

# Geochemistry of the oldest Atlantic oceanic crust suggests mantle plume involvement in the early history of the central Atlantic Ocean

Philip E. Janney\*, Paterno R. Castillo

*Scripps Institution of Oceanography, University of California, San Diego, 9500 Gilman Dr., La Jolla, CA 92093-0220, USA*

Received 21 March 2001; received in revised form 27 June 2001; accepted 16 July 2001

## Abstract

Controversy has surrounded the issue of whether mantle plume activity was responsible for Pangaeian continental rifting and massive flood volcanism (resulting in the Central Atlantic Magmatic Province or CAMP, emplaced around 200 Ma) preceding the opening of the central Atlantic Ocean in the Early Mesozoic. Our new Sr–Nd–Pb isotopic and trace element data for the oldest basalts sampled from central Atlantic oceanic crust by deep-sea drilling show that oceanic crust generated from about 160 to 120 Ma displays clear isotopic and chemical signals of plume contamination (e.g.,  $^{87}\text{Sr}/^{86}\text{Sr}_i = 0.7032\text{--}0.7036$ ,  $\epsilon_{\text{Nd}}(t) = +6.2$  to  $+8.2$ , incompatible element patterns with positive Nb anomalies), but these signals are muted or absent in crust generated between 120 and 80 Ma, which resembles young Atlantic normal mid-ocean ridge basalt. The plume-affected pre-120 Ma Atlantic crustal basalts are isotopically similar to lavas from the Ontong Java Plateau, and may represent one isotopic end-member for CAMP basalts. The strongest plume signature is displayed near the center of CAMP magmatism but the hotspots presently located nearest this location in the mantle reference frame do not appear to be older than latest Cretaceous and are isotopically distinct from the oldest Atlantic crust. The evidence for widespread plume contamination of the nascent Atlantic upper mantle, combined with a lack of evidence for a long-lived volcanic chain associated with this plume, leads us to propose that the enriched signature of early Atlantic crust and possibly the eruption of the CAMP were caused by a relatively short-lived, but large volume plume feature that was not rooted at a mantle boundary layer. Such a phenomenon has been predicted by recent numerical models of mantle circulation. © 2001 Elsevier Science B.V. All rights reserved.

*Keywords:* Atlantic Ocean; mantle plumes; oceanic crust; Mid-ocean ridge basalts; flood basalts; Pangaea

## 1. Introduction

The first major stage in the Mesozoic disintegration of the Pangaeian super-continent was the opening of the central Atlantic Ocean in the Early Jurassic. The opening of this ocean basin was immediately preceded by a brief episode of areally extensive flood volcanism at about 200 Ma (coin-

\* Corresponding author. Present address: Department of Geology, The Field Museum of Natural History, 1400 S. Lake Shore Dr., Chicago, IL 60605, USA. Tel.: +1-312-665-7099; Fax: +1-312-665-7641.

*E-mail address:* pjanney@fmnh.org (P.E. Janney).

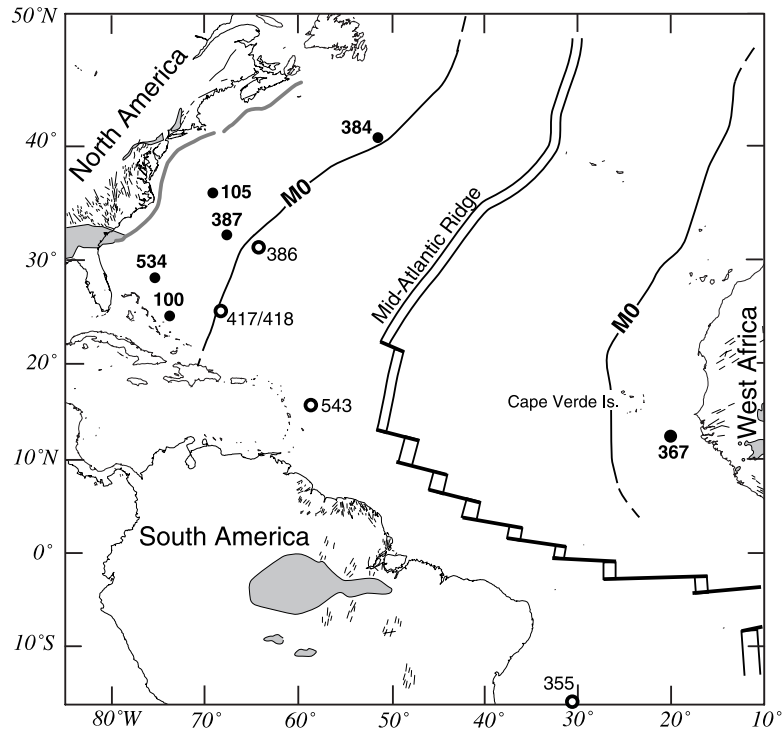


Fig. 1. Location map showing DSDP drill sites examined in this study (solid and open circles indicate crust older and younger than 120 Ma, respectively). Line marked 'M0' represents the location of magnetic anomaly M0 (having an approximate age of 118 Ma [11]). See Table 1 for ages inferred from magnetic anomalies and biostratigraphy. Dikes (black lines), sills and flows (gray fields) and submarine basaltic wedges (gray lines) of the CAMP [1] are shown for reference.

ciding with the Triassic–Jurassic boundary) on what are now the continental margins of eastern North America, southwestern Europe, West Africa and northeastern South America (e.g., [1]). The region originally covered by these volcanic rocks (now largely eroded) has been termed the Central Atlantic Magmatic Province (CAMP) [2]. On the basis of the distribution of dikes and sills, this province may have originally covered an area of  $7 \times 10^6$  km<sup>2</sup>, making it one of the largest continental igneous provinces [3].

The question of whether the volcanism responsible for the CAMP, as well as the opening of the central Atlantic, was the result of passive tectonic forces or the action of a mantle plume, plume-head or some other deep-seated mantle phenomenon is controversial. Arguments for a passive tectonic origin for the CAMP are based on: (1) the widespread and non-centralized distribution

of magmatism (e.g., [4,5]), (2) evidence that the timing of uplift and rifting, which occurred over a  $\sim 35$  Myr interval in the Late Triassic and Early Jurassic, shows little correlation with the rapid pulse of magmatic activity at the Triassic–Jurassic boundary [6], and (3) the fact that no recognized hotspot or linear volcanic chain in the central Atlantic region can be clearly linked to the CAMP in time and space [1]. Proponents of an active, deep mantle origin for the CAMP interpret the huge amount of melt produced over a short, 2–10 Myr interval and the radial orientation of most CAMP dikes as likely requiring involvement of a mantle plume or plume-head (e.g., [2,7,8]). Geochemical investigation of the dominantly tholeiitic CAMP dikes, sills and flows has been of limited use in resolving this controversy because they appear to be largely derived from continental lithospheric mantle and some may

have suffered crustal contamination (e.g., [9,10]). These rocks typically have high initial  $^{87}\text{Sr}/^{86}\text{Sr}$  and low  $\epsilon_{\text{Nd}}(t)$  values that fall outside the isotopic ranges of most ocean island basalts (OIB).

In order to better understand the ultimate causes of the widespread magmatism and continental rifting that occurred in the central Atlantic region in the Early Mesozoic we have collected Sr, Nd and Pb isotopic ratios and trace element data on basalts sampled from early Atlantic oceanic crust at 11 Deep Sea Drilling Project sites in the western and eastern central Atlantic (Fig. 1). On the basis of biostratigraphic and magnetic anomaly ages (Table 1), the sampled crust spans an age range from 160 to 80 Ma. Our objective is to use the radiogenic isotope and trace element record preserved in these basalts to investigate the regional geochemical evolution of the upper mantle beneath the nascent Atlantic ocean basin. Specifically, we wish to determine whether or not a plume-type geochemical signal exists in early Atlantic oceanic crust. The presence or absence of such a signal would support or discredit the hypothesis that plume activity was involved in the generation of the CAMP and may have been a causative factor in the opening of the central Atlantic Ocean.

## 2. Methods

The freshest available basalt samples, mostly free of veins and alteration zones, were selected for analysis. Hypocrystalline basalt was the sample material most commonly used, except for drill cores from Holes 417D, 418A and 543A which contain abundant fresh glass. Fresh glass chips were picked under high magnification to avoid alteration products, then were leached for 20 min in 2 N ultrapure hydrochloric acid to remove surface contamination. Exterior surfaces of whole-rock samples were trimmed and saw marks were removed with silicon carbide grit. Rock samples were broken into  $\sim 0.5 \text{ cm}^3$  chips, pieces free of veins and vesicles were picked and leached for 45 min in 2 N nitric acid. Powders for Sr and Nd isotope and major and trace element analysis were prepared from these chips. Powders used for Sr isotope measurements were further leached four or five times in cold 4 N HCl interspersed with ultrapure water rinses (e.g., [12]). Powders used for Pb isotope measurements were prepared by leaching fresh  $\sim 0.5 \text{ cm}^3$  rock chips for 4 h in warm 4 N nitric acid, followed by rinsing, drying and powdering in a tungsten carbide shatterbox.

Chemical separations of Sr, Nd and Pb were

Table 1  
Basement drilling results in central Atlantic Mesozoic ocean crust

Leg-Hole	Lat. and Long	Oldest age of sediment	Magnetic anomaly	Approx. age (Ma)	Basalt recovery (m)
11–100	24°41'N 73°48'W	Oxfordian–Callovian (J)	M22	155	5
11–105	34°54'N 69°10'W	Oxfordian–Callovian (J)	M25	158	8
39–355	15°42.6'N 30°36'W	Campanian (K)	33	80	8
41–367	12°29'N 20°03'W	Kimmeridgian–Oxfordian (J)	M23	157	6
43–384	40°22'N 51°40'W	Aptian–Barremian (K)	M2	123	2
43–386	31°11'N 64°15'N	Early Albian (K)	KQZ	110	2
43–387	32°19'N 67°40'W	Valanginian–Berrasian (K)	M16	140	3
51/52/53–417A	25°07'N 68°03'W	Aptian–Albian (K)	M0	118	129
51/52/53–417D	25°07'N 68°03'W	Aptian–Albian (K)	M0	118	263
51/52/53–418A	25°02'N 68°04'W	Aptian–Albian (K)	M0	118	391
51/52/53–418B	25°02'N 68°04'W	Aptian–Albian (K)	M0	118	8
76–534	28°21'N 75°23'W	Callovian (J)	M26	160	21
78A–543A	15°43'N 58°39'W	Campanian (K)	33	80	36

Drilling and biostratigraphic age information is summarized from Initial Reports of the Deep Sea Drilling Project, volumes 11, 39, 41, 43, 51–53, 76 and 78A. The abbreviations 'J', 'K' and 'KQZ' stand for Jurassic period, Cretaceous period, and the Cretaceous Quiet Zone, respectively. Numerical age values are estimated from biostratigraphic and magnetic anomaly ages using the timescale of Harland et al. [11].

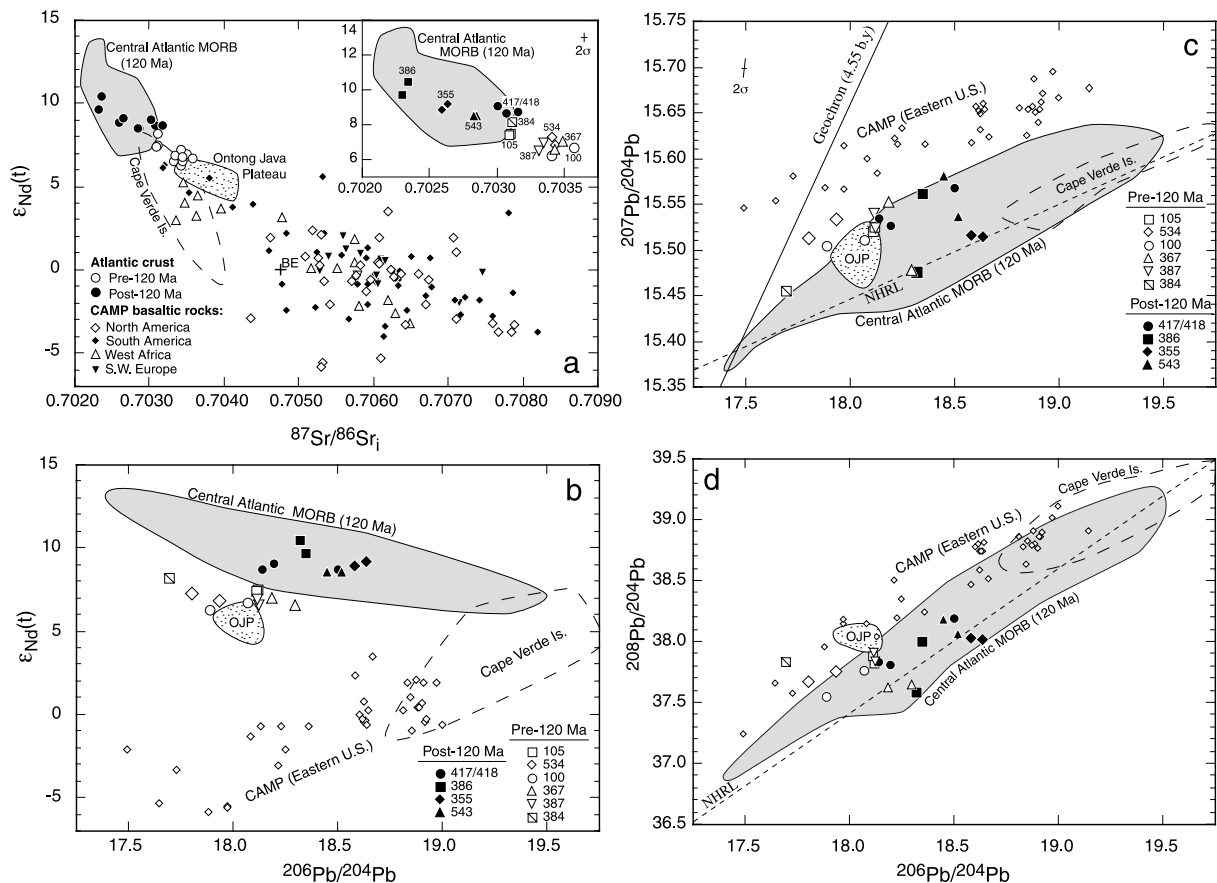


Fig. 2. Isotopic diagrams showing Mesozoic Atlantic ocean crust, young central Atlantic MORB, CAMP basalts and assorted ocean island and oceanic plateau lavas. (a)  $^{87}\text{Sr}/^{86}\text{Sr}_i$  versus  $\epsilon_{\text{Nd}}(t)$ . (b)  $^{206}\text{Pb}/^{204}\text{Pb}_i$  versus  $\epsilon_{\text{Nd}}(t)$ . (c)  $^{206}\text{Pb}/^{204}\text{Pb}_i$  versus  $^{207}\text{Pb}/^{204}\text{Pb}_i$ . (d)  $^{206}\text{Pb}/^{204}\text{Pb}_i$  versus  $^{208}\text{Pb}/^{204}\text{Pb}_i$ . Except where shown, error bars are equal to or smaller than the size of the data symbols. Isotopic data for CAMP basalts are from [4,9,10,17–19], Cape Verde OIB from [20] and Ontong Java Plateau (OJP) basalts from [12]. Pb isotopic data for OJP basalts were age-corrected using mean U/Pb and Th/Pb ratios measured for each main petrologic group (i.e., Site 803, Site 807 Unit A and Site 807 Units C–G). Central Atlantic mid-ocean ridge basalts (MORB) include data from 10°S to 45°N, excluding data from areas affected by modern hotspots (contact first author for list of data sources). Position of the MORB field was corrected for 120 Myr of depleted mantle evolution so as to be directly comparable to Mesozoic Atlantic crust data. Note that Pb isotopic data for CAMP basalts are measured, not age-corrected values. See text for discussion.

performed by ion exchange and isotopic measurements were performed by thermal ionization mass spectrometry, all at the Scripps Institution of Oceanography using standard techniques. Trace element concentration measurements were performed by quadrupole ICPMS at the Massachusetts Institute of Technology, following the technique of Janney and Castillo [13]. In addition to measuring a suite of trace elements from Rb to U on the minimally leached bulk rock powders and glasses, parent–daughter elemental concentra-

tions, used solely for age correction, were measured on the same specially prepared sample powders used for isotopic analysis. The full set of trace element data for early Atlantic crust is included in the accompanying **Background Data Set**<sup>1</sup> and is available from the first author upon request.

<sup>1</sup> <http://www.elsevier.com/locate/epsl>

### 3. Results and discussion

The initial  $^{87}\text{Sr}/^{86}\text{Sr}$  and  $\epsilon_{\text{Nd}}$  values of Mesozoic Atlantic crust cover a significant range, forming a fairly tight linear correlation (Fig. 2; Table 2). The isotopic compositions of the samples can be grouped by age, with crustal basalts generated between 160 and 123 Ma (hereafter termed the ‘pre-120 Ma’ group) having higher  $^{87}\text{Sr}/^{86}\text{Sr}_i$  and lower  $\epsilon_{\text{Nd}}(t)$  values (0.7031–0.7036 and 8.2–6.2, respectively) than basalts generated between 118 and 80 Ma (hereafter the ‘post-120 Ma’ group), which have values of 0.7023–0.7032 and 8.5–10.4, respectively. Moreover, the Atlantic crust samples display a roughly linear decrease with time in  $^{87}\text{Sr}/^{86}\text{Sr}_i$  and a corresponding increase in  $\epsilon_{\text{Nd}}(t)$  (with  $r^2$  values of 0.77 and  $-0.76$ , respectively). Pb isotope ratios are also fairly distinct between the pre- and post-120 Ma groups. The older group has lower  $^{206}\text{Pb}/^{204}\text{Pb}_i$  values ( $<18.3$ ) and higher  $^{207}\text{Pb}/^{204}\text{Pb}_i$  and  $^{208}\text{Pb}/^{204}\text{Pb}_i$  for a given  $^{206}\text{Pb}/^{204}\text{Pb}_i$  value (i.e.,  $\Delta 7/4$  and  $\Delta 8/4$  [14]) than the younger. Only the  $\sim 118$  Ma Site 417–418 basalts have Pb isotope compositions intermediate between these two groups. We note that the highly altered Site 367, 384 and 386 samples appear to have undergone significant U uptake (i.e., they have  $\text{Th}/\text{U} < 1$  and large inter-sample scatter in  $^{207}\text{Pb}/^{204}\text{Pb}$ ), which may be indicative of long-term open system interaction with seawater [15]. Thus, their initial Pb isotopic values should be viewed only as approximations. Overall however, the pre-120 Ma crustal basalts, particularly those from drill sites off the southeastern USA and West Africa (i.e., Sites 100, 367, 387 and 534) tend to display mild EM 1 or ‘Dupal’ isotopic affinities (e.g., [14]), causing them to fall largely outside the isotopic fields of young central Atlantic MORB, whereas the post-120 Ma crust is isotopically indistinguishable from young central Atlantic MORB.

On the whole, Mesozoic Atlantic ocean crust displays much lower  $^{87}\text{Sr}/^{86}\text{Sr}_i$  and higher  $\epsilon_{\text{Nd}}(t)$  values than CAMP sills and dikes, which have means of 0.706 and  $-1$ , respectively. However, the CAMP isotopic data form a fan-shaped array in Sr–Nd isotopic space that converges toward low  $^{87}\text{Sr}/^{86}\text{Sr}_i$  and high  $\epsilon_{\text{Nd}}(t)$  values similar to

those of most pre-120 Ma crustal samples (Fig. 2). Therefore, one source end-member of CAMP magmatism may be either the mild EM 1-type mantle source of the pre-120 Ma Atlantic crust, or a depleted normal MORB (N-MORB) source mantle component that has been significantly diluted even in the most MORB-like CAMP basalts.

Published Pb isotopic data for the CAMP are limited to tholeiitic dike and sill samples from the eastern USA [10], and only measured values (uncorrected for age) are reported for this suite. Nevertheless, it is clear from their present-day Pb isotopic compositions that these tholeiites were characterized by high initial  $\Delta 7/4$  values and (unless their  $^{232}\text{Th}/^{238}\text{U}$  values are exceptionally high) high initial  $\Delta 8/4$  values as well, relative to young Atlantic MORB. Additionally, the eastern USA CAMP basalts likely had relatively unradiogenic initial  $^{206}\text{Pb}/^{204}\text{Pb}$  isotopic compositions, as their current range of  $^{206}\text{Pb}/^{204}\text{Pb}$  ratios extend to only slightly radiogenic values (i.e., 17.5–19.1). Therefore, CAMP basalts from the eastern USA, and presumably other regions, display EM 1-type Sr–Nd–Pb isotopic characteristics that are qualitatively similar to, but much more extreme than, those displayed by the oldest basalts sampled from central Atlantic ocean crust.

The isotope data clearly indicate that Jurassic and Early Cretaceous Atlantic crustal basalts were derived from mantle sources that are isotopically distinct (i.e., having a mild EM 1 affinity) from those tapped by Late Cretaceous to Recent N-MORB from the Mid-Atlantic Ridge. However, it is not immediately clear what is the origin of this distinct source composition. The Sr, Nd and Pb isotope ratios of the older Atlantic crust could be plausibly explained as a mixture between a depleted MORB source and metasomatized continental lithosphere (thought to be the major source component of CAMP basalts [10,19]) that was added to the upper mantle during rifting. However, the Sr–Nd–Pb isotopic compositions of most pre-120 Ma crustal samples also overlap with plume-derived lavas from purely oceanic settings, such as those from the Early Cretaceous Ontong Java Plateau (OJP) of the western Pacific

Table 2  
Sr, Nd and Pb isotope ratio and parent–daughter elemental data for Mesozoic Atlantic crust

Leg–Hole	Core, section interval (cm)	Rb (ppm)	Sr (ppm)	$\frac{87\text{Sr}}{86\text{Sr}}$	$\frac{87\text{Sr}}{86\text{Sr}}$	Sm (ppm)	Nd (ppm)	$\frac{145\text{Nd}}{144\text{Nd}}$	$\frac{143\text{Nd}}{144\text{Nd}}$	$\epsilon_{\text{Nd}}(t)$	U (ppm)	Th (ppm)	Pb (ppm)	$\frac{206\text{Pb}}{204\text{Pb}}$	$\frac{207\text{Pb}}{204\text{Pb}}$	$\frac{208\text{Pb}}{204\text{Pb}}$	$\frac{206\text{Pb}}{204\text{Pb}}$	$\frac{207\text{Pb}}{204\text{Pb}}$	$\frac{208\text{Pb}}{204\text{Pb}}$
11–100	12–1, 129–132	11.5	91	0.70430	0.70341	2.3	6.3	0.512980	0.512758	6.2	0.078	0.30	0.61	18.079	15.513	37.793	17.890	15.504	37.552
	13–2, 99–101	0.7	82	0.70362	0.70357	1.7	4.2	0.513034	0.512782	6.7	0.041	0.15	0.47	18.197	15.518	37.921	18.067	15.511	37.767
11–105	41–3, 146–149	0.9	48	0.70323	0.70311	2.3	5.8	0.513066	0.512823	7.5	0.025	0.10	0.29	18.251	15.531	37.990	18.118	15.525	37.818
	43–1, 82–85	1.2	90	0.70318	0.70310	2.4	5.9	0.513065	0.512819	7.4	0.028	0.12	0.58	18.189	15.524	37.990	18.114	15.520	37.885
39–355	21–1, 100–103	3.7	94	0.70272	0.70259	4.0	10.9	0.513105	0.512990	8.9	0.14	0.16	0.39	18.863	15.529	38.133	18.586	15.516	38.026
	22–5, 61–67	0.92	103	0.70267	0.70264	4.2	11.4	0.513121	0.513005	9.2	0.072	0.16	0.48	18.755	15.519	38.111	18.637	15.513	38.023
41–367	39–1, 63–67	1.8	116	0.70359	0.70349	3.7	10.2	0.513019	0.512803	7.0	0.68	0.29	0.26	22.335	15.755	38.198	18.185	15.551	37.616
	40–1, 66–72	0.71	97.3	0.70348	0.70343	4.3	11.8	0.512995	0.512781	6.6	0.75	0.46	0.43	21.000	15.610	38.184	18.299	15.477	37.647
43–384	22–2, 17–22	1.6	197	0.70316	0.70311	4.0	12.8	0.513050	0.512896	8.2	0.32	0.27	0.48	18.515	15.494	38.061	17.697	15.454	37.839
43–386	66–1, 101–105	9.3	91	0.70277	0.70230	3.6	10.0	0.513148	0.512992	9.7	0.040	0.14	0.61	18.416	15.566	38.076	18.344	15.562	37.998
	66–CC, 137–140	0.09	132	0.70235	0.70234	2.9	8.2	0.513187	0.513031	10.4	0.042	0.13	0.30	18.473	15.485	37.756	18.321	15.478	37.605
43–387	50–1, 20–23	0.82	118	0.70336	0.70332	2.0	5.6	0.512988	0.512791	6.5	0.14	0.25	0.32	18.774	15.555	38.203	18.128	15.523	37.836
	50–2, 107–113	0.18	95	0.70336	0.70335	2.0	5.5	0.513007	0.512811	6.9	0.12	0.29	0.38	18.585	15.562	38.258	18.116	15.539	37.901
51–417D	54–4, 10–13 (g)	1.7	89	0.70316	0.70307	3.7	9.5	0.513112	0.512928	8.7	0.028	0.09	0.35	18.594	15.572	38.293	18.498	15.568	38.189
	66–6, 44–47 (g)	1.4	109	0.70323	0.70316	3.6	9.3	0.513117	0.512931	8.7	0.073	0.11	0.36	18.380	15.545	37.945	18.142	15.534	37.832
51–418A	42–3, 25–28 (g)	1.1	92	0.70306	0.70300	2.6	6.5	0.513133	0.512947	9.0	0.016	0.061	0.25	18.268	15.529	37.905	18.192	15.526	37.810
76–534A	129–1, 26–29	0.94	85	0.70349	0.70342	2.0	5.5	0.513033	0.512808	7.2	0.065	0.21	0.25	18.207	15.532	38.094	17.805	15.512	37.666
	129–4, 136–140	0.28	84	0.70346	0.70344	2.1	5.6	0.513017	0.512788	6.8	0.052	0.18	0.19	18.369	15.554	38.229	17.936	15.533	37.748
78A–543A	13–4, 3–7 (g)	1.19	111	0.70289	0.70285	3.4	10.0	0.513081	0.512972	8.5	0.062	0.14	0.44	18.638	15.541	38.143	18.520	15.535	38.059
	16–1, 72–79 (g)	1.42	124	0.70287	0.70284	4.0	11.7	0.513079	0.512971	8.5	0.067	0.14	0.48	18.571	15.587	38.265	18.452	15.581	38.181

All isotope ratio measurements were performed on a VG Sector 54 thermal ionization mass spectrometer at the Scripps Institution of Oceanography. Our measured values for the NBS 987 (Sr) and La Jolla (Nd) isotopic standards are  $\frac{87\text{Sr}}{86\text{Sr}} = 0.71026$  and  $\frac{143\text{Nd}}{144\text{Nd}} = 0.511859$  with total measured ranges of  $\pm 0.000026$  and  $\pm 0.000014$ , respectively. A 1‰/amu fractionation correction was applied to Pb isotope ratios, based on repeat measurements of NBS 981, to conform to the values of Todt et al. [16]. External uncertainties for  $\frac{206\text{Pb}}{204\text{Pb}}$  and  $\frac{207\text{Pb}}{204\text{Pb}}$  are less than  $\pm 0.01$ , and for  $\frac{208\text{Pb}}{204\text{Pb}}$  are less than  $\pm 0.03$ . Total procedural blanks for Sr, Nd and Pb isotope measurements are 20, 5 and 50 pg, respectively. Parent–daughter elemental concentrations were obtained by quadrupole ICP-MS, on the same specially treated sample material used for isotope ratio analysis. Replicate analysis of rock standards indicates that Sr concentrations are precise to within 2.5%, Rb, Sm and Nd are precise to within 5% and U, Th and Pb are precise to within 8%. Initial (age-corrected) isotope ratios were calculated assuming the numerical sample ages given in Table 1.  $\epsilon_{\text{Nd}}(t)$  values are calculated for age-corrected sample and CHUR. Nd isotope ratios ( $^{142}\text{Nd}/^{144}\text{Nd}$ ) were calculated for age-corrected sample and CHUR ( $t = 0$ ) = 0.512638,  $^{147}\text{Sm}/^{144}\text{Nd}_{\text{CHUR}} = 0.1967$ . A (g) sample suffix indicates that fresh glass chips were used for analysis.

Table 3  
Mean trace element concentrations for basalts from central Atlantic Mesozoic oceanic crust

DSDP Hole:	100	105	355	367	384	386	387	417D (g)	418A (g)	543A (g)	534A	BHVO-1		
No. of samples:	3	5	3	2	2	2	2	5	1	4	3	Mean	S.D.	Ref. val.
Rb (ppm)	2.97	10	6.4	1.1	2.3	5.7	1.4	1.9	1.1	1.3	3.1	9.5	0.049	9.5
Sr	98.3	114	98.7	121	159	138	142	97.0	92.0	118	159	397	9.9	390
Y	22.5	24.4	41.4	44.6	28.5	29.1	20.3	36.7	30.7	35.6	23.5	30.0	0.72	28
Zr	44.9	57.5	112	103	127	90.7	50.1	87.7	63.2	108	49.4	180	1.3	180
Nb	2.9	1.4	3.0	5.9	5.5	2.5	3.8	2.1	1.2	3.0	3.0	19.6	0.15	19.5
Ba	12.7	23.7	7.33	108	29.8	37.8	24.6	11.2	5.22	9.38	16.1	133	2.4	133
La	1.84	1.53	3.39	4.37	5.07	2.98	2.64	2.47	1.60	3.58	2.26	15.6	0.17	15.5
Ce	5.11	5.18	11.1	12.1	14.8	9.26	6.66	8.46	5.70	11.4	6.16	39.7	1.5	38
Pr	0.88	0.91	2.00	2.02	2.45	1.66	1.07	1.60	1.12	2.01	1.06	5.46	0.09	5.45
Nd	4.9	5.2	11.0	11.0	12.7	9.1	5.6	9.1	6.5	10.8	5.7	24.7	0.39	24.7
Sm	1.9	2.1	4.0	4.0	4.0	3.3	2.0	3.5	2.6	3.7	2.1	6.10	0.06	6.17
Eu	0.78	0.76	1.4	1.4	1.5	1.2	0.78	1.3	1.0	1.3	0.79	2.05	0.05	2.06
Gd	2.9	3.1	5.7	6.0	4.9	4.6	2.8	5.0	3.8	5.2	3.0	6.08	0.10	6.22
Tb	0.55	0.60	1.1	1.1	0.85	0.81	0.53	0.93	0.73	0.91	0.56	0.93	0.01	0.95
Dy	3.6	3.9	6.8	7.1	5.1	5.0	3.4	6.2	4.9	5.9	3.7	5.12	0.06	5.25
Ho	0.80	0.90	1.5	1.6	1.1	1.1	0.76	1.4	1.1	1.3	0.83	0.97	0.07	1
Er	2.4	2.7	4.3	4.6	2.9	3.0	2.2	4.1	3.2	3.8	2.5	2.47	0.12	2.56
Tm	0.36	0.41	0.65	0.70	0.41	0.42	0.33	0.60	0.49	0.55	0.37	0.245	0.004	n/a
Yb	2.4	2.6	4.1	4.4	2.4	2.5	2.1	3.8	3.1	3.5	2.4	1.91	0.09	1.98
Lu	0.36	0.39	0.60	0.68	0.35	0.36	0.32	0.57	0.46	0.51	0.36	0.265	0.003	0.278
Hf	1.2	1.6	3.0	2.8	3.2	2.4	1.4	2.5	1.8	2.8	1.4	4.30	0.034	4.3
Ta	0.16	0.08	0.17	0.33	0.30	0.15	0.21	0.12	0.07	0.18	0.17	1.20	0.007	1.2
Pb	0.44	0.46	0.41	0.42	0.58	0.44	0.30	0.35	0.25	0.45	0.29	2.12	0.056	2.1
Th	0.20	0.11	0.17	0.41	0.36	0.14	0.26	0.10	0.061	0.14	0.20	1.27	0.032	1.26
U	0.11	0.032	0.10	0.77	0.38	0.28	0.12	0.044	0.016	0.063	0.064	0.417	0.007	0.42

Data were collected in peak-jumping mode by inductively coupled plasma mass spectrometry on a VG PlasmaQuad 2+ instrument at the Massachusetts Institute of Technology. Analyses were performed using the method of Janney and Castillo [13], except that dissolved rock standards (BIR-1, DNC-1, W-2 and an in-house MORB glass standard) were used instead of elemental standard solutions for calibration. Data are reported relative to the USGS rock standard concentrations reported by Eggins et al. [24]. The BHVO-1 values represent 10 replicate analyses interspersed with the samples, reference values are from [24]. Data indicated with a (g) were obtained on glass chips. The full data set is available as an EPSL Online Background Data Set or by request from the first author.

[12] (although OJP basalts have, on average, slightly higher  $^{87}\text{Sr}/^{86}\text{Sr}$ ,  $\Delta 8/4$  and lower  $\epsilon_{\text{Nd}}(t)$  values). Pre-120 Ma Atlantic crust does not, interestingly, show any isotopic similarity to young central Atlantic ocean island basalts (cf. [20–22]). Both continental lithospheric and plume contamination, therefore, appear to be plausible explanations of the distinct isotopic compositions of the pre-120 Ma Atlantic crust.

Trace element data provide more definitive evidence of the origin of this distinct geochemical signature in the oldest sampled Atlantic crust (Table 3). All pre-120 Ma crustal basalts are less depleted in highly incompatible elements (e.g., Th, Nb and the light rare earth elements (REE))

than young Atlantic N-MORB and all of the older samples, except those from Site 105, display moderate to strong positive Nb anomalies on primitive mantle-normalized diagrams (Fig. 3a). In contrast, the post-120 Ma crustal samples all display strongly incompatible element-depleted patterns essentially identical to young Atlantic N-MORB (Fig. 3b). CAMP tholeiites typically have mild to strong enrichments in most incompatible elements but these basalts are almost universally characterized by negative anomalies in Nb and often in other high field-strength elements, such as Ti and Zr (e.g., [9,17]; Fig. 3d). These features of CAMP basalts, as well as their radiogenic Sr and unradiogenic Pb and Nd iso-

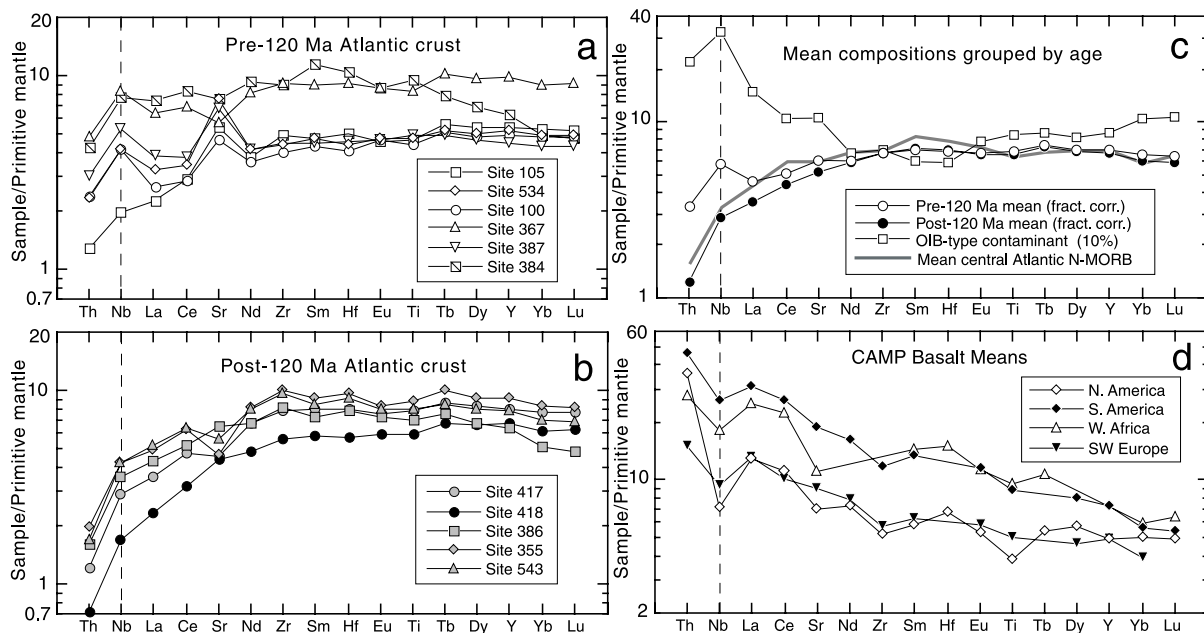


Fig. 3. Primitive mantle-normalized incompatible element patterns for (a) pre-120 Ma and (b) post-120 Ma Atlantic crust, (c) mean fractionation-corrected (to 8 wt% MgO) Atlantic crust compositions grouped by age and (d) mean CAMP tholeiite compositions from North and South America, West Africa and southwestern Europe. Pattern for the 'OIB-type contaminant' (panel c) was calculated assuming that the trace element differences between mean pre- and post-120 Ma Atlantic crust are due to addition of 10% of a contaminant material to the mean post-120 Ma crustal composition (the enrichment in the heavy REE is most likely an artifact of differences in mean depths and degrees of melting between the two groups of Atlantic crust). The pattern for mean central Atlantic MORB was calculated from a compilation of recently collected data, all included samples having chondrite-normalized La/Sm < 1 (main data sources were [23,25]). Sources of data for CAMP basalts are the same as in the Fig. 2 caption, with the addition of [26]. Nb data were not measured on West African CAMP basalts [19], so Ta concentrations were substituted (multiplied by a chondritic Nb/Ta ratio of 17).

tope ratios, have been widely attributed to derivation from mantle sources dominated by ancient continental lithospheric mantle having a history of metasomatism by subduction zone fluids, possibly occurring during previous ocean basin closure events (e.g., [4,10,19]). If these trace element and isotopic characteristics of CAMP magmas are indeed lithospheric mantle source features, then the mild EM 1 isotopic signatures and positive Nb anomalies of pre-120 Ma Mesozoic Atlantic crust cannot be the result of contamination by continental lithosphere.

The trace element characteristics of the pre-120 Ma Atlantic crust are best explained by the variable addition of an OIB-type component enriched in highly incompatible elements, and having a mild positive niobium anomaly, to a depleted, N-MORB-type mantle source (Fig. 3c). This hy-

pothesis is supported by fairly strong correlations between Nd isotopes and Nb/La and Nb/Zr ratios ( $r^2 \approx -0.7$ ) in the older Atlantic crust samples. The trace element characteristics of this component, interestingly, are distinct from those of OJP basalts, which have relatively flat primitive mantle-normalized trace element patterns and no Nb anomalies [12]. The OIB-like contaminant is not detectable in the trace element or isotopic compositions of the post-120 Ma basalts (with the possible exception of samples from Sites 417/418 (118 Ma), which have slightly elevated  $^{87}\text{Sr}/^{87}\text{Sr}$  and  $\Delta 7/4$  values). Most likely, this component had largely dissipated in the Atlantic upper mantle by 110–120 Ma, due to dilution by normal, depleted upper mantle flowing into the expanding sub-Atlantic region from beneath the neighboring continents.

#### 4. Conclusions and implications

Unlike the South and far North Atlantic, where pre- and syn-rifting flood basalt volcanism can clearly be traced to the action of the still-active Tristan and Iceland mantle plumes, respectively (e.g., [27,28]), there is no direct evidence linking the CAMP magmatism to modern plume activity [1]. The apparent center of magmatism, as indicated by the radial orientation of CAMP dikes (located between the former positions of northernmost Florida and Senegal), is very near the location of the Cape Verde hotspot in the mantle reference frame [26], but neither this hotspot, nor any other in the central Atlantic, appears to have produced volcanism older than the latest Cretaceous [1,20]. The chemical and isotopic plume signature detected in early Atlantic crust is strongest in this area (i.e., at Sites 100, 367, 387 and 534), but the  $^{206}\text{Pb}/^{204}\text{Pb}$  ratios of these samples are too low, relative to their moderately high Sr and low Nd isotope ratios, to be directly attributable to mixing with the Cape Verde or other central Atlantic plume sources (e.g., [20–22]).

The lack of clear temporal/spatial and geochemical connections between CAMP magmatism and long-lived central Atlantic hotspot volcanism is difficult to reconcile with classical mantle plume theory, and with the ‘starting plume head-plume tail’ mechanism (e.g., [29]) widely invoked to explain the formation of continental flood basalts and oceanic plateaus. However, the passive tectonic explanations for CAMP magmatism and super-continent breakup, specifically thermal insulation of the upper mantle producing shallow upwelling and volcanism aided by edge-driven upper mantle convection (e.g., [1,30,31]), cannot alone explain the presence of a plume-like component over more than 2500 km of the central Atlantic upper mantle in the Late Jurassic and Early Cretaceous.

A possible solution to this quandary may be found in models of mantle circulation incorporating an endothermic phase transition at the 660 km boundary (e.g., [32,33]), which may act as a partial barrier to convection. The numerical model of Liu et al. [33] predicts that the negative Clapeyron slope of the phase transition at 660 km should

impede plumes arising from the core–mantle boundary (CMB), with the effect of either entirely separating an upwelling plume-head from its plume conduit or attenuating the conduit to such an extent that it would be unlikely to produce significant volcanism at the surface. One such plume-head or diapir separated from a large central Atlantic plume might account for the absence of a low-volume volcanic trace following the widespread burst of CAMP volcanism at 200 Ma. However, this does not explain why the Tristan and Iceland plumes apparently did not suffer plume-head separation (although the Iceland plume may have temporarily stalled at the 660 km boundary [34]) and have long-lived, temporally continuous volcanic traces following flood basalt emplacement. Moreover, an experimental investigation of the plume-head separation hypothesis [35] predicts that an abandoned plume conduit should form a second, smaller plume-head which would impact the lithosphere and produce volcanism at least 10 Myr after the first. There is little evidence for such a second pulse of magmatism on this timeframe in CAMP basaltic rocks exposed on land [2,3] (however, the sparsely sampled complex of seaward-dipping basaltic reflectors off the east coast of North America may be up to 17 Myr younger than the main phase of CAMP activity [36]).

A second, and likely more appropriate, model for Early Mesozoic plume activity in the central Atlantic involves large convective upwellings generated in the upper part of the lower mantle. In the numerical simulation of Cserepes and Yuen [32], general convective transport of warm material from the lower to the upper mantle is impeded by the endothermic phase transition at 660 km. However, when a sufficiently large mass of warm, low-density mantle accumulates in an area beneath this boundary layer, its positive buoyancy anomaly becomes great enough to break through the boundary layer in a catastrophic manner, forming a large, plume-head-like feature (a ‘mid-mantle plume’ [32]) in the upper mantle. This phenomenon is complementary to the cold ‘avalanches’ of accumulated subducted oceanic lithosphere envisioned to fall through the 660 km boundary at irregular intervals (e.g., [37]) and

are distinct from conventional mantle plumes in that they are not rooted at a mantle boundary layer, such as the CMB.

Upon breaking into the upper mantle, mid-mantle plumes may constitute features as large as or larger than the plume-heads believed to be formed by starting plumes originating from the CMB [32,38], but since these features are not rooted at a boundary layer, their lifetimes would be relatively short compared to plumes arising from the CMB (L. Cserepes, personal communication, 2001). Because the mid-mantle plume mechanism is most consistent with the temporally short but spatially broad pattern of Early Mesozoic magmatism in the central Atlantic region, we propose that this phenomenon is the most likely cause of the widespread contamination of the nascent Atlantic upper mantle in the Early Mesozoic, and was likely the key factor in the formation of the CAMP and possibly in the initiation of Pangaean continental rifting.

It must be pointed out, however, that a major difficulty exists in generating the roughly 3500 by 2000 km CAMP by a single mid-mantle plume feature originating from a depth only on the order of 1000 km. For plume-heads or diapirs impinging on continental lithosphere, significant ( $> 1\%$ ) pressure-release melting will only occur over an area of approximately the same diameter as the plume feature before it undergoes flattening at the base of the lithosphere [39], which is on the order of 500–1000 km for a mid-mantle plume [32]. Indeed, this constraint makes the nearly instantaneous creation of such a large igneous province purely by decompression melting implausible even for the largest classical starting plumes arising from the CMB [38].

More high-precision radiometric dating studies of the CAMP must be conducted to determine whether the entire province formed simultaneously, or if there was a spatial progression of volcanism over a few million years. This information, in combination with detailed geophysical modeling, will be necessary to determine if a mid-mantle plume is indeed the best explanation for the long history of uplift and rifting and the short interval of widespread, effusive magmatism associated with continental breakup in the central

Atlantic. Additionally, a more complete (and age-corrected) Pb isotopic data set for CAMP basalts is needed to determine whether CAMP basalts have sampled the same ‘central Atlantic plume’ component expressed in pre-120 Ma Atlantic oceanic crust.

### Acknowledgements

This work was supported by NSF Grants OCE 9617676 and 9731045 to P.R.C. We thank Angelo De Min for sharing unpublished data in a preprint and Fred Frey and the Ocean Drilling Program, East Coast Repository for providing sample materials. We are grateful for the assistance of Chris MacIsaac and Barry Grant in the laboratory. Discussions with Fred Anderson, Munir Humayun, David Rowley and Igor Puchtel at a University of Chicago seminar on this topic were very helpful. Peter Olson, Godfrey Fitton and John Mahoney provided careful reviews that improved the accuracy and clarity of the manuscript. [AH]

### References

- [1] J.G. McHone, Non-plume magmatism and rifting during the opening of the central Atlantic Ocean, *Tectonophysics* 316 (2000) 287–296.
- [2] A. Marzoli, P.R. Renne, E.M. Piccirillo, M. Ernesto, G. Bellieni, A. DeMin, Extensive 200-million-year-old continental flood basalts of the Central Atlantic Magmatic Province, *Science* 284 (1999) 616–618.
- [3] P. Olsen, Giant lava flows, mass extinctions and mantle plumes, *Science* 284 (1999) 604–605.
- [4] A. DeMin, E.M. Piccirillo, A. Marzoli, G. Bellieni, P.R. Renne, M. Ernesto, L.S. Marques, The Central Atlantic Magmatic Province (CAMP) in Brazil: Petrology, geochemistry,  $^{40}\text{Ar}/^{39}\text{Ar}$  ages, paleomagnetism and geodynamic implications, in: W.E. Hames, J.G. McHone, P.R. Renne, C. Ruppel (Eds.), *The Central Atlantic Magmatic Province*, Geophys. Monogr. AGU, Washington, DC, in press.
- [5] W.S. Holbrook, P.B. Kelemen, Large igneous province on the US Atlantic margin and implications for magmatism during continental breakup, *Nature* 364 (1993) 433–436.
- [6] P.E. Olsen, Stratigraphic record of the early Mesozoic breakup of Pangea in the Laurasia-Gondwana rift system, *Annu. Rev. Earth Planet. Sci.* 25 (1997) 337–401.
- [7] R.S. White, D. McKenzie, Magmatism at rift zones: the

- generation of volcanic continental margins and flood basalts, *J. Geophys. Res.* 94 (1989) 7685–7729.
- [8] M. Wilson, Thermal evolution of the central Atlantic passive margins: continental breakup above a Mesozoic super-plume, *J. Geol. Soc.* 154 (1997) 491–495.
- [9] C. Alibert, A Sr–Nd isotope and REE study of late Triassic dolerites from the Pyrenees (France) and the Messenjana Dyke (Spain and Portugal), *Earth Planet. Sci. Lett.* 73 (1985) 81–90.
- [10] W.J. Pegrarn, Development of continental lithospheric mantle as reflected in the chemistry of the Mesozoic Appalachian Tholeiites, USA, *Earth Planet. Sci. Lett.* 97 (1990) 316–331.
- [11] W.B. Harland, R.L. Armstrong, A.V. Cox, L.E. Craig, *A Geologic Time Scale 1989*, Cambridge University Press, New York, 1990.
- [12] J.J. Mahoney, M. Storey, R.A. Duncan, K.J. Spencer, M. Pringle, Geochemistry and age of the Ontong Java Plateau, in: M.S. Pringle, W.W. Sager, W.V. Sliter, S. Stein (Eds.), *The Mesozoic Pacific: Geology, Tectonics and Volcanism*, *Geophys. Monogr.* 77, AGU, Washington, DC, 1993, pp. 233–261.
- [13] P.E. Janney, P.R. Castillo, Basalts from the Central Pacific Basin: evidence for the origin of Cretaceous igneous complexes in the Jurassic western Pacific, *J. Geophys. Res.* 101 (1996) 2875–2893.
- [14] S.R. Hart, A large-scale anomaly in the Southern Hemisphere mantle, *Nature* 309 (1984) 753–757.
- [15] J.D. Macdougall, R.C. Finkel, J. Carlson, S. Krishnaswami, Isotopic evidence for uranium exchange during low-temperature alteration of oceanic basalt, *Earth Planet. Sci. Lett.* 42 (1979) 27–34.
- [16] W. Todt, R.A. Cliff, A. Hanser, A.W. Hofmann, Evaluation of a  $^{202}\text{Pb}$ – $^{205}\text{Pb}$  double spike for high-precision lead isotope analysis, in: A. Basu, S.R. Hart (Eds.), *Earth Processes: Reading the Isotopic Code*, *Geophys. Monogr.* 95, AGU, Washington, DC, 1996, pp. 429–437.
- [17] G. Bellieni, M.H.F. Macedo, R. Petrini, E.M. Piccirillo, G. Cavazzini, P. Comin-Chiaromonti, M. Ernesto, J.W.P. Macedo, G. Martins, A.J. Melfi, I.G. Pacca, A. DeMin, Evidence of magmatic activity related to Middle Jurassic and Lower Cretaceous rifting from northwestern Brazil (Ceará-Mirim): K/Ar age, palaeomagnetism, petrology and Sr–Nd isotope characteristics, *Chem. Geol.* 97 (1992) 9–32.
- [18] J. Dostal, M. Durning, Geochemical constraints on the origin and evolution of early Mesozoic dikes in Atlantic Canada, *Eur. J. Mineral.* 10 (1998) 79–93.
- [19] C. Dupuy, J. Marsh, J. Dostal, A. Michard, S. Testa, Asthenospheric and lithospheric sources for Mesozoic dolerites from Liberia (Africa): trace element and isotopic evidence, *Earth Planet. Sci. Lett.* 87 (1988) 100–110.
- [20] D.C. Gerlach, R.A. Cliff, G.R. Davies, M.J. Norry, N. Hodgson, Magma sources of the Cape Verdes archipelago: Isotopic and trace element constraints, *Geochim. Cosmochim. Acta* 52 (1988) 2979–2992.
- [21] K.A. Hoernle, G. Tilton, H.U. Schminke, Sr–Nd–Pb isotopic evolution of Gran Canaria: evidence for shallow enriched mantle beneath the Canary Islands, *Earth Planet. Sci. Lett.* 106 (1991) 44–63.
- [22] B.D. Taras, S.R. Hart, Geochemical evolution of the New England seamount chain: isotopic and trace-element constraints, *Chem. Geol.* 64 (1987) 35–54.
- [23] L. Dosso, H. Bougault, J.-L. Joron, Geochemical morphology of the north Mid-Atlantic Ridge, 10°–20°N: trace element–isotope complementarity, *Earth Planet. Sci. Lett.* 120 (1993) 443–462.
- [24] S.M. Eggins, J.D. Woodhead, L.P.J. Kinsley, G.E. Mortimer, P.J. Sylvester, M.T. McCulloch, J.M. Hergt, M.R. Handler, A simple method for the precise determination of >40 trace elements in geological samples by ICPMS using enriched isotope internal standardisation, *Chem. Geol.* 134 (1997) 311–326.
- [25] K. Lawson, R.C. Searle, J.A. Pearce, P. Browning, P.D. Kempton, Detailed volcanic geology and geochemistry of the MARNOCK area (Mid-Atlantic Ridge north of Kane Transform), in: C.J. MacLeod, P.A. Taylor, C.L. Walker (Eds.), *Tectonic, Magmatic, Hydrothermal and Biological Segmentation of Mid-Ocean Ridges*, *Geol. Soc. Spec. Publ.* 118, Blackwell Scientific., Oxford, 1996, pp. 61–102.
- [26] E.P. Oliviera, J. Tarney, X.J. João, Geochemistry of the Mesozoic Amapá and Jari dyke swarms, northern Brazil: plume-related magmatism during the opening of the central Atlantic, in: A.J. Parker, P.C. Rickwood, D.H. Tucker (Eds.), *Mafic Dykes and Emplacement Mechanisms*, Balkema, Rotterdam, 1990, pp. 173–183.
- [27] D.W. Peate, The Paraná-Etendeka Province, in: J.J. Mahoney, M.F. Coffin (Eds.), *Large Igneous Provinces*, *Geophys. Monogr.* 100, AGU, Washington, DC, 1998, pp. 217–246.
- [28] A.D. Saunders, J.G. Fitton, A.C. Kerr, M.J. Norry, R.W. Kent, The North Atlantic Igneous Province, in: J.J. Mahoney, M.F. Coffin (Eds.), *Large Igneous Provinces*, *Geophys. Monogr.* 100, AGU, Washington, DC, 1998, pp. 45–94.
- [29] M.A. Richards, R.A. Duncan, V.E. Courtillot, Flood basalts and hot-spot tracks plume heads and tails, *Science* 246 (1989) 103–107.
- [30] S.D. King, D.L. Anderson, An alternative mechanism of flood basalt formation, *Earth Planet. Sci. Lett.* 136 (1995) 269–279.
- [31] P.J. Tackley, Mantle convection and plate tectonics: toward an integrated physical and chemical theory, *Science* 288 (2000) 2002–2007.
- [32] L. Cserepes, D.A. Yuen, On the possibility of a second kind of mantle plume, *Earth Planet. Sci. Lett.* 183 (2000) 61–71.
- [33] M. Liu, D.A. Yuen, W. Zhao, S. Honda, Development of diapiric structures in the upper mantle due to phase transitions, *Science* 252 (1991) 1836–1839.
- [34] J.G. Fitton, A.D. Saunders, M.J. Norry, B.S. Hardarson, R.N. Taylor, Thermal and chemical structure of the Iceland plume, *Earth Planet. Sci. Lett.* 153 (1997) 197–208.

- [35] D. Bercovici, J.J. Mahoney, Double flood basalts and plume head separation at the 660-km discontinuity, *Science* 266 (1994) 1367–1369.
- [36] M.A. Lanphere,  $^{40}\text{Ar}/^{39}\text{Ar}$  ages of basalt from the Clubhouse Crossroads Test Hole #2, near Charleston, South Carolina, USGS Prof. Paper 1313B (1983) 1–8.
- [37] P.J. Tackley, D.J. Stevenson, G.A. Glatzmaier, G. Schubert, Effects of an endothermic phase transition at 670 km depth in a spherical model of convection in the Earth's mantle, *Nature* 361 (1993) 699–704.
- [38] R.W. Griffiths, I.H. Campbell, Stirring and structure in mantle starting plumes, *Earth Planet. Sci. Lett.* 99 (1990) 66–78.
- [39] P. Olson, Mechanics of flood basalt magmatism, in: M.P. Ryan (Ed.), *Magmatic Systems*, Academic Press, New York, 1994, pp. 1–18.