

LEAD ISOTOPIC COMPOSITION OF THE OLDEST VOLCANIC ROCKS OF THE EASTERN GREATER ANTILLES ISLAND ARC

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Abstract

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Pb isotopic analyses for rocks from the early Cretaceous Los Ranchos Formation, which is the basal volcanic assemblage in the Dominican Republic, fall on a short linear array of shallow slope in a $^{207}\text{Pb}/^{204}\text{Pb}$ vs. $^{206}\text{Pb}/^{204}\text{Pb}$ plot. The array is interpreted to have formed by decay of local U since the time of ore and rock formation ~ 110 Ma ago. Previously reported analyses of sulfide samples from the Pueblo Viejo Au-Ag deposit in the upper part of the Los Ranchos Formation fall on the lower half of the same linear array. In a $^{208}\text{Pb}/^{204}\text{Pb}$ vs. $^{206}\text{Pb}/^{204}\text{Pb}$ plot the Los Ranchos Formation can be divided into the lower member with an apparent Th/U ratio of 1.2 and the upper three members with an apparent Th/U ratio of 2.6. A cluster of analyses falls at the least radiogenic end of these arrays and is interpreted to represent the initial Pb isotopic composition of the Los Ranchos magmas. This cluster falls close to the Stacey-Kramers growth curve. For the Los Ranchos Pb, a homogeneous source with $\mu = 9.6$ and Th/U near 3.8, since 3.7 Ga ago, is implied.

Previously reported analyses for correlative rocks of the Water Island Formation, the basal volcanic complex in the Virgin Islands at the east end of the Greater Antilles, suggest a steep linear array extending downwards from the Los Ranchos cluster, after correction of the ratios for U decay since formation of the rocks. This array has a slope of 0.250 ± 0.023 , which could have resulted from magma derivation from sources that had been isolated for ~ 3.0 Ga. Younger rock Pb, from the island of Martinique, has a similar slope, although with larger $^{206}\text{Pb}/^{204}\text{Pb}$ and $^{207}\text{Pb}/^{204}\text{Pb}$ ratios. The data points extend upward from the radiogenic end of the Los Ranchos array.

These relationships indicate a complex derivation of magmas in the area. In the Greater Antilles, the proportion of continental material decreases eastward, while even farther east in the Lesser Antilles at Martinique, the proportion of crustal material, as indicated by Pb isotopic composition, is again very high.

1. Introduction

The Los Ranchos Formation, which is located in the Dominican Republic (eastern Hispaniola, Fig. 1), is one of the oldest volcanic units in the central Greater Antilles island arc. It appears to correlate with the Water Island Formation in the Virgin Islands (Donnelly, 1966),

which is comprised of generally similar lithologies and is also one of the oldest stratigraphic units exposed in the eastern Greater Antilles. Pb isotopic data for four rock samples from the Water Island Formation (Donnelly et al., 1971) are relatively low in radiogenic ^{207}Pb in comparison to other rock and ore Pb from the Greater Antilles (Donnelly et al., 1971; Cum-

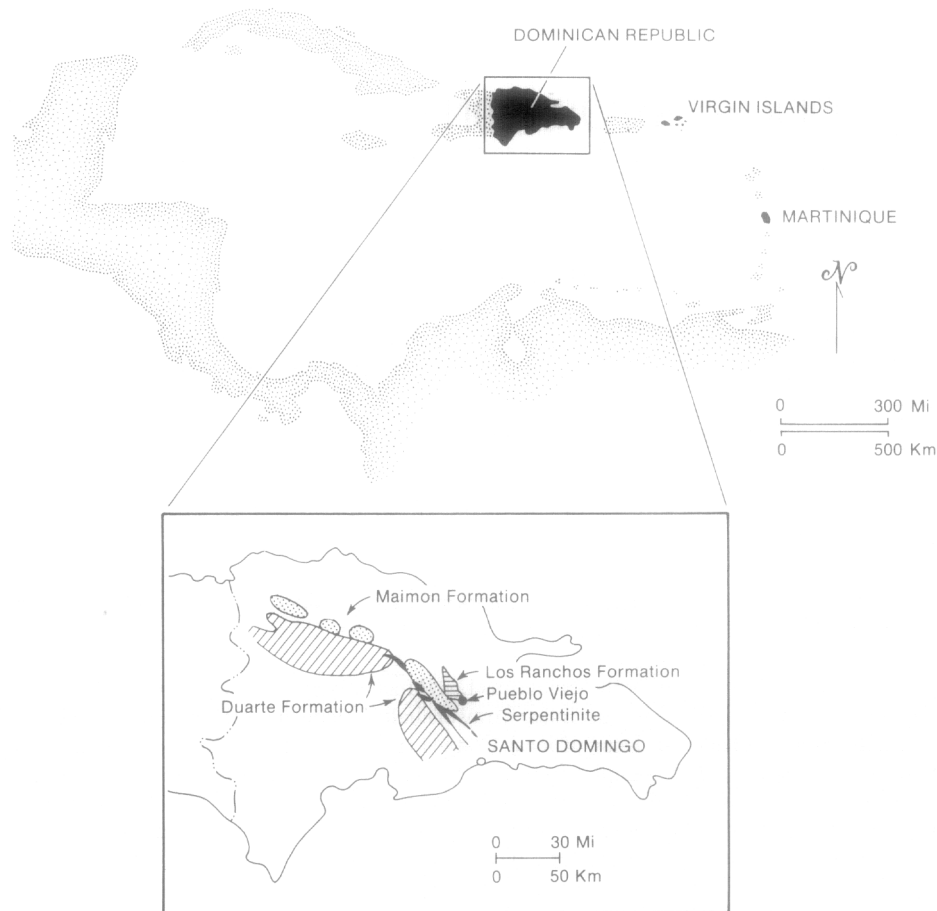


Fig. 1. Location of the Dominican Republic and Virgin Islands in the Greater Antilles. Martinique in the Lesser Antilles is also shown. The *inset* shows the regional geologic setting of the Los Ranchos Formation in relation to the pre-Albian Duarte metabasalt (sea floor?) and Maimon (-Amina) felsic to mafic metavolcanic rocks (island arc?).

ming et al., 1981). Ore Pb from the Pueblo Viejo Au-Ag deposit, which is in the upper part of the Los Ranchos Formation, is also somewhat low in radiogenic ^{207}Pb (Cumming and Kesler, 1976; Cumming et al., 1982) but form a short array of shallow slope. In this paper, we have attempted to evaluate the relationship between the Water Island and Los Ranchos Formations more fully by obtaining new Pb isotope data from rocks of the Los Ranchos Formation. Using these data along with those from the Water Island Formation and ore Pb from the Pueblo Viejo deposit (Cumming et al., 1982), we have assessed the early characteristics of sources for magmagenesis in the Greater Antilles.

2. Geologic setting and characteristics of the Los Ranchos and Water Island Formations

Both the Los Ranchos and Water Island Formations consist largely of a bimodal volcanic suite including basalt and dacite or rhyolite that have undergone extensive chemical interaction with seawater. The Water Island Formation contains abundant siliceous quartz keratophyre flows with smaller amounts of flow breccias and tuffs and spilite flows (Donnelly, 1966). All of these rocks have been altered by extensive interaction with seawater to assemblages containing chlorite, calcite, albite, epidote, pumpellyite and prehnite. The Water

TABLE I

Lithology of major members of Los Ranchos Formation

Pueblo Viejo Member	volcaniclastic, carbonaceous and pyritic sediments with spilitic flows(?) and keratophyre agglomerates in local basin or maar
Platanal Member	spilitic flows and volcaniclastic sediments with lenses of fossiliferous limestone, marl and shale
Zambrana Member	volcaniclastic rocks with keratophyre fragments
Quita Sueño Member	keratophyre flows and stocks, some fine-grained keratophyre tuffs and local spilitic flows

Island Formation is overlain by the Louisenhoj Formation, which consists of augite-andesite breccias that have undergone somewhat less interaction with seawater. The absence of terrigenous sediment and explosive textures was cited by Donnelly (1966) as evidence that volcanic rocks of the Water Island Formation were emplaced at great water depths, whereas the fragmental Louisenhoj Formation was thought to have been emplaced in a shallow water or even a terrestrial environment.

The Los Ranchos Formation, which has been divided into four members (Table I), shows a generally similar stratigraphic distribution of fragmental units (Kesler et al., 1981). The lowermost member, the Quita Sueño, consists largely of keratophyre flows and stocks with some fine-grained keratophyre tuffs and local spilitic flows. The overlying members of the Los Ranchos Formation (Table I) include tuffs and agglomerates that contain terrigenous sediments and are capped by a thick sequence of spilitic flows and spilitic volcaniclastic sediments. The uppermost unit in the Los Ranchos Formation, the Pueblo Viejo Member, hosts a large Au-Ag deposit that formed contemporaneously with the host volcanic pile (Kesler et al., 1981). All of these rocks have been altered

extensively by reaction with seawater to assemblages containing albite, epidote, calcite and chlorite. The abrupt increase in abundance of fragmental material above the Quita Sueño Member resembles that seen at the Water Island-Louisenhoj contact, suggesting that the upper Los Ranchos members actually correlate with the Louisenhoj Formation.

The age of the Water Island Formation is known from fossil evidence to be approximately Aptian-early Albian or ~110-115 Ma (Donnelly, 1966; T.W. Donnelly, pers. commun., 1984). Whole-rock K/Ar ages of $58-62 \pm 5$ Ma and $106-110 \pm 10$ Ma reported for the Water Island Formation (Donnelly, 1966) probably in part represent reheating (Kesler et al., 1977). For the Los Ranchos Formation, the best age estimate is provided by terrestrial plant fossils found in the upper part of the formation, which suggest an early Cretaceous age (G.J. Smiley, pers. commun., 1978). Bowin (1966) reported an age of middle Aptian to middle Albian for marine fossils from one locality in the upper Los Ranchos Formation. As noted by Cumming et al. (1982), the Pb isotopic data for ore samples in the Pueblo Viejo deposit in the upper part of the Los Ranchos Formation support this age assignment. Younger $^{39}\text{Ar}/^{40}\text{Ar}$ ages of 61.6 and 67.8 Ma for alunite in the Pueblo Viejo ore deposit (Kesler et al., 1981) are thought to reflect reheating. Thus, it appears that the Water Island and Los Ranchos Formations are indeed coeval and are the oldest units in the eastern Greater Antilles.

3. Analytical methods

Los Ranchos Formation rock samples were dissolved by refluxing overnight in Teflon® beakers using 10 ml of concentrated HF and a little HNO₃. Pb was separated in an anion-exchange column using HBr-HCl. The same column in nitrate form was used to separate U. Pb blanks were 10-12 ng and the data have not been corrected for the blanks since the effect is negligible for these samples.

TABLE II

Pb/U concentrations				
No.	Sample I.D.	Pb (ppm)	U (ppm)	$^{238}\text{U}/^{204}\text{Pb}$
<i>(a) Quita Sueño member:</i>				
5	77-104 (leached)	0.525	0.500	61.07
6	73-604	1.316	0.323	15.77
<i>(b) Upper Los Ranchos members:</i>				
11	73-617	11.39	0.631	3.54
14	73-511	1.747	0.275	10.09

All analyses were carried out on a Micro-mass[®] MM30 mass spectrometer. At the time these analyses were carried out, measuring precision for Pb was as described in Cumming et al. (1982), that is, close to 0.04% per mass unit difference (2σ) as determined by 40 replicate analyses of N.B.S. SRM981. Data were normalized to near absolute values using the measured ratios of the standard.

4. U/Pb ratios

Reconnaissance measurements of Pb and U concentrations were carried out on four samples and the data are shown in Table II. One sample (No. 5) was leached in HCl to test the relative solubilities of the Pb and U in the rocks. The Pb abundance is lower in the leached sample compared to the other samples, while little change is evident in the U concentration.

The U/Pb systems in the three other samples have clearly been disturbed since the original rock formation at ~ 110 Ma ago. In Fig. 2 we have indicated the position of an isochron beginning with an initial $^{206}\text{Pb}/^{204}\text{Pb}$ ratio of the average of the cluster of points in Fig. 3., as discussed below, with a slope corresponding to 110 Ma. The three sample points on the diagram are substantially depleted in U, lying above the line, while the leached sample (No. 5) is well below the isochron line, indicating substantial differential Pb loss, presumably due to the leaching. It seems doubtful to us that a detailed

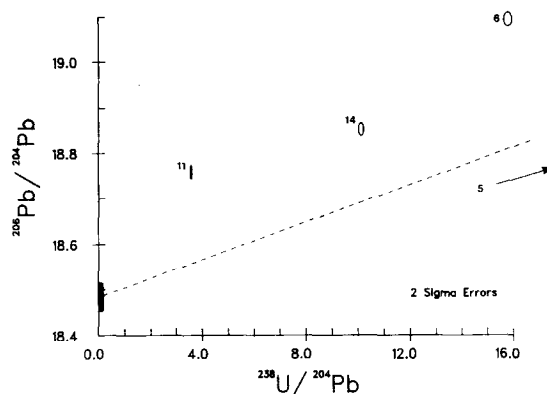


Fig. 2. U/Pb isochron diagram for rocks of the Los Ranchos Formation. Numbering as in Tables II and III. The *dashed line* indicates the expected position of a 110-Ma isochron beginning at the estimated initial $^{206}\text{Pb}/^{204}\text{Pb}$ ratio indicated by the *solid bar*.

study of the U–Pb systematics can produce useful results, bearing in mind the altered nature of these rocks, and we have not pursued this problem any further.

5. Pb/Pb isotope ratios

Table III contains the analytical results of our measurements of 17 rocks, as well as the 11 sulfides from the Pueblo Viejo deposit that were reported in Cumming et al. (1982). The data are plotted in Fig. 3 with different symbols for each class of material as indicated on the figure. Four additional analyses for the Water Island Formation (Donnelly et al., 1971) are also given in Table III and Fig. 3.

There are two data arrays on this plot that require explanation. The simplest of these is formed by the sulfides and rocks of the Los Ranchos Formation. Note that seven of the 11 sulfide samples from the Pueblo Viejo deposit as well as two samples of Pueblo Viejo member rocks form a tight cluster only slightly larger than the experimental error as indicated in Fig. 3. The remaining rock and sulfide data points fall along a linear array in the $^{207}\text{Pb}/^{204}\text{Pb}$ vs. $^{206}\text{Pb}/^{204}\text{Pb}$ plot, with an indication that rock Pb data, particularly from samples of the Quita Sueño member, tend to be somewhat more radi-

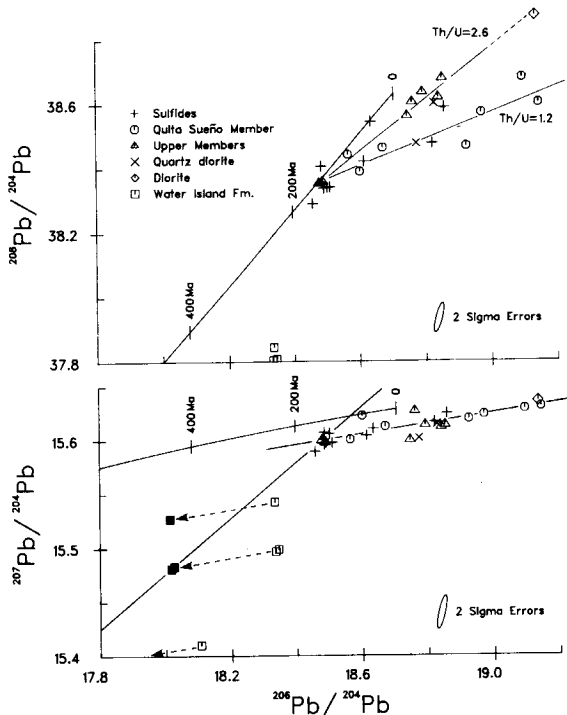


Fig. 3. Pb/Pb diagram for Pueblo Viejo sulfide samples and rocks of the Los Ranchos Formation of the Dominican Republic. Rocks from the Water Island Formation of the Virgin Islands are shown as *open squares* (Donnelly et al., 1971). *Solid squares* are after corrections of the latter ratios for 110 Ma of Pb evolution. One corrected value falls out of the field of the diagram. Two isochrons are discussed in the text. *Error ellipse* indicates the measuring error for the data presented here. The errors for the *squares* are ~ 4 times larger. The growth curves, shown for reference, are those of Stacey and Kramers (1975).

ogenic than the sulfides. On the plot of $^{208}\text{Pb}/^{204}\text{Pb}$ vs. $^{206}\text{Pb}/^{204}\text{Pb}$ in Fig. 3, the Quita Sueño member values are clearly on a line of shallow slope with some scatter, while the data for samples from the upper members closely define a line with distinctly steeper slope. Pb isotope ratios from the sulfides in the Pueblo Viejo deposit are scattered on and between the two lines.

Since the ore deposit and the volcanic rocks are contemporaneous (Kesler et al., 1981), we can treat the rocks and sulfides as a single system. We therefore interpret these data as representing an initial Pb with a composition close to the cluster of data points, to which has been

added radiogenic Pb from U and Th decay since the time of rock and ore formation. Some of the ore sulfides may have U associated with them, which would provide the radiogenic additions and enable us to treat the ore sulfides in the same way as the rock Pb. Thus the slope of the linear array of the $^{207}\text{Pb}/^{204}\text{Pb}$ vs. $^{206}\text{Pb}/^{204}\text{Pb}$ diagram may be used to determine the age of the ore and rocks from the usual isochron equation (Faure, 1977). This relation yields a line of slope $R = 0.048 \pm 0.0065$ with mean squared weighted deviates of $\text{MSWD} = 0.92$, as calculated by the method of Cumming et al. (1972). The age of the ore-rock system as determined from the slope of the regression line (on the assumption that the distribution of points is simply related to the time interval since formation) is 130 Ma, in good agreement with the fossil evidence as discussed above. Such agreement must, however, be considered as fortuitous, since the slope age is not closely defined due to the short line and the magnitude of the experimental errors.

Confirmation for the essential validity of the slope age is provided by the composition of the cluster of points for the ore Pb. The cluster lies just below the Stacey and Kramers (1975) growth curve for which they determined a second-stage U/Pb ratio of $\mu = 9.74$. Changing this value to 9.66 generates a growth curve that passes through the ore Pb cluster at an age of 110 Ma, in excellent agreement with the fossil indications and the slope age. Using this age, we have calculated Th/U ratios from the slopes of the $^{208}\text{Pb}/^{204}\text{Pb}$ vs. $^{206}\text{Pb}/^{204}\text{Pb}$ arrays in Fig. 3. The slope of the line for the Quita Sueño data indicates an average Th/U ratio of only 1.2 ($\text{MSWD} = 6.4$), lower than the line through the data for the upper members, which yields a Th/U ratio of 2.6 ($\text{MSWD} = 1.7$). It is important to note that the two lines intersect close to the Stacey and Kramers (1975) growth curve, indicating that the Th/U ratio in the source of all units of the Los Ranchos Formation was nearly that of the "average earth" (3.8), while the Th/U ratio in the rocks after their forma-

TABLE III

Pb isotope ratios — Los Ranchos Formation and related rocks and minerals

No.	Sample i.d.	$^{206}\text{Pb}/^{204}\text{Pb}$	$^{207}\text{Pb}/^{204}\text{Pb}$	$^{208}\text{Pb}/^{204}\text{Pb}$
<i>(a) Quita Sueño member (lower Los Ranchos):</i>				
1	72-71	18.565	15.601	38.442
2	78-30-B(H)	18.602	15.623	38.389
3	78-1	18.671	15.613	38.463
4	78-24	18.925	15.620	38.468
5	77-104	18.971	15.624	38.572
6	73-604	19.095	15.629	38.681
7	78-15	19.145	15.631	38.602
<i>(b) Upper Los Ranchos members:</i>				
8	DDH86A-11M	18.477	15.600	38.353
9	165-91M-RC(PQK)	18.488	15.598	38.357
10	73-383	18.745	15.600	38.561
11	73-617(H)	18.760	15.627	38.604
12	73-538	18.792	15.614	38.635
13	77-37	18.841	15.612	38.619
14	73-511	18.853	15.614	38.678
<i>(c) Intrusives:</i>				
15	73-599 quartzdiorite	18.773	15.602	38.476
16	78-17 quartzdiorite	18.828	15.615	38.601
17	72-82 Diorite	19.136	15.636	38.873
<i>(d) Water Island Formation:</i>				
18	68-1-20	18.336	15.497	37.844
19	61-80C	18.106	15.410	37.461
20	63-76A	18.345	15.499	37.807
21	425	18.333	15.543	37.804
<i>(e) Pueblo Viejo sulfide samples:</i>				
22*	DDH162-171-G-V	18.457	15.590	38.290
23*	DDH100-16-Py-L	18.482	15.601	38.406
24*	T-6-2-Py-V	18.485	15.608	38.356
25*	DDH71-107-Py-L	18.486	15.597	38.355
26*	T-2-5-Py-L	18.490	15.599	38.338
27*	DDH92-46-G-V	18.500	15.607	38.340
28*	DDH174-56M-Py-V	18.509	15.598	38.341
29*	DDH162-83M-Py-L	18.615	15.605	38.421
30	DDH101-21M-Py-L	18.635	15.611	38.544
31	DDH163-83M-En-V	18.821	15.617	38.477
32*	DDH161-123M-En-V	18.859	15.625	38.589

(a), (b) and (c): New analyses; *(d):* Donnelly et al. (1971); *(e):* Cumming et al. (1981).

*Average of two analyses.

tion was very significantly reduced. This may be due to some complex history of interaction with seawater or to some geochemical process associated with the differentiation of the rocks from their ultimate source.

Table III contains data on three samples of intrusive rocks. The quartz diorite is believed to be comagmatic with the extrusive rocks. On the other hand, there is evidence that the diorite is considerably younger (Bowin, 1966). The

samples have Pb isotopic compositions which are very similar to the volcanic units, indicating closely related sources for the intrusive and extrusive rocks.

Analysis of the steeper array suggested by the Los Ranchos Formation cluster and the compositions for the Water Island Formation is facilitated by correcting the latter data to their original, early Cretaceous compositions. U/Pb ratios necessary to do this are not available so we have estimated an average value of 16, based on the distribution of points on the linear array formed by the Pb/Pb data from the Los Ranchos Formation (Fig. 3). The measured U/Pb ratios were not considered to be a large enough population to obtain a valid average. The fact that the corrected ratios do not fall precisely on a single array after this correction may be a reflection of the variation in the actual U/Pb ratios of these samples. Notwithstanding this problem, the use of any reasonable U/Pb ratio would yield a steep array extending upwards from Water Island compositions to the Los Ranchos cluster, and it is this array that requires explanation.

This Los Ranchos–Water Island array indicates a substantial variation in initial $^{207}\text{Pb}/^{204}\text{Pb}$ ratios for the Water Island and Los Ranchos magmas, which could reflect either a corresponding heterogeneity in the mantle source region for the magmas or mixing between mantle Pb and some other Pb during magma-genesis. Davidson (1983) has observed a similar steep array (line 5, Fig. 4) for volcanic rocks of Martinique in the Lesser Antilles and suggested that it represents a mixing line between mantle and terrigenous sediment. Whereas some information is available on the isotopic composition of sediments that might have been involved in magmagenesis in the Lesser Antilles (Thirlwall and Graham, 1984), no such information is available for early Cretaceous and older sediments that might have been consumed during the Greater Antilles magmagenesis.

We note that the upper limit of the Los Ran-

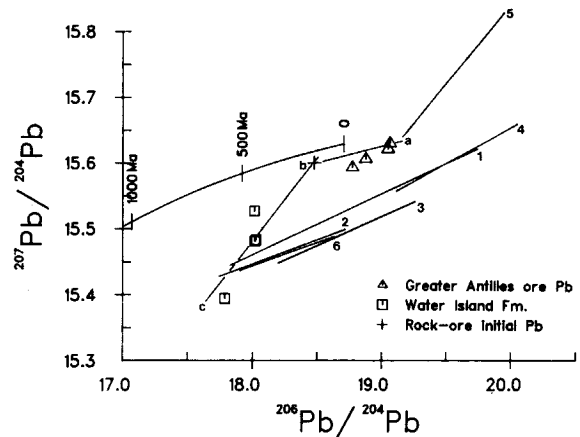


Fig. 4. Relationship of Pueblo Viejo linear array (*a-b*) to other oceanic arrays. 1 = North Atlantic (Dupré and Allègre, 1980); 2 = Mid-Atlantic (Tatsumoto, 1978); 3 = Iceland and Reykjanes Ridge, (Sun and Jahn, 1975; Sun et al., 1975); 4 = Canary Islands (Sun, 1980); 5 = Martinique (Davidson, 1983); 6 = Hawaii (Tatsumoto, 1978); *b-c* = Water Island Formation (Donnelly et al., 1971). Greater Antilles ore Pb from Cumming et al. (1981) are shown as *triangles*. Growth curve as in Fig. 3.

chos–Water Island array, as defined by the Los Ranchos cluster, is generally similar to the composition of pelagic sediments (Doe and Zartman, 1979; Zartman and Doe, 1981). It does not extend to compositions typical of cratons or craton-derived sediment which would be substantially above the growth curve shown. This is in agreement with the fact that the eastern Greater Antilles does not include continental fragments and was not adjacent to a craton in early Cretaceous time (Pindell and Dewey, 1982). The fact that the Water Island rocks exhibit a greater heterogeneity and a less radiogenic character than the Los Ranchos Formation, with the least radiogenic Water Island samples showing compositional similarity to MORB (lines 1–3, Fig. 4), suggests that the degree of mixing would have been greatest for the Los Ranchos magmas.

If the steep Los Ranchos–Water Island array were generated by long-lived heterogeneities in the mantle, then the slope of the array ($R = 0.251 \pm 0.022$) with $\text{MSWD} = 1.05$ and $t_2 = 110 \text{ Ma}$ indicates a t_1 of $3200 \pm 300 \text{ Ma}$ (2σ)

for the two-stage model of Gale and Mussett (1973). This calculation depends greatly on the U/Pb ratios assigned above and would range between 2500 and 4200 Ma for U/Pb ratios of $\mu=32$ and $\mu=0$, respectively. These ages suggest the possibility that the first major separation of crustal material from the mantle from which the Water Island and Los Ranchos magmas were derived, may have occurred very early in the history of the region.

The relation between the isotopic composition of the Los Ranchos–Water Island magmas and later volcanic rocks in the Greater Antilles can be estimated from the limited data available on ore deposits in these younger rocks (Cumming and Kesler, 1976; Cumming et al., 1982). Present information on the origin of these deposits indicates that the metals in the deposits were scavenged from the surrounding volcanic and volcanoclastic rocks by circulating hydrothermal solutions that were active at approximately the time of formation of the volcanic rocks (Kesler, 1978). Accordingly, the isotopic composition of the ores should reflect the isotopic composition of their host volcanic pile. Note that the Pb in late Cretaceous ore deposits from Haiti and Puerto Rico, as well as from an Eocene deposit in Jamaica, fall generally along the shallow array of the Los Ranchos Formation (Fig. 4). It is clear that these younger Greater Antilles volcanic rocks were derived from a source that was more similar to the Los Ranchos Formation than to the Water Island Formation and that, at least in terms of Pb isotopic characteristics, there was not a major change in magmagenesis between Water Island–Los Ranchos time and later volcanism in the Greater Antilles, as has been suggested on the basis of major and minor element data (Donnelly and Rogers, 1978). Rather, it appears that there was significant variation in magmagenesis during the early stages of evolution of the Greater Antilles and, at least as far as can be determined from the limited data, less variation later.

6. Conclusions

The distribution of Pb isotope ratios from the Los Ranchos Formation is consistent with the Cretaceous age for the rocks, both from the slope of the Pb/Pb isochron and from the initial Pb isotopic compositions. The ore Pb is very similar in composition to the host rocks, supporting the view that the ore was derived from the surrounding rocks. The initial Pb composition indicates a source with $\mu=9.66$, and a Th/U ratio close to 3.8. The isotope ratios are thus similar to those predicted by the Zartman and Doe (1981) plumbo-tectonic model for the average orogene. The data indicate a substantial reduction of the Th/U ratio for the rocks, and this must have occurred close to the time of formation since the ore Pb also shows the same reduced values.

On a broader scale, the isotopic data suggest a substantial well-homogenized crustal component in the Cretaceous rocks of Hispaniola while the data from the Virgin Islands, further from the continent, are more variable and generally much lower in ^{207}Pb , indicating a larger mantle component in the rocks. Still further east in the Lesser Antilles at Martinique, ^{207}Pb is again elevated and variable, indicating a greatly increased crustal component, presumably derived from terrigenous sediments subducted during the development of the Lesser Antilles arc.

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