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U–Th isotopes in Hainan basalts: Implications for sub-asthenospheric origin of EM2 mantle endmember and the dynamics of melting beneath Hainan Island

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ABSTRACT

Extensive (about 5000 km³) basaltic magmas erupted on the Hainan Island, south China, mostly during the past 1 million years. U–Th disequilibrium data as well as Nd, Sr, Pb isotopes and major and trace element concentrations were measured on the youngest lavas from Maanling volcano and Leihuling volcano of the Hainan Island. All the Holocene Hainan basalts display light rare earth element (LREE) enriched patterns and ocean island basalt (OIB)-type incompatible element distributions. Their $\varepsilon_{\rm Nd}$ values range from +4.1 to +4.8, ⁸⁷Sr/⁸⁶Sr ratios vary from 0.7039 to 0.7042, and ²⁰⁶Pb/²⁰⁴Pb ratios range from 18.63 to 18.71. The Hainan lavas are characterized by their depleted Sr–Nd isotopic compositions and Dupal-like EM2 (enriched mantle 2) Pb isotope signatures with time-integrated high Th/U and ²³⁵U/Pb.

The olivine tholeiites from Maanling display 18–20% ²³⁰Th excesses and the alkali olivine basalts from Leihuling show 22–32% ²³⁰Th excesses. The pronounced ²³⁰Th excesses in the Holocene basalts indicate that the Holocene Hainan lavas were produced by melting of a mantle source in the garnet stability field (>75 km). Since the lithosphere thickness beneath the Hainan Island is thin (55 km), the garnet peridotite mantle source for the Hainan basalts is not located in the lithospheric mantle. The Nd isotopic compositions do not indicate a highly depleted asthenospheric mantle source. We thus suggest that the EM2 mantle source for the young Hainan basalts is in the mantle transition zone or more likely lower mantle, which is consistent with a plume origin. The significant ²³⁰Th excesses also suggest slow (<1 cm/year) upwelling, possibly indicative of a weakly buoyant mantle plume. The older EM2 Cenozoic basalts from Hainan, South China Sea Basin and adjacent areas may also originate from partial melting of lower mantle materials in the rising Hainan plume.

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1. Introduction

The volcanic eruptions in the northern Hainan Island (20°N, 110°E), south China, produced about 5000 km³ basaltic magmas (Fig. 1). Although the eruptions may have initiated more than 1 million years ago, most of the basalts were produced over a time interval of 200,000 to 500,000 years (Flower et al., 1992). The average magma supply of 0.1 to 0.25 km³/year approaches that of a major flood basalt episode (Flower et al., 1992). The Hainan eruptions are part of a regional magmatic episode surrounding the South China Sea Basin and nearby regions, such as Vietnam (Hoang and Flower, 1998), Thailand (Zhou and Mukasa, 1997) and SE China (Zou et al., 2000). These volcanic rocks surrounding the South China Sea Basin, and those from seamounts within the South China Sea Basin, are younger than the South China Sea Basin (at 30 to 16 million years ago).

The Hainan lavas, located at the northern edge of the South China Sea Basin, may present a microcosm of the volcanic activity surrounding the South China Sea Basin (Flower et al., 1992). It has been recognized (Tu et al., 1991; Flower et al., 1992) that (1) chemically the Hainan basalts resemble the oceanic island basalts (OIB) with enriched OIB-type incompatible element distributions, and (2) isotopically the Hainan basalts are characterized by the intriguing Dupal-like Pb isotopic signatures and depleted Sr-Nd isotopic compositions. There are still debates about the origins of the Hainan volcanoes, with implications for the origins of the 'Southern Hemisphere' Dupal anomaly in the Northern Hemisphere Hainan basalts (Tu et al., 1991; Tu et al., 1992; Liu, 1999; Zhao, 2007). On a global scale, it is not clear whether such Dupal mantle reservoirs are mainly derived from shallow subcontinental lithospheric mantle (Hawkesworth et al., 1990; Tu et al., 1991) or from deep mantle (e.g., lower mantle) (Hart, 1984; Castillo, 1988; Hart et al., 1992).

To provide new insights into the origins of the Hainan basalts, additional geological tools (in addition to Nd, Sr and Pb isotopes and seismic images) are needed. Young basalts erupted ~9000 years (9 ka) ago at Maanling volcano and Leihuling volcano (Fan et al., 2004) in



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Fig. 1. Distribution of late Cenozoic basalts in the northern Hainan Island and Leizhou Peninsula, and sample locations. Shaded areas are late Cenozoic basalts. The dark shaded area represents locations of Holocene lavas.

Hainan Island. These young basalts provide ideal opportunities to apply short-lived U–Th disequilibrium to investigate the origins of the Hainan basalts. U–Th disequilibrium has been demonstrated as an effective tool for investigating the origins of young basalts (Asmerom and Edwards, 1995; Reid, 1995; Asmerom, 1999). Although EM2-like Cenozoic basalts are widespread in the South China Sea Basin and nearby regions, such as Vietnam (Hoang and Flower, 1998) and Thailand (Zhou and Mukasa, 1997) and SE China (Zou et al., 2000), few basalts are young enough for the short-lived U–Th disequilibrium studies.

Here we carry out a systematic U–Th disequilibrium, Nd, Sr and Pb isotope as well as major and trace element study of young lavas from Hainan Island. Our aims are (1) investigate the origins of the Hainan Holocene lavas and the possible link with plume volcanism and (2) constrain the melting conditions below Hainan Island. Obviously this U–Th disequilibrium study builds on pioneering Nd–Sr–Pb isotope studies of the Hainan basalts (Tu et al., 1991; Flower et al., 1992) and recent seismic tomographic imaging (Lebedev and Nolet, 2003; Huang and Zhao, 2006; Lei and Zhao, 2009).

2. Analytical methods

Thorium isotopic compositions of most samples were measured using a VG 54 mass spectrometer equipped with a WARP energy filter (VG54-WARP) and an ion counting system, except for 3 samples (HN106, HN119 and HN11) that were measured by a Cameca IMS 1270 ion microprobe at UCLA. The mass resolution for the ion probe IMS 1270 work is at 5000. The revised chemical method (Zou et al., 2008) to separate and purify thorium involves two columns (1) a 400 µl TRU Spec column to separate Th from other major and trace elements, and (2) a 100 µl anion resin column to purify Th. Major and trace elements were measured by ICP-MS at the GeoAnalytical Center at the Washington State University and the Institute of Geology and Geophysics, Chinese Academy of Sciences. Sample powders were prepared with an agate mill. Nd, Sr and Pb isotopic compositions as well as U and Th concentrations were measured using VG 54 WARP at UCLA. Analytical details on Nd, Sr and Pb isotopes and U and Th concentrations at UCLA using TIMS have been documented in Zou et al. (2003). Nd and Sr isotopic compositions were normalized to ¹⁴⁶Nd/ 144 Nd = 0.7219 and 86 Sr/ 88 Sr = 0.1194, respectively. The measured Nd and Sr isotope standard values are 143 Nd/ 144 Nd = 0.511843 ± 13 (*n*=24) for La Jolla and 87 Sr/ 86 Sr = 0.710239 ± 16 (*n*=13) for NBS 987. Replicate analyses of Pb isotope standard NBS 981 give 206 Pb/ 204 Pb = 16.896 ± 0.013, 207 Pb/ 204 Pb = 15.435 ± 0.014, and 208 Pb/ 204 Pb = 36.525 ± 0.041. Relative to the following values for NBS981: 206 Pb/ 204 Pb = 16.9356, 207 Pb/ 204 Pb = 15.4891, and 208 Pb/ 204 Pb = 36.7006 (Todt et al., 1996), Pb isotopic data in samples were corrected for mass fractionation of 0.118% per atomic mass unit (AMU) for 206 Pb/ 204 Pb, 0.117% per AMU for 207 Pb/ 204 Pb, and 0.119% per AMU for 208 Pb/ 204 Pb.

3. Results

We selected 10 samples for major, trace element and isotope analyses, including 3 Holocene samples from Maanling, 5 Holocene samples from Leihuling and 2 Pleistocene samples from Sanjiaoyuan (HN14) and Bopian (HN119) (Fig. 1). Major and trace element concentrations (in wt.%) are presented in Table 1. According to CIPW norms (Table 1), the Maanling samples are olivine tholeiites, and the Leihuling samples are mostly alkali olivine basalts with minor olivine tholeiites. As for the two older Pleistocene rocks, the Sanjiaoyuan sample (HN14) is olivine tholeiite and the Bopian sample (HN119) is quartz tholeiite.

The MgO content varies from 6.5% to 7.5% for Maanling tholeiites, and 9.6% to 12.8% for Leihuling alkali basalts, 9.1% for Sanjiaoyuan and 6.8% for Bopian. Their mg numbers (molar Mg/(Mg + Fe²⁺)) are 0.59 to 0.61 for Maanling, 0.65–0.66 for Leihuling, 0.67 for Sanjiaoyuan and 0.61 for Bopian. The Holocene basalts from Leihuling and Maanling have high TiO₂ contents, ranging from 2.3 to 2.8%. The Pleistocene lavas have lower TiO₂ (2.2–1.6%).

All basaltic rocks from Hainan Island are enriched in light rare earth elements (LREEs) over heavy rare earth elements (HREEs) (Fig. 2). None of the samples displays any negative Eu anomalies. In the spider diagram (Fig. 2), except for HN119, all samples show appreciable enrichment in high field strength elements (HFSEs), such as Nb and Ta.

The Sr, Nd and Pb isotope data show highly restricted range within each locality (Table 2). The $\varepsilon_{\rm Nd}$ values are +4.5 to +4.8 for Maanling, and +4.1 to +4.4 for Leihuling. The ${}^{87}{\rm Sr}/{}^{86}{\rm Sr}$ values range from 0.70385 to 0.70392 for Maanling, and 0.70418 to 0.70427 for Leihuling. The ${}^{206}{\rm Pb}/{}^{204}{\rm Pb}$ ratios are 18.10 to 18.63 for Maanling, and 18.66 to 18.70 for Leihuling.

The Nd isotope compositions indicate that source rocks for the Maanling and Leihuling lavas are moderately depleted and are homogeneous. The Hainan basalts are less depleted as compared to the Central Indian Ridge MORBS or some basalts from central East China (Fig. 3). In the ²⁰⁷Pb/²⁰⁴Pb vs. ²⁰⁶Pb/²⁰⁴Pb and ²⁰⁸Pb/²⁰⁴Pb vs. ²⁰⁶Pb/²⁰⁴Pb diagrams (Fig. 4), all samples plot above the Northern Hemisphere Reference Line (NHRL) (Hart, 1984) and thus display Dupal Pb anomalies (Tu et al., 1991; Flower et al., 1992). The Nd–Sr–Pb isotope data suggest that they are EM2 like basalts (Figs. 3 and 4).

The U–Th isotope results are presented on a $(^{230}\text{Th}/^{232}\text{Th})$ vs. $(^{238}\text{U}/^{232}\text{Th})$ equiline diagram in Fig. 5. All Holocene samples (Maanling and Leihuling) display significant ^{230}Th excesses (Fig. 5). The $(^{230}\text{Th}/^{238}\text{U})$ values are 1.18 to 1.20 for Maanling, and 1.23 to 1.32 for Leihuling. One Pleistocene basaltic sample (HN14) has $(^{230}\text{Th}/^{238}\text{U})$ value of 1.10 and another Pleistocene quartz tholeiite displays U–Th equilibrium.

4. Discussion

The Hainan samples appear to be relatively primitive basalts without significant crustal contamination or fractional crystallization. All the samples have high mg numbers (0.59 to 0.67) (Table 1). Their high mg numbers suggest that the samples closely reflect source compositions and mantle processes, and are not significantly affected

Table 1

Major element (in wt.%) and trace element (ppm) concentrations of Hainan basalts.

Sample	HN9901	HN9902	106B1	HN9907	HN9907*	HN9908	HN9910	HN9911	HN9912	HN9912*	HN9914	119B1
	OT	OT	OT	OT		AOB	AOB	AOB	AOB		OT	QT
Locality	Maanling	Maanling	Maanling	Leihuling		Leihuling	Leihuling	Leihuling	Leihuling		Sanjiaoyuan	Bopian
SiO ₂	51.74	50.77	50.99	49.10		45.32	45.47	45.14	45.27		49.87	53.14
TiO ₂	2.34	2.37	2.27	2.30		2.81	2.76	2.76	2.75		2.17	1.62
Al_2O_3	14.32	15.12	14.33	13.36		12.83	12.98	12.99	12.76		13.86	14.36
$Fe_2O_3^*$	10.64	10.42	10.97	11.42		12.79	12.67	11.52	12.63		10.41	10.07
MnO	0.15	0.25	0.14	0.27		0.20	0.31	0.19	0.20		0.15	0.14
MgO	6.53	6.81	7.47	9.63		10.29	10.17	9.90	10.29		9.08	6.80
CaO	8.92	9.24	9.28	9.46		10.57	10.80	10.43	10.46		9.37	8.53
Na ₂ O	3.41	2.82	2.71	2.55		2.85	2.42	2.95	3.12		3.12	3.03
K ₂ O	1.68	1.46	1.38	1.53		1.64	1.43	1.61	1.63		1.53	1.00
P_2O_5	0.47	0.47	0.47	0.48		0.86	0.82	0.75	0.79		0.42	0.23
Total	100.19	99.73	100.02	100.11		100.15	99.84	98.24	99.88		99.97	98.93
mg#	0.59	0.60	0.61	0.66		0.65	0.65	0.66	0.65		0.67	0.61
CIPW												
Quartz	0	0	0	0		0	0	0	0		0	2.55
Plagioclase	48.01	48.73	46.19	42.47		30.06	35.58	31.64	27.99		46.14	49.24
Orthoclase	9.99	8.75	8.27	9.1		9.81	8.57	9.81	9.75		9.1	6.03
Nepheline	0	0	0	0		6.41	3.11	6.49	8.1		0	0
Diopside	18.43	15.27	16.52	19.02		24.11	22.93	24.45	25.29		20	15.17
Hypersthene	10.04	17.51	19.74	7.24		0	0	0	0		1.01	21.85
Olivine	6.4	2.57	2.2	14.95		20.35	20.7	18.72	19.9		17.07	0
Ilmenite	4.48	4.56	4.35	4.43		5.37	5.32	5.39	5.28		4.16	3.13
Magnetite	1.55	1.52	1.61	1.67		1.87	1.86	1.73	1.86		1.52	1.49
Apatite	1.09	1.09	1.11	1.14		2.02	1.92	1.78	1.83		0.97	0.56
Cs	0.57	0.61	nd	0.42	0.40	0.46	0.71	0.82	0.71	0.71	0.39	0.52
RD	35.6	30.0	28.8	32.4	32.41	36.5	35.0	35.2	38.8	39.0	30.0	23.6
Ba	396	423	351	396	396	5/3	5/4	535	536	541	353	216
In	3.87	3.90	4.29	4.70	4.13	8.36	8.29	7.63	/./6	6./1	3.38	2.73
U	0.91	0.88	0.99	1.19	1.02	2.00	1.96	1.81	1.81	1.54	0.79	0.59
ND	33.61	33.70	41.06	46.11	51.09	84.08	83.90	/7.05	/9.05	86./1	33.78	16.40
la	2.04	2.04	2.44	2.80	3.51	5.20	5.25	4.//	4.79	5.61	2.05	0.98
La	24.93	24.94	29.52	33.26	35.91	64.82	64.09 125 5	58.88	59.40	63.74	22.30	14.02
Ce	49.4	49.6	57.6	65.2	66.3	125.7	125.5	114.8	116.5	117.1	43.4	27.5
PD Dr	3.03	3.33	4.06	3.33	3.39	5.19	D.27	3.98	4.84	4.91	2.48	2.79
PI Cr	6.20	0.20	/.38	7.83	7.87	14.79	14./1	13.55	13.38	13.30	5.40	3.44
SI	240	229	489	220	263	δ1/ 57.2	820	709	811 52.2	843 51 56	525 22.0	318 150
7.	20.9	27.1	109	32.0	105	200	37.0	32.5	32.2	202	25.0	10.2
	100	109	196	191	195	309	309	209	294	502	102	2.02
ni Sm	4.05	4.92	4.60	4.07	4.72	10.80	10.77	10.26	7.10	0.92	5.95	5.02
5111	0.94	0.85	0.04	7.07	2.10	2 25	2 27	2 15	2.24	9.9J 2 97	2.05	1 / 9
Cd	6.70	6.85	6.44	2.55	6.19	0.12	0.15	0.05	9.65	2.07	2.05	1.40
Th	1.02	1.01	0.44	0.52	0.18	5.12 1.20	1.20	0.05	1.00	1 1 2	0.88	4.50
Dv	5.52	5.51	5.22	5.25	0.87	6.96	6.94	6.65	6.53	5.80	0.00	0.75 //18
Но	0.08	0.96	0.01	0.04	0.82	1.23	1.21	1.18	1.16	1.02	0.86	0.78
Fr.	0.98	2.20	2.10	0.94	2.02	2.07	1.21	1.10	2 72	2.54	0.80	1.20
Vh	1.61	1.53	1.78	1.66	1.61	2.37	2.07	2.77	2.72	2.04	1.46	1.05
V	23.4	1.55	25.5	22.7	24.4	2.17	2.15	2.04	2.00	2.05	21.40	1.44
I III	0.22	23.5	23.5	0.24	0.24	0.32	0.32	0.30	0.31	0.31	0.22	0.21
Eu*	1.03	1.05	0.20	1.03	0.24	1.00	1.01	0.00	1.03	0.01	1.06	1.04
Eu Sr/Th	1.05	1.05	114	1.05	1.97	1.00	1.01	101	1.05	126	1.00	1.04
Da/Th	141	145	114 92	24	06	50	59	70	60	91	104	70
Dd/111	105	108	02	04	50	09	09	70	09	01	104	19

Eu anomaly is given by $Eu^* = 2Eu_N/(Sm_N + Gd_N)$, where N denotes normalized values by CI chondrite.

OT = olivine tholeiite; AOB = alkali olivine basalt; QT = quartz tholeiite. HN9907* and HN9912* are duplicates. $mg# = mg/(Mg + Fe^{2+})$, assuming $Fe^{3+}/Fe^{2+} = 0.15$. $Fe_2O_3^*$: total iron as Fe_2O_3 .

by continental contamination during magma ascent. In addition, strong ²³⁰Th excesses in Hainan basalts cannot be produced from crustal contaminations because bulk assimilation of crustal rocks with $(^{230}\text{Th}/^{238}\text{U}) = 1.0$ can only reduce the extent of ^{230}Th excesses.

4.1. Evidence against subduction-related fluid-induced melting

Our U–Th disequilibrium data, together with trace element data, do not indicate possible influence of subduction-related fluids in the mantle. Uranium is highly mobile in oxidizing aqueous fluids whereas thorium behaves as an immobile HFSE. The subduction-related fluids are thus highly enriched in U relative to Th. Lavas formed by recent subduction-related fluid-induced melting often display ²³⁸U excesses or U–Th equilibrium (Gill and Williams, 1990; Elliott et al., 1997; Turner et al., 2003), whereas the Hainan lavas display significant ²³⁰Th excesses. High field strength elements such as Nb and Ta behave similarly to the immobile Th during slab dehydration. The typical fluidinduced melting along magmatic arcs generates lavas with negative anomalies in Nb and Ta (e.g., Gill, 1981; Elliott, 2003). Hainan basalts display positive Nb and Ta anomalies (Fig. 2), which do not indicate the involvement of recent subduction-related fluids or sediments.

To sum up, the significant ²³⁰Th excesses and the positive anomalies in Nb and Ta, consistently argue against the involvement of subduction-related fluids to generate the Hainan lavas.



Fig. 2. Primitive mantle normalized trace element diagrams for Hainan lavas. Note positive Nb and Ta anomalies and steep REE patterns for the Holocene basalts. Sun and McDonough, 1989.

4.2. Depth and location of melting: evidence for plume melting

After ruling out the possibility of fluid-induced melting, we suggest that decompressional melting is the most likely mechanism to generate the Hainan basalts. An important question regarding decompressional melting is the depth and location of melting, which is critical for understanding the origin of the Hainan basalts.

The significant (18-32%) ²³⁰Th excesses in all samples from Maanling and Leihuling require mantle melting in the garnet stability field. This is due to the fact that garnet is the dominant mineral phase in the mantle where thorium is significantly more incompatible than uranium during mantle melting (Beattie, 1993; LaTourrette et al., 1993; Salters and Longhi, 1999). The steep REE patterns (Fig. 2) also indicate that garnet is a residual phase during mantle melting, in addition to the initial REE patterns of the source. In comparison, U-Th disequilibrium in basalts is independent of source composition as long as the source is in secular equilibrium $((^{230}\text{Th}/^{238}\text{U}) = 1)$ prior to melting. The secular equilibrium in the mantle prior to melting can be easily achieved in 0.35 million years (i.e., five half lives of ²³⁰Th). Thus the significant ²³⁰Th excesses in the young Hainan basalts significantly strengthen the argument of garnet involvement. Because the pressure for the spinel peridotite to garnet peridotite transition in the mantle is at 75 km, melting beneath Hainan Island took place at the depth of > 75 km.

Garnet signature from U–Th disequilibrium may not be exclusively limited to garnet peridotite, as garnet pyroxenites (with olivine content ranging from 0 to 40%) also contains garnet. Although tholeiites cannot be generated by garnet pyroxenites, it has been demonstrated that alkali basalts can be generated by melting of silica-poor garnet pyroxenite (Hirschmann et al., 2003). However, alkali basalts produced by melting of garnet pyroxenite at low pressure (1–2 Gpa) have consistently higher Al₂O₃ (15.2 to 16.2%) (Hirschmann et al., 2003) as compared to Hainan (12–13%; Table 1) alkali basalts. To generate alkali basalts with Al₂O₃ similar to Hainan alkali basalts (12–13%) (Table 1), melting of garnet pyroxenite has to take place at high pressure (5 Gpa, or 165 km) (Kogiso et al., 2003). Thus the significant²³⁰Th excess in the Hainan lavas require deep (>75 km) melting, and even deeper if the source is garnet pyroxenite.

The lithosphere beneath the Hainan Island is relatively thin (55 km) (Wu et al., 2004). The identification of deep melting in the garnet stability field by U–Th disequilibrium indicates that the Hainan basalts were generated below the base of the subcontinental lithosphere at Hainan.

Possible deep mantle sources below the base of mantle lithosphere include the asthenosphere, transition zone and lower mantle. Since the Nd isotopic compositions of the Hainan lavas are significantly lower than the asthenospheric values represented by the Central

Table	2					
U–Th	disequilibrium and	Nd, Sr	and Pb	isotopic	compositi	ons

Samples	HN01	HN02	HN106	HN07	HN08	HN10	HN11	HN12	HN14	HN119
	OT	OT	OT	OT	AOB	AOB	AOB	AOB	OT	QT
Locations	Maanling	Maanling	Maanling	Leihuling	Leihuling	Leihuling	Leihuling	Leihuling	Sanjiaoyuan	Bopian
U	0.876	0.855	0.906	1.15	1.89	3.18	1.72	1.72	0.739	0.552
Th	3.58	3.54	3.77	4.44	7.81	12.30	7.07	7.18	3.04	2.29
U/Th	0.245	0.241	0.240	0.260	0.242	0.258	0.244	0.240	0.243	0.241
(²³⁸ U/ ²³² Th)	0.744	0.733	0.731	0.789	0.736	0.785	0.740	0.729	0.737	0.734
$(^{230}\text{Th}/^{232}\text{Th})$	0.895	0.870	0.863	0.968	0.936	0.979	0.961	0.962	0.810	0.736
$(^{230}\text{Th}/^{238}\text{U})$	1.203	1.186	1.181	1.227	1.272	1.246	1.299	1.321	1.099	1.003
⁸⁷ Sr/ ⁸⁶ Sr	0.703853	0.703919		0.704180	0.704230	0.704273		0.704182		
2SE	0.000011	0.000009		0.000015	0.000014	0.000011		0.000013		
143Nd/144Nd	0.512868	0.512884		0.512848	0.512869	0.512861		0.512862		
2SE	0.000010	0.000008		0.000009	0.000008	0.000010		0.000008		
€ _{Nd}	4.5	4.8		4.1	4.5	4.4		4.4		
²⁰⁸ Pb/ ²⁰⁴ Pb	38.846			38.874	38.881	38.872		38.853		
²⁰⁷ Pb/ ²⁰⁴ Pb	15.605			15.633	15.647	15.630		15.622		
²⁰⁶ Pb/ ²⁰⁴ Pb	18.631			18.655	18.692	18.696		18.705		



Fig. 3. ¹⁴³Nd/¹⁴⁴Nd vs. ⁸⁷Sr/⁸⁶Sr diagram for Hainan lavas. Data sources: Central Indian Ridge (CIR) (Mahoney et al., 1989); South China Sea (Flower et al., 1992; Tu et al., 1992); previous Hainan data (Tu et al., 1991); and SE China (Zou et al., 2000). Arrow pointing to EM2 composition (Zindler and Hart, 1986).

Indian Ridge MORB or some basalts from central east China (Fig. 3), we suggest that asthenosphere may not be the main source. The mantle source for the Hainan basalts may come from regions below the asthenosphere, such as the transition zone or the lower mantle. This conclusion is made possible owing to the thin lithosphere beneath Hainan and the presence of significant ²³⁰Th excesses and moderately depleted Nd isotopic compositions in the Holocene basalts erupted at Maanling and Leihuling on this Island.

The thin lithosphere beneath Hainan Island provides suitable initial conditions for substantial decompressional melting of rising plume materials. It has been demonstrated that thick and cold lithosphere (in



Fig. 4. (a) ²⁰⁷Pb/²⁰⁴Pb vs. ²⁰⁶Pb/²⁰⁴Pb diagram, and (b) ²⁰⁸Pb/²⁰⁴Pb vs. ²⁰⁶Pb/²⁰⁴Pb diagram for the lavas from Hainan Island. All data plot above the Northern Hemisphere Reference Line (NHRL) (Hart, 1984). EM2 composition (Zindler and Hart, 1986).



Fig. 5. U-Th disequilibrium data for Hainan lavas.

cratons, such as Wyoming craton above Yellowstone hotspot) may hinder decompressional melting of rising plume materials (Manea et al., 2009). If a typical craton lithosphere is initially more than 200 km thick, then thinning by at least a factor of two is required to allow decompressional melting of rising plume materials. If lithospheric thinning is insufficient, the lithospheric lid hinders substantial melting of plume materials. Under this condition of still thick lithosphere, substantial melting of the lithospheric mantle occurs, which is the case for Yellowstone hotspot (Leeman et al., 2009; Manea et al., 2009).

Origins of the Hainan lavas from the transition zone or lower mantle indicated by U-Th disequilibrium and Nd isotopes are consistent with the model of plume melting beneath the Hainan Island (Zhao, 2007). It was previously proposed that the Hainan volcano in south China is a manifestation of a hotspot (Liu, 1999). Recent support for the hypothesis of a plume below Hainan comes from geophysical observations. A high-resolution seismic tomography (Huang and Zhao, 2006) and a waveform tomography (Lebedev and Nolet, 2003) revealed prominent low-velocity anomalies in the upper and lower mantle, down to 1300-km depth under Hainan. On a global scale, Hainan plume is regarded as one of the 12 deep plumes (including Hawaii, Tahiti, Louisville, Iceland, Cape Verde, Reunion, Kerguelen, Amsterdam, Afar, Eifel, Hainan, and Cobb hotspots) with visible continuous low-velocity (low-V) anomalies in both the upper and lower mantle. These deep plumes may be whole-mantle plumes originating from the core-mantle boundary (CMB) (Zhao, 2004; Zhao, 2007).

4.3. Melting conditions

Although U–Th disequilibrium in basalts cannot provide quantitative estimate of maximum depth of melting, it can provide new insight into the rate of melting that is related to the plume upwelling rate. Fast plume upwelling (e.g., Hawaii) can generate basalts with small (a few percent) ²³⁰Th excesses, whereas slow plume upwelling (e.g., Iceland) can generate basalts with significant ²³⁰Th excesses.

4.3.1. U-Th disequilibrium and rate and degree of melting

In the case of fast plume upwelling and thus high melting rate, ²³⁰Th behaves like a long-lived isotope ²³²Th or a stable element during fast partial melting. (230 Th/ 238 U) in the melt produced by fast partial melting is mostly controlled by U–Th elemental fractionation. The elemental fractionation is very limited for highly incompatible elements U and Th. During dynamic partial melting, the concentration of a trace element in the magma is (Zou, 1998)

$$C_{\text{melt}} = \frac{C_0}{X} \Big[1 - (1 - X)^{1/[\Phi + (1 - \Phi)D_0]} \Big]$$
(1)

where C_0 is the source concentration, *X* is the fraction of extracted melt (or can be regarded as effective degree of melting), Φ is the critical mantle mass porosity, and D_0 is the bulk partition coefficient. The Th/U ratio in the melt is thus related to the Th/U ratio in the source by

$$\frac{\mathrm{Th}_{\mathrm{melt}}}{\mathrm{U}_{\mathrm{melt}}} = \frac{\mathrm{Th}_{0}}{\mathrm{U}_{0}} \times \frac{1 - (1 - X)^{1/\left[\Phi + (1 - \Phi)D_{0}^{\mathrm{Th}}\right]}}{1 - (1 - X)^{1/\left[\Phi + (1 - \Phi)D_{0}^{\mathrm{U}}\right]}}$$
(2)

For practical values $D_{\text{Th}} = 0.003$ and $D_{\text{U}} = 0.005$ and $\Phi = 0.4\%$, we have $(\text{Th}/\text{U})_{\text{melt}} = 1.11(\text{Th}/\text{U})_0$ when X = 1%, $(\text{Th}/\text{U})_{\text{melt}} = 1.04(\text{Th}/\text{U})_0$ when X = 1.5%, and $(\text{Th}/\text{U})_{\text{melt}} = 1.02(\text{Th}/\text{U})_0$ when X = 2%.

Thus Th/U elemental fractionation factor

$$\delta_{\text{Th/U}} = \left(\frac{\text{Th}_{\text{melt}}}{\text{U}_{\text{melt}}} \middle/ \frac{\text{Th}_{0}}{\text{U}_{0}}\right) - 1 \tag{3}$$

is 11% when X = 1%, 4% when X = 1.5%, and 2% when X = 2%. Note that the fractionation factor δ does not change even if Φ varies in a range of 0.1% to 1%.

The calculation here also indicate that low degree partial melting (1-2%) can generate some ²³⁰Th excesses (2-10%) but not significant ²³⁰Th excesses (30%). In addition, 1-2% partial melting is usually too low for basalts. The degrees of partial melting are usually 2–5\% for alkali basalts and 8–11% for tholeiites (Frey et al., 1978; Zou and Zindler, 1996).

It has been mentioned in a hallmark paper (Williams and Gill, 1989) that low degree (5%) of partial melting is capable of generating significant ²³⁰Th excesses. However, the partition coefficients (D_U = 0.015 and D_{Th} = 0.005) and D_U/D_{Th} ratio of 3 assumed in their calculation are too high for mantle melting, resulting in higher Th/U fractionation. Experimental results on U and Th partitioning during mantle melting (Beattie, 1993; LaTourrette et al., 1993; Salters and Longhi, 1999) became available only after the publication of their theoretical paper.

In the case of slow melting conditions, ²³⁰Th also has time to grow in the mantle owing to the decay of ²³⁸U. These newly produced ²³⁰Th can also be preferentially partitioned into the melt during slow melting, resulting in higher (²³⁰Th/²³⁸U) in the melt. Another way to visualize this is to think that ²³⁰Th in the melt has an additional supply from ²³⁸U decay in the mantle. Note that the slow melting increases the (²³⁰Th/²³⁸U), but does not influence (²³²Th/²³⁸U) or Th/U elemental ratio, because ²³²Th (unlike ²³⁰Th) has no additional supply.

In summary, both U–Th elemental fractionation and ingrowth contribute to the ²³⁸U–²³⁰Th disequilibrium in magmas. However, due to the small magnitude of the U–Th elemental fractionation, dominant contributions from the ingrowth of ²³⁰Th in the mantle during slow melting is needed in order to produce significant ²³⁰Th excess.

4.3.2. Upwelling rate for Hainan plume

Ideally, melting rate and mantle porosity can be constrained using $(^{230}\text{Th}/^{238}\text{U})$ and $(^{226}\text{Ra}/^{230}\text{Th})$ in young basalts (e.g., Chabaux and Allegre, 1994; Sims et al., 1999; Pietruszka et al., 2001). $(^{230}\text{Th}/^{238}\text{U})$ produced by decompression melting is more sensitive to the rate of melting and $(^{226}\text{Ra}/^{230}\text{Th})$ is more sensitive to the porosity in the melting (e.g., Cohen and O'Nions, 1993; Zou and Zindler, 2000). Unfortunately, the Hainan Holocene lavas are about 9000 years (9 ka) old, and thus are too old for the preservation of $(^{226}\text{Ra}/^{230}\text{Th})$ disequilibrium, as the half-life of ^{226}Ra is only 1.6 ka.

Nevertheless, the $(^{230}\text{Th}/^{238}\text{U})$ disequilibrium alone can place robust constraint on melting rate when the disequilibrium is significant, which is the case for the Hainan Holocene lavas. The explicit relationship between the melting rate and the observed extent of $(^{230}\text{Th}/^{238}\text{U})$ disequilibrium (Eq. 3 in Zou et al., 2003) is

$$\dot{M} = \frac{(\lambda_{238} + \lambda_{230})[\rho_{\rm f}\phi + \rho_{\rm s}(1-\phi)D_{\rm U}] - Z\lambda_{230}[\rho_{\rm f}\phi + \rho_{\rm s}(1-\phi)D_{\rm Th}]}{(Z-1)[1 + \rho_{\rm f}\phi / [\rho_{\rm s}(1-\phi)]]}$$
(4)

where Z represents measured $(^{230}\text{Th}/^{238}\text{U})$ activity ratios, ϕ is the melting volume porosity, \dot{M} is the melting rate, ρ_{f} and ρ_{s} represent the density of melt (2800 g/cm³) and solid (3300 g/cm³), respectively. Using the above relationship, Fig. 6 is made to show a plot of melting porosity against melting rate for (²³⁰Th/²³⁸U) ranging from 1.18 to 1.32. The maximum $(^{230}\text{Th}/^{238}\text{U})$ value in the Hainan basalts of 1.32 can be achieved at slow melting rate (<1.0E-4 kg/m³/year) and low porosity (<0.4%) (Fig. 6). In a one-dimensional melting column, the solid mantle upwelling rate (W) is related to the melting rate (\dot{M}), melt productivity (df/dz), and the density of the solid (ρ_s) by $W = \frac{\dot{M}}{\rho_s(df/dz)}$ (Spiegelman and Kenyon, 1992). If the melt productivity $df/dz = 0.003 \text{ km}^{-1}$ (McKenzie and Bickle, 1988) and density $\rho_{\rm s} = 3300 \text{ kg/cm}^3$ are used, then for melting rate of $< 1.0\text{E} - 4 \text{ kg/m}^3/$ year, the mantle upwelling rate is <1 cm/year. This indicates that the mantle upwelling is very slow in the Hainan. The plume upwelling beneath Hainan is estimated as significantly slower than the solid mantle upwelling rates for tholeiites from Hawaii plume ranging from 10 to 30 cm/year (Sims et al., 1999). If slow upwelling is usually indicative of dying plumes, the Hainan plume upwelling might be slowing down and might be close to exhausting its source region.

4.4. EM2 Dupal anomaly in Hainan Island

The Dupal isotopic anomaly in some oceanic island and ridge basalts and continental basalts is characterized by high ²⁰⁸Pb/²⁰⁴Pb and ²⁰⁷Pb/²⁰⁴Pb ratios (at a given ²⁰⁶Pb/²⁰⁴Pb ratio) and high ⁸⁷Sr/⁸⁶Sr ratio, indicating a reservoir with high time-integrated values of Th/U, ²³⁵U/Pb, and Rb/Sr (Dupre and Allegre, 1983; Hart, 1984). Since



Fig. 6. Estimate of the maximum melting rate from U–Th disequilibrium data. Bulk partition coefficients are $D_U = 0.005$ and $D_{Th} = 0.003$ for garnet peridotites. Bulk partition coefficients are given by $D_i = \sum K_i x_i$, where K_i is the mineral/melt partition coefficient, and x_i is its mineral proportion in the source. Mineral/melt partition coefficients are: $K_{Th}(gt) = 0.019$, $K_U(gt) = 0.041$, $K_{Th}(opx) = 0.0002$, $K_U(opx) = 0.0005$ (Salters and Longhi, 1999), $K_{Th}(cpx) = 0.015$, $K_U(cpx) = 0.010$ (Lundstrom et al., 1994), and $K_{Th}(ol) = K_U(ol) = 0.0001$ (assumed). Mineral proportions are: $x_{ol} = 61\%$, $x_{opx} = 22\%$, $x_{gt} = 10\%$, and $x_{cpx} = 7\%$. Mineral abbreviations: gt = garnet, opx = orthopyroxene, opx = clinopyroxene, ol = olivine.

recognition of the Southern Hemisphere Dupal anomaly, its origin remains a geochemical and geophysical enigma. There has been considerable debate about whether Dupal signatures reflect deep or shallow mantle phenomena (Hawkesworth et al., 1986; Castillo, 1988; Sun and McDonough, 1989). The majority of Dupal basalt suites satisfying the requirements of being old, deep and (semi-)global in character clearly occur in the Southern Hemisphere. The South China Sea Basin, including Hainan Island (20°N), appears to be one of the few Dupal-like domains reported from Northern Hemisphere. The significant ²³⁰Th excesses in the young Hainan basalts support for deep mantle reservoirs, instead of local shallow lithospheric mantle origin. If this is also true for older basalts around South China Sea Basin and SE China, then the EM2 domains in South China Sea Basin and SE China may also have deep mantle source.

It has been shown that the source of EM2 ocean island basalts (OIBs) at Samoan islands can be subducted upper continental crust (UCC) materials, owing to the extreme ⁸⁷Sr/⁸⁶Sr isotope compositions (0.720469) and negative Nb and Ta anomalies in some Samoan lavas (Jackson et al., 2007). The EM2 Samoan lavas indicate the presence of an ancient recycled UCC component in the Samoan mantle plume and reflect the return of subducted UCC in Samoan lavas (Jackson et al., 2007).

In comparison with the Samoan lavas, the Hainan lavas have notso-high ⁸⁷Sr/⁸⁶Sr isotopic compositions (0.7038 to 0.7043) and display positive Nb or Ta anomalies. Thus the UCC component in the Hainan plume, even if present, is most likely to be insignificant.

Our preferred interpretation is that EM2 characters from Hainan basalts reflect the inherent signatures of the upwelling lower mantle materials. Another possibility is that the source of EM2 from Hainan basalts may have been delaminanted continental (Hainan or south China) lithospheric mantle. The delaminated pieces sank deeper in the mantle where they melted in the seismically anomalous region, and then the melt mixed with depleted material.

The northern boundary of this deep EM2 Dupal source probably extends to SE China basalts with EM2 characters in Fujian and Guangdong provinces (Zou et al., 2000). In contrast, the basalts from NE China, including potassic basalts from Wudalianchi and Jingbohu, show EM1 characters (Basu et al., 1991; Zhang et al., 1991). The EM1 Dupal Pb isotope signatures in these NE China basalts may have originated from the local thick lithosphere beneath NE China.

4.5. The ages of the Pre-Holocene lavas

Our U-series data of older samples provide information on their ages. Note that the U/Th ratio in HN14 is identical to the olivine tholeiites in Maanling (Fig. 6). Assuming the olivine tholeiite from Sanjiaoyuan (HN14) had initial (230 Th/ 238 U) ratio at eruption within the range (1.18 to 1.20) of the olivine tholeiites from Maanling, the eruption age of HN14 is 65,000 years (ka) to 77 ka. This 65–77 ka eruption age of HN14 is within the K-Ar age range (60 to 110 ka) (Fan et al., 2004) of the Late Pleistocene volcanic rocks (Daotang Formation) at Hainan.

The quartz tholeiite from Bopian (HN119) displays U–Th equilibrium, suggesting that the age of the older quartz tholeiite is equal to or greater than 375 ka (the five half lives of ²³⁰Th). Since quartz tholeiites at Hainan are restricted to the earliest eruptions at Hainan Island (Flower et al., 1992), their secular U–Th equilibrium in HN119 confirms their old age and provided the minimum eruption age (375 ka) for quartz tholeiites. The quartz tholeiites are not Late Pleistocene (10 to 130 ka) volcanic rocks, but may be the Middle to Early Pleistocene or earlier volcanic rocks.

5. Conclusions

1) The Holocene lavas from Maanling and Leihuling volcanoes display strong ²³⁰Th excesses (18 to 32%). Their $\varepsilon_{\rm Nd}$ values are +4.5 to

+ 4.8 for Maanling lavas, and + 4.1 to + 4.5 for the Leihuling lavas. All the Hainan lavas show LREE enriched patterns and positive Nb and Ta anomalies.

- 2) The EM2-like isotopic characteristics in the Hainan basalts were not generated by melting of or contamination by the subcontinental lithosphere. The U–Th disequilibrium data and Nd isotopic compositions support for a model of deep (sub-asthenospheric) melting in the transition zone or more likely the lower mantle (plume melting). It is likely that EM2 component from Hainan basalts represents the isotopic signature of lower mantle component.
- 3) The upwelling of the Hainan plume is slow (<1 cm/year), which might indicate that Hainan plume is a dying plume that is close to exhausting its source region. Deep melting in the transition zone or lower mantle generates the Hainan Island basalts with Dupallike EM2 Pb isotope signatures. The older EM2 Cenozoic basalts from South China Sea Basin and in SE China areas might also have been derived from decompressional melting of lower mantle materials associated with the rising Hainan plume.

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