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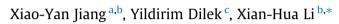
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## Cretaceous magmatic arc in Hainan and the peri-South China Sea as evidenced by geochemical fingerprinting of granitoids in the region



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Mesozoic magmatic rocks occur widely in the South China Block and are generally interpreted as the manifestations of the subduction of the Paleo-Pacific oceanic lithosphere beneath Asia. Subductiondriven magmatism in southeast (SE) China continued from the Late Permian through the Late Cretaceous with an inferred lull between 125 Ma and 115 Ma that is known in the literature as the Cretaceous "magmatic quiescence". We report in-situ zircon U-Pb ages, Hf-O and whole-rock Sr-Nd isotopes, and whole-rock geochemistry of Cretaceous granitoids on Hainan Island and discuss their magmatic evolution within the framework of the Late Mesozoic geodynamics of SE China. We recognize two main stages of the emplacement of Cretaceous granitoids on Hainan, first around 120 Ma and then around 100-95 Ma, displaying high-K calc-alkaline, I-type geochemical affinities. Granites in both age groups are enriched in LILE and LREE, but depleted in Nb, Ta, Ba, Sr, and Eu. The 120 Ma granites have zircon  $\varepsilon_{Hf}(t)$  values of -2.6 to 2.3 corresponding to Hf crustal model ages, ranging from 0.79 Ga to 1.03 Ga, and  $\delta^{18}$ O values ranging from 6.9% to 7.7%. Zircons from 100–95 Ma granites have  $\varepsilon_{Hf}(t)$  values of -4.2 to 1.1 corresponding to Hf crustal model ages of 1.08 Ga to 1.42 Ga, and  $\delta^{18}$ O values ranging from 6.7% to 8.4%. Increasing  $\epsilon_{Hf}(t)$  values of the Cretaceous Hainan granites with younger crystallization ages indicate addition of more juvenile components and reworking of crustal material into their melt evolution. The  $\varepsilon_{Nd}(t)$  values of the 120 Ma and 100–95 Ma granitoids range between -4.1 to -0.4 and -7.7 to -4.0, respectively. The calculated two-stage model age of the 100-95 Ma granitoids clusters between 1.25 Ga and 1.53 Ga. These isotopic data suggest that magmas of the Cretaceous granitoids were produced by partial melting of Mesoproterozoic metabasaltic rocks, which make up much of the crystalline basement of the southern Cathaysia block. The geochemical and isotopic characteristics of the Cretaceous granitoids on Hainan resemble those of magmatic arcs in the Circum-Pacific orogenic belts and identical to those of nearly coeval granitoid intrusions in the continental fragments within the South China Sea basin. We interpret these Cretaceous granitoids in the Peri-South China Sea region as the remnants of a once contiguous Late Mesozoic magmatic arc system that bounded the southern margin of the entire continental Southeast Asia. Our findings do not support the existence of an episode of magmatic quiescence in the geological record of SE China during the Aptian.

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## 1. Introduction

The widespread occurrence of Mesozoic magmatic rocks in South China is one of the most conspicuous geological features of continental East Asia (Fig. 1) (e.g., Zhou and Li, 2000; Li and Li, 2007) and has been widely used to suggest the operation of an Andean-type active margin during the Mesozoic till the Late Cretaceous due to the subduction of the Paleo-Pacific Plate beneath the continent (e.g., Jahn et al., 1990; Charvet et al., 1994; Zhou and Li, 2000; Zhou et al., 2006; Li and Li, 2007; Li et al., 2012; Jiang and Li, 2014; Suo et al., 2019; Cui et al., 2021). Along with voluminous magmatism, structural grabens, local rift systems, and sedimentary basins developed in the South China Block because of the Mesozoic active margin tectonics (Dilek and Tang, 2021; Liu et al., 2021). However, this magmatic activity seems to have had a lull during the Aptian (125–115 Ma) that is known in the literature as the Cretaceous "magmatic quiescence" in SE China

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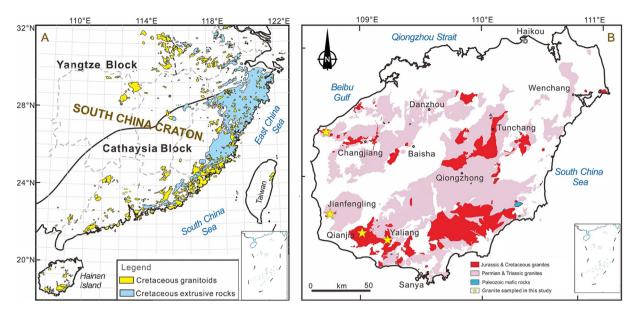
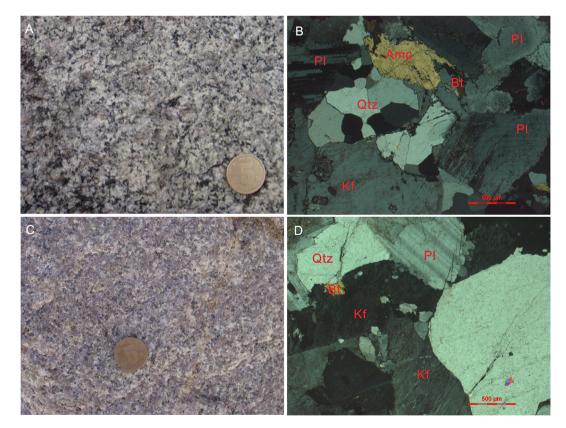


Fig. 1. Distribution of (A) Cretaceous igneous rocks in SE China (modified after Li et al., 2012), and (B) Indosinian-Yanshannian igneous rocks on Hainan Island (modified after Li et al., 2006).

(Li et al., 2010a, 2015; Mao et al., 2013; Wei et al., 2023). The cause (s) and the mode of this Early Cretaceous event are not well established.

Hainan Island is situated at the southern margin of the continental South China Block, separated from the mainland by the Qiongzhou Strait (Fig. 1). The distribution of lithological units, the structural architecture, and the regional geology of the Hainan Island reflect the geological character of the Mesozoic geology of SE China, and hence it is considered as a Mesozoic microcosm of SE China (Dilek and Tang, 2021). However, in comparison to numer-



**Fig. 2.** Field and photomicrographs of the Cretaceous granitoid rocks on Hainan Island. (A) Field photograph of granodiorite 12HN56; (B) thin section photograph of the granodiorite sample 12HN56, showing the rock-forming minerals quartz, potassium feldspar, plagioclase, amphibole, and biotite (cross-polarized light); (C) field photograph of syenogranite sample 12HN80; (D) photomicrograph of the syenogranite sample 12HN80, showing the rock-forming minerals of quartz, plagioclase, and biotite (cross-polarized light). Mineral abbreviations: Qtz = quartz, Kfs = potassium feldspar, Pl = plagioclase, Amp = amphibole, Bt = biotite.

ous studies of magmatic rocks in the South China Block in mainland China, less attention has been paid to the Cretaceous igneous rock assemblages on Hainan Island. Consequently, the geochronological framework, and lithological and isotopic characteristics of the Cretaceous magmatic rock units on Hainan are not well documented. Previous studies have reported the occurrence of several  $\sim$  100 Ma and  $\sim$  73 Ma granitic plutons, and some 130– 80 Ma mafic dike intrusions (Ge et al., 2003; Wang et al., 2012a; Jiang and Li, 2014; Tang et al., 2014; Zhou et al., 2015; Xu et al., 2016; Dilek and Tang, 2021).

We have investigated the Cretaceous granitoid intrusions on Hainan to: (i) document their melt sources and magmatic evolution; (ii) determine the timing of their emplacement and crystallization; (iii) present a systematic correlation of the Cretaceous magmatism and tectonic evolution of Hainan with those of the continental SE China and the adjacent continental fragments (i.e., Taiwan, Palawan, SW Borneo, SW Vietnam and S Cambodia) in the South China Sea region; and (iv) find out what the new data may tell us about the occurrence of the so-called "Cretaceous magmatic guiescence" in SE China. To this end, we undertook in-situ U-Pb and Hf-O isotope analyses of zircons from the Cretaceous granites, conducted whole-rock major and trace element geochemistry analyses of the dated samples, and obtained their Sr-Nd compositions. In the first part of the paper, we summarize the regional geology of South China and the occurrence of Cretaceous granitoids on Hainan. We then present our new geochemical, geochronological, and isotopic data on those granitoids. In the last part of the paper, we discuss the melt evolution and multi-stage development of the Cretaceous magmatism on Hainan, present a model of magmatic arc origin of this Cretaceous magmatism on Hainan and along the southern margin of continental SE China, and evaluate the validity of the Aptian "magmatic quiescence" concept in SE China. Our tectonomagmatic model has strong implications for the latest Mesozoic paleogeography of the continental margin of SE Asia and the westernmost Paleo-Pacific Ocean.

## 2. Regional geology background of South China Block

Separated from the North China Craton by the Qinling-Dabie Orogen to the north, the South China Block further developed when the Cathaysia block became welded into the Yangtze block during the Jiangnan orogeny in the Neoproterozoic (Charvet et al., 1996; Zhao and Cawood, 2012). The basement of the Cathaysia block contains predominantly the early Neoproterozoic volcanic and sedimentary rock successions, which were metamorphosed to greenschist-lower amphibolite facies at  $\sim$  980 Ma. These crystalline basement rocks are overlain by the late Neoproterozoic to Paleozoic terrestrial and marine sedimentary rocks (Li et al., 2010b; Shu et al., 2011; Xia et al., 2015). The Pre-Mesozoic rocks are mainly developed to the west of the Zhenghe-Dapu Fault, generated by multiple stages of magmatism, metamorphism, and deformation at  $\sim$  1.9–1.8 Ga,  $\sim \! 1.4$  Ga,  $\sim \! 1.0 \text{--} 0.7$  Ga, and 0.46– 0.42 Ga (e.g., Li et al., 2002, 2005; Wang et al., 2012b, 2013; Shu et al., 2021). The Cathaysia block experienced extensive tectonic and magmatic events in the Mesozoic due to the subduction of the Paleo-Pacific oceanic slab beneath East Asia and acquired voluminous igneous rock suites, which can be subdivided into three groups according to their age: Permo-Triassic (~260-210 Ma), Jurassic (~190-150 Ma) and Cretaceous (~145-73 Ma) (e.g., Li and Li, 2007; Li et al., 2007, 2014a, 2020; Jiang and Li, 2014; Xu et al., 2020; Liu et al., 2021; Chen et al., 2022). The Permo-Triassic magmatism produced calc-alkaline I-type granites, peraluminous S-type granites, and minor alkaline rocks distributed in the continental hinterland and on Hainan Island that are interpreted to be the manifestations of flat-slab subduction of the Paleo-Pacific Plate or a collision between South China and other blocks (Li et al., 2006, 2012; Zhou et al., 2006; Li and Li, 2007; Wang et al., 2012b, 2013; He et al., 2020). The Jurassic igneous rocks, which are composed of I- and A-type granites and minor basaltic rocks, are mainly scattered in the Wuyishan Mountains–Nanling Range and in the adjacent regions (Zhou et al., 2006; Li et al., 2007; Guo et al., 2012a; Huang et al., 2015). Various models exist regarding their tectonomagmatic origin: foundering of a flat-slab beneath the continent (Li and Li, 2007; Li et al., 2012; Huang et al., 2015), deepening of the subduction slab angle (Zhou and Li, 2000), or intraplate rifting (Gilder et al., 1991).

Manifestations of the Late Mesozoic magmatism occur principally along the eastern and southern margins of the Cathaysia block and display a spatial pattern of oceanward (towards the SE) migration through time (Zhou et al., 2006; Zhu et al., 2014; Cao et al., 2022). The Cretaceous igneous rocks distributed along the coastal area of SE China are composed mainly of high-K calcalkaline, I-type and subordinate A-type granites, as well as their volcanic counterparts (Fig. 1A) (Li, 2000; Zhou and Li, 2000; Zhou et al., 2006; Li and Li, 2007; Li et al., 2014b; Zhao et al., 2019; Xu et al., 2020; Chen et al., 2022). The Cretaceous granitoids in SE China comprise A- and I-type granites with minor S-type granites (Lapierre et al., 1997; Li, 2000; Zhou and Li, 2000; Zhou et al., 2006). Coeval volcanic rocks exhibit bimodal compositions dominated by > 90% felsic rocks with minor amounts (<10%) of basaltic rocks, which include the  $\sim$  140–130 Ma Nanyuan Formation (and its equivalents) volcanic rocks and  $\sim$  110-90 Ma Shimaoshan Group (and its equivalents) volcanic rocks (unconformably covering the foliated and metamorphosed Nanyuan Formation) (Charvet et al., 1994; Lapierre et al., 1997; Zhou et al., 2006; Guo et al., 2012b; He and Xu, 2012; Li et al., 2012, 2014a; Meng et al., 2012). In addition, Cretaceous mafic dikes locally occur within some intraplate rift basins (Li, 2000; Zhou and Li, 2000).

## 3. Field occurrence and mineralogy of the Cretaceous granitoids of Hainan

Located in the northwestern margin of the South China Sea (Fig. 1A), Hainan Island occurs tectonically at the intersection of the Pacific oceanic plate, the Indochina Block, and the southern margin of the South China Block (Li, 2002; Metcalfe, 2009). Because of the similarities of their geology, the Hainan Island is commonly considered as part of the Cathaysian block (Li et al., 2002, 2008). The metamorphic basement exposed on the island consists of:  $(i) \sim 1.43$  Ga amphibolite-facies gneissic granitoids and minor metasedimentary and metavolcanic rocks of the Baoban complex; (ii)  $\sim~$  1.44–1.43 Ga greenschist-facies metasedimentarymetavolcanic rocks of the Shilu Group; and (iii) unconformably overlying  $\sim$  1.2–1.0 Ga quartzite and quartzo-schists of the Shihuiding Formation (Li et al., 2002, 2008; Yao et al., 2017). Phanerozoic sedimentary rock sequences (except for the Devonian and Jurassic strata) and Cenozoic basalts also crop out on Hainan (Wang et al., 1991; Liu et al., 2015; Tian et al., 2021).

Granitoid intrusions are widespread on Hainan Island (Fig. 1B), accounting for ~ 40% of the island's surface area (Wang et al., 1991). Apart from the deformed Middle Permian gneissic granites at Wuzhishan in the central part of the island (Li et al., 2006), Late Permian to Triassic granitoids (ca. 270–230 Ma) are composed mainly of massive and porphyritic syenogranite, monzogranite, and granodiorite (e.g., Wang et al., 1991, 2012a; Ge, 2003; Xie et al., 2005, 2006; Li et al., 2006; Tang et al., 2013; Jiang and Li, 2014; Dilek and Tang, 2021). Subordinate Yanshanian (mainly Cretaceous) granitoid intrusions and their volcanic counterparts are recognized throughout the island. With the exceptions of several

well-dated plutons, high-precision geochronological studies of the Yanshannian granitoid rocks are yet to be done.

In this study, we have investigated a series of granitic plutons in the Yaliang, Qianjia, Jianfengling, and Changjiang areas of the island (Fig. 1B). The Yaliang and Jianfengling plutons in the south are intrusive into the Permian–Triassic granitoids and are relatively small in size. The Qianjia and Changjiang plutons are intrusive into Lower Paleozoic strata or Upper Paleozoic rock bodies in the NE and SW parts of Hainan, respectively, cover large areas, and are overlain by Quaternary deposits.

The Yaliang granodiorite in the south–central part of the island is a medium- to coarse-grained pluton, consisting mainly of euhedral to subhedral tabular plagioclase and alkaline feldspar (Fig. 2). Xenomorphic, granular quartz and minor, platy dark-green biotite also occur in these rocks, with accessory minerals of zircon and apatite.

A medium-grained porphyritic biotite-amphibole monzogranite collected from the central part of the Qianjia Pluton in the south (Fig. 1B) shows a perthitic texture in alkaline feldspar phenocrysts ( $\sim$ 30 vol%) and contains quartz ( $\sim$ 30 vol%) and plagioclase ( $\sim$ 30 vol%) as the main components of the matrix. Plagioclase laths are tabular, exhibiting polysynthetic twinning. Xenomorphic granular quartz grains are interspersed with feldspar and mafic minerals (Fig. 2)). Minor minerals include biotite ( $\sim$ 3 vol%) and amphibole ( $\sim$ 5 vol%). Apatite, zircon, magnetite, and sphene form accessory mineral phases.

The Jiangfengling pluton in the SW part of the island is a subrounded, composite granitic intrusion (Fig. 1B). The main part of this pluton consists of Triassic massive biotite-syenogranite and monzogranite. A small exposure of a coeval diorite intrusion occurs along the outer rim of the Jiangfengling pluton. The mineral assemblage of the dioritic rock includes quartz, potassium feldspar, plagioclase, and biotite. The accessory minerals are zircon, apatite, sphene, and magnetite/ilmenite.

The Changjiang granite porphyry in the west-central part of the island occurs in the form of dikes in a pluton, which displays a typical porphyritic texture with large crystals of idiomorphic and tabular, pink K-feldspar and anhedral, granular quartz grains. Minor plagioclase, sphene and biotite are also observed in thin section. The accessory minerals include apatite, zircon, magnetite, and ilmenite.

## 4. Analytical methods

#### 4.1. SIMS zircon U-Pb dating

Zircons were separated from rock samples using standard density and magnetic separation techniques. Approximately two hundred zircon grains from each sample, together with zircon standards, were loaded onto an epoxy mount. The mount was then polished to section the zircon grains in half. Zircons were imaged in transmitted and reflected-light photomicrographs, as well as in cathodoluminescence (CL) images, to reveal their internal structures. The zircon mount was then vacuum-coated with highpurity gold, prior to SIMS analysis.

Zircon U–Pb isotopes were analyzed on a Cameca 1280 SIMS at the Institute of Geology and Geophysics, Chinese Academy of Sciences (IGG–CAS), using standard operating conditions (7-scan duty cycle, ~8 nA primary  $O_2^-$  beam, 20 µm × 30 µm analytical spot size, mass resolution ~ 5400). A single electron multiplier was used in ion-counting mode to measure secondary ion beam intensities by peak jumping. U–Pb ratios were determined relative to the standard zircon Plešovice ( $^{206}$ Pb/ $^{238}$ U = 0.05369, corresponding to 337.1 Ma; Sláma et al., 2008), and the absolute abundances were determined relative to the standard zircon M257 (U = 840 ppm, Th/U = 0.27; Nasdala et al., 2008). Analyses of Plešovice were interspersed with those of unknown grains, using operating and data processing procedures described by Li et al. (2009). A long-term uncertainty of 1.5% (1RSD) for <sup>206</sup>Pb/<sup>238</sup>U measurements of the standard zircons was propagated to the unknowns (Li et al., 2010b), even though the measured <sup>206</sup>Pb/<sup>238</sup>U error during this study is  $\sim 1\%$  (1 RSD). Measured Pb isotopic compositions were corrected for common Pb, using the <sup>204</sup>Pb-method. An average Pb of present-day crustal composition (Stacey and Kramers, 1975) is used for the common Pb correction, assuming that it is largely due to surface contamination introduced during sample preparation. Uncertainties on individual analysis in the data table are reported at  $1\sigma$  level; mean ages for pooled U/Pb analyses are quoted with 95% confidence intervals. Data reduction was carried out using the Isoplot/Exv. 3.70 program (Ludwig, 2008). SIMS zircon U-Pb age data are given in **Supplementary Data Table S1**.

In order to monitor the external uncertainties of SIMS U-Pb measurements, analyses of an in-house zircon standard Qinghu were interspersed with unknowns. Forty-nine analyses of this standard zircon yielded a Concordia age of  $159.8 \pm 0.7$  Ma (MSWD of concordance = 0.16), identical within a margin of error to its reported age of  $159.5 \pm 0.2$  Ma (Li et al., 2013).

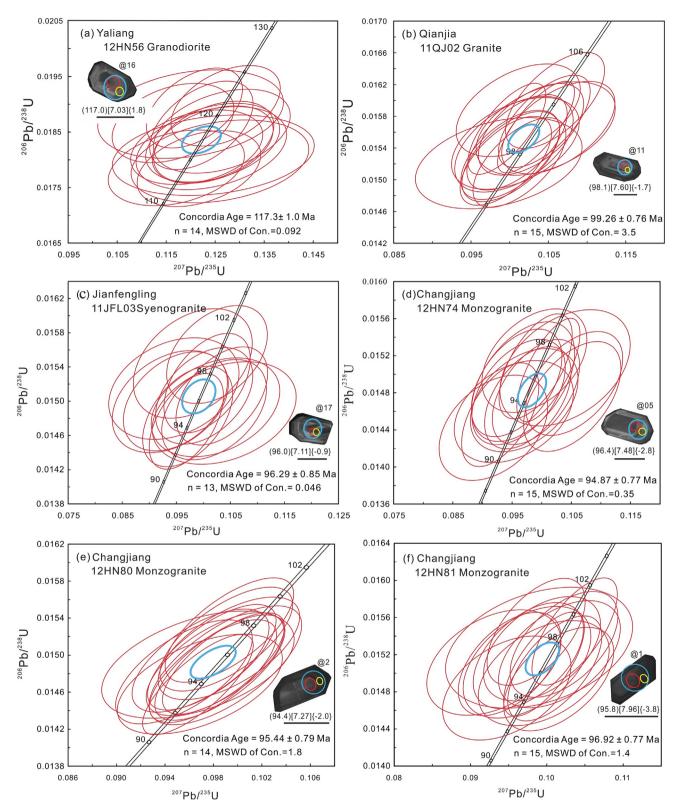
#### 4.2. Major and trace element analyses

Geochemical analyses of our granite samples were carried out at the Guangzhou Institute of Geochemistry, Chinese Academy of Sciences (GIG-CAS). Major element oxides were measured by a Rigaku RIX 2000 X-ray fluorescence spectrometry (XRF) in the form of fused glass beads. Precision for the analytical results is typically 1%–5%. An Agilent 7500a quadruple ICP–MS (inductively coupled plasma mass spectrometry) was used for determining trace elements. The signal drift of the spectrometer was monitored by an internal standard Rh solution. Analytical uncertainties for most elements are generally < 5%. The operational and calibrated procedures for the major and trace element analyses are similar to those detailed by Li (2002). Major and trace element analyses are listed in **Supplementary Data Table S2**.

## 4.3. In-situ zircon Hf-O isotopic analyses

In-situ Hf-O isotope analyses were conducted on zircons with obtained crystallization ages from the dated samples. Before SIMS oxygen isotope analysis was done, the sample zircon mount was reground to ensure that any oxygen implanted in the zircon surface from the  $O_2^-$  beam used for U-Pb analysis was removed. LA-MC-ICPMS zircon Hf-isotope was analyzed following the SIMS oxygen isotope measurement. This sequence of measurements enables U-Pb age determinations and Hf-O isotopic compositions to be done on the sites shown in Fig. 3.

In-situ zircon Lu-Hf isotope analyses were carried out on a Neptune Plus MC-ICPMS equipped with a Geolas 193 nm excimer ArF laser-ablation system at the IGG-CAS. The analytical procedures and calibration methods are similar to those described by Wu et al. (2006). Lu-Hf isotopic analyses were obtained on the same zircon grains that were previously analyzed for U-Pb and O isotopes, with ablation pits of 60 µm in diameter. The ablation time was 26 s for each measurement at a repetition rate of 8 Hz at 10 J/cm<sup>2</sup>. Measured <sup>176</sup>Hf/<sup>177</sup>Hf ratios were normalized to <sup>179</sup>- $Hf/^{177}Hf = 0.7325$ . Standard zircon 91500 was analyzed along with unknowns to monitor the reproducibility of the data and the stability of the instrument, and it yielded a weighted average <sup>176</sup>Hf/<sup>177</sup>Hf ratio of 0.282301 ± 0.000004 (2SE, n = 90) in good agreement (within errors) with the reported value of 0.282307 ± 0.000031 (2SD) (Wu et al., 2006). Zircon Hf isotope data are listed in Supplementary Data Table S3.



**Fig. 3.** SIMS U–Pb concordia age plots for zircons from the Cretaceous granitoids on Hainan Island and representative cathodoluminescence (CL) images. Photomicrographs of representative zircons analyzed for U-Pb ages, and Lu-Hf and O isotopes. Small ellipses indicate the SIMS analytical spots for U-Pb and O isotopes, and the large circles denote the LA-MC-ICP-MS analytical spots for Lu-Hf isotopes. Numbers near the analytical spots are the U-Pb ages (within parentheses),  $\delta^{18}$ O values [within square brackets], and  $\varepsilon_{Hf}(t)$  values [within large brackets].

Zircon oxygen isotopes were measured using the same Cameca 1280 at SIMS laboratory in IGG-CAS. The Cs<sup>+</sup> ion beam was accelerated to 10 kV, with an intensity of  $\sim$  2 nA. The analysis site was the same as for U-Pb dating, and the spot size is about 20  $\mu m$  in diam-

eter. The normal incidence electron flood gun was used to compensate for sample charging. The NMR (nuclear magnetic resonance) was used for stabilizing magnetic field. Oxygen isotopes were measured in multi-collector mode with two off-axis Faraday cups. Analytical procedures are the same way as described by Li et al. (2010c). The instrumental mass fractionation factor (IMF) was corrected using Penglai zircon standard with a  $\delta^{18}$ O value of 5.31% (Li et al., 2010d). The internal precision of a single analysis was generally better than 0.20% (1 $\sigma$  standard error) for the <sup>18</sup>O/<sup>16</sup>O ratio. The external precision, measured by the reproducibility of repeated analyses of Penglai standard during this study, is 0.40% (2SD, n = 58). During this study, an in-house zircon standard Qinghu was also measured as an unknown together with other unknowns. Twenty-seven measurements of Qinghu zircon yield a weighted mean of  $\delta^{18}$ O = 5.40% ± 0.50% (2SD), which is consistent within errors with the reported value of 5.4% ± 0.2% (Li et al., 2013). Zircon O-isotope data are listed in **Supplementary Data Table S3**.

#### 4.4. Whole-rock Sr-Nd isotopes

Chemical separation of Sr and Nd from the sample matrix was undertaken by conventional ion-exchange techniques. The detailed procedures are the same as those reported by Yang et al. (2011), and only a brief description is given here. For Sr–Nd isotopic analysis, about 100 mg of rock samples were dissolved using mixed HF + HNO<sub>3</sub> acid in Teflon bombs at ca. 195 °C for 2-d. Strontium and rare earth elements (REEs) were separated using AG50W × 12 (Bio-Rad) cation-exchange resin, and Nd was further separated by EICHROM © Ln-resin.

Whole-rock Sr isotopic compositions were analyzed using a Finnigan MAT 262 thermal ionization mass spectrometer at IGG-CAS. Measured <sup>87</sup>Sr/<sup>86</sup>Sr ratios were normalized to <sup>86</sup>Sr/<sup>88</sup>Sr = 0.1194. The replicated analyses of <sup>87</sup>Sr/<sup>86</sup>Sr ratio of NBS SRM 987 standard solution was 0.710233  $\pm$  0.000026 (2SD, n = 6), consistent with the recommended value of 0.710248  $\pm$  0.000023 (Thirlwall, 1991). Whole-rock Nd isotopic measurements were performed on a Thermo Scientific Neptune MC–ICPMS at the IGG–CAS. Measured <sup>143</sup>Nd/<sup>144</sup>Nd ratios were normalized to <sup>146</sup>Nd/<sup>144</sup>Nd = 0.7219 using the exponential law. The measured <sup>143</sup>Nd/<sup>144</sup>Nd ratio of the Shin Etsu JNdi-1 standard solution was 0.512108  $\pm$  0.000014 (2SD, n = 6), consistent within errors with the recommended value of 0.512115  $\pm$  0.000007 (Tanaka et al., 2000). The whole-rock Sr and Nd isotopic data of the studied samples are given in **Supplementary Data Table S4**.

## 5. Results

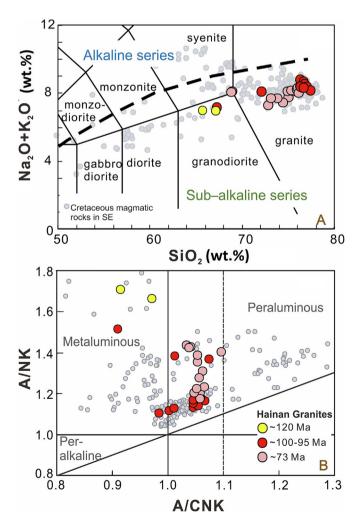
#### 5.1. Zircon U-Pb ages

Zircons separated from the six granite samples are euhedral, transparent, and prismatic in crystal form, ranging in length at  $30-200 \mu$ m, with aspect ratios of 1:1 to 4:1. Oscillatory zoning is common as seen in CL images (Fig. 3). The analyzed zircons have variable abundance of U (157–1720 ppm) and Th (87 – 2867 ppm) contents, with relatively high Th/U ratios (0.26–2.13), indicating their magmatic origin. Values for  $f_{206}$  are 0.00%–0.94%. All the analyses are concordant in  $^{206}$ Pb/ $^{238}$ U and  $^{207}$ -Pb/ $^{235}$ Pb within analytical errors. Based on the results of our zircon analyses, we can separate the Cretaceous granitoid rocks into two age subgroups of 117 Ma and ca. 100–90 Ma.

The Yaliang granodiorite (12HN56) belongs to the 117 Ma subgroup. The analyses conducted on the sample yielded a Concordia age of  $117.3 \pm 1.0$  Ma (MSWD of concordance = 0.09) (Fig. 3a) that represents the crystallization age of this granitoid. Three samples that are dated between 100 Ma and 95 Ma include granites from the Qianjia (11QJ02), Jianfengling (11JFL03), and Changjiang (12HN74, 12HN80 and 12HN81) plutons. Fifteen zircons from the Qianjia pluton gave consistent U–Pb ages of 99.3  $\pm$  0.8 Ma (MSWD of concordance = 3.5) (Fig. 3b). Thirteen grains from the Jianfengling pluton yielded a Concordia age of 96.3  $\pm$  0.9 Ma (MSWD of concordance = 0.046) (Fig. 3c). Analysis of 15 zircon grains from the 12HN74 (Changjiang) produced a Concordia age of 94.9  $\pm$  1.0 Ma (MSWD of concordance = 0.35) (Fig. 3d). 12HN80 present Concordia ages of 95.4  $\pm$  0.8 Ma (n = 15, MSWD of concordance = 1.8) (Fig. 3e). Another granite (12HN81) from Changjiang has the concordant U–Pb age 96.9  $\pm$  0.8 Ma (n = 15, MSWD of concordance = 1.4) (Fig. 3f). The ages between 100 Ma and 95 Ma are representative of the emplacement ages of these three Cretaceous intrusive bodies.

## 5.2. Whole-rock geochemistry

The Cretaceous granitoid rocks exhibit a relatively wide range of chemical compositions, with SiO<sub>2</sub> = 65.7-77.5 wt.%, Al<sub>2</sub>O<sub>3</sub> = 11.5 – 15.7 wt.%, CaO = 0.46-4.39 wt.%, and P<sub>2</sub>O<sub>5</sub> = 0.01-0.18 wt.%. They are high in total alkalis, with K<sub>2</sub>O = 3.59-5.10 wt.% and Na<sub>2</sub>O = 2.79-4.09 wt.%, but low in MgO = 0.11-2.02 wt.% and Fe<sub>2</sub>-O<sup>T</sup><sub>3</sub> = 0.39-4.02 wt.%. On the total alkali-silica (TAS) diagram, these rocks fall into the granite and granodiorite fields, belonging to the sub-alkaline series (Fig. 4A). They are metaluminous to weakly per-



**Fig. 4.** (A) Rock classification diagram for the Cretaceous granitoids. (B) A/NK vs. A/ CNK diagram. A/NK = Al/(Na + K) (molar ratio). A/CNK = Al/(Ca + Na + K) (molar ratio). Data for the Late Cretaceous granitoids on the Hainan Island are from Jiang and Li (2014). Data for the Cretaceous magmatic rocks in SE China are from: He et al. (2009), Jiang et al. (2011), He and Xu (2012), Chen et al. (2013, 2014a), Li et al. (2013, 2014b, 2015), Deng et al. (2014), Zhao et al. (2015).

aluminous in compositions, with A/CNK values between 0.91 and 1.07 (Fig. 4B). All samples fall into the high-K calc-alkaline series field in the SiO<sub>2</sub> vs. K<sub>2</sub>O diagram (Fig. 5). According to the classification scheme of Frost et al. (2001), the samples plot in the alkalicalcic and calc-alkaline, magnesian fields (Fig. 5). The varied TiO<sub>2</sub>, MgO, CaO, and P<sub>2</sub>O<sub>5</sub> contents decrease with increasing SiO<sub>2</sub> contents (Fig. 5).

In the Chondrite-normalized REE diagram (Fig. 6A), the Cretaceous granitic rocks exhibit steep slopes, with the enrichment of the LREE relative to HREE, and mildly to strongly negative Eu anomalies . On the primitive mantle-normalized incompatible element spidergram (Fig. 6B), they appear enriched in LILE (e.g., Rb, Th and U) and LREE, but depleted in Nb, Ta, Ba, Sr and Eu. The zircon saturation temperatures calculated by using the equation proposed by Watson and Harrison (1983) fall between 700 °C and 800 °C.

#### 5.3. Zircon Hf-O isotopic compositions

Zircon grains from the ca. 117 Ma granite have variable initial <sup>176</sup>Hf/<sup>177</sup>Hf ratios ranging from 0.282632 to 0.282767, corresponding to  $\varepsilon_{\rm Hf}(t)$  of –2.6 to 2.3 (Fig. 7). The crustal Hf model age ( $T_{\rm DM}^{\rm C}$ ) varies from 0.79 Ga to 1.03 Ga. Their measured  $\delta^{18}$ O values range from 6.9‰ to 7.7‰ (Fig. 7). Zircon grains of the ca. 100–95 Ma granitoid rocks have variable initial <sup>176</sup>Hf/<sup>177</sup>Hf ratios from 0.282617 to 0.282745, corresponding to  $\varepsilon_{\rm Hf}(t)$  of –4.2 to 1.1 (Fig. 7), with average crustal Hf model age ( $T_{\rm DM}^{\rm C}$ ) of 1.08–1.42 Ga. In-situ oxygen isotopic analyses show relatively heterogeneous O isotopic compositions, with  $\delta^{18}$ O<sub>zircon</sub> values ranging from 6.7‰ to 8.4‰ (only two spots showing values > 8.0‰).

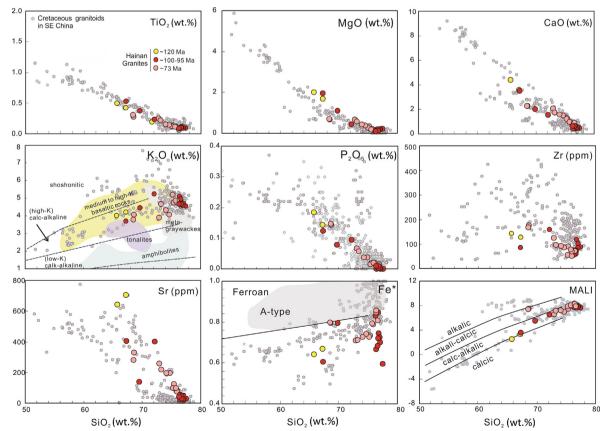
#### 5.4. Sr-Nd isotopic compositions

Eight samples were analyzed for Sr-Nd isotopes. Both Sr and Nd isotopic compositions vary over a large range (Fig. 8). The samples from ca. 117 Ma granitic rocks have revealed  ${}^{87}$ Rb/ ${}^{86}$ Sr = 0.60–1.32 and <sup>87</sup>Sr/<sup>86</sup>Sr = 0.70689–0.70954, corresponding to initial <sup>87</sup>Sr/<sup>86</sup>Sr ratios ( $I_{Sr}$ ) = 0.7057–0.7081. Their measured <sup>147</sup>Sm/<sup>144</sup>Nd ratios are 0.1135-0.1245 and <sup>143</sup>Nd/<sup>144</sup>Nd ratios are 0.51237-0.51256, with calculated  $\varepsilon_{Nd}(t)$  values of -0.4 to -4.1. Their calculated two-stage Nd model age  $(T_{2DM})$  clusters around 0.94–1.24 Ga. The granitoid samples with ages of 100-95 Ma have <sup>87</sup>Rb/<sup>86</sup>Sr ratios of 1.39–41.18 and <sup>87</sup>Sr/<sup>86</sup>Sr ratios of 0.71020–0.78144, corresponding to more evolved and higher initial  ${}^{87}$ Sr/ ${}^{86}$ Sr ratio ( $I_{Sr}$ ) values of 0.7026-0.7344. However, three granitoid samples (12HN74, 12HN80 and 12HN81) have higher Rb/Sr ratios (>10), leading to higher and less reliable I<sub>Sr</sub> ratios (0.7026, 0.7225 and 0.7344). Excluding those values, the obtained  $I_{\rm Sr}$  ratios range from 0.7057 to 0.7101. The measured  $^{147}$ Sm/ $^{144}$ Nd values are 0.0845–0.1204 and <sup>143</sup>Nd/<sup>144</sup>Nd values are 0.51219-0.51239, with calculated  $\varepsilon_{\rm Nd}(t)$  values between -7.7 and -4.0. The calculated two-stage Nd model age  $(T_{2DM})$  of these three samples clusters around 1.25-1.53 Ga.

## 6. Discussion

6.1. Genetic types and melt origin of the Cretaceous granitoids on Hainan

Since the introduction of the terms I- and S- type granites by Chappell and White (1974) and A-type granites by Loiselle and



**Fig. 5.** Harker-type major and trace element plots for the Cretaceous granitoids. Data for the Late Cretaceous granitoids on the Hainan Island are from Jiang and Li (2014), and data for the Cretaceous intrusive rocks in SE China are from: He et al. (2009), Jiang et al. (2011), He and Xu (2012), Chen et al. (2013, 2014a), Li et al. (2013, 2014b, 2015), Deng et al. (2014), Zhao et al. (2015). Correlation between K<sub>2</sub>O and SiO<sub>2</sub> contents for the Cretaceous granitoids as compared with the experimental melts by partial melting of different protoliths. Data . Sources for different protoliths: medium to high-K basaltic rocks from Sisson et al. (2005), tonalites from Singh and Johannes (1996), metagraywackes from Montel and Vielzeuf (1997), amphibolites from Rapp et al. (1991), Wolf and Wyllie (1994) and Patiño Douce (1999)

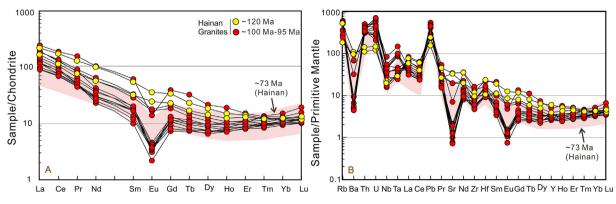
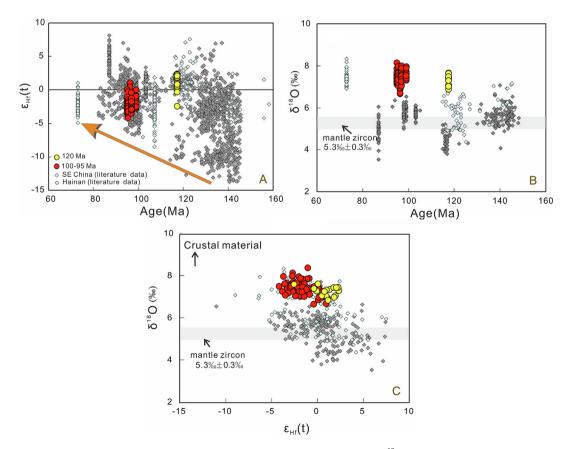


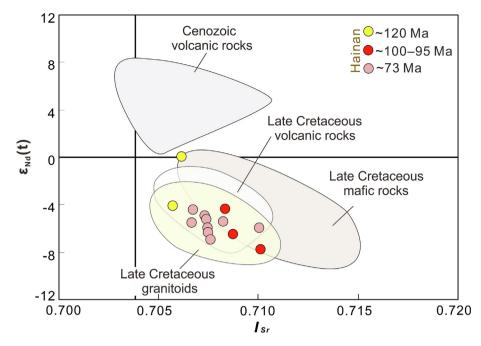
Fig. 6. (A) Chondrite-normalized REE diagram, and (B) primitive mantle-normalized trace element spidergram for the Cretaceous granitoids. Normalization values are from Sun and McDonough (1989). Data for the ca. 73 Ma granites on the Hainan Island are from Jiang and Li (2014).

Wones (1979), granites have commonly been divided into these three types according to their mineralogical and geochemical features. However, differences among these types are not always clear, especially for the case of slightly peraluminous and differentiated granites, which tend to have chemical and mineral compositions converging to those of haplogranites (e.g., Chappell and White, 2001; King et al., 2001; Chappell et al., 2004). The petrogenesis of such peraluminous, differentiated granites is commonly attributed to variable mantle–crust interactions (DePaolo, 1981; Altherr et al., 1999) or recycling of continental crystalline basement material in the melt column, followed by fractional crystallization of their magmas (Patiño Douce and McCarthy, 1998). We discuss below different parameters and variables that may have controlled the physiochemical conditions of the magmatic formation of the Cretaceous granitoids on Hainan.

The ca. 117 Ma Yaliang amphibole-bearing granodiorite samples have relatively low SiO<sub>2</sub> contents (<70 wt.%) and A/CNK values (0.91–0.97), and plot in the high-K calc-alkaline field (Fig. 5). All these characteristics are identical to those of typical I-type granitoids in the Lachlan Fold Belt (Chappell and White, 1992). Moreover, their relatively low  $\delta^{18}$ O values (6.9%–7.7%) also favor an I-type affinity (Valley, 2003; Kemp et al., 2006). On the other hand, most of the ca. 100–95 Ma granites have high SiO<sub>2</sub> contents (>70 wt.%). The absence of alkaline minerals (riebeckite,



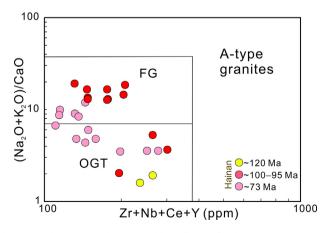
**Fig. 7.** (A) Crystallization ages vs.  $\varepsilon_{\text{Hf}}(t)$  values of zircons with concordant ages; (B) crystallization ages versus  $\delta^{18}$ O values of zircons with concordant ages; (C)  $\varepsilon_{\text{Hf}}(t)$  vs.  $\delta^{18}$ O values of zircons of the Cretaceous granitoids on Hainan. The Hf-isotope evolution line for Depleted Mantle and New Crust are after Griffin et al. (2000) and Dhuime et al. (2011), respectively. Mantle zircon  $\delta^{18}$ O value = 5.3‰ ± 0.6‰ (2SD) (Valley et al., 1998; Page et al., 2007). Data. Sources: He et al. (2010), Guo et al. (2012b), Wang et al., (2012a), Chen et al. (2013, 2014a), Jiang and Li (2014), Liu et al. (2014), Li et al. (2015), Jiang et al. (2015).



**Fig. 8.** Initial <sup>87</sup>Sr/<sup>86</sup>Sr ( $I_{Sr}$ ) vs.  $\varepsilon_{Nd}(t)$  plot for the Cretaceous granitoids. The data of Cenozoic volcanic rocks in southeast China are from Chung et al., (1994,1995,1997), Ho et al. (2003), Zhu et al. (2004), Zhou et al. (2009), Huang et al. (2012); the data of Late Cretaceous volcanic rocks in southeast China are from Zhou and Chen (2001) and Chen et al. (2004); the data of Late Cretaceous granitoids are from Lan et al. (1995), Chen et al. (2004, 2013), Mao et al. (2006), Wang et al. (2012a); the data of Late Cretaceous mafic dikes are from Ge (2003), Qi et al.(2012), Wang et al. (2012a); the data of 73 Ma granites of Hainan Island are from Jiang and Li (2014).

arfvedsonite and aegirine) in these rocks precludes the possibility of their origin as alkaline A-type granites. Although several granodiorite samples display 10,000 × Ga/Al ratios > 2.7, the lower HFSE (Zr + Nb + Ce + Y < 330 ppm) (Fig. 9) values also exclude the possibility of their aluminous A-type origin. The observed petrological features (presence of amphibole) and geochemical characteristics (metaluminous to weakly peraluminous, and the negative correlation between SiO<sub>2</sub> and P<sub>2</sub>O<sub>5</sub>) suggest that the 100–95 Ma granitoids represent I-type granites. The  $\delta^{18}$ O ratios of these granites are lower than those of the S-type granites, which are characteristically higher than 8‰ (Valley, 2003). All these features collectively indicate that these 100–95 Ma granitoids have close geochemical affinity to I-type, rather than A- or S-type granites.

Magmatic temperatures have significant implications for delineating the petrogenesis of granites. A prevalent method for the



**Fig. 9.**  $Zr + Nb + Ce + Y vs. (Na_2O + K_2O)/CaO diagram for the Cretaceous granitoids on the Hainan Island. Data for the Late Cretaceous granitoids on the Hainan Island are from Jiang and Li (2014).$ 

estimation of magmatic temperatures of granites is the zircon saturation thermometer  $(T_{Zr})$ , proposed by Watson and Harrison (1983). The Cretaceous granites on Hainan show a relatively wide range of T<sub>Zr</sub> between 700 °C and 800 °C. Because xenocrystal zircons are rare to absent in our rock samples and because there is a tight negative correlation between their Zr and SiO<sub>2</sub> values (Fig. 5), the maximum  $T_{Zr}$  temperature of each pluton provides a minimum estimate of the magmatic temperature prior to its crystallization. Therefore, the initial magmatic temperatures might have exceeded 800 °C, reminiscent of the "high temperature" and/or "hot" I-type granites (Chappell et al., 1998, 2000; Miller et al., 2003; Collins et al., 2016), which require advective heat input into the crust. Besides temperature, the oxygen fugacity is also an important factor for evaluating the physiochemical conditions of granitic magma formation (Dall'Agnol et al., 2005; Jiang et al., 2018a,b). Previous studies of the Cretaceous granites on Hainan have shown the accessory mineral assemblage of magnetite-tita nite-allanite-apatite (Chen et al., 2008, 2014b), and mineralogical compositions reflecting oxidized conditions. These features are common in magmas produced in subduction-related environments (Wones, 1989).

High-K, calc-alkaline, I-type granites are generally formed by partial melting of medium- to high-K, calc-alkaline, mafic to intermediate rocks (Rapp et al., 1991; Roberts and Clemens, 1993; Rapp and Watson, 1995). Sisson et al. (2005) obtained high-K granitic melts reaching high-silica contents (SiO<sub>2</sub> > 65 wt.%) and Na<sub>2</sub>O/ K<sub>2</sub>O < 1 by using medium- to high-K basaltic compositions as starting materials, similar to the compositions of those low-Si Cretaceous granites on the Hainan Island. Therefore, we consider medium- to high-K metabasaltic rocks as the best candidates for the melt source of the Cretaceous granites on Hainan (Fig. 5). Moreover, the zircon Hf isotopic compositions, which indicate their progressive change with time, suggest an increasing input of juvenile components with a younging trend (Fig. 7). Considering the two-stage Nd model and Hf model ages, we posit that magmas of the Cretaceous granites on Hainan were most likely generated by X.-Y. Jiang, Y. Dilek and X.-H. Li

partial melting of the regional Mesoproterozoic metabasaltic basement rocks with an input of juvenile components.

6.2. Multi-stage development of Cretaceous magmatism on Hainan and comparison with coeval magmatism in SE China

The temporal patterns of Cretaceous magmatism in mainland SE China show a nearly continuous record from 145 Ma to 80 Ma with two major peaks at  $\sim$  130 Ma and  $\sim$  100 Ma, and a short period of apparent quiescence from 125 Ma to 115 Ma (Fig. 10**A**; Guo et al., 2012b; He and Xu, 2012; Jiang et al., 2015, 2021; Li et al., 2015; Chen et al., 2022). Jiang and Li (2014) identified the youngest Mesozoic granite in South China as the ca. 73 Ma Longlou granite. Furthermore, Xu et al. (2016) reported three stages of magma

emplacement for the origin of the Cretaceous, Mo-hosting granitoids during ca. 113–108 Ma, ca. 100–94 Ma, and ca. 90–70 Ma, respectively. Cretaceous magmatic rocks in SE China are made vastly of silicic intrusive and volcanic rock associations. Intrusive rocks are mainly gabbro, syenite, and I-type and A-type granites (Dong et al., 1997; Wong et al., 2009, 2011; He and Xu, 2012; Liu et al., 2012; Li et al., 2015a; Zhang et al., 2019; Peng et al., 2021; Chen et al., 2022).

The compilation of zircon age data reveals that magmatic rocks on Hainan Island were developed through multiple stages during the period of ca. 135–70 Ma, but mainly between 120 Ma and 90 Ma with a major peak at ca. 100 Ma (Fig. 10B). In addition to the 117 Ma Yaliang granodiorite that has been identified and described in this study for the first time, a  $\sim$  133 Ma biotite-

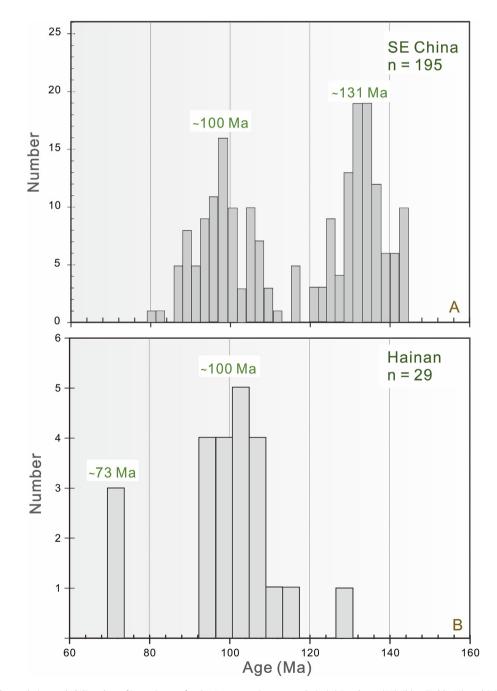


Fig. 10. Histograms and cumulative probability plots of isotopic ages for the Cretaceous igneous rocks in (A) Southeast (SE) China (Table S5), and (B) Hainan Island (Ge, 2003; Tang et al., 2010, 2013, 2014; Wang et al., 2012a; Chen et al., 2014b; Jiang and Li, 2014; Zhou et al., 2015; Li et al., 2016; Xu et al., 2016).

granite was reported in the Shilu deposit (Li et al., 2016), a 107 Ma granodiorite was found in Tunchang (Wang et al., 2012a), and some other plutons (composed of monzogranite and granodiorite) dated at ca. 107–105 Ma were discovered in the southern part of Hainan (Yan et al., 2017). All these Cretaceous plutons that formed during the 100–90 Ma time interval are distributed in the southwestern and eastern parts of the island (Chen et al., 2008; Tang et al., 2010; Wang et al., 2012a; Yan et al., 2017; Sun et al., 2018). Based on our new age data and the available geochronological information, it appears that the Cretaceous granitic magmatism on Hainan Island occurred mainly in three stages: ~135 Ma, ca. 120–90 Ma, and ca. 70 Ma.

In addition to intrusive rocks, there are also limited Cretaceous hypabyssal intrusions and volcanic rocks on the Hainan Island. Mafic dikes that are mainly distributed in the eastern and southern parts of the island are dated at  $\sim$  135 Ma, ca. 117 Ma, ca. 105 Ma, ca. 102–90 Ma, ca. 86 Ma, and ca. 81 Ma (Ge et al., 2003; Wang et al., 2012a; Tang et al., 2014; Dilek and Tang, 2021). A suite of basalt–andesite–rhyolite volcanic rock associations has been recently identified in the southern part of the island and dated at 102 Ma (Zhou et al., 2015). Therefore, the Cretaceous hypabyssal intrusive and volcanic rocks on Hainan can be roughly divided into two broad age groups of ca.135 Ma and ca.120–80 Ma.

Compared with the temporal and lithological characteristics of the Cretaceous rock suites in mainland SE China, their counterparts on Hainan appear different. Mafic plutonic rocks are rare on Hainan and volcanic rocks are limited in exposure. The Cretaceous intrusive rocks on Hainan belong to high-K, calc-alkaline I-type granites.

#### 6.3. Magmatic arc origin of the Cretaceous plutonic rocks on Hainan

The following observations and findings from Hainan support an extended magmatic arc origin of the Cretaceous granitoids on this island: (i) Cretaceous granitoids on Hainan (in this study and previous work) are all high-K, calc-alkaline granites, representing I-type affinities, which commonly occur in active continental margins and in continental arcs (Roberts and Clemens, 1993; Barbarin, 1999); (ii) these Cretaceous granites on Hainan are geochemically akin to granites formed in "syn-collisional" and "volcanic-arc" settings (Fig. 11), and are broadly consistent (geochemically) with the Cretaceous calc-alkaline granitic rocks in SE China; (iii) a progression of increasing  $\varepsilon_{Hf}(t)$  values of granitic zircons through time in the Cretaceous (Fig. 7) suggests a systematic increase of juvenile components into the magmatic plumbing system, analogous to subduction-related magmatism in the circum–Pacific arcs and orogenic belts (Kemp et al., 2009; Collins et al., 2011); and, (iv) the geochemistry of nearly 81 Ma mafic dike swarms in southern Hainan resembles that of magmas formed by partial melting of subduction–metasomatized mantle sources in extensional tectonic settings (Ge et al., 2003).

The widespread occurrence of Cretaceous magmatic rocks, structural domes and basins, metamorphic core complexes, and sedimentary basins in SE China represent the manifestations of extensional deformation in a backarc setting above the subducting Paleo-Pacific slab and inland from this inferred magmatic arc (Li and Li, 2007; Li et al., 2014a, 2020; Zhao et al., 2017; Chu et al., 2019; Dilek and Tang, 2021). The development of the magmatic arc must have followed the trench migration and the slab retreat into the Paleo-Pacific Ocean realm to the E–SE. The  $\sim$  100 Ma basalt, andesite, and rhyolite volcanic rock suites in SE China represent the results of subduction-related magmatism in the backarc environment behind the magmatic arc (Zhou et al., 2015).

## 6.4. Regional extent of the Cretaceous magmatic arc

Granitic plutonic rock associations with ages and compositions similar to those in Hainan and SE China and coeval volcanic rocks have been also reported from farther south in Southeast Asia, including the islands of Palawan and Borneo bounding the southernmost part of the South China Sea, in the Pearl River and the

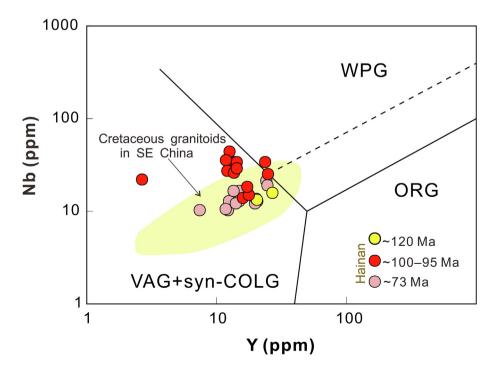


Fig. 11. Tectonic discrimination diagram (Pearce et al., 1984) for the Cretaceous granitoids that plot in the field of VAG (volcanic arc granites) and syn-COLG (syn-collisional granites). Data for the Cretaceous granitic rocks in southeast China are from Chen et al. (2004, 2013), Lan et al. (1995), Li (2000), Qiu et al. (2008) and Xing et al. (2009). Data for the Late Cretaceous granitoids on Hainan Island are from Jiang and Li (2014).

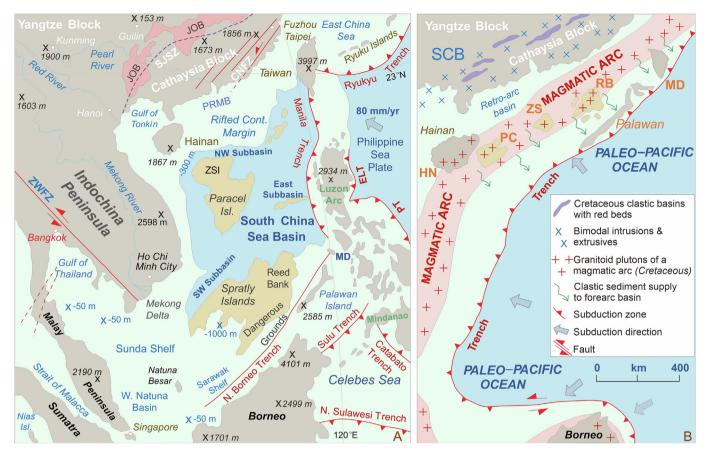


Fig. 12. (A) Simplified tectonic map of modern SE Asia, South China Sea, and its environs. (B) Reconstructed paleogeography of SE Asia and the Cretaceous (~120 Ma) magmatic arc located outboard of the coastal SE Asia (data from: Li et al., 2014a; Zahirovic et al., 2014; Shao et al., 2015; Jiang et al., 2015).

Beibu Gulf Basins to the east and the west of the Hainan Island, and on Taiwan in the NE end of the South China Sea (Fig. 12A) (Yui et al., 1996; Yan et al., 2014; Hennig et al., 2017; Shao et al., 2017; Shao et al., 2019; Cao et al., 2021; Cui et al., 2021; Jin et al., 2022; Xu et al., 2022; Wang et al., 2023). Metamorphosed plutonic rocks with ages of 130-80 Ma in SW Borneo represent the products of the Paleo-Pacific oceanic plate subduction beneath the Indochina-East Malaysia Block during the Cretaceous (Hennig et al., 2017; Wang et al., 2023). Nearly 130-110 Ma granites and doleritic dike intrusions have also been reported from microcontinental blocks in the South China Sea (Nanshan and Zhongsha Islands, and Borneo) (Yan et al., 2010, 2014; Cai et al., 2019; Miao et al., 2021) that were probably once part of the South China Block. The  $\varepsilon_{Nd}(t)$  and  $\varepsilon_{Hf}(t)$  isotopic values, and geochemical characteristics of Jurassic-Cretaceous mafic to felsic, intrusive and extrusive rock associations in Sabah, in western Borneo, SE Vietnam, and Coastal South China Sea display spatial variations (particularly in their  $\varepsilon_{Nd}(t)$  and  $\varepsilon_{Hf}(t)$  values, coupled with younging age progression from the NW to the SE (Wang et al., 2023). These features are interpreted to indicate the existence of a major active continental margin arc as a result of the westward (landwarddipping) subduction of the Paleo-Pacific slab (Fig. 12B; Breitfeld et al., 2020; Batara and Xu, 2022; Wang et al., 2023; van de Lagemaat et al., 2024). Trench and slab retreat caused the eastward migration of this magmatic arc and created microcontinental blocks that were rifted away from the mainland Asia and drifted southwards with the opening and spreading of the South China Sea (Suggate et al., 2014; Cui et al., 2021). Detrital fingerprints (including geochemistry, heavy mineral, and zircon U-Pb geochronology) of Mesozoic to Cenozoic strata from Hainan and Palawan Islands, the Pearl River Mouth Basin, and the South China Sea Basin all reveal similar provenance signatures (with age peaks at ca. 120 Ma and ca. 115 Ma), suggesting that there is a geological record of arc magmatism in the region around 120 Ma (Knittel, 2010; Yokoyama et al., 2012; Suggate et al., 2014; Jiang et al., 2015; Padrones et al., 2017; Shao et al., 2017; Yan et al., 2018; Cao et al., 2021; Cui et al., 2021).

Previous studies have proposed that a ca. 120 Ma magmatic arc existed on the margin of the South China Block and possibly outboard (Jiang et al., 2015). The late Mesozoic igneous rocks found along the South China Sea margins can be geochemically and geochronologically correlated with those widely distributed in the surrounding landmasses, representing a possible continuous continental arc system outboard of Southeast Asia (Fig. 12B and Supplementary Data Fig. S1; Yan et al., 2014; Hennig et al., 2017; Shao et al., 2017; Xu et al., 2017; Cui et al., 2021). Combined with the extant geochronological and geochemical data from Cretaceous magmatic rocks on Hainan and SE China, our new results support the occurrence of an Andean-type magmatic arc along and offshore of SE China throughout the late Cretaceous (Fig. 12B). Lithological assemblages, sedimentary structures, provenance signatures, and the geochronology of detrital minerals indicate, for example, that the West Philippines microcontinent was attached to and was contiguous with mainland China until the Oligocene, when the opening of the South China Sea began (Fig. 12B; Shao et al., 2017; Chen et al., 2019; Cao et al., 2021;

Wei et al., 2023). Thus, the geological evidence from the mainland South China and other continental fragments within the South China Sea points to continued subduction–related magmatism in this region, including Hainan and other microcontinental blocks in the South China Sea.

# 6.5. Was there a magmatic quiescence stage in SE China and Hainan during the Aptian (125–115 Ma)?

Based on the available magmatic zircon U-Pb ages from the continental SE China (Fig. 10A) some geologists have proposed a nearly 10-million year-long magmatic quiescence period (or magmatic lull) between 125 Ma and 115 Ma in the Early Cretaceous. The timing of this inferred magmatic quiescence seems to be consistent with the absence of  $\sim$  120 Ma detrital zircons in all major river systems in South China (Jiang et al., 2015). Some of the current interpretations for the lack of magmatism during this time include: slab window opening due to the break-off of the subducting slab (Li et al., 2014a, 2020) and the collision between a hypothetical micro-continent (e.g., the West Philippine microcontinent) and the SE China continental margin (Faure et al., 1989; Chu et al., 2020; Wei et al., 2023). Considering the deformation and metamorphism that occurred in the Changle-Nan'ao Metamorphic Belt (CNMB) as documented by macroscopic and microscopic structures (Wei et al., 2015, 2023), the emplacement of syn-tectonic plutons (with oriented minerals) during 130-105 Ma (Li et al., 2015; Wei et al., 2023; Zhou et al., 2023), the angular unconformity between ca. 140-130 Ma and ca. 110-90 Ma volcanic rocks in southeast China (Guo et al., 2012b; Li et al., 2018), and the Early Cretaceous ophiolites exposed in the Mindoro Island (Yumul, 2007) and involved in S-directed thrusting, it seems likely that the collision between the South China block and the West Philippines microcontinent caused the NW-SE shortening regime around CNMB and halted magmatism temporarily and creating a pause in magmatic output (Faure et al., 1989; Wei et al., 2015, 2023). More field-based, regional geological, geochronological, isotopic, and geophysical studies have to be conducted to test these two hypotheses. The question of the real cause of the apparent Aptian gap in magmatism in the region is beyond the scope of our current study.

However, it is important to point out that the 120 Ma granites and detrital zircons on Hainan appear to overlap in time with the inferred  $\sim$  125 Ma to 115 Ma "magmatic quiescence" period in mainland southern China. Furthermore, our discovery of  $a \sim 117$  Ma high-K, calc-alkaline I-type granite on Hainan suggests that there was some magmatic activity in the region during this time. Thus, our data from Hainan does not support the possible existence of a magmatic quiescence period in SE China during the Aptian. It is possible that the apparent lack of Aptian magmatic rocks in parts of SE China may also be either due to their local absence or poor documentation in the geological record. Alternatively, this 10 m.y. period of magmatic lull was a result of accelerated, trench-normal slab velocity of the Paleo-Pacific Plate, causing flattening of the subduction angle that, in turn, might have inhibited hydrous melting and associated magmatism in the upper plate (Dilek and Tang, 2021), or the collision between the South China Block and the West Philippines microcontinent (Wei et al., 2023). Hainan Island and the micro-continental blocks in the peri-South China Sea are outside the range of the flattening subduction angle or crustal shortening regime due to the arrival of a microcontinent carried by the subduction slab, the continuing subduction of the Paleo-Pacific plate leading to the continuous magmatic activity. Further field-based structural mapping and precise geochronological studies of Cretaceous magmatic rocks in SE China and on the islands in the South China Sea are needed to test all currently available hypotheses.

## 7. Conclusions

We draw the following conclusions based on the results of our integrated in-situ zircon U-Pb and Hf-O isotope analyses, and whole-rock major, trace element, and Sr-Nd isotope analyses of Cretaceous granites on Hainan Island, and on our synthesis of the compilation of the extant geological, geochemical, and geochronological data from SE China and from the continental fragments in the South China Sea.

- (i) Cretaceous granitic magmatism on the island of Hainan developed during three discrete pulses at 135 Ma, 120– 90 Ma, and around 70 Ma. The Hainan granitoids are all high-K, calc-alkaline granites with an I-type geochemical affinity, typical of magmatic arc granites. Their  $\varepsilon_{\rm Hf}(t)$  values show increasing trends with younging in crystallization ages, indicating more contribution of juvenile components into their melt regime through time. These geochemical signatures and the correlation between the  $\varepsilon_{\rm Hf}(t)$  and  $\delta^{18}$ O values point to the effects of crustal reworking in the late Mesozoic magmatism in SE China.
- (ii) The occurrence of Early to Late Cretaceous granitoid rocks in mainland SE China, on the Hainan, Taiwan, and Palawan Islands, and in continental fragments near the northern margin of the South China Sea points to the development of a magmatic arc above the Paleo-Pacific oceanic slab, which was dipping northward beneath the Asian continent during the Cretaceous. Much of the geological record of this arc was fragmented and obliterated as a result of tectonic processes associated with the opening of the South China Sea during the Cenozoic.
- (iii) The identification of ca. 117 Ma granodiorite pluton on Hainan Island and other magmatic rocks and nearly coeval detrital zircon ages from the West Philippines microcontinent, Taiwan, and other continental blocks in the northern part of the South China Sea (i.e., Zhongsha and Nansha Blocks) indicate that subduction zone magmatism was ongoing in SE Asia during the Aptian (~125 Ma to 115 Ma), a time period that has been widely considered as magmatically null (so-called "magmatic quiescence"). In light of our new findings and a regional correlation of magmatic events presented in this study, we deduce that the subduction of the Paleo-Pacific oceanic plate beneath SE Asia and the spatially associated flux-melting magmatism must have continued well into the Late Cretaceous. Further precise geochronological studies of Cretaceous plutonic and volcanic rocks in the continental SE China and in the microcontinents within the periphery of the South China Sea are needed to rule out the purported, Aptian "magmatic quiescence" event.

#### **CRediT authorship contribution statement**

**Xiao-Yan Jiang:** Formal analysis, Funding acquisition, Investigation, Methodology, Software, Visualization, Writing – original draft, Conceptualization, Project administration. **Yildirim Dilek:** Data curation, Visualization, Writing – review & editing, Funding acquisition. **Xian-Hua Li:** Funding acquisition, Resources, Supervision, Writing – review & editing, Conceptualization.

## **Declaration of Competing Interest**

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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## Appendix A. Supplementary data

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#### X.-Y. Jiang, Y. Dilek and X.-H. Li

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