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# The geochemical characteristics of Cretaceous volcanics in southern Hainan Island and implications for tectonic evolution in the South China Sea

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The southeastern Eurasian plate, where the South China Sea (SCS) is located, lies in a complex tectonic setting between the Pacific and Tethys tectonic belts. It is widely accepted that the tectonics of the SCS area were influenced by subduction in the late Mesozoic, but there is still controversy over whether it was paleo-Pacific subduction or Tethyan subduction. Volcanic activity in the south of Hainan Island was intense during the Cretaceous, and geochemical analysis of the collected basaltic andesite, andesite and rhyolite samples in this study indicate those intermediate-acid series igneous rocks are high-K calc-alkaline or calc-alkaline. Some andesites have high MgO contents and Mg<sup>#</sup> values (2.04–5.34 wt% and 36.83–55.29; Mg<sup>#</sup> = 100 × Mg<sup>2+</sup> / (Mg<sup>2+</sup> + TFe<sup>2+</sup>)). Light rare earth elements (LREEs) and large ion lithophile elements (LILEs) are enriched in all the samples, but high field strength elements (HFSEs) are depleted. The negative Eu anomalies are more obvious in the rhyolites than andesites. The geochemical characteristics of the volcanic arc igneous rocks show that the mid-Cretaceous tectonic setting of Hainan Island can be classified as an Andean active continental margin. During the mid-Cretaceous, intermediate volcanism occurred in Hainan Island and its adjacent areas. The zircon-saturation temperatures of the acid volcanic rocks in study area exhibit relatively low values (ranging from 746°C–790°C). Unlike igneous rocks forming in the coastal area east of the South China Block at the same time, no A-type granitoids with alkaline dark minerals appear in Hainan Island. During the late Mesozoic, the western SCS, where Hainan Island was located, may not have been affected by the subduction of the paleo-Pacific Plate, but rather Neotethyan subduction which dominated the Cretaceous magmatic and tectonic activities along the western margin of the SCS. This finding helps to understand the late Mesozoic tectonic evolution of the southeastern edge of the Eurasian plate.

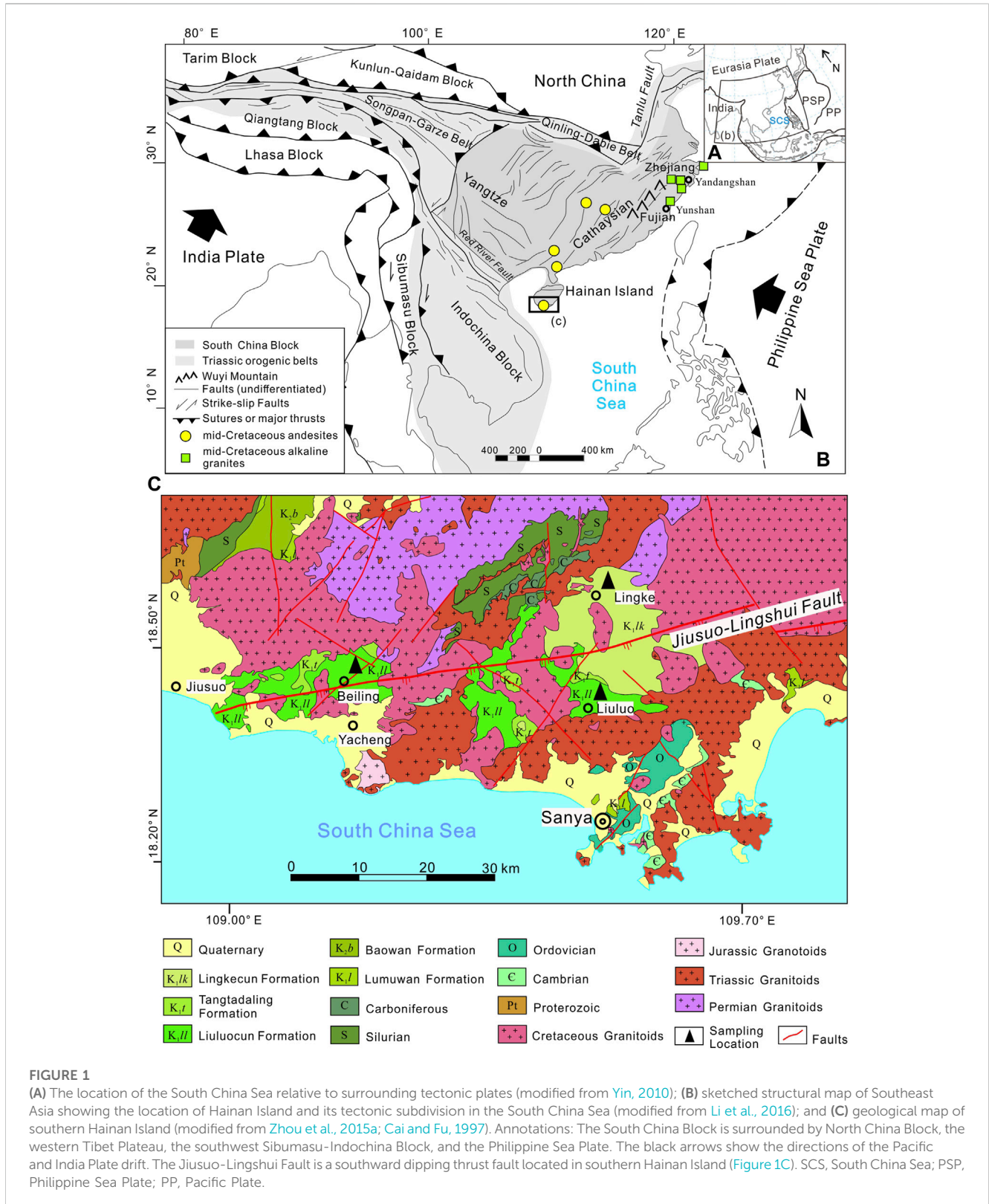
## KEYWORDS

andesites, rhyolites, geochemical characteristics, active continental margin, Neotethyan subduction

# 1 Introduction

The southeastern Eurasian Plate lies among the Pacific Plate, Philippine Sea Plate and the India-Australia Plate (Figure 1A), and between the circum-Pacific belt and the Tethys belt, which implies a

complicated tectonic evolution. The Cenozoic opening and growth of the South China Sea (SCS) is a remarkable tectonic event in this region. However, there are many disputes regarding the tectonics of this area before the opening of the SCS and the extension of the continental margin of the South China Block (SCB). In the late



Mesozoic, this region is often regarded as the southwestern extension of the magmatic belt of the eastern SCB, followed by a transition from the Tethys orogenic regime to the paleo-Pacific tectonic regime (Zhou and Li, 2000; Wang et al., 2001; Xia and Zhao, 2014; Zhou et al., 2015a; Zahirovic et al., 2016). Because of the subduction and rollback of the paleo-Pacific Plate, the western margin of the SCS experienced a regional lithospheric extension, and large-scale magmatism occurred in this region (Li et al., 1999; Thuy et al., 2004; Geng et al., 2006; Yan et al., 2010; Wang et al., 2012; Wang et al., 2019; Tang et al., 2014; Zhou et al., 2015a; Xu et al., 2016; Sun et al., 2017; Yan et al., 2017; Nong et al., 2021; Yang et al., 2022). On the other hand, some studies suggest that tectonic and magmatic activities with related ore deposits were still affected by the Tethys orogenic regime during the Cretaceous (Fang, 2016; Sun, 2016; Zhang et al., 2017; Zhu et al., 2017; Sun et al., 2018; Huang et al., 2019; Yuan et al., 2019; Liu et al., 2020; 2021). The debates over which subduction affected the SCS have led to differences in the identification of geological features on the southeastern Eurasian Plate during the late Mesozoic, such as magmatic activities and mineralization.

Cretaceous granitoids were extensively studied to explore tectonic evolution during the Mesozoic period because of their numerous outcrop areas (Li et al., 1999; Thuy et al., 2004; Yan et al., 2010; Wang et al., 2012; 2019; Tang et al., 2014; Xu et al., 2016; Sun et al., 2017; Yan et al., 2017; Zhang et al., 2017; Zhu et al., 2017; Sun et al., 2018; Huang et al., 2019; Yuan et al., 2019; Nong et al., 2021), which implies these studies give less consideration of other magmatic rocks, such as andesites, from this period. Considering the region where the SCS lies was an active continental margin, the volcanic rocks need to be studied comprehensively, especially intermediate rocks, which will help to better understand the state of the lithosphere and deep mantle, and the tectonic setting of this region.

Located in the northwestern SCS, Hainan Island has a large outcrop of Cretaceous volcanic rocks and is one of the most important exposed areas of Cretaceous andesite on the SCB. Therefore, a detailed study of the Cretaceous volcanic rocks in Hainan Island will help to characterize the type of volcanic activity along the margin of the SCS during the Cretaceous period, and improve our understanding of the tectonic evolution of the southeast edge of the Eurasian plate during the late Mesozoic.

## 2 Geological setting and sample information

Hainan Island, located at the intersection of the Eurasian, Indian-Australian, Philippine Sea and Pacific plates, is separated from the Chinese mainland by the Qiongzhou Strait (Figure 1B). Hainan Island has experienced multi-stage tectonic movements, and an E-W trending tectonic system is the main structural system. The Baoban Group, about 1.43 Ga (Li et al., 2008), is the oldest known basement exposed on the island. The strata outcrop on Hainan Island is very limited, accounting for only 18.6% of the island area, and the remaining area generally has magmatic rocks (Figure 1C), especially granitoid rocks covering ca. 40% area of the island (Zhou et al., 2015b).

Extensive Cretaceous volcanism resulted in a >5200 m thick volcanic sequence, which is divided, from the base to the top, into the Liuluocun, Tangtadaling and Lingkecun formations (Guangdong BGMR, 1988). The rocks consist predominantly of intermediate-felsic compositions, with minor occurrences of mafic volcanic rocks, and are primarily distributed along the Jiusuo-Lingshui Fault, a southward dipping thrust fault located in southern Hainan Island (Figure 1C). The Liuluocun Formation consists of rhyolitic tuff lava, flow-banded rhyolite on the top, rhyolitic ignimbrite, eruptive breccia with stratified basalt, and andesite in the lower part and purplish red siliciclastic rock at the bottom. The Liuluocun Formation is unconformably overlain by the Tangtadaling Formation, which is mainly composed of dacitic tuff. The Lingkecun Formation, on the top of this volcanic sequence, is mainly composed of rhyolitic lava and pyroclastic rocks, with several layers of dacites. According to zircon geochronology, rhyolitic and andesitic rocks of the Liuluocun Formation formed in 107–101.5 Ma, and the age of the Lingkecun Formation was about 98 Ma (Cai and Fu, 1997; Zhou et al., 2015a). The Cretaceous magmatic activities in the south of Hainan Island occurred about the mid-Cretaceous (110–90 Ma) (Tang et al., 2010; Sun et al., 2018; Yuan et al., 2019).

The samples were collected from Beiling, Liuluo and Lingke, respectively. The Liuluocun Formation was sampled in Beiling and Liuluo, while the Lingke samples were from the Lingkecun Formation (Figures 2A, B). Based on hand specimens, these samples are andesite or rhyolite. Andesite samples are dark gray in color, massive, and porphyritic (Figures 2C, E, G). Andesites from Beiling and Liuluo contain 10%–30% phenocrysts, of which are mostly plagioclase with minor amounts of dark minerals. The dark minerals are mainly hornblende with a small amount of pyroxene (Figures 2C, E). But phenocrysts of andesites from Lingke are much less common (~10%), and they are all pyroxene (Figure 2G). Dark minerals have been altered to chlorite. The matrix of andesites consists of microcrysts of plagioclase, dark minerals, Fe-Ti oxides, zircon and apatite.

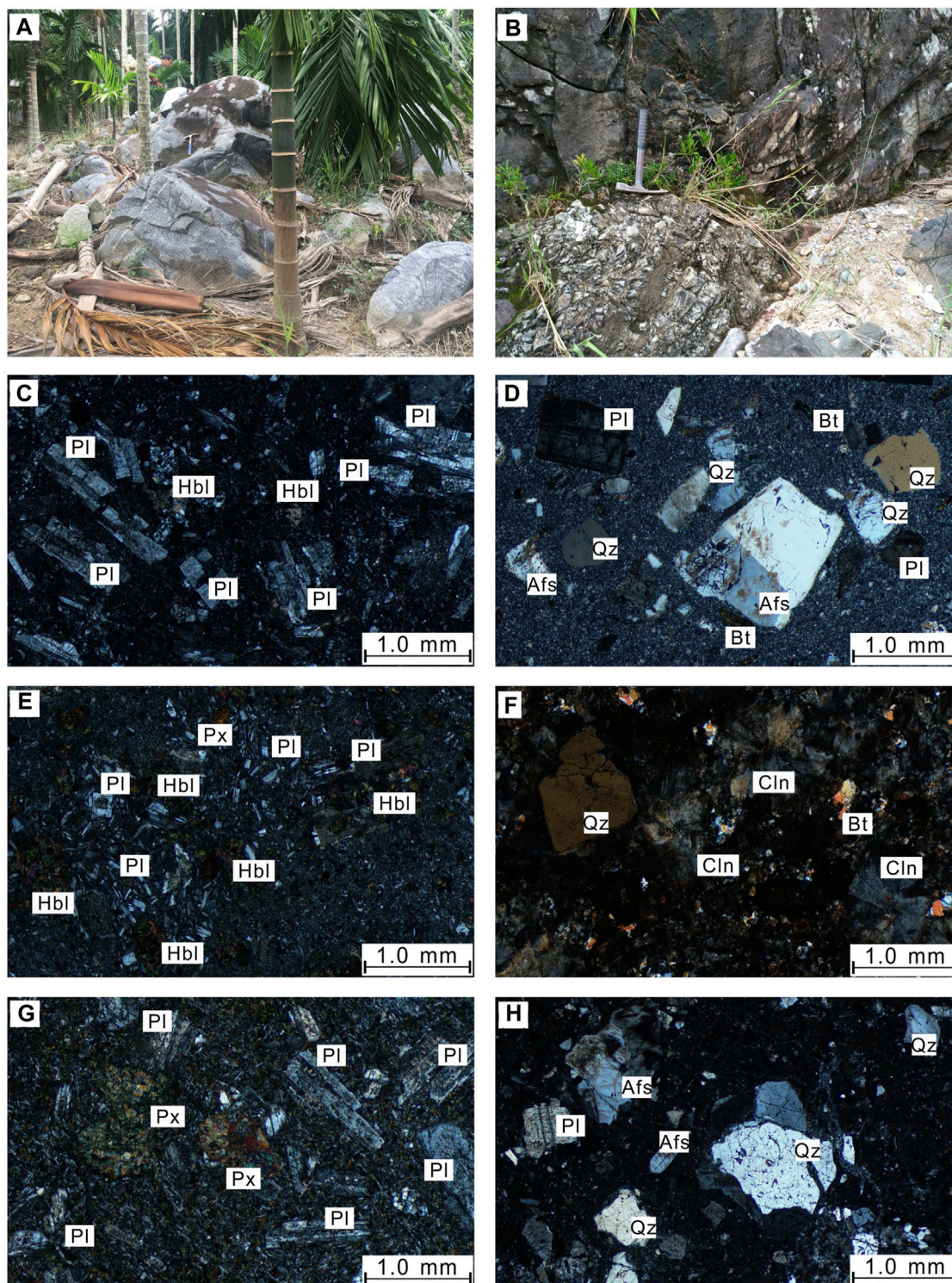
Rhyolite samples are light purple or gray in color, massive, and porphyritic (Figures 2D, F, H). The phenocrysts include quartz (10–15 vol%), plagioclase (3–8 vol%), alkali feldspar (5–10 vol%) and biotite (~3%). Those phenocrysts are slightly altered. Chalcedony can be seen in some rhyolites (Figure 2F). The matrix consists of microcrysts and opaque minerals.

## 3 Methods

The samples were ground to 74- $\mu$ m using an agate mortar and pestle. To determine the oxides of major elements, samples were analyzed according to the Chinese National Standard GB/T14506.28-2010 (National Standard of P.R. China, 2011b). About 0.7 g of powder and 7 g of latent solvent ( $\text{Li}_2\text{B}_4\text{O}_7 + \text{LiF} + \text{NH}_4\text{NO}_3$ ) were weighed out and stirred together in a Pt-(Au) crucible. Then, 1 mL BrLi was added in the crucible at 1200°C for 20 min. The liquid melt was used to make a matrix for analysis. About 1 g sample was added in a crucible ( $W_1$ ) and weighed ( $W_2$ ). The crucible was placed in a muffle furnace at 1000°C for 2 h and then dried and weighed ( $W_3$ ). The loss on ignition (LOI) was determined using the following equation:

$$\text{LOI} = (W_2 - W_3) / (W_2 - W_1)$$





**FIGURE 2**

Field photograph (A, B) and photomicrographs (C–H) of Cretaceous volcanic rocks from southern Hainan Island. Annotations: (A, C, D) from Beiling; (E, F) from Liuluo; (B, G, H) from Lingke. (C–F) from Liuluo Formation; (G, H) from Lingkecun Formation. (C, E, G) andesitic samples; (D, F, H) rhyolitic samples. Abbreviations: Pl, plagioclase; Hbl, hornblende; Px, pyroxene; Qtz, quartz; Afs, alkali feldspar; Bt, biotite; Cln, Chalcedony.

Major element oxides except FeO were analyzed on fused glass discs using a Rikagu RIX 2100 X-ray fluorescence spectrometer (XRF). FeO contents were analyzed by wet chemical analysis based on the [National Standard of P.R. China \(2011a\)](#). The Chinese national reference material

GBW07104 was used for quality control. The method detection limit (MDL) for each element was calculated as three times the standard deviation of the average from the blank samples ( $n = 10$ ). The uncertainties of major element analyses were better than  $\pm 5\%$ .

TABLE 1 Major element (wt%) and trace element (ppm) compositions of the Cretaceous volcanic rocks of southern Hainan Island.

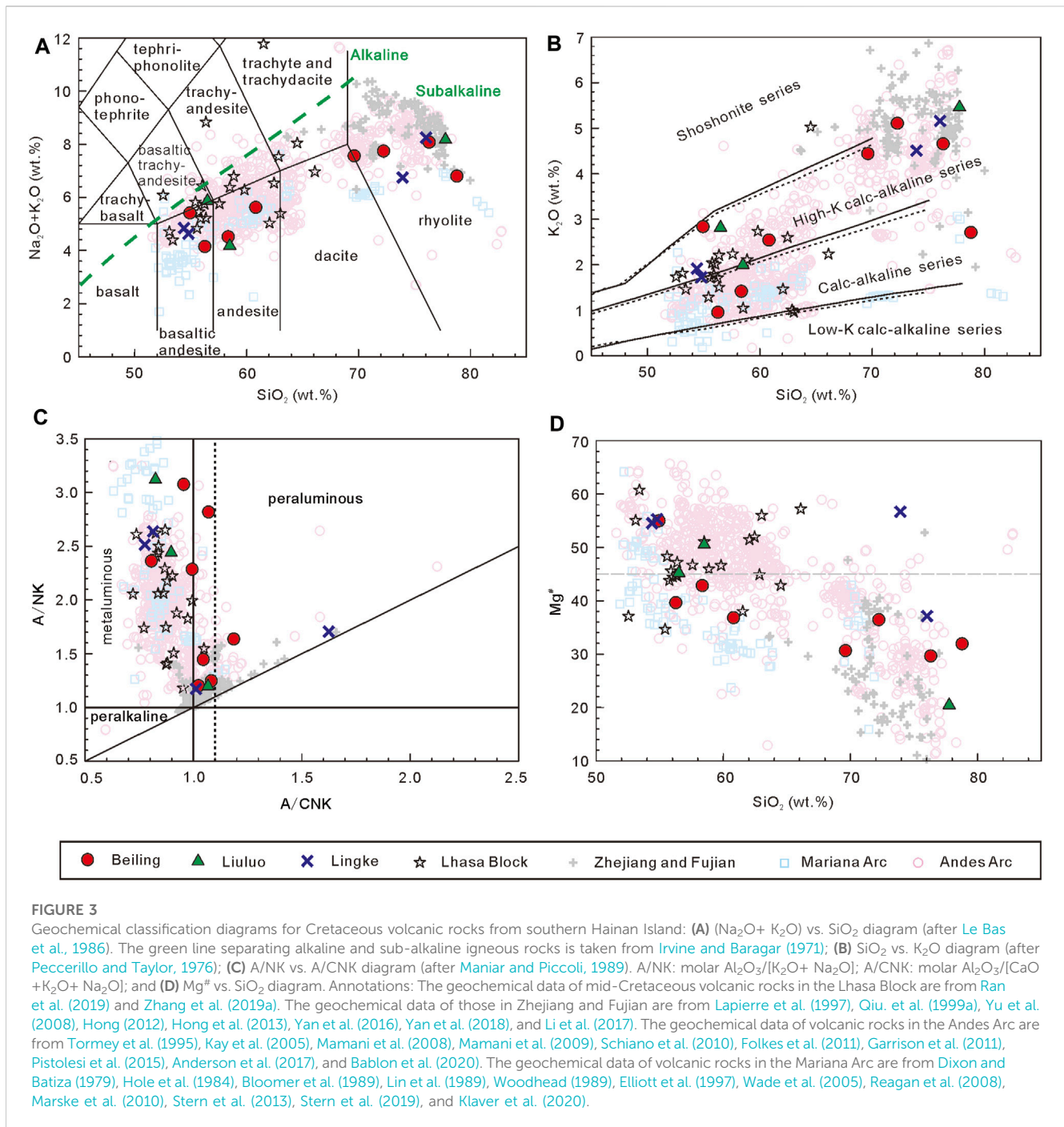
Samples	Beiling								Liuluo			Lingke			
	BLRR1	BLRR501	16SY05-3R	16SY06-1	BLRR201	16SY01-2	16SY03-2	16SY06-5	16SY10-6-1	16SY10-5-1	16SY10-2	16SY08-3-1	16SY08-2	16SY08-1	16SY08-5-1
SiO <sub>2</sub>	52.90	56.73	55.31	59.64	68.20	76.04	78.14	71.52	57.56	55.65	77.38	53.84	53.35	75.41	72.41
Al <sub>2</sub> O <sub>3</sub>	16.63	18.17	19.00	17.52	15.92	12.78	11.95	14.12	17.65	19.40	12.37	17.22	17.01	12.41	14.30
TiO <sub>2</sub>	1.13	0.88	1.06	0.81	0.45	0.16	0.07	0.32	0.71	0.92	0.10	0.97	0.99	0.15	0.30
Fe <sub>2</sub> O <sub>3</sub>	2.74	5.81	4.74	5.00	2.52	0.48	0.71	1.19	3.25	3.13	0.53	1.86	1.89	0.63	1.32
FeO	4.65	1.99	3.35	1.73	0.57	0.76	0.39	1.22	2.75	2.92	0.18	6.02	6.08	0.91	1.02
CaO	7.47	5.80	7.54	5.47	2.03	1.02	0.80	2.06	8.65	7.52	0.70	8.04	8.33	0.93	0.23
MgO	4.88	3.04	2.81	2.04	0.71	0.28	0.27	0.74	3.25	2.66	0.09	5.34	5.24	0.49	1.63
K <sub>2</sub> O	2.73	1.38	0.94	2.49	4.35	4.64	2.69	5.06	1.96	2.77	5.43	1.70	1.88	5.12	4.41
Na <sub>2</sub> O	2.48	3.01	3.14	3.02	3.05	3.40	4.05	2.61	2.14	3.00	2.70	2.85	2.87	3.06	2.19
MnO	0.16	0.13	0.16	0.10	0.05	0.06	0.07	0.11	0.15	0.16	0.02	0.15	0.13	0.05	0.11
P <sub>2</sub> O <sub>5</sub>	0.54	0.29	0.26	0.25	0.12	0.03	0.04	0.09	0.32	0.36	0.01	0.27	0.27	0.03	0.02
LOI	3.45	2.61	1.52	1.77	1.86	0.31	0.73	0.85	1.41	1.28	0.42	1.59	1.78	0.75	1.98
Total	99.73	99.84	99.82	99.84	99.84	99.96	99.91	99.86	99.81	99.78	99.94	99.85	99.84	99.94	99.92
A/CNK	0.60	0.99	1.61	1.43	1.50	8.48	4.81	2.49	1.14	1.66	5.99	1.42	1.31	3.70	1.04
FeO*	7.11	7.22	7.61	6.24	2.84	1.19	1.03	2.29	5.68	5.74	0.65	7.69	7.79	1.48	2.21
Mg <sup>#</sup>	55.01	42.85	39.65	36.83	30.65	29.65	31.94	36.46	50.54	45.21	20.45	55.29	54.56	37.12	56.69
La	46.13	30.45	26.1	32.0	55.04	30.6	39.9	54.9	28.0	24.7	28.1	24.3	30.5	42.1	20.2
Ce	91.44	56.98	55.8	56.0	88.14	60.1	65.9	88.7	48.1	54.2	48.5	43.5	48.9	83.1	31.1
Pr	12.06	7.28	6.76	6.76	11.46	6.59	7.38	10.9	5.98	6.70	5.66	5.55	6.14	9.82	3.74
Nd	48.18	28.56	27.5	26.8	40.56	23.6	25.4	40.8	24.3	28.1	19.3	23.2	26.2	37.3	14.2
Sm	9.12	5.64	5.32	5.11	6.98	4.71	4.04	7.59	4.17	5.46	3.37	4.63	5.29	7.08	2.73
Eu	2.50	1.71	1.51	1.30	1.51	0.37	0.76	1.52	1.24	1.68	0.43	1.27	1.43	0.49	0.52
Gd	7.75	5.16	4.61	4.50	6.02	4.48	3.94	7.19	3.57	4.62	3.29	4.14	4.67	6.12	2.62
Tb	1.11	0.82	0.70	0.71	0.88	0.76	0.52	1.11	0.53	0.69	0.48	0.68	0.74	0.90	0.47
Dy	5.89	4.86	3.82	3.96	5.04	4.81	2.73	6.27	2.88	3.62	2.81	4.02	4.06	4.96	3.13

(Continued on following page)

TABLE 1 (Continued) Major element (wt%) and trace element (ppm) compositions of the Cretaceous volcanic rocks of southern Hainan Island.

Samples	Beiling								Liuluo			Lingke			
	BLRR1	BLRR501	16SY05-3R	16SY06-1	BLRR201	16SY01-2	16SY03-2	16SY06-5	16SY10-6-1	16SY10-5-1	16SY10-2	16SY08-3-1	16SY08-2	16SY08-1	16SY08-5-1
<b>Ho</b>	1.08	0.94	0.67	0.72	0.95	0.95	0.50	1.13	0.53	0.64	0.53	0.73	0.71	0.92	0.63
<b>Er</b>	2.89	2.43	1.87	2.04	2.60	2.93	1.52	3.18	1.48	1.76	1.65	1.93	1.99	2.80	1.89
<b>Tm</b>	0.45	0.41	0.32	0.34	0.46	0.51	0.24	0.48	0.23	0.30	0.28	0.29	0.33	0.43	0.30
<b>Yb</b>	2.51	2.39	1.99	2.12	2.83	3.36	1.60	2.99	1.41	1.87	1.83	1.79	2.07	2.76	1.87
<b>Lu</b>	0.57	0.50	0.28	0.30	0.55	0.57	0.27	0.49	0.23	0.27	0.31	0.28	0.30	0.47	0.31
<b>Y</b>	33.53	27.08	24.6	26.9	25.30	27.0	15.6	31.4	15.1	22.2	16.9	19.7	23.8	25.0	20.6
<b>Ba</b>	879.3	792.8	523	559	1062	159	411	666	671	844	275	347	402	232	346
<b>Rb</b>	57.2	52.0	39.0	89.8	162.2	256	83.1	157	54.0	81.0	196	57.8	57.6	192	251
<b>Th</b>	6.99	5.28	4.43	3.99	19.64	29.2	16.4	20.2	2.73	2.48	20.8	4.37	3.68	21.0	11.2
<b>Nb</b>	14.46	12.81	12.7	12.8	11.00	22.4	14.1	12.9	13.3	14.4	16.0	12.2	10.3	16.5	11.4
<b>U</b>	1.84	1.55	0.87	1.74	5.28	11.2	5.36	4.59	0.83	1.23	4.63	1.07	1.16	4.42	2.33
<b>Ta</b>	0.85	0.86	0.59	0.77	1.29	2.23	1.22	0.99	0.48	0.92	1.70	0.48	1.22	1.36	1.04
<b>Sr</b>	924.6	652.5	700	581	411.7	76.5	151	318	788	790	77.8	690	576	103	49.6
<b>Pb</b>	18.1	17.0	12.51	15.97	24.0	25.4	25.1	24.7	7.84	16.21	23.8	13.3	14.67	12.5	12.9
<b>Zr</b>	254.0	164.1	172	176	152.1	102	115	149	151	196	89.9	145	173	143	131
<b>Hf</b>	7.25	4.56	4.82	5.29	8.39	3.81	3.74	4.08	4.22	5.77	3.61	4.13	5.29	5.19	4.11
<b>Cr</b>	74.3	9.4	18.9	3.05	11.8	3.60	4.98	5.02	23.4	32.7	2.67	111	101	6.69	9.41
<b>Ni</b>	49.8	6.8	7.10	2.15	3.9	0.55	0.99	1.97	14.9	12.6	0.41	44.8	38.8	2.68	5.65
<b>Ga</b>	20.29	24.00	23.9	20.9	19.44	14.9	7.95	16.2	20.9	24.0	11.5	19.6	20.8	15.3	17.9
<b>∑REE</b>	231.67	148.12	137.15	142.60	223.02	144.38	154.68	227.23	122.58	134.53	116.54	116.25	133.31	199.36	83.74
<b>LREE/HREE</b>	3.76	2.93	3.16	3.08	4.56	2.78	5.32	3.77	4.31	3.36	3.76	3.05	3.06	4.05	2.28
<b>(La/Yb)<sub>N</sub></b>	13.19	9.14	9.38	10.84	13.94	6.53	17.90	13.16	14.28	9.47	11.00	9.75	10.56	10.94	7.76
<b>δEu</b>	0.89	0.95	0.91	0.81	0.69	0.24	0.57	0.62	0.96	0.99	0.39	0.87	0.86	0.22	0.59
<b>Sr/Y</b>	27.58	24.10	28.46	21.64	16.27	2.83	9.63	10.14	52.36	35.65	4.60	34.96	24.18	4.12	2.40



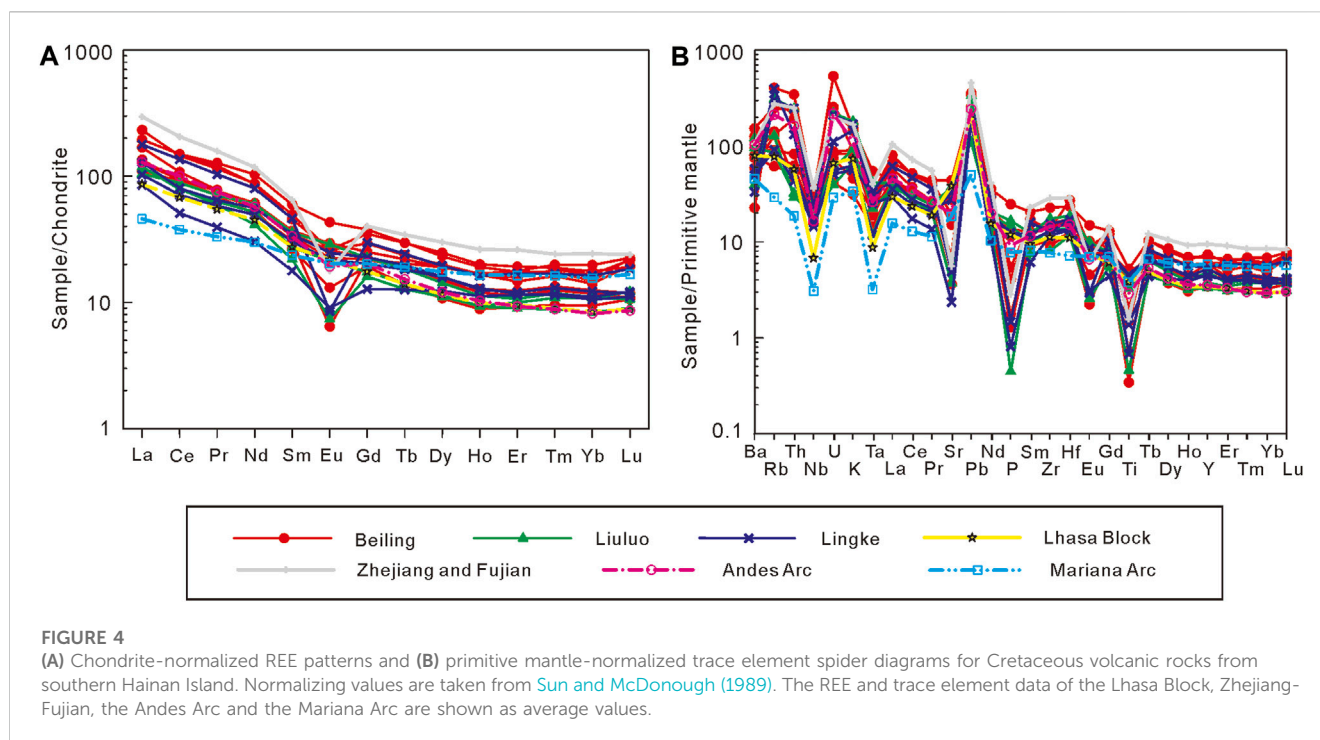


The sample preparation for trace element analysis was according to the [National Standard of P.R. China GB/T14506.30-2010 \(2011\)](#). Powdered samples of about 50 mg were weighed and then dissolved in  $\text{HNO}_3$  (0.5 mL)-HF (1 mL) in Teflon bombs at  $185^\circ\text{C}$  for 24 h, dried, and then treated with  $\text{HNO}_3$  (0.5 mL). All samples underwent acid digestion twice and were then treated with  $\text{HNO}_3$  (5 mL) at  $130^\circ\text{C}$  for 3 h. Dissolved samples were diluted to 50 mL in a clean PET bottle prior to the analyses.  $^{103}\text{Rh}$  and  $^{185}\text{Re}$  were used as the internal standard references. The trace elements were measured using an Agilent 7500a inductively coupled plasma mass spectrometer (ICP-MS). The precision of all trace elements was estimated to be  $\pm 10\%$ .

Major and trace elements of the samples were analyzed at the Hebei Institute of Geology and Mineral Resources, China Geological Survey.

## 4 Results

Major and trace element data for the mid-Cretaceous volcanic rocks in southern Hainan Island are presented in [Table 1](#). In the  $\text{Na}_2\text{O} + \text{K}_2\text{O}$  vs.  $\text{SiO}_2$  diagram ([Figure 3A](#)), volcanic samples fall within the basaltic andesite-andesite-rhyolite field. The andesite/basaltic andesite samples are characterized by relatively low  $\text{SiO}_2$ ,



$\text{Na}_2\text{O}$  and  $\text{K}_2\text{O}$  contents (52.90–59.64 wt%, 2.14–3.14 wt% and 0.94–2.77 wt%). Some basaltic andesites and andesites display elevated  $\text{MgO}$  contents as well as high  $\text{Mg}^\#$  values [ $\text{Mg}^\# = 100 \times \text{Mg}^{2+}/(\text{Mg}^{2+} + \text{TFe}^{2+})$ ]. Thus, they can be classified as high- $\text{Mg}$  andesite according to Kelemen (1995) (54–65 wt%  $\text{SiO}_2$ ;  $>45 \text{ Mg}^\#$  values) (Figure 3D). The rhyolites possess relatively high  $\text{SiO}_2$ ,  $\text{Na}_2\text{O}$  and  $\text{K}_2\text{O}$  contents (68.20–78.14 wt%, 2.19–4.05 wt% and 2.69–5.43 wt%). The moderate  $\text{K}_2\text{O}$  content suggests that the volcanics are calc-alkaline or high-K calc-alkaline (Figure 3B). Relatively high  $\text{Al}_2\text{O}_3$  contents (12.05–19.70 wt%) with elevated  $\text{A/NK}$  values (1.20–3.12;  $\text{A/NK} = \text{molar Al}_2\text{O}_3/[\text{K}_2\text{O} + \text{Na}_2\text{O}]$ ) and moderate  $\text{A/CNK}$  values (0.81–1.62;  $\text{A/CNK} = \text{molar Al}_2\text{O}_3/[\text{CaO} + \text{K}_2\text{O} + \text{Na}_2\text{O}]$ ) indicate mid-Cretaceous volcanics are metaluminous to peraluminous (Figure 3C). These samples exhibit a geochemical composition similar to the volcanic rocks from the Andes Arc.

The volcanic samples have total rare earth element ( $\Sigma\text{REE}$ ) contents ranging from 83.70 ppm to 231.70 ppm, and obvious fractionation between light and heavy REEs with  $(\text{La}/\text{Yb})_N$  values of 6.53–17.90 (Table 1; Figure 4A). The chondrite-normalized REE patterns show variable negative Eu anomalies. The  $\delta\text{Eu}$  values [ $\delta\text{Eu} = 2 \times \text{Eu}_N/(\text{Sm}_N + \text{Gd}_N)$ ] range from 0.99 to 0.22 as a result of plagioclase fractionation, while the Eu anomalies of the rhyolites are more negative (Figure 4A). The primitive mantle-normalized multi-element diagram (Figure 4A) shows both andesite and rhyolite samples share features similar to arc igneous rocks, such as enrichment of LILEs (e.g., Rb, U, K and Pb) and HFSEs (e.g., Nb, Ta, P and Ti) (Figure 4B). The variable negative Ba and Sr suggest that there was a relict of plagioclase and hornblende in the magma source or that these two minerals fractionated during an earlier magmatic stage. These characteristics are more remarkable in rhyolites on the primitive mantle-normalized diagram compared to the andesites in Hainan Island and volcanic rocks in the Andes

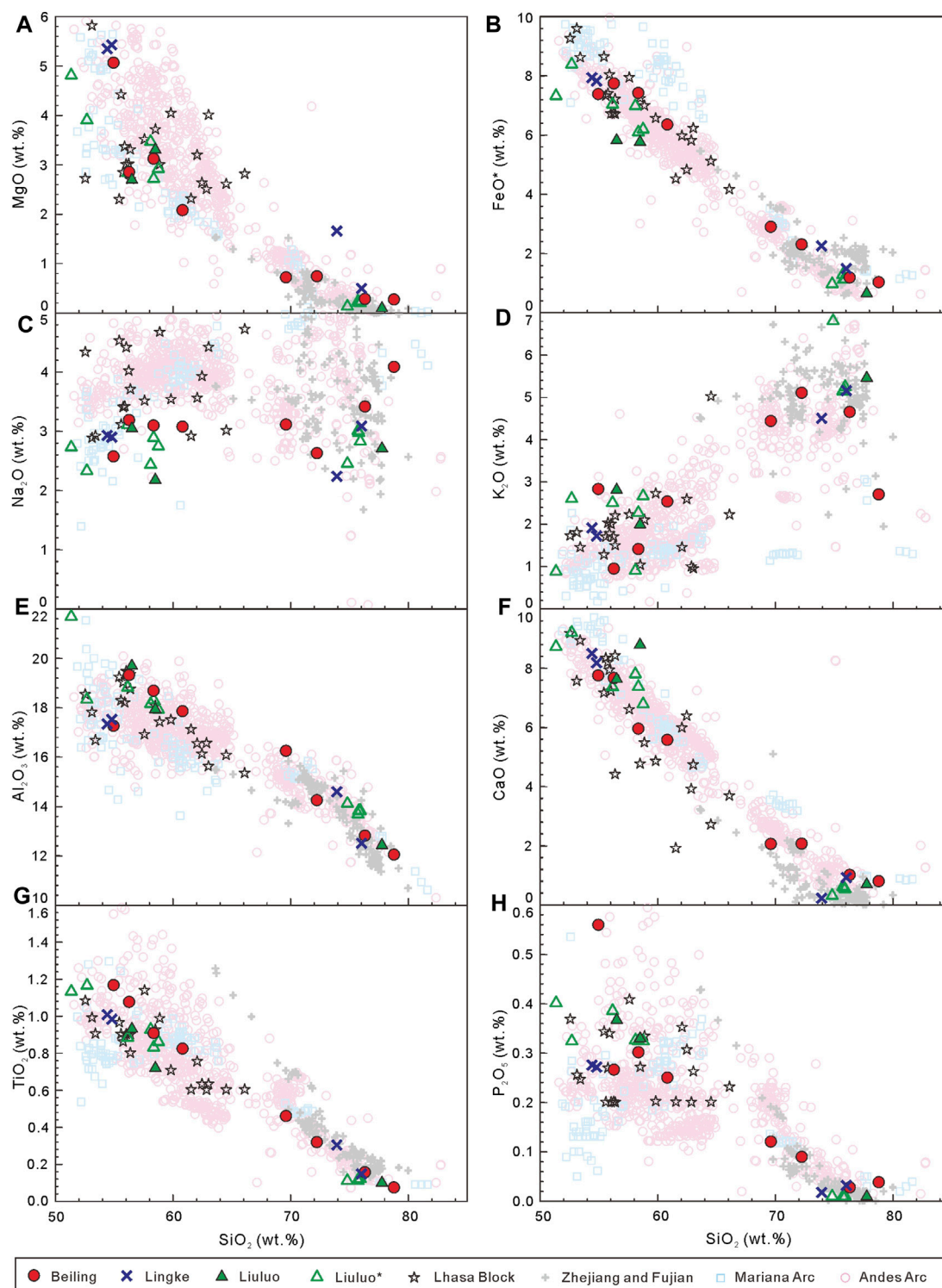
Arc (Figure 4; the average geochemical data of volcanic rocks in the Andes Arc are based on the data from Tormey et al. (1995); Kay et al. (2005); Mamani et al., 2008; Mamani et al., 2009; Schiano et al. (2010); Folkes et al. (2011); Garrison et al. (2011); Pistolesi et al. (2015); Anderson et al. (2017); Bablon et al. (2020)).

## 5 Discussion

### 5.1 Magmatic evolution and source of the mid-Cretaceous volcanic rocks in southern Hainan Island

The  $\text{FeO}^*$ ,  $\text{MgO}$ ,  $\text{TiO}_2$ ,  $\text{Al}_2\text{O}_3$ ,  $\text{P}_2\text{O}_5$  and  $\text{CaO}$  contents of volcanics in southern Hainan Island decrease with increasing  $\text{SiO}_2$  contents (Figure 5), but  $\text{K}_2\text{O}$  contents increase, similar to the relationships found in volcanic rocks from the Andes Arc (Tormey et al., 1995; Kay et al., 2005; Mamani et al., 2008; Mamani et al., 2009; Schiano et al., 2010; Folkes et al., 2011; Garrison et al., 2011; Pistolesi et al., 2015; Anderson et al., 2017; Bablon et al., 2020). The significant linear relationships between concentrations of the major elements in all samples of the study area imply that the magmatic sources of basic-intermediate-acid volcanic rocks are relatively consistent, and the volcanic magmas underwent similar magmatic evolution processes. The fractionation of dark minerals, plagioclases and apatites contributed to the decrease in most of major element contents, such as  $\text{FeO}^*$ ,  $\text{CaO}$  and  $\text{P}_2\text{O}_5$ . In addition, Zhou et al. (2015) determined that the  $^{87}\text{Sr}/^{86}\text{Sr}(t)$  ratios of volcanics in the Liuluocun Formation range from 0.707532 to 0.7089651, and  $\epsilon_{\text{Nd}}(t)$  values range from  $-4.09$  to  $-2.35$ . These narrow isotopic ranges indicate that the mid-Cretaceous magmatic sources in southern Hainan Island were relatively uniform and close to an EM-II type source.



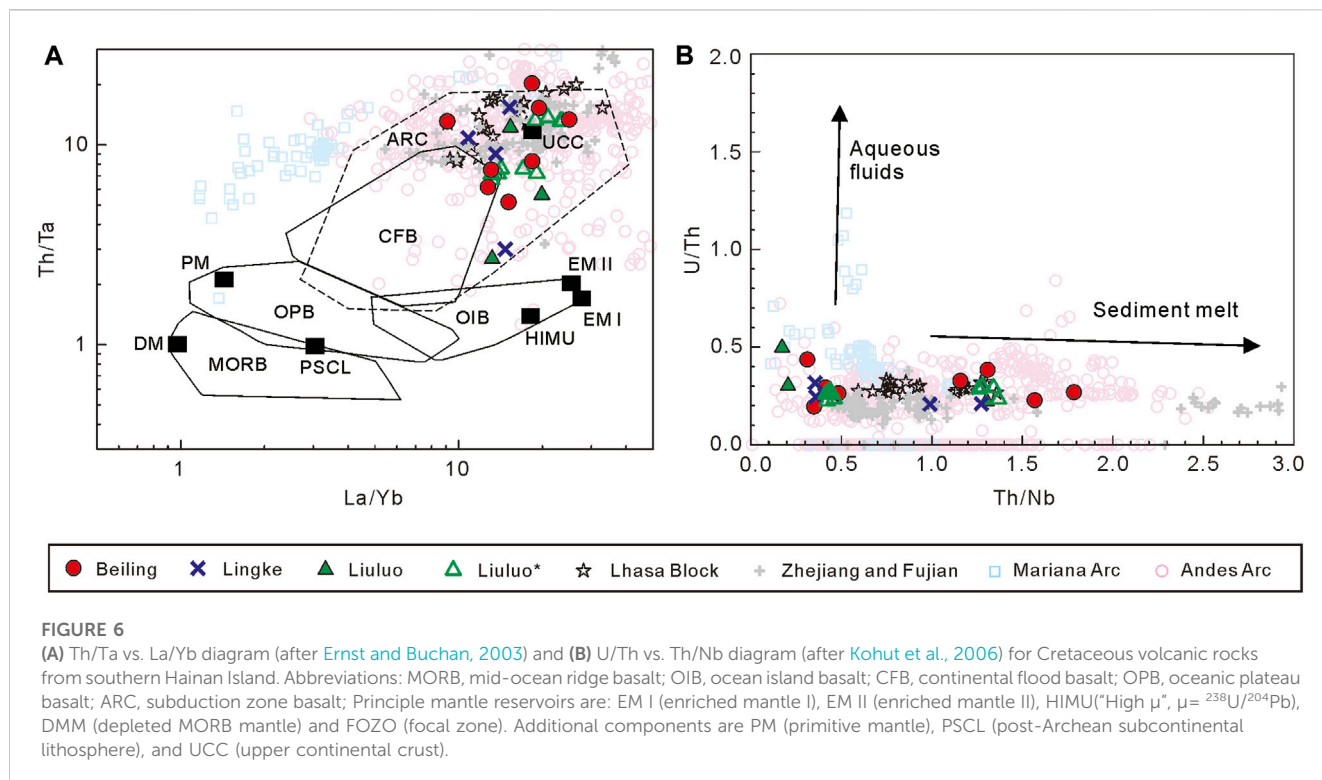


**FIGURE 5**

Selected major element oxides versus  $\text{SiO}_2$  plots for Cretaceous volcanic rocks from southern Hainan Island. The geochemical data of volcanic rocks of Liuluo\* are from Zhou et al. (2015a).

The high La/Yb and Th/La ratios of the volcanic rocks indicate an arc-type magmatic source during the mid-Cretaceous in southern Hainan Island (Figure 6A). The narrow ranges of La/Yb and U/Th ratios, combined with the significantly elevated Th/Ta and Th/Nb

ratios (Figures 6A, B), suggest that the magmas originated from the EM-II type source with inputs of continental crust material, as in the Andes Arc. Liu et al. (2020, 2021) showed that the andesites in Guangdong and Guangxi, north of Hainan Island, also had similar



geochemical characteristics, and the mid-Cretaceous magmatic sources were altered by the melt of continental sediments. In addition, zircon  $\varepsilon_{\text{Hf}}(t)$  data range from  $-9.73$  to  $+0.47$  ([Zhou et al., 2015a](#)), which is the result of the addition of continental crust materials to the magmatic source.

The input of continental materials into a magmatic source can occur during subduction. A large number of mid-Cretaceous andesites indicate that a subduction tectonic environment impacted Hainan Island during the Cretaceous. These andesites exhibit geochemical characteristics like Andean andesites, including remarkably high La/Yb ratios, high Th/Yb ratios and low Sc/Ni ratios ([Figures 7A, B](#)). These geochemical features also imply that the western margin of the SCS represented an Andes-type active continental margin during the Cretaceous period. Additionally, the presence of negative Eu anomalies, relatively flat chondrite normalized HREE patterns, low Y contents and low Sr/Y ratios observed in high-Mg andesites bear resemblance to sanukitic high-Mg andesites ([Kamei et al., 2004; Tatsumi, 2006; Tang and Wang, 2010](#)). These high-Mg andesites are believed to originate from the mantle peridotite that has undergone metasomatism by melts derived from subducted slab and sediments ([Tatsumi, 2001; 2006](#)). Moreover, the formation of these andesites can be attributed to ridge subduction tectonics ([Tatsumi, 2001; 2006; Tang and Wang, 2010](#)). At the same time, rhyolites in the study area have low  $(\text{Zr} + \text{Nb} + \text{Ce} + \text{Y})$  values and  $\text{FeO}^*/\text{MgO}$  ratios ([Figure 7C](#)), which are similar to I-type or S-type granites that mainly form under a compressional tectonic setting ([Lu and Sang, 2004](#)). The lower Y+Nb values and Rb contents of magmatic rocks also imply that the rhyolites were formed in the volcanic arc tectonic environment ([Figure 7D; Pearce et al., 1984](#)).

In summary, the geochemical characteristics of the andesites and rhyolites of southern Hainan Island indicate that the volcanic rocks

were formed in a volcanic arc tectonic setting. The high but relatively similar Zr contents, combined with the low Ti contents which exhibited a wide range in values ([Figure 8A](#)), not only show that these volcanic magmas are of the arc type, but also indicate a magmatic evolution similar to those in Mexico, Ecuador, Chile and other regions ([Pearce and Norry, 1979](#)). Furthermore, Hainan Island experienced an active continental margin tectonic setting during the mid-Cretaceous that was an Andes-type active continental margin. Because of the fluids or melts in subduction zones, the Th/Yb ratio increases ([Figure 8B](#)), leading to enrichment of the geochemical characteristics due to subduction of volcanic rocks in the northwestern SCS during the mid-Cretaceous.

## 5.2 Tectonic setting of the western SCS margin during the mid-Cretaceous

The Cretaceous tectonics of Hainan Island and its surrounding areas are often regarded as the southwestern part of the magmatic belt in the eastern SCB, which resulted from the westward subduction of the paleo-Pacific Plate ([Li et al., 1999; Thuy et al., 2004; Geng et al., 2006; Yan et al., 2010; Wang et al., 2012; Tang et al., 2014; Zhou et al., 2015a; Xu et al., 2016; Sun et al., 2017; Yan et al., 2017; Nong et al., 2021](#)). However, many differences were found by analyzing the late Mesozoic magmatic activities on Hainan Island and the eastern SCB as detailed below.

First, the mid-Cretaceous (c.a. 100 Ma) volcanic rocks in the Zhejiang-Fujian area are characterized as rhyolite-dominated bimodal volcanic suits, with few records of andesite ([Chen et al., 2008; Li et al., 2014; Liu et al., 2016](#)). In contrast, intermediate acid volcanic rocks dominate in southern Hainan Island, and andesites have also been reported in southeastern Guangxi, western

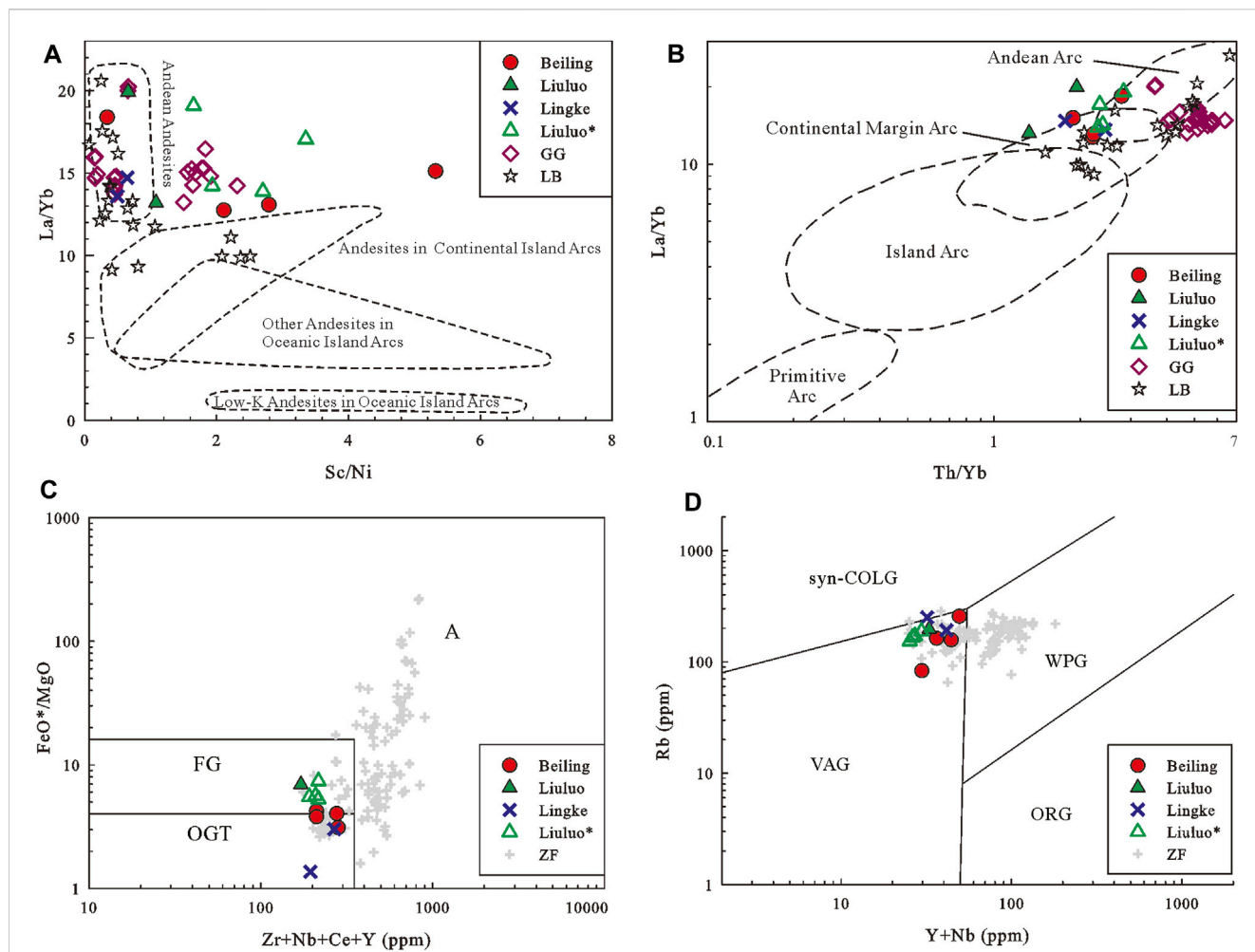


FIGURE 7

(A) La/Yb vs. Sc/Ni diagram (after Bailey, 1981) and (B) La/Yb vs. Th/Yb diagram (after Condie, 1986) for Cretaceous andesitic rocks from southern Hainan Island; (C) FeO\*/MgO vs. (Zr+Nb+Ce+Y) diagram (after Whalen et al., 1987) and (D) Rb vs. (Y+Nb) diagram (after Pearce et al., 1984) for Cretaceous rhyolitic rocks from southern Hainan Island and its adjacent areas. Annotations: The geochemical data of mid-Cretaceous volcanic rocks in Guangxi and Guangdong are from Liu et al. (2021) and Jiang et al. (2015). The geochemical data of those in the Lhasa Block are from Ran et al. (2019) and Zhang et al. (2019a). The geochemical data of those in Zhejiang and Fujian are from Lapierre et al. (1997), Yu et al. (2008), Yan et al., 2016, Yan et al., 2018, Hong (2012), Hong et al. (2013), Li et al. (2017), and Qiu et al. (1999a). Abbreviations: FG, fractionated felsic granites; OGT, unfractionated M-, I- and S-type granites; VAG, volcanic arc granites; syn-COLG, syn-collision granites; WPG, within plate granites; ORG, ocean ridge granites; GG, Guangxi and Guangdong; LB, Lhasa Block; ZF, Zhejiang and Fujian.

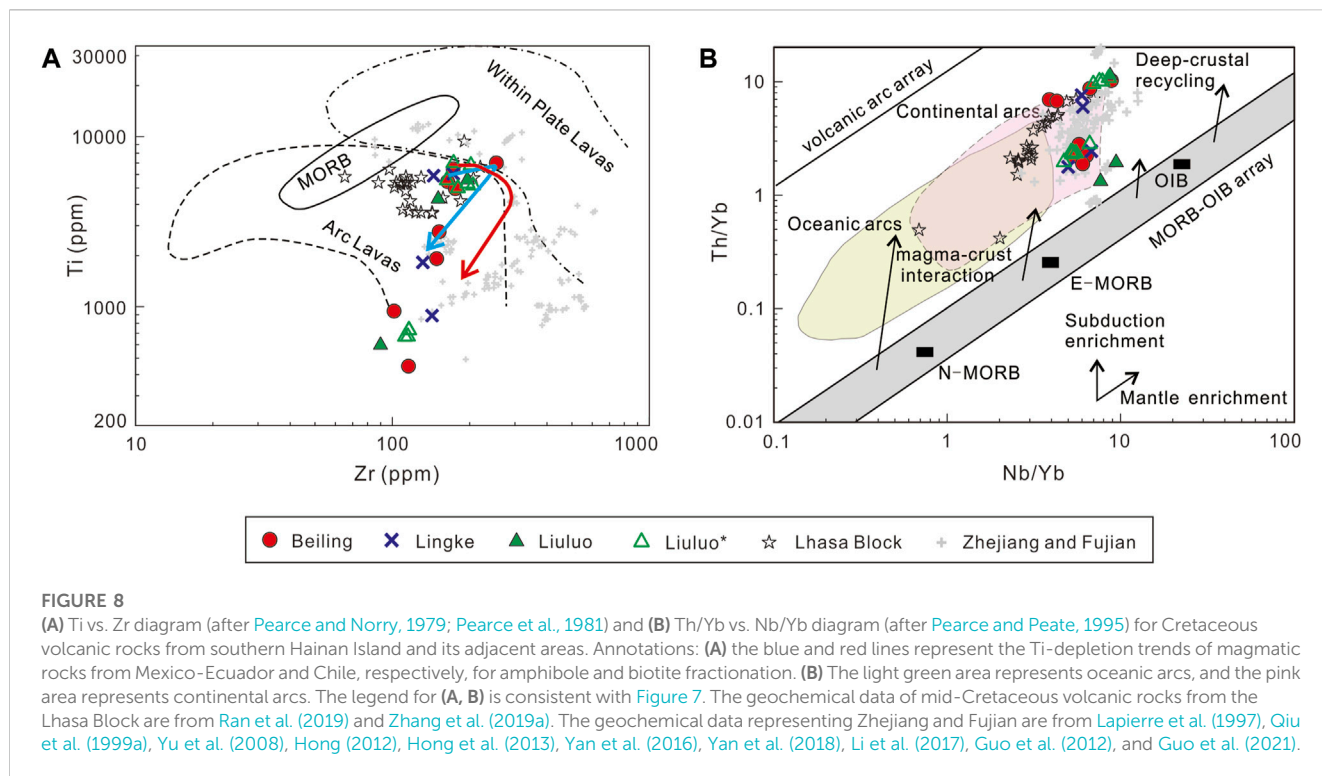
Guangdong and southern Vietnam, surrounding Hainan Island (Jiang et al., 2015; Liu et al., 2020; Liu et al., 2021). Besides, there are contemporaneous diorites reported in Hainan Island (Dilek and Tang, 2020). Intermediate magmatic activities were strong on the western margin of the SCS during the mid-Cretaceous. In addition, rhyolites in Hainan Island contain a small amount of K<sub>2</sub>O and (Zr+Nb+Ce+Y) (Figure 3B; Figure 7C), showing volcanic arc settings (Figure 7D). While some mid-Cretaceous rhyolites in Zhejiang and Fujian are more alkaline and indicate extensional settings (Figures 3A, B; Figure 7D), namely, back-arc extensional environment (Yan et al., 2018; Guo et al., 2021).

Secondly, the rhyolites from Zhejiang and Fujian exhibit higher temperatures (determined using the method described in Watson and Harrison, 1983), such as those from Yandangshan (786°C–864°C, Yan et al., 2016) and Yunshan (861°C–930°C, Yan et al., 2018). The Paleo-Pacific plate subduction changed from a low-angle subduction during the Jurassic period to a high-angle

subduction during the Cretaceous (Zhou and Li, 2000; Zhou et al., 2006). Rhyolites had high zircon-saturation temperatures in the mantle-upwelling tectonic setting due to the back-arc extension in Zhejiang and Fujian (Yan et al., 2016; Yan et al., 2018). However, saturation temperatures of acid volcanic rocks in southern Hainan Island are much lower, primarily ranging from 746°C–790°C with a temperature outside of this range (808°C) in only one sample. Thus, the magmatic activities in the southwestern SCB did not produce rocks with the characteristics of those experiencing lithospheric extension and high magmatic temperatures as the result of mantle upwelling, like in the eastern coastal area of the SCB.

Thirdly, from the central to the coastal areas of the SCB, the age of magmatic rocks gradually decreases, indicating the magmatic arc migrated south-eastward during the Yanshanian period (around the Jurassic-Cretaceous) (Zhou and Li, 2000; Zhou et al., 2006; Li et al., 2007; Liu et al., 2012; Zhou et al., 2015a). As a result, Cretaceous





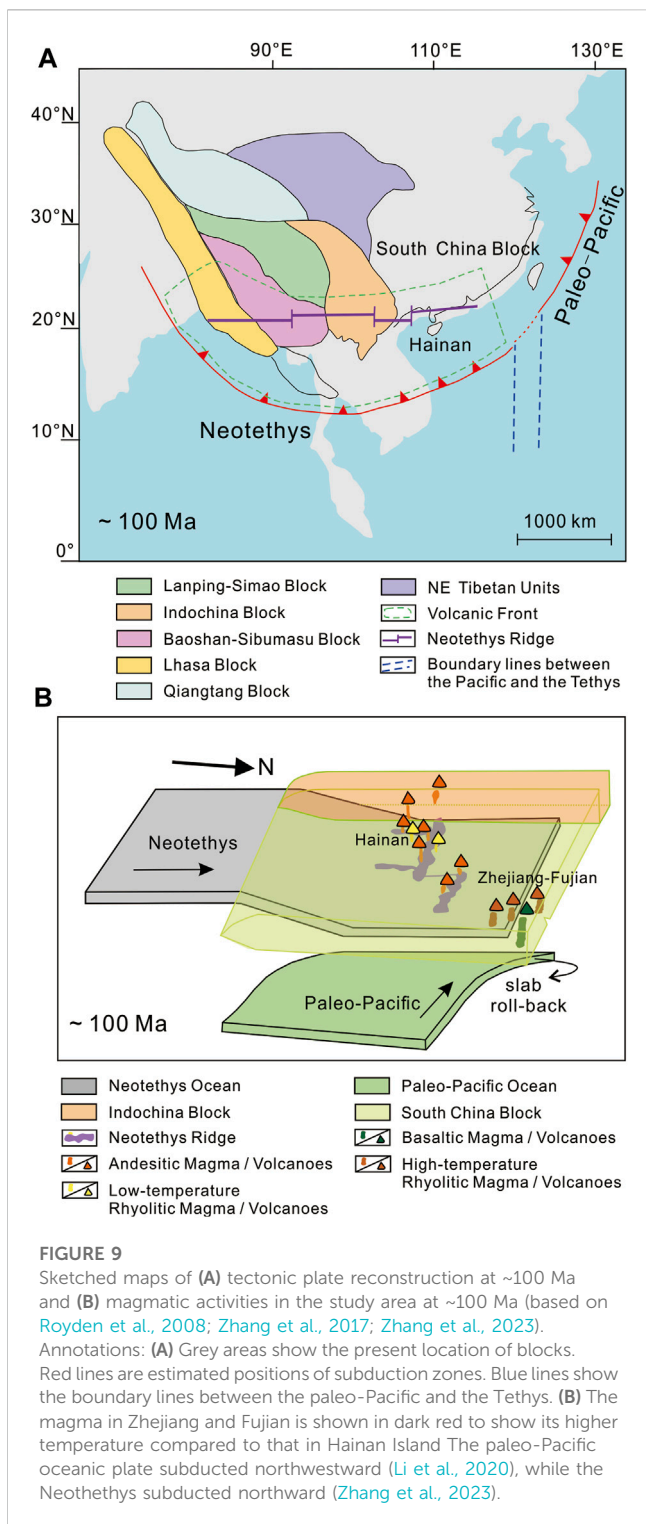
magmatic activity was concentrated in the eastern coastal area of Zhejiang and Fujian (Zhou and Li, 2000; Zhou et al., 2006). According to the retreat model of paleo-Pacific subduction (Zhou and Li, 2000; Zhou et al., 2006), the magmatic activity of Hainan Island in the mid-Cretaceous should be significantly weaker than during the Jurassic, due to its southwestern location in the SCB. However, the Jurassic strata in Hainan Island are not developed, and there were no Jurassic volcanic rock records. The exposed area of Jurassic granitoids is about 572.2 km<sup>2</sup>, while the Cretaceous granitoids, mainly concentrated around the ages of 110–90 Ma, have an exposed area of more than 2900 km<sup>2</sup> (Hainan Geological Survey, 2004; Yichang IGMR and Hainan GS, 2004; Hainan Geological Survey, 2007). Therefore, the magmatic activities in the mid-Cretaceous were much stronger than during the Jurassic.

Finally, mid-Cretaceous alkaline granites, containing alkaline dark minerals, have been found in coastal areas of Zhejiang and Fujian (Qiu et al., 1996; Qiu et al., 1999b; Qiu et al., 2000; Xiao et al., 2007; Zhao et al., 2018) (Figure 1B), and have very high Ga/Al values and (Zr + Ce + Nb + Y) contents. They are typical A-type granites and provide powerful evidence of an extensional environment in the eastern part of the SCB, which resulted from the rollback of the paleo-Pacific Plate or the oblique subduction (Zhou and Li, 2000; Zhou et al., 2006; Li et al., 2007; Li and Li, 2007; Wong et al., 2009). In contrast, on Hainan Island and its adjacent areas on the western margin of the SCS, I- and S-type granitoids are reported widely, without reports of typical A-type granitoids from the mid-Cretaceous.

In summary, the western margin of the SCS, where Hainan Island is located, was not influenced by the paleo-Pacific subduction, which controlled the tectonic and magmatic evolution of the eastern SCB in areas such as Zhejiang and Fujian. Previous work has shown that the subduction of the paleo-Pacific to East Asia in the

Cretaceous mainly affected the area east of Wuyi Mountain (Figure 1B; Chen et al., 2005; Zhang et al., 2017). Previous studies (Yan et al., 2014; Hennig et al., 2017; Ye et al., 2018; Guo et al., 2021) have reported that contemporaneous magmatic records in the eastern and southern microblocks around Hainan Island, emphasizing the more western interior of Hainan Island that was unlikely to be within the subduction range of the paleo-Pacific plate. At that time, the paleo-Pacific subduction could not have contributed to the arc-type magmatic activities on Hainan Island and its adjacent areas, and the western margin of the SCS was not within the affected range of the paleo-Pacific subduction. In the view of the fact that the southeastern edge of the Eurasian plate, where Hainan Island is located, is adjacent to the Tethys tectonic belts to the west, more attention should be paid to the impact of the Tethys on the western SCS.

At ca. 100 Ma, the Neotethyan ridge subducted northward, and upwelling of the hot asthenosphere along the slab window caused the partial melting of the subducted oceanic slab and mantle wedge, resulting in strong magmatism (Zhang et al., 2019a; Zhang et al., 2019b). As a result, adakitic rocks and the high-Mg andesites of this period were exposed in southeastern Lhasa Terrane (Zhang et al., 2019a; Zhang et al., 2019b). At similar times on Hainan Island, the granitoids from Qianjia and Tunchang also show the geochemical characteristics of adakites, such as high Sr/Y ratios with low Y and Yb contents (Wang et al., 2012; Sun et al., 2018). Kelemen (1995) limited the Mg<sup>#</sup> value of high-Mg andesite to 45, which means that some andesites of the Liuluo and Lingke Formations are high-Mg andesite. There were high-Mg andesites-adakitic rocks in the mid-Cretaceous assemblage on Hainan Island. This mid-Cretaceous assemblage is also found in Guangxi and Guangdong, to the north of Hainan Island (Sun et al., 2017; Zhang et al., 2017; Liu et al., 2020; Liu et al., 2021). In the modern subduction zones, this



type of assemblage is often related to the subduction of the ocean ridge (Sajona et al., 2000; Bourdon et al., 2003; Castillo, 2008). The same assemblage occurs both in the western margin of the SCS and the eastern Lhasa Terrane and is the same age, which indicates that the two areas were in the special tectonic setting of the subducting Neotethyan ridge (Figure 9). Moreover, the basalts in southern Hainan Island exhibit Nb contents ranging from 9.20–10.79 ppm (Zhou et al., 2015a), while the Nanxiong basalts (96 Ma) in

Guangdong Province, north of Hainan Island display Nb contents between 22.61–24.28 ppm (Shu et al., 2004), indicating their enriched-Nb characteristics. Additionally, the Cretaceous mafic dikes (101–93 Ma) on Hainan Island contains Nb concentrations of 12.36–20.22 ppm (Tang et al., 2010), further supporting the existence of Nb-enriched basaltic magma in this region and its surrounding during this time as well as ridge subduction processes (Wang et al., 2020). During the mid-Cretaceous period, Neotethyan ridge began to subduct northward into southern Asia. Consequently, young oceanic slab and terrigenous sediments melted upon subduction and metasomatized the mantle wedge (Tatsumi, 2001; Tatsumi, 2006). As a result of these geological processes occurring simultaneously across different regions within Southeastern Lhasa Terrane and Hainan Island high-Mg andesites-adakitic rocks were formed.

Due to the opening of the SCS and subsequent extrusion tectonic movement in Asia (Tapponnier et al., 1982), Hainan Island is geographically distant from the Neotethys suture zone located southwest of Sumatra Island and Borneo Island (Zahirovic et al., 2016). According to a paleomagnetic study of the Khorat Basin on the Indochina Block, the collision between India and Asia caused the Khorat Basin to move  $950 \pm 150$  km southward and rotate clockwise by  $16^\circ$ – $17^\circ$  relative to the SCB (Charusiri et al., 2006). Another study by Singsoupho et al. (2014) suggests the southern transport was 750–1700 km. Thus, the paleolatitude of the southern margin of the Indochina Block and the northern margin of the SCS were roughly the same in the late Mesozoic, leading to the formation of a nearly E-W trending magmatic belt from Hainan Island to the Lhasa Block. Cretaceous granitoids are found in many places in the south of this belt, such as the Xisha Islands, Mekong Basin, South Con Son Basin, Natuna Islands, Malay Peninsula, and Singapore (Areshev et al., 1992; Li et al., 1999; Yan et al., 2010; Yan et al., 2014; Oliver et al., 2011; Ng et al., 2015; Zhu et al., 2017; Gillespie et al., 2019), which also resulted from the northward subduction of the Neotethys. Considering different generations of the Tethys Ocean may be characterized as land (islands) alternating with seas (just as Indonesian Archipelago; Fang, 2002), the possibility cannot be ruled out that the area of sea affecting the northwestern part of the SCS was a branch of the Neotethys. This does not conflict with the tectonic framework of the western margin of SCS, which was affected by the subduction structure of the Neotethys in the Cretaceous.

## 6 Conclusion

- (1) The volcanic rocks of southern Hainan Island are an assemblage of basaltic andesite-andesite-rhyolite, belonging to high-K calc-alkaline series or calc-alkaline series, with metaluminous-peraluminous characteristics. The andesites and rhyolites are enriched in LREEs with positively skewed REE distribution curves, and negative Eu anomalies of the rhyolites are obvious. Additionally, they are enriched in LILEs and depleted in HFSEs.
- (2) There are a large number of mid-Cretaceous andesites on Hainan Island, and acid volcanic rocks formed at low temperatures. No A-type granite of the same age was found, indicating the tectonics of Hainan Island were not influenced by the subduction of the paleo-Pacific Plate in the mid-Cretaceous.

(3) The geochemical characteristics of the mid-Cretaceous andesites and rhyolites indicate an active continental margin tectonic setting on Hainan Island. The high-Mg andesites and adakitic rocks of Hainan Island and its adjacent area can also be found in the Lhasa Block. During the mid-Cretaceous, the western margin of the SCS was mainly affected by the subduction of the Neotethys.

## Data availability statement

The original contributions presented in the study are included in the article/Supplementary material, further inquiries can be directed to the corresponding author.

## Author contributions

YL, NF, and ZW designed the research. YL and NF conducted fieldwork, collecting samples. YL, NF, and ZW conducted the lab work. YL and ZW wrote the paper. NF revised the paper critically for important intellectual content. All authors contributed to the article and approved the submitted version.

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## Conflict of interest

The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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