

Hafnium isotopes in basalts from the southern Mid-Atlantic Ridge from 40°S to 55°S: Discovery and Shona plume-ridge interactions and the role of recycled sediments

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[1] Using high-precision multiple collector inductively coupled plasma-mass spectrometry (MC-ICP-MS), ¹⁷⁶Hf/¹⁷⁷Hf was measured in 64 mid-ocean ridge basalts (MORB) from the Mid-Atlantic Ridge (MAR) between $40^{\circ}S$ and $55^{\circ}S$ and in one basalt from the Discovery Tablemount. ϵ Hf of these basalts range from -2.8 to +16.6. The low end of this range exceeds that previously reported for MORB by 3 epsilon units and is only 5 epsilon units higher than the lowest reported ocean island basalt (OIB) value. Besides showing evidence for large-scale DUPAL-like pollution, the South Atlantic depleted upper mantle is also a region of localized plume-ridge interactions. ¹⁷⁶Hf/¹⁷⁷Hf from this study confirms the interaction of the Discovery and Shona plumes with the MAR as has been proposed previously on the basis of Pb, Sr, and Nd isotope ratios in these basalts. $\Delta \in Hf$ deviations from the global $\in Hf$ versus $\in Nd$ mantle array show that Shona anomaly basalts tend to fall below the mantle array while most Discovery and LOMU anomaly basalts fall above the array. ¹⁷⁶Hf/¹⁷⁷Hf versus ²⁰⁶Pb/²⁰⁴Pb for basalts from the Shona anomaly indicate that the ridge-centered heterogeneous Shona plume is mixing with the ambient upper mantle, which is locally fairly homogeneous. The Shona anomaly basalts can be modeled as three-component mixtures of ambient depleted upper mantle (ADM) with a recycled plume component consisting of old oceanic crust (ca. 1.5 Ga) and varying proportions of ancient sediment. The proportion of sediment present in the recycled Shona plume component increases stepwise from north to south along the MAR, but calculated sediment fractions are model dependent. In contrast, according to this three-component model, ancient sediment is probably not a significant component in the intraplate Discovery plume. Delaminated continental material, either from the subcontinental lithosphere or from the lower crust, may be present as the EM1 or LOMU-type component in the Discovery plume. In addition, both Shona and Discovery plumes likely contain a primitive rare gas component entrained from the lower mantle in order to account for the high ${}^{3}\text{He}/{}^{4}\text{He}$ and the solar neon isotope compositions of these MORB.

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1. Introduction

[2] The South Atlantic upper mantle is characterized by complex mixing processes involving heterogeneities which are compositionally highly varied. Previous geochemical and isotopic studies of fresh basalt glasses from the Mid-Atlantic Ridge (MAR) between 40°S and 55°S have revealed two distinct scales of mixing: 1) the localized interaction of the Shona and Discovery mantle plumes with the depleted upper mantle and 2) the ubiquitous largescale isotopic enrichment of the South Atlantic upper mantle relative to normal upper mantle.

[3] The ridge-centered Shona plume has a HIMU isotopic character and high water content, whereas the off-axis Discovery plume has an EM1 affinity and relatively low water content, suggesting distinct recycled components in these two plumes [Douglass et al., 1999; Leist et al., 2001] (Dixon et al., Hot spots, wetspots and dampspots: Evidence for efficient dehydration of recycled crust and sediments observed in plume influenced consult, submitted to Nature, 2002, hereinafter referred to as Dixon et al., submitted manuscript, 2002). Yet both plumes have high ³He/⁴He and solar-like neon isotope compositions which are usually attributed to entrainment of more pristine lower mantle by the rising mantle plumes [Moreira et al., 1995; Sarda et al., 2000]. A component, referred to as LOMU [Douglass et al., 1999; Kamenetsky et al., 2001], with an unusually strong continental signal and DUPAL-like rare-gas isotopic composition [Sarda et al., 2000] is also apparently present either as part of the Discovery plume or directly in the South Atlantic upper mantle.

[4] In addition to the plume-ridge interactions, the ambient LILE-depleted upper mantle shows some

unusual isotopic and geochemical characteristics whose origin remains uncertain, but may be related to large-scale pollution of the depleted upper mantle. Proposed pollution mechanisms include: 1) plume-head dispersion, 2) delamination of continental material from thermal erosion by plumes, 3) dispersal of continental material during the break-up of Gondwana, 4) redistribution of mantle material previously metasomatized by sediment melts or fluids over an ancient subduction zone, or 5) a transition zone from the mid-ocean ridge basalts (MORB) source that is normal Pacific/ North Atlantic-type to a source that is DUPALtype upper mantle [Hart, 1984; Hanan et al., 1986; Douglass et al., 1999; Fontignie and Schilling, 1996; Douglass and Schilling, 2000; Sarda et al., 2000; le Roux et al., 2002c].

[5] In order to further constrain mantle mixing processes and investigate the nature and origin of these various heterogeneities, we report Hf isotope systematics on the same fresh MORB glasses that were previously studied for major and trace elements, Pb, Sr and Nd isotopes, and rare gases [*Douglass et al.*, 1995, 1999; *Moreira et al.*, 1995; *Sarda et al.*, 2000; *le Roux et al.*, 2002a, 2002b, 2002c].

2. Analytical Techniques

[6] The 64 MORB included in this study (Figure 1) are all glass rims, except for six of the AG32 basalts, which are pillow interiors. In addition, one basalt from the Discovery Tablemount (BM 1954 152) is included in the study [*Kempe and Schilling*, 1974; *le Roux et al.*, 2002c]. This sample is a crushed whole rock powder.

[7] The two-column separation method of *Blichert-Toft et al.* [1997] was carried out at the





Figure 1. Map of dredge station locations. Inset shows a map of the South Atlantic region with the study area outlined in red and Tristan, Gough, Discovery, Shona and Bouvet plume locations marked with stars [*Douglass et al.*, 1999].

University of Rhode Island to isolate the Hf for isotope ratio measurement. Subsequently, ¹⁷⁶Hf/¹⁷⁷Hf was measured at the Ecole Normale Supérieure de Lyon using high precision magnetic sector multiple collector inductively coupled plasma mass spectrometry (MC-ICPMS, VG Plasma 54). The internal precision of the Plasma 54 for Hf isotope ratio measurement is better than 20 ppm and the reproducibility is better than 35 ppm. Standards were run between every second or third sample to monitor machine performance. During the isotope analyses (March 2000–May 2001), the JMC-475 Hf standard gave 176 Hf/ 177 Hf = 0.282160 with an external reproducibility of ±0.000010.

[8] Trace element concentrations reported here (Lu, Hf, Ba, and Nb) are from *Douglass* [2000]. These measurements were carried out on dissolved sam-

Table 1. Data for South	Atlantic Basalts											
Sample ID ^a	Latitude, $^{\circ}S$	Longitude, °W	Depth, m	$^{176}{\rm Hf}/^{177}{\rm Hf}^{\rm b}$	$\pm 2\sigma_m$	εHf°	Lu ^d	Hf^{d}	$^{176}\mathrm{Lu}/^{177}\mathrm{Hf}$	Ba^{d}	Nb^{d}	Ba/Nb
All 107-7 07-3g	40.44	16.75	2612	0.283181	9 ∓	14.5	0.494	1.88	0.0358	10	1.9	5.17
EW9309 45D-3g	40.59	16.77	3203	0.283104	± 16	11.7	0.368	0.96	0.0521	5	1.1	4.54
EW9309 44D-3g <i>FW</i> 0300 44D-3g	40.86	16.79	3067	0.283122 0.283118	8 + + 8 +	12.4 12.2	0.489	1.99	0.0336	13	2.2	5.90
АП 107-7 06-200	41.25	16.60	2394	0.283081		10.9	0.477	1.77	0.0368	<u>(1</u>	2.4	5.54
BM 1954 152	42.07	0.06	-	0.282812	£9 ∓	1.4	0.28	4.83	0.0079	372	27.7	13.43
EW9309 43D-1g	42.19	16.08	2925	0.283114	₩	12.1	0.512	2.06	0.0340	16	2.4	6.66
EW9309 43D-2g	42.19	16.08	2925	0.283128	十7	12.6	0.507	1.99	0.0347	15	2.3	6.72
AII 107-7 04-4g	42.92	16.37	2802	0.283165	±7	13.9	0.489	1.9	0.0343	24	3.4	7.03
EW9309 42D-1g	43.43	16.17	3449	0.282985	9∓	7.5	0.313	1.47	0.0291	87	7.6	11.46
EW9309 41D-1g	44.02	16.08	3522	0.283191	±8	14.8	0.493	1.87	0.0360	18	2.3	8.04
EW9309 40D-1g	44.41	15.91	3488	0.283242	±7	16.6	0.460	1.91	0.0330	24	2.6	9.24
EW9309 37D-1g	45.23	15.07	3534	0.283037	9∓	9.4	0.506	2.75	0.0252	96	8.7	11.06
EW9309 39D-1g	45.23	15.07	3534	0.283105	± 10	11.8	0.488	2.23	0.0298	25	3.2	7.78
EW9309 36D-1g	45.57	14.71	3114	0.283128	±7	12.6	0.507	2.24	0.0309	15	2.4	6.36
EW9309 34D-1g	45.85	14.19	3443	0.283028	6∓	9.1	0.494	1.57	0.0429	13	1.6	8.43
EW9309 33D-1g	45.99	14.08	3381	0.282954	7	6.4	0.450	3.02	0.0204	182	15.7	11.58
All 107-7 02-53g	46.21	13.64	2699	0.283128	十7	12.6	0.470	1.86	0.0345	21	2.7	7.85
EW9309 32D-1g	46.23	13.55	3108	0.283102	=6	11.7	0.522	1.90	0.0375	21	2.4	8.63
EW9309 32D-2g	46.23	13.55	3108	0.283036	9∓	9.3	0.462	2.19	0.0288	68	6.4	10.60
EW9309 31D-1g	46.39	13.45	2822	0.283024	#8	8.9	0.389	2.26	0.0235	113	9.2	12.23
EW9309 30D-1g	46.59	13.38	3153	0.283095	#8	11.4	0.420	1.77	0.0323	29	2.9	9.88
EW9309 28D-1g	46.90	13.45	3417	0.283178	±7	14.4	0.419	1.52	0.0377	14	1.4	9.94
EW9309 27D-2g	47.11	13.40	3081	0.283080	#8	10.9	0.399	1.26	0.0431	10	1.2	8.40
EW9309 25D-1g	47.35	10.32	2032	0.282700	十7	-2.5	0.217	1.58	0.0187	69	5.3	13.03
EW9309 25D-5g	47.35	10.32	2032	0.282694	±7	-2.8	0.221	1.55	0.0195	99	5.0	13.11
EW9309 26D-1g	47.35	13.40	3857	0.283114	#8	12.1	0.644	3.01	0.0292	20	2.4	8.15
EW9309 26D-2g	47.35	13.40	3857	0.283159	±7	13.7	0.635	3.02	0.0287	20	2.5	8.02
EW9309 02D-1g	47.55	10.18	2494	0.282891	9∓	4.2	0.389	3.64	0.0146	199	15.0	13.25
EW9309 02D-3g	47.55	10.18	2494	0.282913	#8	5.0	0.343	2.51	0.0187	100	7.8	12.83
EW9309 03D-3g	47.80	10.15	2549	0.283038	7	9.4	0.447	2.33	0.0262	77	6.0	12.71
EW9309 04D-2g	47.97	10.08	2895	0.282965	± 10	6.8	0.472	3.54	0.0182	145	11.5	12.65
EW9309 04D-3g	47.97	10.08	2895	0.282994	十7	7.9	0.422	2.53	0.0228	105	8.3	12.59
EW9309 05D-5g	48.24	9.99	3453	0.282909	#8	4.8	0.399	3.61	0.0151	176	13.6	12.97
EW9309 06D-1g	48.55	10.17	3267	0.282811	±11	1.4	0.311	2.27	0.0187	161	10.6	15.19
EW9309 07D-1g	48.76	10.07	3218	0.282728	± 10	-1.6	0.273	3.76	0.0099	338	19.8	17.09
EW9309 07D-6g	48.76	10.07	3218	0.282743	± 10	-1.0	0.273	3.16	0.0118	331	19.8	16.71
EW9309 07D-2g	48.76	10.07	3218	0.282990	#8	7.7	0.721	4.72	0.0209	166	12.7	13.14
EW9309 08D-1g	48.96	9.97	3894	0.282979	±7	7.3	0.513	3.10	0.0226	102	7.3	13.98
EW9309 09D-1g	49.15	9.91	3892	0.282908	#8	4.8	0.517	2.98	0.0237	104	7.9	13.23
EW9309 09D-3g	49.15	9.91	3892	0.282971	#8	7.0	0.456	1.02	0.0610	38	2.7	13.82
EW9309 10D-5g	49.25	8.14	3860	0.283158	#8	13.7	0.531	2.43	0.0297	11	1.9	5.69
EW9309 10D-3g	49.25	8.14	3860	0.283187	#8	14.7	0.499	2.26	0.0302	11	1.6	7.14



Table 1. (continued)												
Sample ID ^a	Latitude, $^{\circ}$ S	Longitude, °W	Depth, m	$^{176}\mathrm{Hf}^{177}\mathrm{Hf}^{\mathrm{b}}$	$\pm 2\sigma_m$	${}^{\mathrm{eHf}^{\mathrm{c}}}$	Lu ^d	hf ^d	$^{176}{\rm Lu}/^{177}{\rm Hf}$	Ba^{d}	Nb^{d}	Ba/Nb
EW9309 11D-1g	49.44	7.97	3868	0.283140	τŦ	13.0	0.500	2.04	0.0335	6	1.1	6.50
EW9309 12D-3g	49.76	8.03	3874	0.283152	L	13.4	0.399	1.21	0.0452	5	1.0	4.99
EW9309 13D-1g	49.99	7.88	3555	0.283151	± 11	13.4	0.395	1.37	0.0394	9	1.0	6.31
EW9309 14D-1g	50.27	7.06	3347	0.283133	$^{\mp 0}$	12.8	0.545	2.26	0.0320	13	2.6	5.08
EW9309 14D-4g	50.27	7.06	3347	0.283051	± 14	9.9	0.486	2.56	0.0260	59	7.7	7.67
EW9309 24D-3g	50.46	6.53	3183	0.283086	$^{\mp 0}$	11.1	0.575	2.61	0.0301	31	5.7	5.54
EW9309 15D-1g	50.58	6.43	2980	0.283132	7	12.7	0.530	2.24	0.0324	27	4.6	6.00
EW9309 15D-1g(repl.)				0.283116	± 8	12.2						
EW9309 17D-1g	50.76	6.34	2943	0.283047	± 10	9.7	0.480	2.25	0.0291	35	5.1	6.88
EW9309 18D-1g	51.05	6.20	1991	0.283024	#8	8.9	0.345	1.67	0.0281	49	5.4	8.95
EW9309 19D-1g	51.06	6.16	1743	0.282962	6∓	6.7	0.355	1.85	0.0262	21	3.1	6.72
EW9309 19D-1g(repl.)				0.282956	6^{\pm}	6.5						
EW9309 19D-3g	51.06	6.16	1743	0.283052	6∓	9.6	0.380	1.70	0.0305	52	6.6	7.83
AG32-10-1	51.28	5.85	2025	0.282974	8∓	7.1	0.436	2.45	0.0243	71	8.2	8.68
EW9309 20D-1g	51.43	5.78	1719	0.282980	L∓	7.4	0.461	2.90	0.0218	80	8.9	9.00
AG32-9-1	51.62	5.60	1675	0.282997	L	8.0	0.475	2.73	0.0237	83	9.0	9.31
EW9309 21D-1g	51.82	5.50	2025	0.282958	6∓	6.6	0.463	2.71	0.0233	86	9.3	9.25
EW9309 23D-1g	52.16	5.34	2609	0.283027	十7	9.0	0.772	4.57	0.0231	61	8.1	7.53
AG32-7-1	52.27	4.83	3400	0.282997	L	8.0	0.520	2.60	0.0273	54	5.9	9.21
EW9309 22D-3g	52.46	4.57	3059	0.283008	± 10	8.3	0.891	6.00	0.0203	120	14.7	8.20
AG32-6-2g	52.65	4.38	2325	0.283042	±7	9.5	0.486	2.15	0.0308	47	5.2	9.19
AG32 5-1	52.98	4.33	1700	0.282999	± 13	8.0	0.375	2.18	0.0235	93	12.4	7.50
AG32-4-111	54.03	2.30	2500	0.283097	6 ∓	11.5	0.702	3.47	0.0276	44	6.0	7.32
AG32-3-99	54.12	1.97	2250	0.283098	#8	11.5	0.592	2.54	0.0318	28	4.2	6.69
AG32-2-3g	54.37	1.20	2250	0.283067	± 10	10.4	0.549	2.78	0.0269	56	6.8	8.25
				Standards								
BCR-1							0.510	5.01		681	12.7	
rel. standard deviation, $n = 14$							1.8%	1.8%		0.9%	1.5%	
BHVO-1							0.268	4.35		132	19.0	
rel. standard deviation, $n = 14$							1.4%	1.6%		1.6%	0.4%	
ENO26 10D-3g												
rel. standard deviation, $n = 42$							1.7%	2.5%		2.8%	1.8%	
^a Samples are glass rims, unless the ^b For all analyses, JMC-475 Hf star	e Sample ID doe indard gave ¹⁷⁶ H	s not end in "g". Re f^{177} Hf = 0.282160 ±	plicates are denc 0.000010. ¹⁷⁶ Hf	ted as <i>repl.</i> ⁄ ¹⁷⁷ Hf was normaliz	ced for mas	s fractiona	tion relativ	e to ¹⁷⁹ Hi	f'^{177} Hf = 0.7325.			
`EHf is calculated using (```Ht/```]	Hf) _{Bulk} silicate Far	$h_{\rm h} = 0.282772.$										

^d Element concentrations are given in ppm and are from *Douglass* [2000], except for BM 1954 152 element concentrations, which are from *le Roux et al.* [2002c]. ^eURI in-house MORB standard from the Mohns Ridge [*Schilling et al.*, 1999].

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ples using a Finnegan MAT High Resolution ICP-MS at the University of Rhode Island. The method is adapted from that of *Eggins et al.* [1997] using internal and external standardization. Values for BCR-1 and BHVO-1 obtained during the period of measurements of these samples are listed in Table 1 with reproducibility in percent standard deviation. Reproducibility for the University of Rhode Island in-house standard is also listed in Table 1. Numerous other trace element concentrations in these basalts have been measured by LA-ICP-MS. These are reported and discussed in-depth elsewhere [*le Roux et al.*, 2002a, 2002b, 2002c].

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3. Results

[9] ¹⁷⁶Hf/¹⁷⁷Hf results with 2 sigma errors are listed in Table 1. Replicate analyses performed on three of the South Atlantic MORB are in good agreement with one another. Measured ε Hf in the South Atlantic data set range from -2.8 (¹⁷⁶Hf/¹⁷⁷Hf = 0.282694) to +16.6 (¹⁷⁶Hf/¹⁷⁷Hf = 0.283242). The low end of this ε Hf range exceeds the range so far reported for MORB [*Salters*, 1996; *Chauvel and Blichert-Toft*, 2001] by 3 epsilon units. This range covers about 60% of the range documented for ocean island basalt (OIB) which fall approximately between -8 and +24 (*Salters* [1996, Figure 1], corrected to JMC-475 = 0.282160).

3.1. Isotope Profiles

[10] The Discovery plume is located about 425 km east of the ridge at 44.45°S, 6.45°W (Figure 1) [Douglass et al., 1995]. Previously collected Pb, Sr and Nd isotope data indicate that MORB influenced by the Discovery plume lie along the MAR between 40°S and 48.2°S. Since the Discovery plume is EMtype, the Discovery anomaly is most apparent in the ⁸⁷Sr/⁸⁶Sr profile (Figure 2c). The mildly HIMU or C-type Shona plume is ridge centered at 51.5°S (Figure 1) [Small, 1995; Douglass et al., 1995] and influences MORB compositions from 49.2°S-52.65°S. This HIMU influence is most clearly seen on the ²⁰⁶Pb/²⁰⁴Pb profile (Figure 2d). Further south, the MORB may also be influenced by the mildly HIMU or C-type Bouvet plume (Figure 1) [le Roex et al., 1987; Douglass et al., 1999].

[11] As expected, the ¹⁷⁶Hf/¹⁷⁷Hf profile along the MAR between 40°S and 55°S resembles that of ¹⁴³Nd/¹⁴⁴Nd (Figures 2a and 2b). In regions of plume–ridge interaction, both ¹⁷⁶Hf/¹⁷⁷Hf and ¹⁴³Nd/¹⁴⁴Nd are lower (less radiogenic) than in the surrounding NMORB population. This indicates that these plume-influenced basalts arise from a source that shows time-integrated enrichment in incompatible elements (i.e., low Sm/Nd and low Lu/Hf) which is typical of mantle plumes.

[12] Basalts from 40°S to 45.2°S are largely NMORB with relatively high (non plume-like) ¹⁷⁶Hf/¹⁷⁷Hf (Figure 2a). ε Hf in these basalts ranges from +10.9 to +16.6 which is at the low end of the range reported for North Atlantic and Pacific MORB (ε Hf = +9 to +20) [*Chauvel and Blichert-Toft*, 2001]. However, one of the two EMORB from this region (EW9309 42D-1g) does have moderately low (moderately plume-like) ε Hf of +7.5. This is probably due to mixing of the upper mantle with a small amount of the nearby Gough plume (Figure 1) as suggested by *Douglass et al.* [1999] to account for elevated Sr and Pb isotope ratios.

[13] The Hf isotope data collected in this study support the model of Discovery and Shona plume-ridge interactions along the MAR between 45°S and 53°S deduced previously from Pb-Sr-Nd isotope space [Douglass et al., 1995, 1999]. The ¹⁷⁶Hf/¹⁷⁷Hf profile (Figure 2a) shows two broad regions of low ¹⁷⁶Hf/¹⁷⁷Hf due to the influences of the Discovery plume from 45.2°S to 48.2° S (ε Hf = -2.8 to +14.4) and the Shona plume from 49.2° S to 52.6° S (ε Hf = +6.6 to +14.7). In addition to these two broad regions of low ¹⁷⁶Hf/¹⁷⁷Hf, there is one narrow region of low ¹⁷⁶Hf/¹⁷⁷Hf just south of the main Discovery anomaly from 48.5° S to 49.1° S (ϵ Hf = -1.6 to +7.7). This region has been called the LOMU segment and has been attributed to delaminated subcontinental Gondwanan lithosphere which may be present as passive heterogeneities in the upper mantle [Douglass et al., 1999]. According to this model, even though these heterogeneities may be widely dispersed throughout the region, since they are refractory, they are only melted and incorporated in MORB when plumes are available to provide the necessary excess heat [Douglass et



Figure 2. Isotope profiles along the southern MAR. Horizontal dashed–dotted lines represent the baseline isotope values for the ambient South Atlantic depleted upper mantle. Grey dashed lines show where MORB are offset by fracture zones. These are, from north to south: Agulhas FZ, 48.5° Offset, 49° FZ and 50° FZ. ε Hf errors in (a) are smaller than symbols.

al., 1999; *Mahoney et al.*, 1992]. However, if the Discovery plume is heterogeneous, the LOMU ridge segment may simply be part of the Discovery plume anomaly.

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[14] It is interesting to note that ¹⁷⁶Hf/¹⁷⁷Hf of the two basalts from dredge station EW9309 25D along the MAR are even lower (more plume-like)

than ¹⁷⁶Hf/¹⁷⁷Hf in the basalt from the Discovery Tablemount. The Discovery Tablemount (42°S, 0°) is northeast of the present location of the Discovery plume [*Douglass et al.*, 1995]. The Discovery Tablemount basalt is 25 Ma old and was probably formed by intraplate Discovery plume volcanism [*Kempe and Schilling*, 1974; *Douglass et al.*, 1999; *le Roux et al.*, 2002c].



Figure 3. ε Hf $-^{206}$ Pb/²⁰⁴Pb. Ellipses outline the Discovery, LOMU and Shona ridge anomalies. Inset shows data contours relative to mantle end-members. C composition is based on *Hanan and Graham* [1996] with ε Hf calculated from their ε Nd and the equation for the mantle array: ε Hf = 1.4 ε Nd + 2.8 (based on published and unpublished Hf and Nd isotope data from J. Blichert-Toff for 1500 MORB and OIB samples of global distribution). ²⁰⁶Pb/²⁰⁴Pb for DMM, EM1, EM2, and HIMU are from *Hart et al.* [1992]. ε Hf for EM1, EM2 and HIMU are from *Salters and White* [1998]. ε Hf for DMM is calculated from DMM ε Nd [*Hart et al.*, 1992] and the equation of the mantle array. LOMU composition is from basalt S18-60/1 [*Kamenetsky et al.*, 2001] with ε Hf calculated from ε Nd and the equation of the mantle array.

3.2. Isotope Space

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[15] The three mixing trends of Discovery ($45.2^{\circ}S-48.2^{\circ}S$), LOMU ($48.5^{\circ}S-49.1^{\circ}S$) and Shona ($49.2^{\circ}S-52.6^{\circ}S$) material with upper mantle material are revealed best in ϵ Hf $-^{206}$ Pb/ 204 Pb space (Figure 3). The three mixing trends converge on the high ϵ Hf (non plume-like) end but they diverge toward different "enriched" (plume-like) end-members. The Shona anomaly basalts trend toward ϵ Hf = +6.7, 206 Pb/ 204 Pb = 18.9 and 208 Pb/ 204 Pb = 38.8 (EW9309 19D-1g). This points toward a mildly HIMU or C-type end-member. The Discovery anomaly basalts trend toward ϵ Hf = -2.8, 206 Pb/ 204 Pb = 18.2 and 208 Pb/ 204 Pb = 38.7 (EW9309 25D-5g). The LOMU segment basalts

trend toward ε Hf = -1.6, ²⁰⁶Pb/²⁰⁴Pb = 17.8 and ²⁰⁸Pb/²⁰⁴Pb = 38.2 (EW9309 7D-1g). Basalts from both the Discovery and LOMU segments trend toward EM-type end-members. While basalts from the Discovery and LOMU segments overlap in terms of ε Hf values, for a given ε Hf the LOMU basalts have lower ²⁰⁶Pb/²⁰⁴Pb and ²⁰⁸Pb/²⁰⁴Pb than the Discovery basalts.

[16] The Discovery, LOMU and Shona mixing vectors are difficult to distinguish in ε Hf– ε Nd space (Figure 4) since these basalts all fall quite close to the Hf–Nd mantle array [*Patchett et al.*, 1984; *Blichert-Toft and Albarède*, 1997; *Vervoort et al.*, 1999]. $\Delta \varepsilon$ Hf is a measure of the deviation of ε Hf from this array at a given ε Nd, where $\Delta \varepsilon$ Hf =



Figure 4. ε Hf– ε Nd. Dashed line is the mantle array where ε Hf = 1.4 ε Nd + 2.8 (based on published and unpublished Hf and Nd isotope data from J. Blichert-Toft for 1500 MORB and OIB samples of global distribution). Inset shows Discovery, LOMU and Shona ridge anomalies (grouped by ellipses) relative to mantle end-members. ε Nd of C is from *Hanan and Graham* [1996]. ε Nd of DMM, EM1, EM2 and HIMU are from *Hart et al.* [1992]. ε Nd of LOMU is from basalt S18-60/1 [*Kamenetsky et al.*, 2001]. End-member ε Hf sources are listed in the caption of Figure 3. Global OIB and MORB data are from *Chauvel and Blichert-Toft* [2001], *Salters and White* [1998], *Salters* [1996], and *Salters and Hart* [1991].

 $\epsilon H f_{measured} - \epsilon H f_{mantle array}$ [Johnson and Beard, 1993] and the best fit to the mantle array is ε Hf = $1.4 \in Nd + 2.8$ (based on published and unpublished Hf and Nd isotope data from J. Blichert-Toft for 1500 MORB and OIB samples of global distribution). MORB from this study have $\Delta \in Hf$ ranging from -2.6 to +3.4 (Figure 5a). Discovery and LOMU MORB tend to fall above the mantle array with positive $\Delta \in Hf$. In some cases, positive $\Delta \in Hf$ has been interpreted as evidence for the presence of ancient pelagic sediments in the source of basalts since pelagic sediments have relatively high Lu/Hf for a given Sm/Nd [Patchett et al., 1984]. In contrast to Discovery and LOMU MORB, most Shona anomaly basalts fall below the mantle array and thus have negative $\Delta \in Hf$ which is also a characteristic of HIMU basalts from St. Helena, Tubuaii and Rurutu [Salters and White, 1998].

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[17] The short 49°S fracture zone separates the LOMU anomaly from the Shona anomaly. The $\Delta \varepsilon Hf$ profile indicates that there may be some mixing of LOMU and Shona material across this fracture zone. Basalt EW9309 10D-5g lies just south of this fracture zone within the Shona anomaly. Yet its has $\Delta \varepsilon Hf = +3.06$ demonstrating that it falls significantly above the mantle array. Basalt EW9309 9D-3g which lies just north of the fracture zone within the LOMU anomaly has $\Delta \varepsilon Hf = -2.08$ and thus falls significantly below the array. In addition, this basalt has unusually high Lu/Hf (Figure 5b and Figure 6).

[18] Figure 5a shows an overall decrease in $\Delta \epsilon$ Hf from 40°S to 55°S. This may be simply because the EM-type plumes (Tristan, Gough and Discovery) are neighbored to the south by mildly HIMU Shona and Bouvet plumes. However, it may also be part



Figure 5. $\Delta \varepsilon \text{Hf}$, ¹⁷⁶Lu/¹⁷⁷Hf and R/R_A profiles along the southern MAR. In (a) $\Delta \varepsilon \text{Hf} = \varepsilon \text{Hf}_{\text{measured}} - (1.4\varepsilon \text{Nd}_{\text{measured}} + 2.8)$. EW9309 40D-1g lies north of the Discovery anomaly. Its high $\Delta \varepsilon \text{Hf}$ may reflect the influence of material from the Gough plume [*Douglass et al.*, 1999]. EW9309 13D-1g is an outlier; its low $\Delta \varepsilon \text{Hf}$ may reflect analytical uncertainty in the εNd measurement (see Figure 3b where this MORB has significantly higher εNd than the surrounding MORB). Data in (c) is from *Moreira et al.* [1995] and *Sarda et al.* [2000]. R/R_A is ³He/⁴He normalized to the atmospheric ratio, R_A, which is 1.384×10^{-6} . Dashed–dotted line shows NMORB baseline [*Sarda et al.*, 2000]. ³He/⁴He peaks for the Shona and Discovery plumes are much narrower than the peaks on Pb, Sr, Nd and Hf isotope profiles.

of a longer gradient along the entire Mid-Atlantic Ridge from 80°N to 55°S (M. Andres et al., manuscript in preparation, 2002).

3.3. Parent-Daughter Systematics

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[19] ¹⁷⁶Lu/¹⁷⁷Hf of the South Atlantic MORB range from 0.0079 to 0.061 (Figure 5b). The parent–daughter ratios generally correlate positively with ¹⁷⁶Hf/¹⁷⁷Hf (Figure 6). However, due to complex melting conditions of a few samples

(e.g., incomplete pooling of fractional melts and two stage melting such as for 25D [*le Roux et al.*, 2002b; *Douglass*, 2000] (Dixon et al., submitted manuscript, 2002), the relationship between ¹⁷⁶Lu/¹⁷⁷Hf and ¹⁷⁶Hf/¹⁷⁷Hf is somewhat scattered at the high end of the Lu/Hf range (Figure 6). This scatter is also present in the inverse correlation of ¹⁷⁶Lu/¹⁷⁷Hf with La/Sm (not shown). EW9309 9D-3g has particularly high Lu/Hf which is probably due to some unusual fractionation during its gen-



Figure 6. ε Hf $-^{176}$ Lu $/^{177}$ Hf. In general, ε Hf and 176 Lu $/^{177}$ Hf in MORB from this study are positively correlated. Basalts, which likely experienced recent enrichment in Lu/Hf due to unusual melting events [*le Roux et al.*, 2002b], are labeled.

eration by partial melting. Detailed analysis of the trace element systematics and their implications for partial melting and fractional crystallization processes in generating these basalts is beyond the scope of this paper. This is discussed by *le Roux et al.* [2002a, 2002b, 2002c].

4. South Atlantic Upper Mantle Composition

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[20] Each of the three mixing trends identified in ε Hf⁻²⁰⁶Pb^{/204}Pb space (Figure 3) is a mixture between an enriched source (i.e., the Shona plume, Discovery plume or LOMU rafters) and a depleted source (i.e., the ambient depleted upper mantle; ADM). *Douglass et al.* [1999] found that the upper mantle with which the Discovery and Shona plumes are mixing is not the normal North Atlantic/Pacific-type upper mantle. They defined the likely isotopic composition of this ambient

upper mantle from the intersection of the Discovery and Shona mixing trends on isotope–isotope plots. They found that the ambient upper mantle is significantly more radiogenic relative to the North Atlantic and Pacific in terms of Pb and Sr isotopes and unradiogenic in terms of Nd isotopes.

[21] The data collected in the present study allow characterization of the ¹⁷⁶Hf/¹⁷⁷Hf of the ambient upper mantle. Following the method of *Douglass et al.* [1999], the Shona and Discovery trends intersect at roughly ¹⁷⁶Hf/¹⁷⁷Hf = 0.28324 (ε Hf = +16.6) in ¹⁷⁶Hf/¹⁷⁷Hf-²⁰⁶Pb/²⁰⁴Pb isotope space. By comparison, the average ε Hf of 26 North Atlantic NMORB (La/Sm_n < 0.65) from the URI MORB collection is +20 (JGS and JBT, unpublished data). Thus, as one would expect based on Nd isotopes, the ambient depleted upper mantle is relatively unradiogenic in comparison to the North Atlantic in terms of Hf isotopes. The isotopic composition of this ambient upper mantle is com-



Table 2.Ambient Versus Normal Upper MantleCompositions

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	Ambient Upper Mantle ^a	Normal Upper Mantle ^b
²⁰⁶ Pb/ ²⁰⁴ Pb	18	18
²⁰⁷ Pb/ ²⁰⁴ Pb	15.5	15.43
²⁰⁸ Pb/ ²⁰⁴ Pb	37.8	37.3
⁸⁷ Sr/ ⁸⁶ Sr	0.7025	0.7022
143 Nd/ 144 Nd (ϵ Nd)	0.51312 (+9.4)	0.5133 (+12.9)
¹⁷⁶ Hf/ ¹⁷⁷ Hf (ɛHf)	0.283240 (+16.6)	0.283358 (+20.8)

^a Ambient upper mantle is from *Douglass et al.* [1999], except for 176 Hf/ 177 Hf, which is from this study.

^bNormal upper mantle is from *Hart et al.* [1992], except for ¹⁷⁶Hf¹⁷⁷Hf, which is calculated from ¹⁴³Nd/¹⁴⁴Nd and the equation of the Hf–Nd mantle array: ε Hf = 1.4 ε Nd + 2.8 (based on published and unpublished Hf and Nd isotope data from J. Blichert-Toft for 1500 MORB and OIB samples of global distribution).

pared to the normal depleted upper mantle composition of *Hart et al.* [1992] in Table 2.

[22] The ambient depleted upper mantle with which the Shona and Discovery plumes are mixing also has relatively high Ba/Nb. Ba/Nb for typical North Atlantic/Pacific MORB (and by inference the "normal" upper mantle) is ~ 2.8 [*Hofmann*, 1988]. MORB from the Discovery and Shona anomalies, however, converge to an ambient upper mantle composition with Ba/Nb ~ 5 [*Douglass*, 2000; *le Roux et al.*, 2002c].

[23] The cause of this isotopic enrichment and high Ba/Nb remains controversial. Rehkämper and Hofmann [1997] modeled the unusual characteristics of the Indian Ocean MORB source as a mixture of normal upper mantle with recycled ancient sediments and oceanic crust. The same mixing model can also be invoked for the unusual South Atlantic MORB source. However, in both cases, the model is quite unconstrained and a pollution mechanism remains speculative. One possibility is that material from the melt-residue of a plume-head [Fontignie and Schilling, 1996] infiltrated the upper mantle prior to the breakup of Gondwana. Evidence in favor of this comes from plate reconstructions which show that South Africa was lying over the Discovery and Shona plumes prior to the opening of the Atlantic about 130 Ma ago [le Roex, 1986; Hartnady and le Roex, 1985]. Another possibility suggested by le Roux et al. [2002c] is that the South Atlantic upper mantle pollution was caused by subduction

zone fluids and melts. Their arguments are based largely on the unusually low Nb concentration and high Ba/Nb ratio in South Atlantic NMORB. Their view is supported by evidence of a long Mesozoic subduction zone in the southwest coast of Gondwana. This model is appealing from a physical sense since it allows for easy infiltration of pollutant into the mantle.

[24] *Douglass and Schilling* [2000] suggested that the upper mantle beneath the Indian and South Atlantic Oceans was polluted by delamination of continental material during the breakup of Gondwana. This material could be either subcontinental lithosphere [*Douglass and Schilling*, 2000] or lower crustal mafic granulites [*Kamenetsky et al.*, 2001].

[25] he new South Atlantic ε Hf data alone cannot be used to constrain a pollution model for the South Atlantic. In fact, ongoing work along the entire MAR indicates that the low 176Hf/177Hf isotope signal is not merely restricted to the southern South Atlantic. Rather it is part of a remarkable large scale EHf gradient, which extends along the entire MAR from 80°N to 55°S. The cause of this isotope gradient will be explored elsewhere (M. Andres et al., manuscript in preparation, 2002). It is worth noting that while the ε Hf decreases in NMORB along the entire MAR, Ba/Nb does not (JGS, unpublished data). This indicates that there may be different processes affecting the upper mantle composition and that they act over different wavelengths.

5. Shona Ridge Anomaly

5.1. Evidence for a Heterogeneous Plume

[26] In isotope space, the Shona anomaly $(49.25^{\circ}S - 52.65^{\circ}S)$ is quite broad. ¹⁷⁶Hf/¹⁷⁷Hf for these basalts reveal systematic structure within the Shona anomaly that had not been recognized previously based only on Pb–Sr–Nd isotope space. This structure indicates that the Shona anomaly cannot be caused by simple binary mixing of a homogeneous plume with a homogeneous ambient upper mantle. At least three mixing end-members are needed to produce the observed isotopic structure of the Shona anom-





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Figure 7. Plume-upper mantle mixing cartoons viewed along the ridge axis. (a): homogeneous plume mixing with upper mantle containing embedded passive heterogeneities. MORB are produced by three-component mixing of plume, ambient upper mantle and embedded heterogeneities whose density increases to the south (right). (b): zoned heterogeneous plume mixing with a homogeneous ambient upper mantle. MORB are produced by binary mixing of sections of the heterogeneous plume with the uniform ambient upper mantle.

aly. Two different physical models can be used to describe mixing of these three end-members.

5.1.1. Model 1: Homogeneous Plume

[27] In the first model (Figure 7a), a homogeneous plume is mixing with a heterogeneous upper mantle. In this case, the ambient upper mantle has passive heterogeneities embedded in it. The plume, ambient mantle and passive heterogeneities mix in varying proportions to produce Shona anomaly MORB.

5.1.2. Model 2: Heterogeneous Plume

[28] In a second possible model, the local upper mantle is homogeneous but the upwelling Shona plume is heterogeneous (Figure 7b). In this case, the mixing of the three end-members is sequential binary mixing. In Stage 1, two end-members mix to produce the heterogeneous Shona plume. Then in Stage 2 this heterogeneous plume mixes with the homogeneous ambient mantle to produce the Shona anomaly basalts.

[29] The isotopic structure within the Shona anomaly basalts is evident on plots of 206 Pb/ 204 Pb $^{-176}$ Hf/ 177 Hf and 206 Pb/ 204 Pb $^{-Ba/Nb}$ (Figures 8a and 8b). Basalts from the Shona anomaly fall along distinct mixing vectors. The basalts within each of these geochemical groups are also distinct spatially. The Shona anomaly can be subdivided into 4 segments along the MAR (Figure 1): *segment 1*—(49.25°S– 49.99°S), *segment 2*—(50.27°S–51.06°S), *segment 3*—(51.05°S–52.16°S) and *segment 4*—(52.27°S– 52.65°S). However, there is some overlap at the ends of the segments (e.g., basalt EW9309 18D-1g lies within the southern end of *segment 2* but is geochemically like the basalts immediately to the south in *segment 3*).

[30] While Ba/Nb $-^{206}$ Pb/ 204 Pb (Figure 8b) clearly shows that *segments 1* and 2 fall on different mixing trends, in isotope–isotope space (Figure 8a) these two segments appear to lie along the same trend. This may indicate a recent decoupling of trace elements and isotopes as has been suggested by *Douglass et al.* [1999] or it may reflect the presence of some Discovery/LOMU material in these basalts.

[31] The segmentation of the Shona anomaly is also evident from the three mixing vectors in plots of ${}^{206}\text{Pb}/{}^{204}\text{Pb}-\varepsilon\text{Nd}$, ${}^{206}\text{Pb}/{}^{204}\text{Pb}-{}^{87}\text{Sr}/{}^{86}\text{Sr}$ and ${}^{206}\text{Pb}/{}^{204}\text{Pb}-{}^{208}\text{Pb}/{}^{204}\text{Pb}$ (Figures 9a, 9b, and 9c). It is difficult to discern separate mixing trends in ${}^{206}\text{Pb}/{}^{204}\text{Pb}-{}^{207}\text{Pb}/{}^{204}\text{Pb}$ space, possibly because of the lower precision of ${}^{207}\text{Pb}/{}^{204}\text{Pb}$ measurements (Figure 9d). The depleted ends of these mixing vectors converge to the ambient upper mantle composition discussed in section 5. The mixing vectors trend toward different enriched compositions. Since binary mixing of two homogeneous end-members must form a single mixing line in Pb isotope– isotope space, basalts within the Shona anomaly must arise from a more complicated mixing process.

5.2. Mixing Models: Possible Role of Sediments in the Shona Plume

[32] Subducted altered oceanic crust has been identified as a possible source of HIMU plumes

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Figure 8. ε Hf and Ba/Nb versus 206 Pb/ 204 Pb for Shona anomaly basalts. Arrows show segment-confined isotope trends converging to the ambient upper mantle (green circle). Segment 1 basalts lie along same trend as segment 2 basalts in (a) but not in (b). This indicates a recent decoupling of isotope ratios and trace element concentrations, possibly due to fractional melting of the Shona plume as it flows along the MAR. Open circle represents basalt EW9309 14D-4g. While this basalt lies within segment 1 along the MAR, it has a very unusual isotopic composition and is not considered further here. DM is shown for comparison: 206 Pb/ 204 Pb is from *Hart et al.* [1992]; Ba/Nb is from *Hofmann* [1988]; ε Hf is from ε Nd of *Hart et al.* [1992] and the equation of the mantle array.

[e.g., *Hofmann and White*, 1982] where this source is on the order of 1 to 2.5 Ga old [*Chase*, 1981]. Since sediments can be subducted together with oceanic crust [e.g., *Tera et al.*, 1986], sediments may play a role in the isotopic variability of

the mildly HIMU Shona anomaly basalts. Since the composition of sediments varies widely, two mixing models, each with a different sediment type, are presented below. The first sediment type is ancient pelagic sediment. The second is ancient



Figure 9. Isotope–isotope plots for Shona anomaly basalts. Arrows show the segment-confined isotope trends converging to the ambient upper mantle (green circle). The greater scatter observed in (d) is probably related to the poor analytical precision of 207 Pb/ 204 Pb ratios. Open circle represents anomalous basalt EW9309 14D-4g.

sediment based on the average composition of sediments being subducted today.

5.2.1. Pelagic Sediments

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[33] *Rehkämper and Hofmann* [1997] (R&H) successfully modeled the pollution of the Indian Ocean MORB source as a mixture of normal North Atlantic/Pacific upper mantle (DM) with a recycled component. This recycled component was assumed to be 1.5 Ga old altered oceanic crust (OC) that has been subducted together with variable amounts of 1.5 Ga old pelagic sediment (SED). Their model, with some modifications, can also reproduce the Shona anomaly basalts.

[34] Since the ambient upper mantle in the South Atlantic is isotopically enriched relative to R&H's DM end-member, a modified mixing end-member is used here to represent the upper mantle contribution to the Shona anomaly basalts. The isotopic composition of this ambient depleted mantle (ADM) is taken from the intersection of the Shona and Discovery anomalies in isotope–isotope space [*Douglass et al.*, 1999, and this study]. Trace element concentrations in ADM are taken from DM except for Nb. *le Roux et al.* [2002c] show that the unusually high Ba/Nb in the South Atlantic upper mantle is probably due to Nb depletion. ADM with Ba/Nb of 5 is generated by using Nb =

	OC ^a (1.5 Ga Altered Crust)	SED ^a (1.5 Ga Pelagic Sediment)	ADM ^b (Ambient Upper Mantle)	DM ^c	1.5 Ga GLOSS ^d	Modern GLOSS ^e
Rb ^f	1.3	65	0.091	0.091	57.2	57.2
Ba	10	2000	1	1	776	776
Th	0.187	11.3	0.0187	0.0187	6.91	6.91
U	0.082	1.85	0.0071	0.0071	1.68	1.68
Nb	3.51	24	0.2	0.351	8.94	8.94
Pb	0.31	55	0.0489	0.0489	19.9	19.9
Nd	11.2	85	1.12	1.12	27	27
Sr	113	300	11.3	11.3	327	327
Sm	3.75	18	0.375	0.375	5.78	5.78
Lu	0.589	1.375	0.0589	Not reported	0.413	0.413
Hf	2.974	3.8	0.2974	Not reported	4.06	4.06
Ba/Nb	2.8	83	5	2.8	86.8	86.8
⁸⁷ Sr/ ⁸⁶ Sr	0.703	0.7203	0.7025	0.7025	.713587	0.7173
¹⁴³ Nd/ ¹⁴⁴ Nd	0.5129	0.5117	0.51312	.05132	.511977	0.51218
²⁰⁶ Pb/ ²⁰⁴ Pb	22.4	16.7	18	18	17.69	18.913
²⁰⁷ Pb/ ²⁰⁴ Pb	15.92	15.44	15.5	15.43	15.52	15.673
²⁰⁸ Pb/ ²⁰⁴ Pb	41.7	36.8	37.8	37.7		38.899
¹⁷⁶ Hf/ ¹⁷⁷ Hf	0.28295	0.2826	0.283240	Not reported	0.282228	Not reported.

 Table 3.
 Trace Element and Isotope Compositions of Mixing Model Components

^a OC and SED from *Rehkämper and Hofmann* [1997], except for the following: Lu and Hf concentrations in ancient oceanic crust are from present-day NMORB values of *Hofmann* [1988]. Lu and Hf concentrations in pelagic sediment are from *Ben Othman et al.* [1989]. ¹⁷⁶Hf/¹⁷⁷Hf of ancient oceanic crust is estimated from the composition of HIMU basalts [*Salters*, 1996, Figure 1]. ¹⁷⁶Hf/¹⁷⁷Hf of pelagic sediment is from a two-stage model. During Stage 1, from 4.5 to 1.5 Ga, ¹⁷⁶Lu/¹⁷⁷Hf is chondritic (0.0332). During Stage 2, ¹⁷⁶Lu/¹⁷⁷Hf = 0.027, which is the average value for modern-day pelagic sediments [*Patchett et al.*, 1984].

^b ADM trace element concentrations are from R&H's DM, except for the following: Nb is reduced such that ADM's Ba/Nb = 5. Lu and Hf concentrations in ADM are 10% of the N-MORB values [after *Rehkämper and Hofmann*, 1997]. Isotope ratios are from *Douglass et al.* [1999] and this study.

^cDM from *Rehkämper and Hofmann* [1997] is shown for comparison.

^d 1.5 Ga GLOSS trace element ratios are from modern GLOSS. Sr, Nd, and Hf isotope ratios are calculated assuming a two-stage model. Modern GLOSS parent–daughter ratios are used for Stage 2, which begins 1.5 Ga ago. Stage 1 parent–daughter ratios are ${}^{87}\text{Rb}/{}^{86}\text{Sr} = 0.0871$, ${}^{147}\text{Sm}/{}^{144}\text{Nd} = 0.1967$, ${}^{176}\text{Lu}/{}^{177}\text{Hf} 0.0332$. Isotope ratios at 1.5 Ga are ${}^{87}\text{Sr}/{}^{86}\text{Sr} = 0.70298$, ${}^{143}\text{Nd}/{}^{144}\text{Nd} = 0.510699$, and ${}^{176}\text{Hf}/{}^{177}\text{Hf} = 0.281824$. Pb evolution is modeled as a two-stage Stacey–Kramers evolution until 1.5 Ga after which μ equals that in modern GLOSS.

^eModern GLOSS is from *Plank and Langmuir* [1998].

^fConcentrations are in ppm.

0.2 rather than R&H's value of 0.351. In addition, Lu and Hf concentrations and 176 Hf/ 177 Hf isotope ratios, which were not considered in the original R&H model, are included in the present model. The end-member isotope ratios and trace element concentrations used in the calculations are listed in Table 3. R&H's DM composition is shown for comparison.

[35] Isotopic compositions of ADM–OC–SED mixtures are plotted in Figure 10 together with Discovery, LOMU and Shona anomaly basalts. Ancient pelagic sediment (SED) has much lower Sr/Pb, Nd/Pb and Hf/Pb than ancient oceanic crust (OC) or ambient upper mantle (ADM) (e.g., (Sr/Pb)_{SED} = 5, (Sr/Pb)_{OC} = 360, (Sr/Pb)_{ADM} = 230). Therefore, DM–SED and OC–SED binary mixtures have high degrees of curvature in Hf, Nd and Sr versus Pb isotope space. On this basis, the three

binary mixtures in Figure 10 define the allowable area for isotopic compositions of three component ADM-OC-SED mixtures.

[36] Figure 11 shows Shona anomaly basalts and three component mixtures of ADM–OC–SED with dashed lines that represent mixtures in which the OC to SED ratio is constant. If one considers oceanic crust plus sediment a single recycled component, then these lines each represent mixing of ADM with a different recycled component. Mixtures that fall along these lines can be considered binary mixtures of ambient upper mantle with different recycled components (i.e., model 2: heterogeneous plume).

[37] It is evident that *segments 2*, *3* and *4* from the Shona anomaly each fall along a trend of constant sediment–oceanic crust ratio (Figure 11). Thus,

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Figure 10. MORB compositions compared to ADM–OC–SED mixtures. ADM is ambient upper mantle. OC is 1.5 Ga old altered subducted oceanic crust. SED is 1.5 Ga old subducted pelagic sediment. See Table 3 for component compositions. Curvature of the OC–SED binary, k, is annotated on each plot. Modern and 1.5 Ga GLOSS compositions are shown for comparison. Red star in (b) is SED end-member calculated with single stage Pb evolution of $\mu = 9.1$ up to 1.5 Ga (see text). Black line in (e) is the mantle array.

each segment can be considered a binary mixture. The depleted end-member in all three cases is the ambient upper mantle (ADM). In *segment 2*, the recycled end-member is a mixture of approximately 0.85% sediment with 99.15% oceanic crust. (The best fit on ${}^{87}\text{Sr}/{}^{86}\text{Sr}-{}^{206}\text{Pb}/{}^{204}\text{Pb}$ and ${}^{207}\text{Pb}/{}^{204}\text{Pb}-{}^{206}\text{Pb}/{}^{204}\text{Pb}$ plots is 0.4% sediment, while in Nd–

Pb and Hf–Pb space the best fit is for a recycled component with 0.85% sediment.) In *segment 3*, the recycled end-member contains about 1.1% sediment and in *segment 4* this value increases to about 1.6% sediment. These results show that very small amounts of pelagic sediment have a very large effect on the composition of the





Figure 11. Modeling Shona segmentation with ADM-OC-SED mixtures. The dashed lines show compositions of ADM with different recycled (OC + SED) components. The binaries are colored as follows: OC-SED is red, ADM-OC is blue and ADM-SED is green. Red dashed-dotted line in (e) is the mantle array.

resulting mixture. Mixtures of these three recycled components (0.85%, 1.1% and 1.6% sediment) with ADM also successfully reproduce the observed Ba/Nb trends of *segments 2*, *3* and *4* (Figure 11f).

5.2.2. GLOSS

[38] Similar three-component mixing calculations can be carried out using a different sediment type.

Plank and Langmuir [1998] pointed out that sediments entering trenches today are usually not purely "pelagic" or "terrigeneous" but contain layers of different sediment types. They calculated a global average composition of sediments being subducted today (GLOSS). If the composition of sediments returned to the mantle in the past was similar to today's GLOSS composition, one can use trace element concentrations in modern GLOSS to

estimate the isotopic composition of an ancient GLOSS component.

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[39] GLOSS does not take into account sediments that are removed from the downgoing slab by underplating and thus may not be completely representative of sediment recycled into the mantle. In addition, the nature of subducted sediments has likely changed over time. Nevertheless, GLOSS is a useful starting point to extrapolate an ancient sediment component. Modern GLOSS and 1.5 Ga GLOSS trace element concentrations and isotope ratios are listed in Table 3.

[40] Three component mixtures of ADM-OC-1.5 Ga GLOSS can also reproduce compositions of the Shona anomaly basalts. In this model, as with ADM-OC-SED, the Shona basalts fall on mixing lines of constant OC-sediment ratios (not shown). In this case, however, the percentage of sediment present in each recycled component (OC + 1.5 Ga GLOSS) is much higher. The percentages of sediment in the recycled components are 3%, 5% and 11% in *segments 2, 3* and *4* respectively.

[41] Due to the high concentration of Pb in SED (55 ppm) and GLOSS (19.9 ppm) relative to that in OC (0.31 ppm), even a small amount of sediment greatly affects ²⁰⁶Pb/²⁰⁴Pb of the OC-sediment mixture. This leverage effect on ²⁰⁶Pb/²⁰⁴Pb is much stronger than the effect of sediment on Sr, Hf or Nd isotope ratios. Pure recycled oceanic crust (with no sediments) usually is considered the source of HIMU plumes [e.g., Hofmann and White, 1982]. True HIMU plumes are rare (St. Helena, Tubuaii, Mangaia [e.g., White, 1985]). It is much more common to find mildly HIMU or C-type plumes (Shona, Bouvet, Iceland, Azores [e.g., Hanan and Graham, 1996]). This is probably because subduction of oceanic crust that is completely free of sediments is unlikely and even a small amount of sediment quickly erases the high ²⁰⁶Pb/²⁰⁴Pb associated with pure HIMU plumes.

[42] Both ADM-OC-SED and ADM-OC-1.5 Ga GLOSS models are consistent with a Shona plume containing recycled material, namely anciently subducted oceanic crust with sediment. The calculated fraction of sediment in the recycled component is highly dependent on the nature of the sediment. However, regardless of which sediment type is used in the model, the Shona plume is not well mixed since there seem to be three different "flavors" of recycled component (i.e., three different crust/sediment ratios) mixing with a locally homogeneous ambient upper mantle.

[43] It is of course possible that the Shona plume is in fact homogeneous (i.e., model 1), either with a HIMU composition (pure oceanic crust with no sediment present) or C composition (some small, but constant fraction of sediments present). In this case, the ambient upper mantle with which the plume is mixing must be heterogeneous with the sediment fraction in the ambient upper mantle increasing to the south. In such a scenario, however, it is difficult to explain how the sediment/oceanic crust fraction remains constant over a given ridge segment since the oceanic crust resides in the plume and the sediment resides in the ambient upper mantle. There is evidence for heterogeneous plumes in other areas such as Hawaii [Hauri, 1996], Iceland [Chauvel and Hémond, 2000] and the Galapagos [Blichert-Toft and White, 2001]. It is likely that the Shona plume is also an example of a heterogeneous plume rather than a homogeneous plume mixing with a locally heterogeneous ambient upper mantle.

[44] A final point can be made regarding ADM-OC-SED mixtures and Hf-Nd isotope systematics. Patchett et al. [1984] suggested that EHf can be used to evaluate whether pelagic sediment is present in the source of a basalt since ε Hf for those basalts which arise from a source containing pelagic sediment fall above the Hf-Nd mantle array. This is because ε Hf is high in pelagic sediment for a given ENd due to the high time-integrated Lu/Hf (the zircon effect). Blichert-Toft et al. [1999] used ϵHf with ϵNd and $^{206}Pb/^{204}Pb$ to identify pelagic sediment in the source of Hawaiian basalts. From Figure 11e it is clear that ε Hf is a sensitive indicator of pelagic sediment only when the sediment component of the basalt source is greater than about 2%. If smaller sediment fractions are involved, the ε Hf $-\varepsilon$ Nd of the mixture falls very close to (or even below) the mantle array, espe-

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cially if oceanic crust is also present in the basalt source. This seems to be the case for the Shona anomaly data (Figure 11e). While the Shona anomaly data falls within the Hf–Nd isotope space defined by ADM–OC–SED, since the sediment component is so small, the data is clustered near the ADM–OC binary.

6. Discovery and LOMU Ridge Anomalies

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[45] Basalts from the Discovery and LOMU ridge anomalies generally fall above the mantle array as can be seen from their positive $\Delta \epsilon$ Hf values (Figure 5a). Positive $\Delta \varepsilon$ Hf may be interpreted as evidence of pelagic sediments in the basalt source [e.g., Blichert-Toft et al., 1999]. Despite this indication that pelagic sediment may be involved, ADM-OC-SED mixtures used to model the compositions of basalts from the Shona anomaly do not successfully reproduce compositions of basalts from the Discovery or LOMU anomalies (Figure 10). Isotope compositions of many basalts from these two regions fall outside of the range of possible isotope compositions of ADM-OC-SED mixtures for two reasons. The first reason is that the ²⁰⁷Pb/²⁰⁴Pb of the pelagic sediment component (SED) is too low to encompass all of the Discovery and LOMU anomaly basalt compositions on a ²⁰⁷Pb/²⁰⁴Pb-²⁰⁶Pb/²⁰⁴Pb plot (Figure 10b). The second problem is the difference in Sr/ Pb, Nd/Pb and Hf/Pb in the SED and the OC endmembers. These large differences in trace element concentration ratios cause high degrees of curvature in the OC-SED binaries in Pb-Sr, Pb-Nd and Pb-Hf isotope space. Because of this curvature, many Discovery and LOMU anomaly basalt compositions fall outside of the possible range of ADM-OC-SED mixtures.

[46] Different end-member(s) must be involved. Several possibilities for alternatives to the SED end-member can be investigated qualitatively.

6.1. Recycling Oceanic Crust With Sediments

[47] One possibility is that pelagic sediment is in fact mixing with ambient depleted mantle and

oceanic crust to produce Discovery and LOMU anomaly basalts but that SED is an inaccurate representation of this pelagic sediment. If this is true, ²⁰⁷Pb/²⁰⁴Pb of SED is too low and the Pb concentration is too high to accurately represent pelagic sediment. The SED end-member used in the previous ADM-OC-SED model is from Rehkämper and Hofmann [1997]. These authors estimate the ${}^{206}\text{Pb}/{}^{204}\text{Pb}$, ${}^{207}\text{Pb}/{}^{204}\text{Pb}$ and ${}^{208}\text{Pb}/{}^{204}\text{Pb}$ of 1.5 Ga old pelagic sediment from a two-stage Stacey-Kramers Pb evolution model ($\mu_1 = 7.19$ up to 3.7 Ga, $\mu_2 = 9.7$ up to 1.5 Ga) [*Stacey and Kramers*, 1975] followed by evolution with $\mu = 2$ which is typical of ²³⁸U/²⁰⁴Pb in modern pelagic sediments [Patchett et al., 1984]. There is some evidence that the Stacey-Kramers model underestimates U/Pb in the first stage of Pb evolution. If one instead uses a single-stage evolution model until 1.5 Ga with the U/Pb ratio suggested by Allègre et al. [1988] ($\mu =$ 9.1) then pelagic sediment does have the appropriate Pb isotope ratios (i.e., high enough ²⁰⁷Pb/²⁰⁴Pb for a relatively low 206 Pb/ 204 Pb) to encompass all the Discovery and LOMU data on Pb-Pb plots (Figure 10b, red star).

[48] While the Pb isotope evolution of 1.5 Ga old pelagic sediment may be modeled such that it is an appropriate mixing component for Discovery and LOMU anomaly basalts, the Pb concentration of pelagic sediment is more problematic. The SED Pb concentration of Rehkämper and Hofmann [1997] is 55 ppm. While this value is in the range of pelagic sediment concentrations reported in the literature, it falls on the high end of the range of pelagic sediment data from Plank and Langmuir [1998] or Ben Othman et al. [1989]. The average of Pb concentrations in pelagic sediment samples from these two sources is about 35 ppm (the range is 16-64 ppm). However, neither 35 ppm nor 16 ppm (the lowest value reported by Ben Othman et al. [1989]) is low enough to reduce the curvature in the SED-OC mixing binaries enough to encompass the Discovery and LOMU anomaly data completely. Thus, because of the relatively high Pb concentration in pelagic sediment, it is not likely that pelagic sediment is involved in the mixtures that give rise to Discovery anomaly and LOMU section basalts.

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[49] A final argument against the presence of ancient pelagic sediment in the source of Discovery and LOMU anomaly basalts is based on the Hf-Nd isotope systematics of ADM-OC-SED mixtures. Figure 10e shows that the Discovery and LOMU anomaly basalts that fall squarely outside of the possible ε Hf- ε Nd range have ε Hf values that are too low for a given ε Nd. However, one of the distinguishing features of ancient pelagic sediment is that it has high time-integrated Lu/Hf (and therefore high ϵ Hf) for a given time-integrated Sm/ Nd due to the zircon effect [Patchett et al., 1984]. The trends of Discovery and LOMU data indicate that they arise from a source with a much lower ϵ Hf (and therefore lower time-integrated Lu/Hf) than pelagic sediment. Thus, despite the high $\Delta \varepsilon Hf$ of these basalts, pelagic sediment is probably not a significant component in the source of Discovery and LOMU basalts.

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[50] Sediments based on ancient GLOSS are also not suitable end-members for Discovery and LOMU anomaly basalts. As with SED, three component mixtures including an ancient GLOSS cannot reproduce the compositions of the most isotopically enriched Discovery and LOMU basalts because of the curvature of the OC–GLOSS binary (not shown) and because of the Pb isotope composition of ancient GLOSS (Figure 10b). Ancient sediments based on GLOSS are unsuitable components regardless of their age and regardless of the evolution model used to estimate the Pb isotopes (*Stacey and Kramers* [1975] or *Allègre et al.*'s [1988] single stage).

6.2. Recycling Lower Continental Crust or Subcontinental Lithosphere

[51] It is possible that material from the lower continental crust rather than sediment is a component in the source of Discovery and LOMU anomaly basalts. Whether or not lower crust is a suitable component can possibly be assessed from the isotopic composition of lower crustal material which is brought to the surface as garnet *granulite xenoliths*. The isotopic compositions of such xenoliths, which come from depths between about 25–45 km, vary greatly from one region to another depending on their protolithic nature [*Rudnick and*]

Goldstein, 1990; Rudnick and Fountain, 1995]. The component that is mixing with the ambient depleted mantle to produce Discovery and LOMU anomaly basalts must have ²⁰⁶Pb/²⁰⁴Pb significantly lower than 18. Globally, many garnet granulite xenoliths do not have such low ²⁰⁶Pb/²⁰⁴Pb. However, xenoliths from stable regions (cratons) do have unusually low ²⁰⁶Pb/²⁰⁴Pb [e.g., Rudnick and Goldstein, 1990]. The Kaapvaal Craton in southern Africa, for example, is one such region where xenoliths with low $^{206}\mathrm{Pb}/^{204}\mathrm{Pb}$ are found [Huang et al., 1995]. However, the source of Discovery and LOMU anomaly basalts must also have high ²⁰⁷Pb/²⁰⁴Pb. Of the Kaapvaal Craton granulite xenoliths, only those from the northern Lesotho region have high enough ²⁰⁷Pb/²⁰⁴Pb for a given ²⁰⁶Pb/²⁰⁴Pb [Huang et al., 1995].

[52] While granulites from northern Lesotho do have the Pb isotope characteristics required to produce Discovery and LOMU anomaly basalts from ADM-granulite or ADM-OC-granulite mixtures, they do not have the appropriate ⁸⁷Sr/⁸⁶Sr. Of the 13 xenoliths from the Lesotho region reported by Rogers and Hawkesworth [1982] only one (LT2, a felsic xenolith) has high enough ⁸⁷Sr/86</sup>Sr to be a suitable mixing endmember in the source of Discovery and LOMU anomaly basalts. Xenoliths from regions in South Africa other than Lesotho do have high enough ⁸⁷Sr/⁸⁶Sr but not high enough ²⁰⁷Pb/²⁰⁴Pb for a given ²⁰⁶Pb/²⁰⁴Pb. Apparently, only one reported xenolith from the Kaapvaal Craton region has the combined isotope characteristics required in the source of Discovery and LOMU anomaly basalts. It is possible that some combination of South African lower crustal material might be invoked to contribute to the Discovery and LOMU anomaly basalts, but this remains speculative.

[53] Finally, material from the *subcontinental lithospheric mantle* (SCL) may be considered as a possible component in mixtures that give rise to the Discovery and LOMU anomaly basalts. As with the garnet granulite xenoliths, these SCL xenoliths exhibit great variability in their isotopic compositions. *Hawkesworth et al.* [1990] report isotope and trace element data for SCL xenoliths from Kimberly, South Africa which fall on a 150 Ma secondary isochron caused by modal metasomatism. These samples apparently have not suffered any significant U/Pb contamination by the host kimberlite during emplacement 90 Ma ago. Since most of these xenoliths have 206 Pb/ 204 Pb that is relatively high, SCL metasomatized 150 Ma ago is not a likely component in the Discovery and LOMU basalts. However, a garnet peridotite and a garnet phlogopite peridotite from this group of SCL xenoliths, both with very low μ , do meet the 206 Pb/ 204 Pb $-^{207}$ Pb/ 204 Pb requirements of a Discovery/LOMU component.

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[54] In general, xenoliths from both the SCL and the lower crust exhibit large isotopic variability and the recovered xenoliths may not be completely representative of SCL or lower crust compositions. Thus it is difficult to establish with certainty whether either reservoir is present in the source of Discovery and LOMU anomaly basalts. Although there is no overwhelming evidence in favor of a lower crustal granulite source in the Discovery and LOMU anomaly basalts from xenolith compositions, there is some evidence from the trace element characteristics of the S18-60/1 fresh basaltic glass located just north of the Bouvet triple junction described by Kamenetsky et al. [2001]. This fresh basalt has high 87 Sr/ 86 Sr, 207 Pb/ 204 Pb and low ε Nd, more extreme than any Discovery or LOMU anomaly basalts. Kamenetsky et al. [2001] suggest that mafic granulite from the lower crust is a reasonable source for this basalt. In their view, based on rare earth elements, CaO, Sc, and V, the source of this basalt could have a mineralogy that is consistent with the mineralogy of lower crustal mafic granulites (i.e., the source contains olivine, orthopyroxene and garnet). Thus, although a mafic xenolith with the appropriate Sr and Pb isotope characteristics has not yet been recovered, it remains possible that material from the lower crust is indeed a component in these unusual Discovery and LOMU anomaly basalts.

7. Reconciling Primitive and Recycled Signals

[55] Sr-Nd-Pb-Hf isotope data provide strong evidence for the presence of recycled material in

both the Shona and Discovery plumes. However, Shona and Discovery anomaly basalts have very high ³He/⁴He (up to 12.2R_A and 15.9R_A respectively, Figure 5c) and a solar neon component [Moreira et al., 1995; Sarda et al., 2000; Moreira and Sarda, 2000]. High ³He/⁴He is considered a characteristic of primitive mantle material [Lupton and Craig, 1975]. In contrast, recycled material, which has been degassed as it goes through the noble gas subduction barrier [Staudacher and Allègre, 1988], develops low ³He/⁴He over time through build up of radiogenic ⁴He. Thus, Shona and Discovery anomaly basalts likely contain a more primitive and poorly degassed component in addition to the recycled components [Moreira et al., 1995; Sarda et al., 2000; Moreira and Sarda, 2000]. Presumably, this component is lower mantle material that is entrained as the Shona and Discovery plumes rise either from the core mantle boundary [Kellogg and Wasserburg, 1990; Hauri et al., 1994; Albarède and van der Hilst, 1999] or from the presumed 660 km thermal boundary layer [Allègre, 1997]. If the lower mantle is poorly degassed, its helium concentration should be relatively high in comparison to that of recycled material. Therefore, even if the amount of entrained lower mantle material is relatively small, its helium ratio dominates the helium budget of the plumes and may impart a more primitive ³He/⁴He signal to the plumes.

[56] In contrast to ³He/⁴He, the presence of primitive mantle may not be readily apparent from Pb, Sr, Nd and Hf isotope ratios. This is because the Pb, Sr, Nd and Hf isotope nature of the lower mantle likely falls within the so-called DM–EM1– EM2–HIMU end-member component tetrahedron. Such intermediate compositions have been referred to as FOZO [*Hart et al.*, 1992], PHEM [*Farley and Craig.*, 1992] or C component [*Hanan and Graham*, 1996].

[57] The presence of a relatively more primitive component in the Shona and Discovery anomaly basalts does not invalidate the general conclusions drawn from the ADM–OC–sediment mixing calculations. Even with the presence of a primitive component, Discovery basalts probably do not contain ancient sediment while Shona anomaly

basalts still may arise from mixtures containing ancient sediments and oceanic crust. However, the presence of a primitive component does affect the calculated percentages of ambient mantle, oceanic crust and sediment present in Shona anomaly basalts. With a primitive component the amount of recycled component (OC + SED or OC + 1.5 Ga GLOSS) present in these basalts would simply be lower than that calculated with the ADM–OC– SED or ADM–OC–1.5 Ga GLOSS models because primitive material is less radiogenic for Hf and Nd and more radiogenic for Sr and Pb than ADM.

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8. Conclusions

[58] ¹⁷⁶Hf/¹⁷⁷Hf data collected in this study support the observation that the depleted upper mantle beneath the South Atlantic is transitional to DUPAL composition. Mechanisms of large-scale pollution of the South Atlantic upper mantle remain speculative as is the case for the upper mantle beneath the Indian ocean.

[59] Sr-Nd-Pb-Hf isotope data combined with the high ³He/⁴He of Shona anomaly basalts indicate that two types of heterogeneities coexist in the Shona plume. The heterogeneity that is apparent in the Sr-Pb-Nd-Hf isotope data probably results from recycling of ancient oceanic crust which was subducted with varying amounts of sediment. This heterogeneity is likely on the order of 1.5 Ga old. Superimposed on this effect is a heterogeneity that may have resulted from relatively recent entrainment of primitive material as the plume rose through the lower mantle or detached from the presumed 660 km discontinuity. This heterogeneity is readily detected in the ³He/⁴He and neon isotope compositions of these MORB, but less so with Hf, Nd, Sr and Pb isotope ratios.

[60] The source of the Discovery plume remains enigmatic. *Douglass et al.* [1999] pointed out that this source must have low ²⁰⁶Pb/²⁰⁴Pb and ¹⁴³Nd/¹⁴⁴Nd and high ²⁰⁷Pb/²⁰⁴Pb and ⁸⁷Sr/⁸⁶Sr. While sediment is most likely not a significant component in the Discovery plume, material from the lower crust or subcontinental lithospheric mantle may be present in the Discovery plume. Even though lower crustal or SCL xenoliths exhibiting the required isotopic composition are elusive, there is some evidence that lower crustal granulites may be present in the Discovery plume [*Kamenetsky et al.*, 2001]. In addition, it remains unresolved whether the Discovery and LOMU anomaly basalts arise from two distinct sources or whether both arise from a single heterogeneous Discovery plume.

[61] The mixing models developed here demonstrate that the possible isotopic compositions of upper mantle–ancient sediment mixtures (with or without oceanic crust) are severely restricted by the trace element concentrations in the components. Due to the high concentration of Pb relative to Sr, Nd, and Hf in ancient pelagic or "average" (i.e., GLOSS) sediment, it is not possible to generate mixtures that have high ⁸⁷Sr/⁸⁶Sr (and low ϵ Nd and ϵ Hf) but which do not also have low ²⁰⁶Pb/²⁰⁴Pb and ²⁰⁷Pb/²⁰⁴Pb.

[62] Finally, the mixing models imply that despite the zircon effect, when the concentration of pelagic sediment in a mixture is very low, ε Hf cannot be used to confirm its presence. In these cases, other isotope ratios (such as ²⁰⁶Pb/²⁰⁴Pb) are better indicators of the presence of pelagic sediment. However, when the concentration of pelagic sediment in a mixture is larger than a few percent, ε Hf– ε Nd systematics can be used to establish the presence of pelagic sediment and to estimate its concentration.

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