic differentiation process. Gottfried et al. (19) suggest that the high uranium-thorium ratio is due to assimilation of geosynclinal sediments. This hypothesis, however, implies some change in the uranium-thorium ratio with the changing composition of the volcanic rock, and is thus not entirely consistent with the observations. An alternative hvpothesis is that the high uraniumthorium ratio in the volcanic rocks is due to a similar high ratio in some 11 SEPTEMBER 1964

parts of the mantle. It is perhaps more than coincidence that the uraniumthorium ratio in the mantle, inferred from isotope data for Ascension Island, is similar to that inferred from chemical data in the circumpacific region.

It appears that regional chemical heterogeneities in the upper mantle are the most plausible explanation of observed inter-island variations in the isotope composition of lead and strontium. If the uranium-lead ratio in different parts of the mantle has been differentiated at various times in the last 3 to 4 billion years as we have inferred, the existence of terrestrial lead evolved in a chemically closed system formed at the same time as the planet is open to question. Also, if the isotopic variations observed at these two islands are due to chemical processes occurring in the mantle, the initial isotope ratios of lead and strontium in extrusive igneous rocks may not be used in continental regions as unambiguous indicators of contamination with older crustal rocks.

The hypotheses we have listed offer neither a unique nor a complete explanation of the observed variations. We report these measurements because they indicate a parameter which may be useful in investigating magma forming mechanisms, and because they give some indication of the scale on which isotope measurements must be undertaken in order to yield detailed information on the upper mantle.

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 This work was done while P.W.G. was a research associate of the Carnegie Institution of Washington. We acknowledge the cooperaresearch associate of the Carnegie Institution of Washington. We acknowledge the coopera-tion of Prof. S. Agrell, Cambridge University, who obtained the samples from Gough Island for us, and the help of Prof. J. B. Thompson, Harvard University, who enabled us to ob-tain the Ascension Island samples from the Harvard Department of Geology. We are indebted to Bruce Doe and Zell Peterman of the LIS. Geological Survey for pointing out the U.S. Geological Survey for pointing out the new work on thorium-uranium ratios. The preliminary K-Ar determinations were made by Prof. Peter Signer, University of Minnesota.

31 July 1964

Hydrogen Atom Thermal Parameters

Abstract. Isotropic hydrogen atom thermal parameters for N,N'-hexamethylenebispropionamide have been determined. They show a definite trend and vary from approximately the same as the mean thermal parameters for atoms other than hydrogen near the center of the molecule to appreciably greater for atoms near the end. The indicated trend for this compound, along with other results, provides the basis for a possible explanation of the anomolous values that have been obtained for hydrogen atom thermal parameters.

The techniques of x-ray diffraction have now advanced to the point where hydrogen atom parameters of some significance can be determined for organic molecules in the solid state. Several recent refinements of structures (1) have included isotropic thermal parameters for the hydrogen atoms and, in most instances, have led to results where the apparent parameters are less than those of the atoms to which hydrogen is bonded and in some cases even to negative values (2).

We now report the results of refining hydrogen atom thermal parameters in N,N'-hexamethylenebispropionamide (HMBPA) (Fig. 1) and suggest a possible explanation for the anomolous results that have appeared.

The reported parameters for HMBPA (3) were used as the starting point in further refining the structure by full matrix least squares (4). In the last stage of refinement, variation of both