26. ISOTOPIC AND TRACE ELEMENT COMPOSITION OF BASALTS FROM SITES 556-559 AND 561-564: CONSTRAINTS ON SOME PROCESSES AFFECTING THEIR COMPOSITION

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ABSTRACT

Sr and Nd isotopic composition of 23 basalts from Sites 556-559 and 561-564 are reported. The $^{87}$Sr/$^{86}$Sr ratios in fresh glasses and leached whole rocks range from 0.7025 to 0.7034 and are negatively correlated with the initial $^{143}$Nd/$^{144}$Nd compositions, which range from 0.51315 to 0.51289. The Sr and Nd isotopic compositions (in glasses or leached samples) lie within the fields of mid-ocean ridge basalts (MORB) and ocean island basalts (OIB) from the Azores on the Nd-Sr mantle array/fan plot.

In general, there is a correlation between the trace element characteristics and the $^{143}$Nd/$^{144}$Nd composition (i.e., samples with Hf/Ta > 7 and (Ce/Sm)$_N$ < 1 [normal-MORB] have initial $^{143}$Nd/$^{144}$Nd > 0.51307, whereas samples with Hf/Ta < 7 and (Ce/Sm)$_N$ > 1 [enriched-MORB] have initial $^{143}$Nd/$^{144}$Nd compositions < 0.51300). A significant deviation from this general rule is found in Hole 558, where the N-MORB can have, within experimental limits, identical isotopic compositions to those found in associated E-MORB.

The plume-depleted asthenosphere mixing hypothesis of Schilling (1975), White and Schilling (1978) and Schilling et al. (1977) provides a framework within which the present data can be evaluated. Given the distribution and possible origins of the chemical and isotopic heterogeneity observed in Leg 82 basalts, and some other basalts in the area, it would appear that the Schilling et al. model is not entirely satisfactory. In particular, it can be shown that trace element data may incorrectly estimate the plume component and more localized mantle heterogeneity (both chemical and isotopic) may be important.

INTRODUCTION

The origin of chemical and isotopic heterogeneity in the oceanic mantle is a subject of continuing debate (see, for example, White and Hofmann, 1982; Wood, 1979a; Schilling, 1973). One particular point of contention is whether the origin of the heterogeneity is related to a random distribution of distinct sources—the veined-mantle and/or plum-pudding models (Wood, 1979b; Davies, 1981)—or if the large part of the heterogeneity is related to the presence of mantle plumes (Schilling, 1975; White and Schilling, 1978).

The origin of the geochemical characteristics and their distribution in basalts erupted on the Mid-Atlantic Ridge (MAR) between 30 and 40°N has been explained by Schilling (1975) and White and Schilling (1978) as a result of mixing between an Azores mantle plume/blob (AMP/B) and a depleted low-velocity layer (DLVL). Briefly, the major features of their model can be summarized as follows: (1) presently, there is a major mantle plume centered at 39°N (the Azores mantle plume) and possibly a minor plume at 35°N (the Oceanographer mantle plume); (2) the plume source(s) are characterized by higher abundances of K, Ba, Sr, Cs, and light rare earth elements (LREE) and are enriched in the radiogenic Sr, Nd, and Pb isotopes (White et al., 1976; Hawkesworth et al., 1979; Sun, 1980); (3) mixing of the end-member sources depends on a number of factors, including spreading rate and plume flux (the rate of which can vary considerably and can also be episodic). In support of their model, White (1977) and White and Schilling (1978) have shown that, at least qualitatively, the distribution patterns of Sr isotopic composition, REE, and LIL (large ion lithophile) elements are consistent with a binary mixing model. The authors have also summarized supportive geophysical and morphologic features.

In addition to ascribing the origin of the zero-age geochemical features of MAR basalts to AMP/B-DLVL mixing, Schilling et al. (1977) have extended the model to include older basalts. Based on variations in (La/Sm)$_N$ ratios in basalts from Site 335 (Leg 37, 16 Ma), Site 332 (Leg 37, 3 Ma) (both from Aumento, Melson, et al., 1977), and FAMOUS (zero-age) (Schilling et al., 1977) suggested that the contribution from the AMP/B increases from (approximately) 0% at Site 335 to 30% at Site 332 and to 45% at the FAMOUS site.

At least one aim of the Leg 82 program was to determine if the geochemistry of basalts erupted over the last 35 Ma between 30 and 40°N is related to the presence of the Azores mantle plume. In order to evaluate the hypotheses of Schilling and his co-workers and to determine the origins of the heterogeneity in Leg 82 basalts, we have studied a number of basalts from Leg 82, using a wide variety of analytical techniques and approaches (see Sachtleben et al., in preparation; Hertogen et al., 1979; Sun, 1980).
this volume; Dupré et al., this volume). In this paper, we report Sr and Nd isotopic compositions on 23 samples and REE abundances in 10 samples. The samples were selected to represent the spatial and temporal, as well as geochemical, characteristics of Leg 82 basalts. Discussion of the data is broadly divided into two sections: one concerned with in-site variation, and the second concerned with how the data can be useful in delineating the origin of the heterogeneity.

The correlations between trace, Sr, and Nd isotopic composition and Pb isotopic composition are also briefly discussed. This aspect of the paper comes from joint work with B. Dupré at Mainz, in which 10 samples were analyzed for Sr, Nd, and Pb isotopic composition and REE abundances. For a more complete discussion of Pb techniques and results, see Dupré et al. (this volume).

**METHODS**

Conventional chemical techniques were used for the separation of Sr, Nd, and REE (see White and Patchett [1984] for details). Details of the blanks and mass spectrometry are also given in White and Patchett (1984). In this study, more specifically measured blanks are Sm < 50 pg; Nd < 180 pg; and Sr < 200 pg. Nd isotopic compositions were determined using double-filament, single-collector, and conventional-massspectrometric methods. Sr isotopic compositions were determined using a double-collector system installed on a Finnigan MAT 261 mass spectrometer. Errors reported for isotopic composition (Ta = 7.5) are two standard errors of the mean based on within-run statistics. This statistic slightly underestimates reproducibility.

The 10 samples for which REE data are reported (Table 2) were also analyzed for Pb isotopic composition. The procedure used for dissolution of these samples differed from that used on the other 13 samples. In particular, these samples had a 1-hour leach in warm 6N HCl, followed by Pb separation. The residue after Pb extraction was treated with HClO₄, HF, HCl, and the solution was split for isotope dilution (REE) and isotope composition analysis.

**RESULTS**

Sr and Nd isotopic compositions are reported in Table 1. Nd isotopic compositions have been corrected for age using the criteria given in Table 1. Sm/Nd ratios for samples 2, 3, 4, 9, 10, 12, 13, 17, 18, and 23 (see Table 1 for DSDP designations) are based on the isotope dilution REE measurements reported in Table 2; those for other samples are from the instrumental neutron activation data reported by Hertogen et al. (this volume). Values for CHUR (chondritic uniform reservoir) used in the Nd(T) calculations are based on a present-day value for (143Nd/144Nd)CHUR = 0.51264 and (147Sm/144Nd)CHUR = 0.1966. A summary of important trace element and isotopic data is given in Table 3. Chemical groups given in Table 3 refer to those assigned by the shipboard scientists. These groups do not conform to the subdivision into N (normal)-MORB and E (enriched)-MORB often used. For the purposes of this paper, N-MORB has Hf/Ta > 7, Zr/Nb > 13, and (Ce/Sm)N < 1; E-MORB has Hf/Ta < 7, Zr/Nb < 13, and (Ce/Sm)N > 1. This nomenclature is similar to that used by Wood et al. (1979), Sun et al. (1979), and many others.

**Hole 556.** All the samples analyzed are N-MORB; however, they come from two different shipboard-designated chemical groups. Within the limits of analytical error, all these basalts may be considered to have come from a source that is approximately uniform isotopically (see Table 3). The variation in Nd isotopic composition (εNd(T) = 8.6 to 9.7) may indicate some heterogeneity, but further study is necessary for confirmation.

**Hole 557.** Only one sample was analyzed for isotopic composition. This sample is an E-MORB with an εNd(T) value of 7.4.

**Hole 558.** Eight samples were analyzed from this location. These basalts can show marked heterogeneity in trace and isotopic characteristics; however, the degree of correlation between the two types of heterogeneity is variable. The following points are notable: (1) Sample 8 (558-28-3, 118–120 cm), with N-MORB characteristics, compared to Sample 7 (558-27-3, 69–72 cm), an E-MORB, has essentially indistinguishable Nd (and Pb) isotopic composition; (2) Samples 8 and 13 (558-28-2, 84–92 cm) have N-MORB characteristics and identical Nd isotopic composition; however, they are different with respect to 206Pb/204Pb and 208Pb/204Pb composition; (3) Sample 8 versus Sample 12 (558-33-2, 22–25 cm) (N-MORB versus E-MORB) shows trace element heterogeneity accompanied by both Nd and Pb isotopic heterogeneity; (4) the 143Nd/144Nd isotopic compositions are positively correlated with 147Sm/144Nd (i.e., the data define an isochron/mixochron with an "age" of 224 ± 51 Ma).
son to Hole 558 basalts, the change in isotope characteristics between E-MORB and N-MORB are much more notable.

Holes 562, 563, and 564. All of the basalts analyzed are N-MORB. The initial $^{143}$Nd/$^{144}$Nd ratios are generally higher than those for the other N-MORB samples studied here, but they are more consistent with those suggested as characterizing the N-MORB source region (White and Hofmann, 1982).

**DISCUSSION**

**Isotopic Variability**

Figure 1 shows the variation in the Nd and Sr isotopic composition of Leg 82 basalts. The results include data from fresh glasses, leached whole rocks, and whole rocks. Almost all the samples are olivine-hypersthene normative tholeites and were selected as among the freshest samples available. No leached-unleached whole-rock pairs were tested; however, it appears that at least seven of the whole rocks have undergone significant alteration in Sr isotopic composition because of seawater/basalt interaction. Because of the uncertainty involved in using the whole-rock $^{87}$Sr/$^{86}$Sr ratios, only $^{87}$Sr/$^{86}$Sr data from the glasses and leached whole rocks are used to describe source characteristics.

The range of variation observed in Nd and Sr isotopic composition in Leg 82 basalts is quite large (spanning 5 ε Nd units). The pattern shown is consistent with previous studies on MORB; however, although all Leg 82 basalts would be considered MORB, they overlap in composition with the Azores Islands basalts. It is also somewhat surprising that in spite of having N-MORB with depleted to very depleted trace element abundances and

patterns (see Hertogen et al., this volume), many of the N-MORB recovered during Leg 82 have initial $^{143}$Nd/$^{144}$Nd less than that of the mean N-MORB reservoir (taken to be 0.51315 for zero-age basalts) (Jenner et al., in press).

Variation in $^{87}$Sr/$^{86}$Sr is correlated with $^{206}$Pb/$^{204}$Pb (Fig. 2) in a similar manner as observed in other MORB.

### Table 1. Isotope dilution, rare earth element abundances (in ppm).

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<th>Gd</th>
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**Table 2. Isotope dilution, rare earth element abundances (in ppm).**

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<th>Sm</th>
<th>Eu</th>
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<td>2.76</td>
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**Table 3. Shipboard chemical group; selected trace element and isotopic compositions for analyzed samples.**

<table>
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<tr>
<th>Hole</th>
<th>Sample number</th>
<th>Shipboard chemical group</th>
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Figure 1. Initial $^{143}\text{Nd}/^{144}\text{Nd}$ versus $^{87}\text{Sr}/^{86}\text{Sr}$ for Leg 82 basalts. Solid squares = glasses; half-filled squares = leached whole-rock samples; open squares = whole-rock powders (no leach). Comparison fields from published literature and unpublished data from Max-Planck-Institute. MORB = Mid-ocean ridge basalts.

Figure 2. Comparison of $^{87}\text{Sr}/^{86}\text{Sr}$ versus $^{206}\text{Pb}/^{204}\text{Pb}$ for Leg 82 basalts and MORB. Triangles = data from Cohen and O'Nions (1982) and Dupré and Allegre (1980); dashed line shows trend from Mid-Atlantic Ridge (MAR) basalts (zero age); solid squares = glasses, Leg 82; half-filled squares = leached whole rocks. (Pb isotopic data for Leg 82 from Dupré et al., this volume.)

Figure 3. $\text{La/Ce}$ versus $\text{Tb/Yb}$ for Leg 82 basalts. Open symbols = 17 Ma (Hole 558 = square; Hole 562 = triangle). Closed symbols = 35 Ma (Hole 556 = circles; Hole 558 = square; Hole 559 = triangle). Half-filled symbols = 35 Ma (Hole 563 = square; Hole 564 = triangle). Solid line is calculated mixing curve between samples from Sections 556-2-5 and 559-8-1 (see Hertogen et al., this volume).

Trace Element and Isotope Tests for Binary Mixing

As outlined in the introduction, Schilling and his coworkers (Schilling 1975, and White and Schilling, 1978) have developed a series of hypotheses relating variations in chemical characteristics to mixing between AMP/B and DLVL. As a first approximation, these authors have considered the mixing to be binary and also assumed that trace element ratios can be used to indicate the relative amount of components present. Accepting these conditions, one would hope to find a consistent (predictable) relationship between trace element and isotopic variation. To evaluate this possibility we have tested for mixing using trace elements and trace-element-isotope pairs. Because of our wider data base for initial Nd isotopic composition, only this isotopic system is considered here. Dupré et al. (this volume) discuss qualitatively the results for Pb isotopes versus trace elements.

Initial tests for a binary mixing relationship were made using variations in $\text{La/Ce}$ versus $\text{Tb/Yb}$. These elements and ratios were selected for the following reasons: (1) an extensive base of good data was available (see Hertogen et al., this volume); (2) both ratios were thought to be reasonably free of fractional crystallization effects and thus to reflect source characteristics; (3) the ratio pairs were sufficiently different so as to minimize the introduction of artificially good mixing fits given element similarities. The variation in $\text{La/Ce}$ versus $\text{Tb/Yb}$ is shown in Figure 3. The mixing curve illustrated was calculated for E-MORB and N-MORB samples analyzed by Hertogen et al. (this volume). The scatter about this curve is similar to that found in earlier studies by Schilling (1975) and White and Schilling (1978).

It is easier to evaluate the mixing hypothesis using the reciprocal plot (Langmuir et al., 1978) of $\text{La/Ce}$ versus $\text{Yb/Ce}$. In this plot a straight line should be formed with the data in the same relative order. For the data illustrated (Fig. 4), it is possible to calculate a linear least-squares regression line with $r^2 = 0.87$. This result essentially confirms the results of previous studies and would
ISOTOPIC AND TRACE ELEMENT COMPOSITION

Figure 4. Reciprocal plot of La/Ce versus Yb/Ce for Leg 82 basalts. Mixing line for samples from Sections 556-2-5 to 559-8-1 shown in solid line. Dashed lines are sketched-in possible mixing lines for variations observed at Sites 561 and 558. Symbols same as Figure 3.

model. However, it is informative to consider the data in more detail. Figure 4 shows a calculated mixing line for mixing between the same basalt pair used in Figure 3. Also illustrated are two other possible mixing lines (note: dashed lines drawn between samples from same sites). It is possible then that the trends shown in Figures 3 and 4 illustrate mixing between different sets of binary end-members.

The relationship between Nd isotopic composition and trace elements is illustrated in Figure 5. eNd(T) was chosen to illustrate the Nd isotopic variation so as to remove age differences, minimize effects caused by differential evolution of N-MORB and less-depleted reservoirs over the last 35 Ma, and facilitate easier discussion of the Nd isotopic composition. The Ce/Sm ratio is used to illustrate the trace element variation because of the poor trace element data base for the 10 samples jointly analyzed for Pb isotopic composition. The mixing curve shown was calculated for mixing between Samples 563-25-1 (an N-MORB from this study) and Faial 33 (an Azores Island basalt). Faial 33 was recommended by W. White, and if binary mixing is applicable, then it is a reasonable representation of the Azores Islands. However, this is not strictly true because isotopic heterogeneity in the Azores seems unlikely to be explained by simple binary mixing (Hawkesworth et al., 1979; Dupré et al., 1982; White, pers. comm., 1983).

In general, the correlation observed between eNd(T) and Ce/Sm is the expected correlation for a mixing relationship between AMP/B and DLVL (i.e., E-MORB are characterized by lower eNd(T) values than N-MORB). The correlation is not as well developed as that required by a binary mixing model. Two points are worth noting (see also Table 3 and Fig. 5): (1) the basalts most depleted in trace element (Hole 556) do not have the highest eNd(T) values; and (2) in Hole 558 it is possible to find N-MORB and E-MORB with identical (within error) Nd isotopic compositions (see Results, Hole 558). As in the trace element mixing plots, it is easy to visualize a set of possible mixing curves as illustrated in Figure 5.

To more carefully evaluate the relationship between Nd isotopic composition and trace element characteristics, and its significance for AMP/B-DLVL mixing, we have considered our data plus that from the literature for a possible mantle flow line between the FAMOUS site and Hole 558 (Leg 82). The Hole 558-FAMOUS flow line may be considered to be defined by or sampled by the following points: Hole 558 (Leg 82), 35 Ma; Hole 335 (Leg 37) 16 Ma; Hole 332 (Leg 37), 3 Ma; and FAMOUS, zero-age. Data sources are: this study; White, 1979; Schilling et al., 1977; Cohen and O'Nions, 1982; O'Nions et al., 1977.

Schilling et al. (1977) had suggested on the basis of REE data that the AMP/B contribution increased from 0% at Hole 335 to 30% at Hole 332 and to 45% in FAMOUS. Considered scatter was observed in the trace element characteristics at these sites (Schilling et al., 1977), but nonetheless a significant temporal variation seemed to exist. In contrast to the findings of Schilling et al. (1977), we would suggest the following (see Fig. 6):

1. Based on eNd(T), the plume component in Hole 335, Hole 332, and the FAMOUS site can be considered to be constant at (for this specific model) ~8%.

2. REE data are not a good indication of plume component and give a range of 7-20% plume component.

3. Hole 558 would have the highest plume component. Therefore, if this is due to an AMP/B influence, then there was a marked decrease in intensity between 35 and 16 Ma followed by a relative constancy of plume effect. The effect on Hole 558 is greater than that at Hole 556, although the latter would probably have been closer to the AMP/B source at 35 Ma.

4. Within Hole 558 there are basalts that appear to have trace elements and isotopic composition characteristics correlated, whereas others show marked trace element changes with no marked Nd isotopic variability.
The results presented here demonstrate that, in general, there is a correlation observed between isotopic and trace element characteristics in Leg 82 basalts. Those Leg 82 basalts classified as E-MORB have initial 143Nd/144Nd ratios < 0.51300, whereas, those basalts classified as N-MORB have initial 143Nd/144Nd ratios > 0.51307. A significant deviation from this general rule exists at Hole 558, where basalts defined as N-MORB by trace element characteristics have Nd isotopic characteristics of E-MORBs.

The degree of correlation observed between trace element and Nd isotopic composition is not as well developed as would be predicted by a unique binary AMP/B-DLVL mixing model. Rather it seems likely that there are more localized sets of end-members involved in mixing relationships. It is also clear that trace elements alone are not a reliable indicator of the proportion of components in mixing relationships. This study confirms earlier results, based on work on Leg 37 samples and FAMOUS samples (cf., Blanchard et al. 1976; White, 1979; and Langmuir et al., 1977), that trace elements can be separated from isotopic characteristics and reflect complex processes occurring just before or during mixing/partial melting of the source.

The nature of the processes that lead to significant trace element fractionation without accompanying isotopic changes are not well defined. One possibility discussed for the FAMOUS basalts is the concept of dynamic melting; this model is discussed fully in the paper by Langmuir et al. (1977) and provides an explanation for variations in REE patterns where the samples retain similar ratios of K/Ba, K/Rb, Rb/Sr, and Zr/Nb. This model would not appear to explain the severe trace element fractionation seen in Hole 558, where the changes in REE pattern are also accompanied by significant fractionation of Zr(Hf) and Nb(V). With the presently available limits on distribution coefficients for Zr and Nb, it would appear that no plausible simple partial melting model can explain the variation observed in Hole 558 (cf., Le Roex et al., 1981). Veining of the mantle within < 200 Ma before eruption of the Hole 558 basalts with a nephelinitic melt (cf., Wood, 1979b and Green, 1971) may be a partial solution, because mixes between these types of melts and DLVL sources can generate the right style of geochemical signature. The problem with Hole 558 is that it would appear to have had a reasonable chance of having E-MORB characteristics (eNd(T) = 6 to 7), thus the problem is to generate the N-MORB characteristics. One possibility is that at some stage (< 200 Ma before eruption) there was a local redistribution of elements caused by incipient partial melting, in which there was a volatile phase present in sufficient quantity to fractionate the trace elements in a more complex manner than that resulting from simple batch melting. In contrast to the "normal" veining episode, this melting/veining episode must generate a depleted trace element signature on top of an enriched isotope signature. The Pb isotopic variation observed in Hole 558 basalts suggests a possible redistribution event involving a fluid phase (melt + volatiles). If this recent fluid had a high µ value (µ = 238U/204Pb), it could help explain the relatively constant 207Pb/204Pb ratios and variable 208Pb/204Pb and 206Pb/204Pb ratios observed in Hole 558 basalts (Dupré et al., this volume).

The observations that simple binary mixing is not strictly applicable to Leg 82 basalts, and that trace elements are not a reliable indicator of mixing components necessitate a revision of the hypotheses proposed by Schilling and his co-workers (Schilling, 1975; and White and Schilling, 1978). The distribution of the observed heterogeneities also do not fit any simple AMP/B-DLVL mixing model. If initial 143Nd/144Nd ratios < 0.51305 are taken as evidence for a significant plume component in the source, then we would conclude that there was a plume input at the following: Sites 558 and 559 at 35 Ma; Sites 557 and 561 at 17 Ma; and the Azores at 0-10 Ma. The origin of this plume input at Sites 559 and 561 might be attributed to the Oceanographer plume of White and Schilling (1978). In fact, the data presented in this study may favor the plume explanation for the heterogeneity observed on the MAR in the vicinity of the Oceanographer Fracture Zone rather than the alternative explanations offered by these authors. However, the presence of a "plume" over the last 35 Ma in this area can only be inferred on geochemical grounds and lacks supporting bathymetric evidence. The Azores mantle plume may be the explanation for the characteristic geochemical and isotopic character ("plume input") at Site 557. However, it is difficult to attribute the plume input at Site 558 to the Azores mantle plume, as noted earlier in this paper. It is certainly possible to come up with AMP/B-DLVL explanations for the data. The plume hypothesis has become quite flexible with disconnected blobs, fluctuating intensity, and possibly "plume heterogeneity." At this stage the plume model becomes untestable and potentially unique to one area. We prefer to consider a model in which localized heterogeneity of

**SUMMARY AND CONCLUSIONS**

The observations that simple binary mixing is not strictly applicable to Leg 82 basalts, and that trace elements are not a reliable indicator of mixing components necessitate a revision of the hypotheses proposed by Schilling and his co-workers (Schilling, 1975; and White and Schilling, 1978). The distribution of the observed heterogeneities also do not fit any simple AMP/B-DLVL mixing model. If initial 143Nd/144Nd ratios < 0.51305 are taken as evidence for a significant plume component in the source, then we would conclude that there was a plume input at the following: Sites 558 and 559 at 35 Ma; Sites 557 and 561 at 17 Ma; and the Azores at 0-10 Ma. The origin of this plume input at Sites 559 and 561 might be attributed to the Oceanographer plume of White and Schilling (1978). In fact, the data presented in this study may favor the plume explanation for the heterogeneity observed on the MAR in the vicinity of the Oceanographer Fracture Zone rather than the alternative explanations offered by these authors. However, the presence of a "plume" over the last 35 Ma in this area can only be inferred on geochemical grounds and lacks supporting bathymetric evidence. The Azores mantle plume may be the explanation for the characteristic geochemical and isotopic character ("plume input") at Site 557. However, it is difficult to attribute the plume input at Site 558 to the Azores mantle plume, as noted earlier in this paper. It is certainly possible to come up with AMP/B-DLVL explanations for the data. The plume hypothesis has become quite flexible with disconnected blobs, fluctuating intensity, and possibly "plume heterogeneity." At this stage the plume model becomes untestable and potentially unique to one area. We prefer to consider a model in which localized heterogeneity of
not necessarily confirm or disprove the binary mixing both recent (e.g., Wood, 1979b; Green, 1971) and long-term (> 10^9 Ma) origin (e.g. Davies, 1981) plays a role. This type of heterogeneity may be superimposed upon regional heterogeneity generated by "plume mixing."

Resolution of the relative roles to be played by long-lived local heterogeneity, recent heterogeneity caused by complex partial melting, and regional plume-DLVL mixing is beyond the scope of this paper. We note only that the distribution and nature of the heterogeneities observed in the Azores area require a reevaluation of the hypotheses proposed by Schilling and his co-workers (Schilling, 1975; White and Schilling, 1978; Schilling et al., 1977).

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