Tempo-spatial variation of the late Mesozoic volcanism in Southeast China

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Abstract. The magmatism (including volcanism) in East Asia (/ China) could provide key clues and age constrains for the subduction and dynamical process of the Paleo-Pacific plate. Although lots of absolute isotope ages of extrusive rocks have been published in the 1980s-2000s, large uncertainties and big errors prevent the magmatism in SE China from being well understood. In this study, we investigate the zircon geochronology of extrusive rocks and tempo-spatial variations of the late Mesozoic volcanism in Southeast (SE) China. We reported zircon U-Pb ages of new 48 extrusive rock samples in the Shi-Hang tectonic zone. Together with the published data in recent decade, ages of 291 rock samples from ~ 40 lithostratigraphic units were compiled, potentially documenting a relatively complete history and spatial distribution of the late Mesozoic volcanism in SE China. The results show that the extrusive rocks spanned ~ 95 Myr (177-82 Ma), but dominantly ~ 70 Myr (160-90 Ma), within which the volcanism in the early Early Cretaceous (145-125 Ma) was the most intensive and widespread eruption. We propose that these ages represent the intervals of the Yanshanian volcanism in SE China. Spatially, the age geographic pattern of extrusive rocks shows that both the oldest and youngest age clusters occur in coastal magmatic arc (eastern Zhejiang and Fujian), and the most intensive and widespread age group (145-125 Ma) occurs in back arc / rifting basin (eastern Jiangxi, middle Zhejiang, and northern Guangdong), implying that the late Mesozoic volcanism migrated northwestly and subsequently retreated southeastly. This volcanic migration pattern may imply that the Paleo-Pacific plate subducted northwestward and the roll-back subduction did not begin until the Aptian (~ 125 Ma) of the mid-Cretaceous.

Keywords: geochronology; tempo-spatial variation; volcanism; late Mesozoic; Southeast China

1. Introduction

It is generally believed that an Andean-type active continental margin had been developed during the late Mesozoic in eastern Eurasia along which the Paleo-Pacific plate (PPP) subducted beneath the East Asia (e.g., Taylor and Hayes, 1983; Faure and Natal'in, 1992; Charvet et al., 1994; Zhou and Li, 2000; Chen et al., 2005; Liu et al., 2017; Li SZ et al., 2019). The subduction has exerted profound impacts in Southeast (SE) China (e.g., Taylor and Hayes, 1983; Zhou and Li, 2000; Li CL et al., 2014; Li JH et al., 2014; Jiang YH et al., 2015; Liu et al., 2016;) and many other parts of East Asia (e.g., Stepashko, 2006; Wu et al., 2007; Choi and Lee, 2011; Zhang et al., 2011; Sun et al., 2013; 2015; Dong et al., 2016; Liu et al., 2017), as indicated by the pervasive crustal deformation associated with the Yanshanian orogeny (e.g., Lapierre et al., 1997; Li, 2000; Zhou and Li, 2000) and the widespread magmatism (e.g., Zhou et al., 2006; Sun et al., 2007). Obviously, the study of the magmatism would help to constrain the process of the PPP subduction.

The late Mesozoic volcanism in SE China, as a response to the PPP subduction, has long attracted attention, and lots of dating work has been carried out. However, different time intervals and various episodes / cycles / periods of the volcanism have been proposed (e.g., Li et al., 1989; Feng et al., 1993; Zhang, 1997; Guo et al., 2012; Li CL et al., 2014; Liu et al., 2012, 2014, 2016; Jiang SH et al., 2015; Ji et al., 2018; Zhang et al., 2018; Yang et al., 2018; Zhang et al., 2019). The issue can be attributed to: 1) the published ages were generally based on separate and often limited datasets that were commonly from only several to a dozen of samples from a local region such as a mining field, or a province, or at most from a relatively wide area of two neighboring provinces; 2) age data were obtained using different methods, by which, the Rb-Sr, K-Ar, and Ar-Ar dating of bulk-dominated samples yielded ages with large uncertainties and big errors in the 1980s-1990s; 3) refined zircon U-Pb ages of the volcanism have not been analyzed for the whole SE China.

It is essential to obtain spatially more comprehensive datasets from different parts of SE China and also temporally more expanded datasets from sedimentary basin archives that can document the relatively complete volcanic history to achieve a holistic understanding of the late Mesozoic volcanism and geodynamics in SE China.

In this study, we investigate the geochronology of extrusive rocks in the middle and northern Shi-Hang tectonic belt (SHTB. e.g., Gilder et al., 1996; Jiang et al., 2011; Yang et al., 2012). The SHTB contains thick sedimentary strata, which are interbedded with extrusive rocks, and thus has the advantage of providing a more complete stratigraphic archive that preserves more complete and recognizable volcanic events. We also compile the published zircon U-Pb isotope geochronological data of extrusive rocks from the entire SE China. Obviously, ages of the extrusive rocks can constrain the geochronology of the initiation, evolution, and termination of the late Mesozoic volcanism in SE China. Specifically, we analyze the temporal evolution and the geographical distribution of the late Mesozoic volcanism, which can indirectly help date and better understand the slab subduction between the eastern Asian continent and the western PPP in East Asia (e.g., Gilder et al., 1991, 1996).

2. Geological setting

The South China Block comprises the Yangtze Block and Cathaysia Block. The Yangtze Block has an Archean to Proterozoic basement, whereas the Cathaysia Block has a Proterozoic basement. Yangtze and Cathaysia blocks amalgamated during the early Neoproterozoic Orogeny (e.g., Zhao and Cawood, 1999; Wang et al., 2006; Zheng and Zhang, 2007; Li et al., 2009), forming the Jiangnan orogen. A cover sequence of marine strata from the late Neoproterozoic to the Paleozoic was accumulated on the united South China Block that subsequently underwent the Caledonian orogeny (or the Guangxi movement) in the early Paleozoic (e.g., Guo et al., 1989; Qiu et al., 2000; Charvet et al., 2010) and the Indosinian orogeny in the early Mesozoic (e.g., Carter et al., 2001; Lepvrier et al., 2004).

The major Jiangshan-Shaoxing suture zone separating the Yangtze and Cathaysia blocks (e.g., Jiang et al., 2011; Yang et al.,

2012) had been reactivated during the Indosinian and Yanshanian movements (e.g., Wang et al., 2013). During the Yanshanian, the Andean-type convergent margin was developed along the SE China following the subduction of the PPP (e.g., Taylor and Hayes, 1983; Faure and Natal'in, 1992; Charvet et al., 1994; Zhou and Li, 2000; Chen et al., 2005; Liu et al., 2017; Li SZ et al., 2019). A series of NE-striking back-arc basins associated with widespread and large-scale magmatism were produced (e.g., Zhou and Li, 2000; Li and Li, 2007; Liu et al., 2014, 2016; Xie et al., 2017; Yang et al., 2017). Since the deposition in these basins was concomitant with volcanism, it is fairly common that the sedimentary successions are interbedded with volcanic rocks. On the basis of the abundance of volcanic rocks in the strata, these basins can be grouped into three types (Fig. 1): volcanic (-dominated), volcanic-sedimentary, and sedimentary (e.g., Chen et al., 2005; Shu et al., 2009). These three types of basins are roughly separated by two NE-striking fault zones: the Jiangshan-Shaoxing fault zone and the Zhenghe-Dapu fault zone (Fig. 1). The volcanic basins occur SE to the Zhenghe-Dapu fault zone and were formed on the magmatic arc (Lapierre et al., 1997) along the coastline, i.e., the Coastal zone (CZ). The volcanic-sedimentary basins occur in the SHTB confined between the two fault zones, and volcanic rocks are typically interbedded and / or intercalated with sedimentary strata, which had been constructed in the back arc / rifting basin (e.g., Gilder et al., 1991; Jiang et al., 2009; 2011). Nevertheless, the late Mesozoic volcanic rocks are almost absent east to the Yujiang-Yudu fault zone in sedimentary basins and western SHTB basins (Fig. 1).

The large-scale magmatism is evidenced by the occurrence of granitic plutons in both the SHTB and the CZ stretching over 1000 km along the coastal SE China. These granitic plutons intruded into the Precambrian basement and the overlying Paleozoic strata during the Middle Jurassic-Early Cretaceous (e.g., Jiang et al., 2011; Yang et al., 2012). The intrusions mainly occur as A-type and / or I-type granitic rocks, and together with huge volcanic rocks, strongly support the model of the western subduction of the PPP (e.g., Zhao et al., 2016; Jiang et al., 2011; Yang et al., 2012; Xie et al., 2017; Yang et al., 2017).

3. Material and methods

A total of 48 extrusive rock samples were collected from about 20 lithostratigraphic formations (supplementary data Table S1) in 11 basins / regions within the main SHTB to obtain new zircon U-Pb isotope ages (L1-L10 in Fig. 1; supplementary data Figs. S1-S3 and Table S1). The extrusive rock specimens are volcanic and pyroclastic rocks that are interbedded and intercalated with the sedimentary strata, in which sampling horizons and associated lithologies are marked in the supplementary data figures S4-S12. These samples were collected from volcanic layers in the main type sections of typical basins in SE China (supplementary data Fig. S4-S12). In general, 3-4 rock samples were taken at lower/base, middle and upper/top part when a lithostratigraphic unit has multiple volcanic horizons or a volcanic layer is over 100-200 m thick (see supplementary data Table S1). The locations of these samples were determined with a GPS device and are marked on the geological maps (supplementary data Figs. S1-S3 and Table S1).

Zircon grains were separated using the conventional heavy liquid and magnetic techniques. Single zircon grains were handpicked and mounted on adhesive tapes, embedded in epoxy resin, and then polished to about half to one-third of their thickness and photographed in both reflected and transmitted light. Cathodoluminescence (CL) images were taken at the State Key Laboratory for Mineral Deposits Research, School of Earth Sciences and Engineering, Nanjing University, to examine the internal structures of single zircon grains before U-Pb isotope analysis.

LA-ICP-MS, U-Th-Pb analyses of single zircon grains were performed on a Nd of YAG 213 laser ablation system (Agilent 7500a, New Wave Research, U.S.A.) coupled with VG PQ Excell ICP-MS, which is housed in the State Key Laboratory for Mineral Deposits Research, Nanjing University. General ablation time is ca. 60 s and the ablation pit diameter is at 25-35 μm. The ablation repetition rate is 5 Hz with the incident pulse energy of about 10-20 J/cm^2. Calibrations of mass fractionation were made using the index sample GEMOC/GJ (608 Ma). In each experiment, a total of 11 to 21 zircon grains were

measured, among which 8 to 18 grains yield concordant age data. Prior to each experiment, the standard GJ-1 and Mud Tank samples were measured. Other measurements follow the methods described by Jackson et al. (2004). Analyses of Mud Tank sample yielded a weighted $^{206}Pb/^{238}U$ age of 726 ± 10 Ma - 737 ± 5 Ma (2σ), which is in good agreement with the recommended value (TIMS age = 732 ± 5 Ma, Black and Gulson, 1978).

Data reduction, isotope ratio, age calculation, and Pb correction were conducted with the GLITTER software using Zircon 91500 as an external standard. Data processing and plotting were executed with the Isoplot 3.23 programs (Ludwig, 2001). The uncertainties of age results are quoted at 1σ confidence level, whereas errors for weighted mean ages are quoted at 2σ.

It is worth noting that those aged samples of mafic dykes, basalts and gabbros were not herein compiled for the analysis of volcanic temporal-spatial variation in SE China. This is because: 1) among the magmatic rocks, gabbros and basalts are rare, and diorites and andesites are even less common in South China (Zhou and Li, 2000), leading to a weak significance in statistic of the volcanic samples; 2) those published ages of the dykes, basalts and gabbros were mainly measured using different (Ar-Ar, K-Ar, Rb-Sr) isotopic methods (e.g., Li, et al., 1989; Chen et al., 2008b; Wang et al., 2008; Meng et al., 2012), likely causing chaos of real ages; 3) it is difficult to obtain a good isotopic age for mafic rocks, and particularly, the bulk (basalt) samples ages by K-Ar, Ar-Ar, and Rb-Sr are ~ 10-20 Ma younger than those by zircon U-Pb isotopes (Li et al., 2019); and 4) some basalts predominantly are of the Indosinian orogeny age, instead of the Yanshanian orogeny.

4. Results

4.1 Uncertainty of zircon U-Pb ages

It is necessary to first evaluate the uncertainty of the new age results and other cited age data. The uncertainty depends on three aspects, i.e. origin of zircon, precision, and accuracy (Schoene et al., 2013).

For the origin, all zircons used in this work were microscopically evaluated with CL to ensure that laser ablation positions of zircons are away from the nucleus, cracks, and inclusions. CL images manifest the growth rings. In the concordant 636 zircons of this work, 20 grains (3.1 %) are 0.1-1.0 in Th/U ratio, 615 (96.7 %) are 1.0-10.0 (Table 1). Th/U ratios of 3539 zircons can be available in the age data from published references. Together with published data and this work, 1766 zircon grains (42.3 %) are 0.1-1.0 in Th/U ratio, 2394 grains (57.3 %) are 1.0-10.0, 14 grains (0.3 %) are > 10.0, and only one is less than 0.1 (Table 1). CL images and Th/U ratios of this work combined the collected data demonstrate that predominant (> 99.9 %), if not all, zircons are magmatic origin.

Precision and accuracy uncertainties produced during LA-ICP-MS zircon U-Pb dating have been more and more concerned (e.g., Klötzli et al., 2009; Solari et al., 2010; Li et al., 2015) and come from multiple sources, including the isotopic ratio measurements, the fractionation factor calculation using an external standard, the common lead correction, the external standards, and the data reduction (Li et al., 2015). According to the suggested ~ 4 % (2σ) of precision and accuracy (Li et al., 2015), we used the ~ 2 % and ~ 2-4 % (1σ) to evaluate uncertainties of extrusive rock ages.

A total of 48 rock samples were respectively weighed in mean from 636 concordant zircon U-Pb ages in this work (supplementary data Table S1 and S2). In the samples, 46 (95.8 %) have a < 3 million years (Myr) error in 1σ, in which 36 (75 %) samples have < 2 Myr error in 1σ; 41 samples (85.4 %) have < 2.0 % (error / age) deviation, and 7 (14.6 %) have 2-4 % deviation (Table 1). Similar percentages of sample error and age deviation are comparative with those single zircons analyzed in this work (Table 1).

For zircons from the published data, the literature often provides CL images of zircons showing quite similar nature in source and error. For the zircon U-Pb ages from the previous studies, we carefully examine the experiments described in the literature, re-analyze the concordant ages, and eliminate those that are not concordant and / or greater than ~ 5 % in age deviation (error / age) as well as ages with distinct inheritance, which were not discarded by the original authors. This scrutinizing procedure allows us to identify reliable U-Pb age data from 188 volcanic rock samples from the SHTB and from

103 volcanic rock samples from the CZ (supplementary data Table S2 and S3). Then, results show that in the combined 291 samples, 246 samples (85.5 % = 246/291) are < 2 Myr in 1σ error of age and 39 (13.4 %) are 2-4 Myr; and 264 samples (90.7 %) are < 2.0 % age deviation and 25 (8.6 %) are 2-4 Myr deviation in age (Table 1). Closely, total concordant single zircons 4639 are similar in percentages of 1σ error and age deviation with the weighed-mean age samples (Table 1).

The above relatively low errors in 1σ and deviation of age indicate that samples of both this and previous work have highly proportional age results (> ~ 95 %) with fine precision.

Systematic biases often dominate uncertainty in comparisons between dating methods and between laboratories (Schoene et al., 2013). For measurements of our zircon samples, the internal systematic 2σ error is less than 3 %, which has been verified by reproductive measurements of Mud Tank sample (see Section 3). These systematic biases were mostly met for those zircons from the references. Therefore, small internal systematic 2σ errors allow our zircon date results to be a moderate accuracy in geochronological application.

The internal systematic conditions are same for weighted mean dates of individual samples from both this and previous work (ref. and comp. to supplementary data Table S1-S3). Compiled zircons are predominantly single dates generally within less than 2 Myr in 1σ errors (< 3 % biases) for the Late Jurassic – Early Cretaceous volcanic rocks. The dates are to great degree consistent with the biostratigraphy of pollens-spores, plants, ostracods, and conchostracans in the volcanic-sedimentary basins, SHTB (e.g., Chen and Shen, 1982; Sha, 1990; Jiang et al., 1993; Chen, 2000).

In summary, the zircon origin and the age precision and accuracy indicate the sample weighed-mean ages have relatively low uncertainty and they are eligible for investigating the eruption geochronology of extrusive rocks in SE China.

4.2 U-Pb age spectra of extrusive rocks

Spot analyzing results of this work show that 48 samples have a wide range of (concordant $^{206}Pb/^{238}U$) weighed-mean ages from 162 Ma to 92 Ma (green histogram, Fig. 2a), from which two peaks of weighted mean ages are inconspicuously regressed as 133.3 ± 1.5 Ma and 97.2 ± 1.1 Ma, respectively (Fig. 2a). In addition, 636 concordant single zircons from the samples show similar wide age range (166 Ma to 92 Ma) with three age peaks (Fig. 2a. 135.87 ± 0.42 Ma, 124.71 ± 0.35 Ma, and 98.91 ± 0.57 Ma).

Combining our new results with the published age data from the main SHTB (e.g., Wu et al., 2011a, b; Wu and Wu, 2013; Liu et al., 2012, 2014, 2016; Li CL et al., 2014; Li JH et al., 2014; Ma et al., 2016; Wang et al., 2016; Shu et al., 2017. Locations M1-M22, Table S1 and Fig. 1) yields a similar age pattern (Fig. 2b). A total of 188 rock samples show that the weighed-mean age range from 177 Ma to 92 Ma with distinct age peak 136.11 ± 0.38 Ma and inconspicuous age peak100.0 ± 1.0 Ma (Fig. 2b). Also, a total of 2593 single zircons from the SHTB show the concordant $^{206}Pb/^{238}U$ ages ranging from 180 Ma to 92 Ma with strong age peak 132.07 ± 0.17 Ma and weak peak 101.26 ± 0.23 Ma (Fig. 2b).

The published data of 103 rock samples from the CZ (for Locations N1-N21, see Fig. 1 and supplementary data Table S1 and S3. Chen et al., 2008; Li et al., 2009; Guo et al., 2012; Li CL et al., 2014; Liu et al., 2012, 2016; Zhang et al., 2018) show a wide weighed-mean ages ranging from 174 Ma to 82 Ma and three remarkable age peaks of 143.15 ± 0.82 Ma, 130.96 ± 0.87 Ma, and 98.13 ± 0.55 Ma (Fig. 2c), similar to those from the SHTB (comp. Fig. 2b and 2c). The 2046 single zircons from the 103 samples also display the same range of concordant $^{206}Pb/^{238}U$ ages (Fig. 2c; supplementary data Table S3) with two prominent age peaks (131.04 ± 0.32 Ma and 99.08 ± 0.32 Ma. Fig. 2c).

Further combined and optimized age data of 291 extrusive rock samples of over 40 lithostratigraphic units in both SHTB and CZ illustrate that sample weighed-mean ages mainly vary between 177 Ma and 82 Ma (Fig. 3). Of the ages, two peaks are at 132.86 ± 0.46 Ma (75 samples, 138-130 Ma, MSWD = 2.3) and 98.19 ± 0.47 Ma (25 samples, 100-96 Ma, MSWD = 1.14), respectively. The compilation of age data from all the 4639 concordant single zircons shows that the $^{206}Pb/^{238}U$ ages range between ~ 180 Ma and ~ 76 Ma with two age peaks at 132.90 ± 0.14 Ma and 99.86 ± 0.19 Ma (Fig. 3).

5. Discussion

5.1 Temporal evolution of volcanism

The late Mesozoic extrusive rocks are widespread in SE China and their dating has been conducted extensively. In early times, they have been roughly dated as the (Late) Jurassic and (to the Late) Cretaceous by the confinement of interbedded / intercalated terrestrial fossil-bearing sedimentary strata, and the ages are quite crude. Later on, Rb-Sr, K-Ar, and Ar-Ar dating of bulk-dominated samples yielded ages of ~ 150-65 Ma with large age uncertainties in the 1980s-1990s (e.g., Hu et al., 1982; Li et al., 1989; Feng et a., 1993; Zhang, 1997), much younger than the earlier rough estimates, and ~ 10-20 Myr younger than the zircon U-Pb isotope ages on average (Li et al., 2019).

In the recent decade, though zircon U-Pb age data of the igneous rocks have been reported, rock samples in individual references were taken from separate locations resulting in different age interpretations of volcanic eruption in SE China, and a relative concurrent viewpoint has not been reached. Multiple volcanic age durations are available at different locations or regions, such as 145-129 Ma, 143-98 Ma, and 140-118 Ma in eastern and northwestern Zhejiang (Liu et al., 2014), 140-88 Ma and 136-129 Ma in southeastern (Liu et al., 2012) and central Zhejiang (Li JH et al., 2014), 168-95 Ma in northeastern Guangdong and southeastern Fujian (Guo et al., 2012), 162-130 Ma from two locations in Fujian (Li et al., 2009), 160-99 Ma from northern Fujian (Liu et al., 2016), and 112-99 Ma from Zijingshan Mineral Field of Fujian (Jiang et al., 2013, 2015). Obviously, these ages are incomplete and intermittent, and cannot individually reveal the age of volcanism in the entire SE China.

To investigate the geochronology of extrusive rocks, we conducted zircon U-Pb age analysis in the SHTB and combined the published data from both SHTB and CZ. Then relatively high precise and representative dating results are obtained in entire SE China: the combined and optimized ages from 291 rock samples (4639 concordant zircons) range from ~ 177 Ma to ~ 82 Ma (mainly 160-90 Ma).

As we know, the U-Pb isotope ages of zircons represent the cease time of the crystalline zircon formation when volcanic eruption, therefore, we propose that the age range above is an eligible representation for the duration of volcanism in SE China. That means, the volcanism could have initiated at the late Toarcian (~ 177 Ma) of the late Early Jurassic and terminated at the early Campanian (~ 82 Ma) of the Late Cretaceous, and it has a ~ 95 Myr duration, which shows little discrepancy with those of the single zircon ages (Fig. 3). On the other hand, the volcanism occurred chiefly during the interval of the Late Jurassic-early Late Cretaceous (160-90 Ma = 70 Myr) when only several samples with ages of pre-160 Ma and post-90 Ma are disregarded (e.g., Chen et al., 2007; Guo et al., 2012; Liu et al., 2012). When consider the relationship of the magmatism to the Yanshanian origination, the above age range and duration (~ 177-82 Ma) probably represent the time of the Yanshanian orogeny in East and SE Asia.

Then the temporal evolution scenario of the volcanism in SE China can be summarized as (Fig. 3): 1) during the latest Early Jurassic (late Toarcian)-Latest Late Jurassic (~ 177-145 Ma), the volcanism was sporadic; 2) the early Early Cretaceous (Berriasian-Barremian, ~ 145-125 Ma) volcanic eruption was the most intensive; 3) and the volcanism became fading during the main mid-Cretaceous (Aptian-Turonian, ~ 125-92 Ma); 4) the volcanism almost ceased since then (~ 92-82 Ma). The most extensively volcanic eruption episode (145-25 Ma) seems to correspond to the period of rapid increase in the magmatic flux of both the Mid-ocean ridge and Large Igneous Provinces (Coffin & Eldholm, 1994) during the late Late Jurassic-early Early Cretaceous (Fig. 4) although the relationship between them remains unclear.

It is noted that among the compiled single zircon U-Pb ages of extrusive rocks, the oldest one is from the Maonong Formation in the Songyang Basin, southwestern Zhejiang. The weighted mean age is 177.4 ± 1.0 Ma for the sample MN01 (location M14. Liu et al., 2012). In addition, a weighted mean age of 180 ± 4 Ma from the same horizon (Chen et al., 2007) has also been reported despite that the error is relatively large, up to 6-8 Myr.

Similarly, variable youngest ages of volcanic rocks are reported. The weighted-mean age 82.5 ± 1.0 Ma of the sample ZJ23

(location N2. Chen et al., 2008) from the Taozu section of eastern Zhejiang could be the youngest age. One zircon grain from the section is dated at 74 ± 0.6 Ma and five zircon grains yield concordant ages of 76 ± 0.6 Ma from the same sample (Table S3. Chen et al., 2008), suggesting that it is possible the termination of volcanism was ~ 5 Myr younger than 82.5 Ma. Two hiatuses volcanism at 128-122 Ma and 120-110 Ma were recently proposed in eastern Zhejiang (Liu et al., 2012), and volcanic reticence of 130-115 Ma was reported in northeastern Guangdong and southeastern Fujian (Guo et al., 2012. N17, N19, N20 in figure 1). Similar silence / inactiveness of volcanism seems happened in other parts of SE China. However, this volcanic silence is a gloss, and it would not have happened when we see all the late Mesozoic volcanism in SE China.

5.2 Spatial pattern of volcanism

Though it is well-known that the late Mesozoic magmatic rocks are widespread in SE China, the previous volcanic distributions are to some degree out of date as those ages contain large errors with low preciseness and accuracy by bulk isotope dating (e.g., Li et al., 1989; Wang et al., 2000; Zhou and Li, 2000; Chen et al., 2008b) and detailed age distribution patterns by precise age constraints have not been outlined yet. To delineate the spatial variation of the late Mesozoic volcanism in SE China that are refined by the zircon U-Pb geochronology, we sketched three distribution maps of extrusive rocks by the initial, peak, and terminal ages of volcanism (Fig. 5a, 5b, and 5c).

Firstly, we identified the initial ages of extrusive rocks. The initial age is defined as the earliest age of volcanic eruption in a location, a basin, and / or a region marked as capital letters L, M and N with numbers in figure 1. Three age boundaries ~ 163 Ma, ~ 145 Ma, and ~ 125 Ma are chosen to divide the initial ages into four intervals: 177-163 Ma, 163-145 Ma, 145-125 Ma, and < 125 (-94) Ma, ordinarily corresponding to the epochs of the Middle and Late Jurassic, the early and late Early Cretaceous, respectively. We used the boundary age 177 Ma as the earliest boundary within the first period of the volcanism because it could represent the initiation time of the first Yanshanian orogenic episode in SE China and the corresponding stratal boundary is marked by an unconformity (e.g., Yu et al., 2003; Shu et al., 2009). The boundary between the Upper Jurassic and the Lower Cretaceous is also represented by a widely observed unconformity (e.g., Yu et al., 2003; Shu et al., 2009) and the intensification of volcanism in SE China (Fig. 3). As there are fewer samples with ages of < 125 Ma and the age boundary at ~ 125 Ma marks the rapid waning of volcanism (Fig. 3), we designed the interval 125-94 Ma of volcanism as the latest initial age recognition.

Then, isolines ages are drown by the boundary age 163 Ma, 145 Ma, and 125 Ma, separately. Interpolation ages are used to confine the zones when there are no exact ages same as the boundary age occur in the map. Plotting the initial ages in the geographical map shows four zones of initial volcanism in SE China (Fig. 5a). Zone 1 (Middle Jurassic, 177-163 Ma) marks areas where initial volcanic eruption locally occurs in the northernmost corner of Guangdong and neighboring southern corner of Fujian as well as northeastern corner of Fujian and at one location of southwestern Zhejiang (M14, Songyang, Liu et al., 2012). Zone 2 (Late Jurassic, 163-145 Ma) delineates areas where initial volcanic eruption occurs around Zone 1 in southern and northeastern Fujian with a much larger scope in southern Fujian than Zone 1. Zone 3 (early Early Cretaceous, 145-125 Ma) defines regions where initial volcanic eruption chiefly and largely extends in SE China, and mostly bounded in west of the volcanic area, extending along the eastern Jiangxi, northwestern Fujian, and middle Zhejiang (Fig. 5a). Zone 4 (late Early Cretaceous, 125-94 Ma) locally occupies eastern Zhejiang and limited southeastern Fujian (south of Fuzhou). Same zones can be also recognized in the map made from the single zircon U-Pb ages (comp. the supplementary data Fig. S13), supporting the zonations of the sample weighed-mean ages.

Secondly, the peak eruption age of extrusive rocks can be identified, which is defined as the main age of extensively volcanic eruption in a location, a basin, and / or a region marked with L, M and N with numbers in figure 1. Here we use 145 Ma, 125 Ma, and 100 Ma as three boundary ages to differentiate the most extensive volcanism in SE China. This is because the main ages are much younger than 145 Ma and few samples show ages younger than 100 Ma, for which the main age isolines are more readily made. Similarly, the corresponding age intervals confined by the boundary ages pertain to the

epochs of the Late Jurassic, the early and late Early Cretaceous, and early Late Cretaceous, respectively.

Isolines of boundary ages are delineated by 145 Ma, 125 Ma, and 100 Ma and completed with interpolation ages when no exact ages in the transition zone. Four zones of peak volcanism are then shown in the geographical map, SE China (Fig. 5b). Zone 1 (Late Jurassic, 163-145 Ma) is the area where most intensively volcanic eruption occurred in southeastern and northeastern Fujian and locally at a place in southwestern Zhejiang (Fig. 5b). Zone 2 (early Early Cretaceous, 145-125 Ma) largely extends along the eastern Jiangxi, middle Zhejiang, northwestern Fujian, and northern Guangdong (Fig. 5b) and indicates widespread volcanism in SE China. Zone 3 (late Early Cretaceous, 125-100 Ma) occurs as a band in middle Zhejiang, southern Fujian, and northeastern Guangdong. Zone 4 (early Late Cretaceous, 100-76 Ma) locally distributes along Zone 3.

Thirdly, we use the terminal eruption age of extrusive rocks to represent the termination time of the last volcanism, which is helpful to distinguish the terminal volcanic distribution in SE China. The age boundaries and intervals are the same as the peak eruption. It is noted that only one age is older than 145 Ma in northeastern corner of Fujian and lots of samples are younger than 100 Ma.

With the confinement of boundary age 145 Ma, 125 Ma, and 100 Ma, age isolines are drown separately, and interpolation method was used to confine the zones when there are no exact ages in the map. Four age zones of the terminal volcanism are recognized in the geographical map by the age isolines (Fig. 5c). Zone 1 (> 145 Ma) occurs in northeastern corner of Fujian due to only one location of the terminal age. Zone 2 (145-125 Ma) mainly occurs in eastern Jiangxi and banded boundary of northern Fujian; Zone 3 (125-100 Ma) largely distributes in the boundary region of eastern Jiangxi and western Fujian and in middle and southwestern Zhejiang. Zone 4 (100-83 Ma) widely appears in regions of the middle Fujian, eastern Zhejiang, and northern Guangdong. Similar zonations can be classified in the map sketched by the single zircon U-Pb ages (supplementary data Fig. S14), verifying the zones of the sample weighed-mean ages in SE China.

Zonations of initial, peak, and terminal volcanism indicate a distinct pattern of volcanic extrusion in SE China (Figs. 5): the oldest ages in the eastern SE China, the younger intensive age clusters in the western SE China and the youngest ones in eastern SE China again. Detailed distributional patterns can be observed: 1) the earliest appearance and earliest disappearance of extrusive rocks dominantly occur in southeastern and northeastern Fujian, where the magmatic arc was located (e.g., Lapierre et al., 1997); 2) the most widespread distribution of extrusive rocks is the most intensive volcanism age as 145-125 Ma in eastern Jiangxi, middle Zhejiang, and northern Guangdong, in which a back-arc / rifting basin was developed (e.g., Gilder et al., 1991; Jiang et al., 2009; 2011); 3) the latest appearance and latest disappearance mainly occur in eastern Zhejiang, eastern Fujian, and northern Guangdong.

With the observation of volcanism, two distributional patterns manifest: 1) the migration of the volcanism was from the northwestward to the southeastward, implying that the PPP could have been subducted northwestly during the late Mesozoic time; 2) the first appearance (initial volcanism) area and the first disappearance (terminal volcanism) region are the same region, suggesting that a roll-back subduction of the PPP happened after ~ 125 Ma.

It is surprising that the zone 1 and / or 2 of volcanism look like thermal-dome patterns (Fig. 5) by exhumation and exposure that may be related to the regional magmatic intrusion, likely misleading the migration of volcanism. However, the distribution pattern is not dome-controlled because: 1) The data are derived from extrusive rocks, instead of intrusive rocks; 2) it is impossible that a crater is over 200-300 km wide in diameter; 3) lots of agglomerates representing craters were observed in a variety of strata at locations / basins out of Zone 1. For instance, these agglomerates are widespread in basins of western Zhejiang (L1-L4; M9-M14), eastern Jiangxi (L5-L7; M16-M18b), and western Fujian (L8-L10, M19-M22).

6. Conclusions

We analyzed weighed mean ages of 48 extrusive rock samples (total of 636 concordant single zircons) from ~ 20

lithostratigraphic units at 11 localities in the SHTB. Published ages of 243 rock samples (total of concordant 4003 zircons) from ~ 40 lithostratigraphic units in SE China are compiled and re-examined. Based on a total of refined 291 sample ages (4639 concordant zircon U-Pb ages) from this study and the published literatures, we propose that the late Mesozoic volcanism in SE China initiated at ~ 177 Ma (late Toarcian of the late Early Jurassic) and terminated at ~ 82 Ma (early Campanian of the Late Cretaceous), spanning an ~ 95 Myr interval (mainly ~ 70 Myr = 160-90 Ma), during which the 145-125 Ma (the early Early Cretaceous) volcanism is the most intensive and widespread magmatic eruption. These age range and span may represent the time of the Yanshanian magmatism in SE China.

Isolines of initial, peak, and terminal volcanic ages are drawn to outline the geographic distribution of extrusive rocks in SE China. The volcanic extrusion age spatial change shows a distinct pattern of the late Mesozoic volcanism: both the oldest and youngest ages in eastern (coastal) Zhejiang and Fujian (magmatic arc), and the most intensive and widespread ages in eastern Jiangxi, middle Zhejiang and northern Guangdong (back arc / rifting basin), hundreds of kilometers away from the coast line. The geographical distribution pattern of the volcanic eruption ages indicates a migration process of magmatic extrusion in SE China and implies that a northwestern subduction of the Paleo-Pacific plate happened and a possible roll-back subduction did not begin until the Aptian (~ 125 Ma) of the mid-Cretaceous.

Acknowledgments

We thank Ke Cao, Sijing Liang, Yannan Ji, Sihe Wang for participating the field investigation. We are grateful to the reviewers for their helpful comments and constructive suggestions. This research was supported by National Key R & D Plan (Grant 2017YFC0601405), Natural Science Foundation of China (NSFC) projects 41372106 and 41672097, and National Basic Research Program of China (973 Project) 2012CB822003.

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529 **Tables**

530 Table 1 Percentages of single zircons and rock samples in 1σ error (Myr), error/age ratio, and Th/U ratio of the late
531 Mesozoic extrusive rocks in SE China

Sources	*1*	*2*	1σ error						Error / age						*3* (Th/U)	Th/U		
			Age (Myr)	*3*	%	Age (Myr)	*3*	%	Ratio	*3*	%	Age (Myr)	*3*	%		Ratio	*3*	%
This work in SHTZ	**636**	**48**	<3	570	89.6	<2	**46**	**95.8**	0-3	581	91.4	<2	**41**	**85.4**	**636**	<0.1	1	0.2
			3-5	63	9.9	2-4	**2**	**4.2**	3-5	50	7.9	2-4	**7**	**14.6**		0.1-1.0	20	3.1
			>5	3	0.5	>4			>5	5	0.8	>4				1.0-10	615	96.7
Composed in SHTZ	**2593**	**188**	<3	2066	79.7	<2	**153**	**81.4**	0-3	2212	85.3	<2	**168**	**89.4**	**2503**	<0.1	1	0.0
			3-5	441	17.0	2-4	**31**	**16.5**	3-5	348	13.4	2-4	**18**	**9.6**		0.1-1.0	945	37.8
			>5	86	3.3	>4	**4**	**2.1**	>5	33	1.3	>4	**2**	**1.1**		1.0-10	1543	61.6
Composed in SHTZ + CZ	**4639**	**291**	<3	3543	76.4	<2	**246**	**84.5**	0-3	3798	81.9	<2	**264**	**90.7**	**4175**	<0.1	1	0.0
			3-5	898	19.4	2-4	**39**	**13.4**	3-5	769	16.6	2-4	**25**	**8.6**		0.1-1.0	1766	42.3
			>5	198	4.3	>4	**6**	**2.1**	>5	73	1.6	>4	**2**	**0.7**		1.0-10	2394	57.3

Notes: Numbers of evaluated zircon grains differ from sources in U-Pb age and Th/U ratio due to unavailability of some original dada. CZ, Coastal zone; SHTC, Shi-Hang tectonic zone. ***1***, Con-cordant zircon Number; ***2***, Rock Sample number; ***3***, Zircon number

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Figures

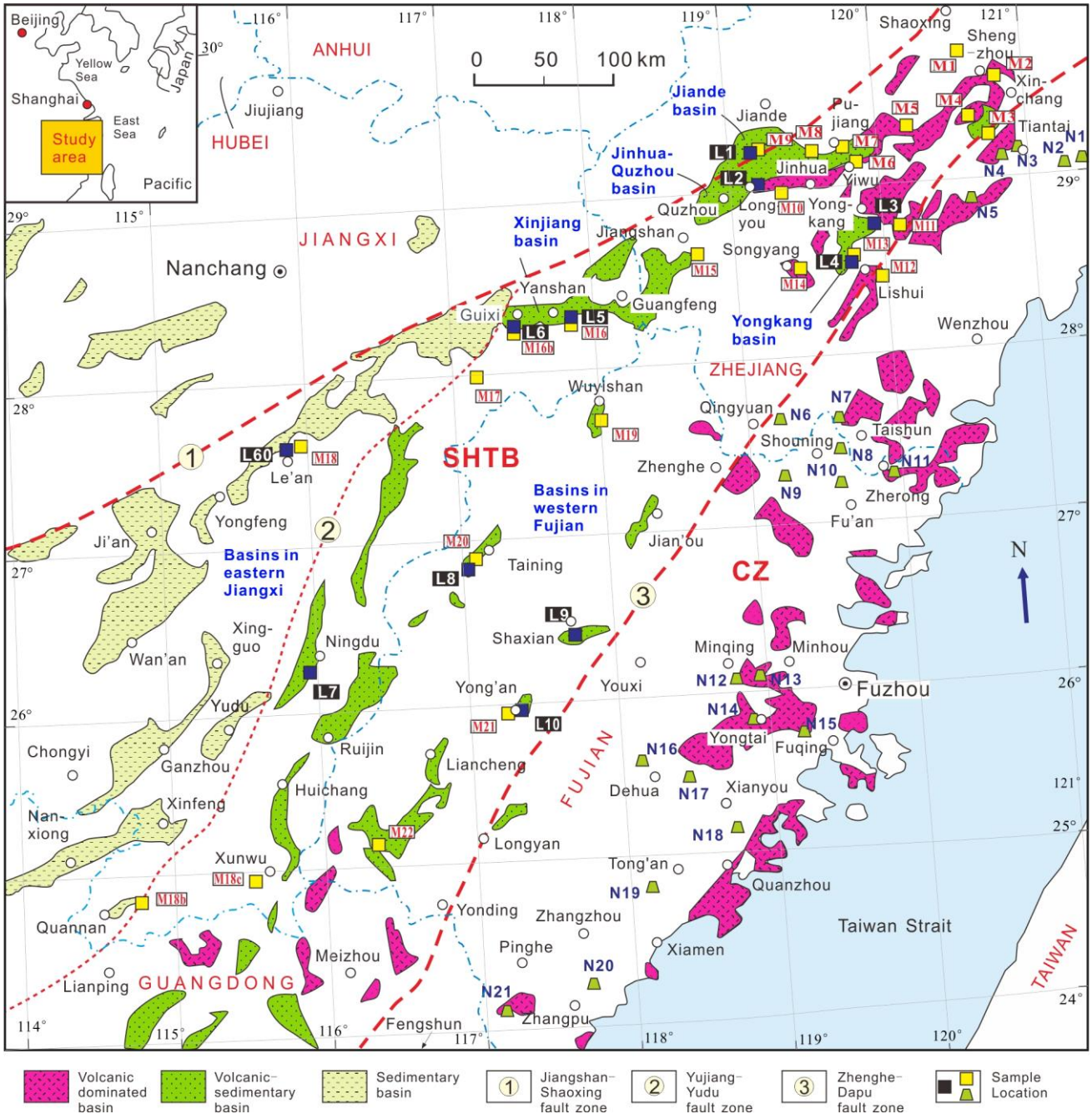
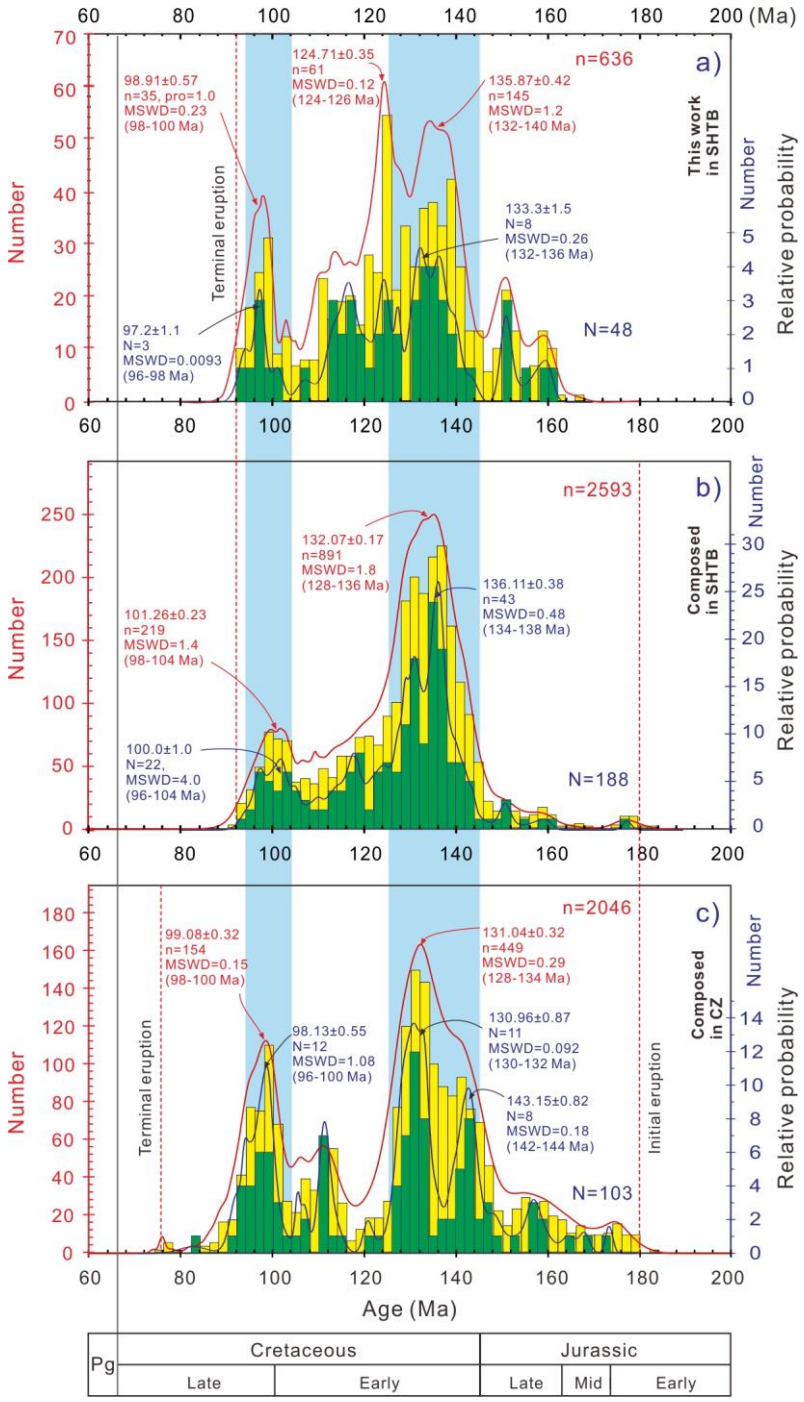


Figure 1 Geological map sketch of South China showing tectonic and basin zonations of the upper Mesozoic and sample locations (map simplified after Shu et al., 2009). In SHTB (Shi-Hang tectonic belt), dark blue squares with white capital letter L + numbers within black rectangles mark the sampling locations of this study (supplementary data figures S1, S2, and S3), and yellow squares with red capital letter M + numbers within white rectangles indicate sampling locations of previous studies (supplementary data Table S1 and S2). In CZ (Coastal Zone), green trapezoids with bold capital letter N + numbers are sample locations of previous studies (supplementary data Table S1 and S3).



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547 **Figure 2 Relative probability and histogram diagrams of concordant zircon U-Pb isotope and sample weighed–mean ages of**
548 **extrusive rocks from SE China (details see in supplementary data Table S1, S2, and S3). a), this study in SHTB; b), combined this**
549 **and previous studies in SHTB; c), published data in CZ. N = number of rock samples, n = total number of zircon grains.**

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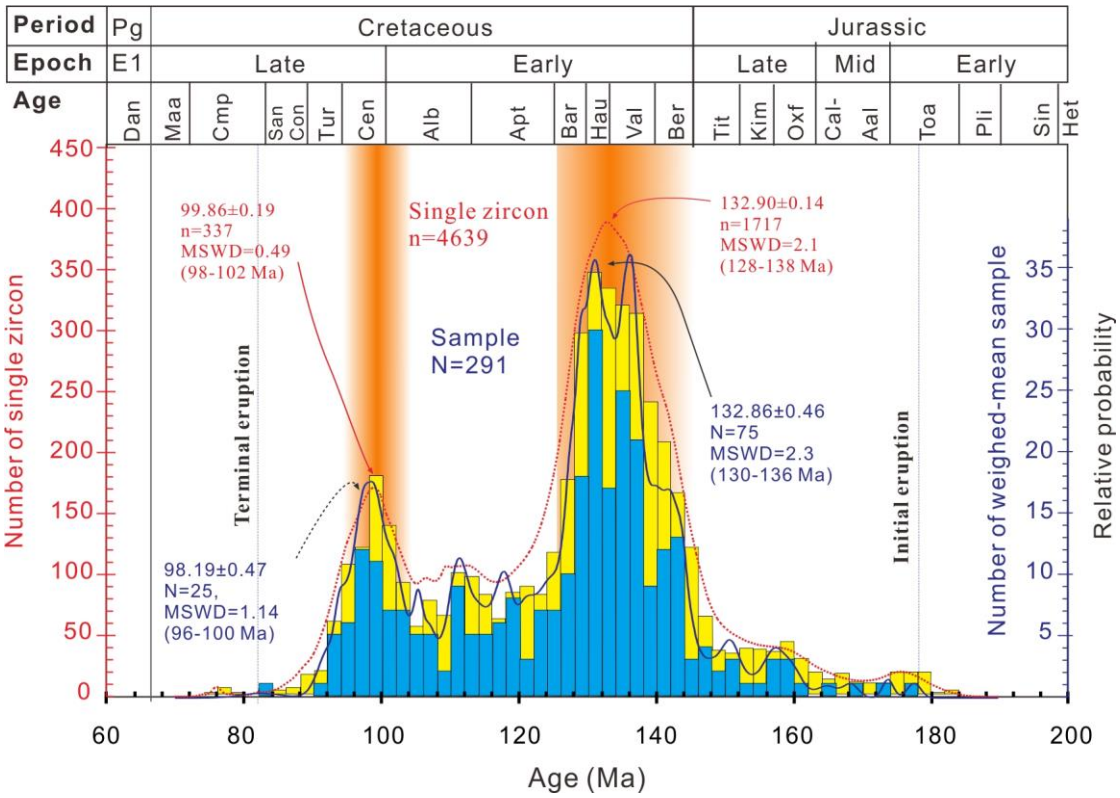

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553 **Figure 3 Diagram showing U-Pb isotope age relative probability and histogram of both single zircon and individual sample**
554 **weighed-mean zircons from all extrusive rock samples in SE China. N = number of rock samples, n = total number of zircon**
555 **grains.**

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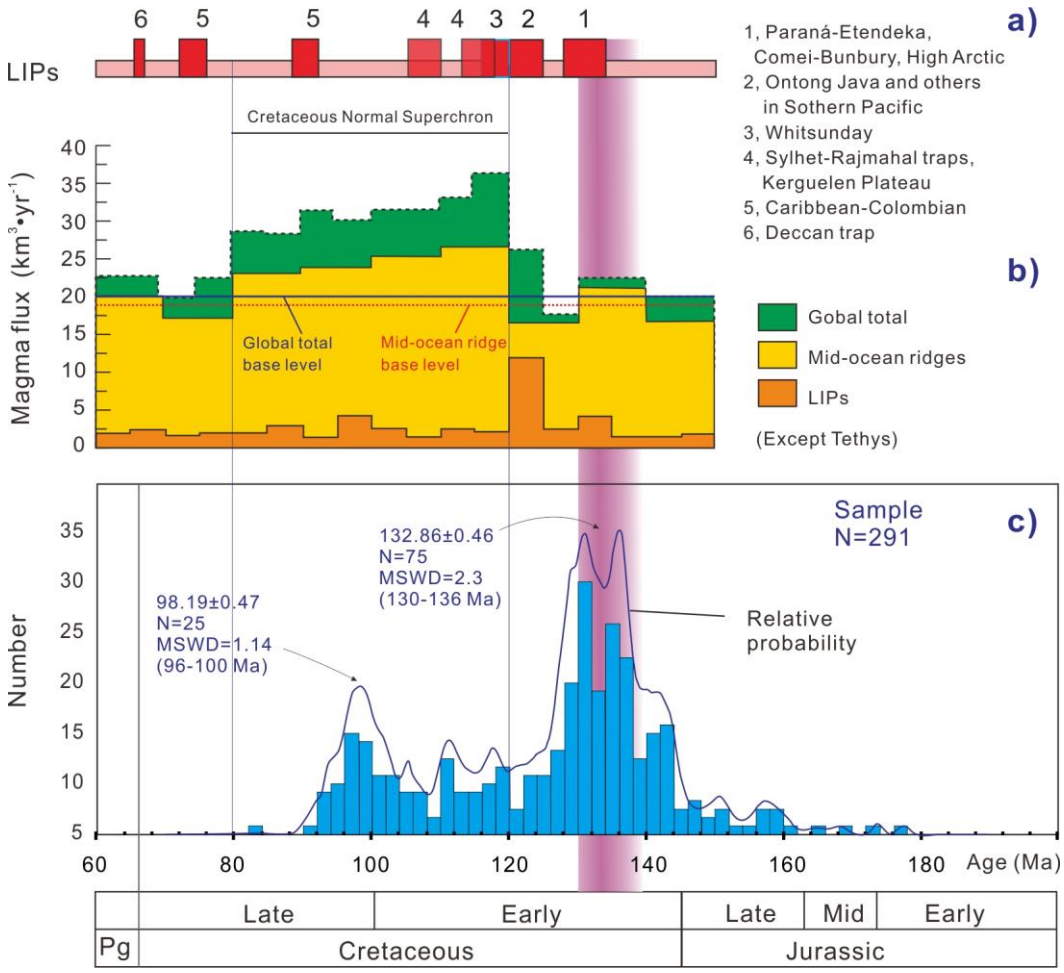
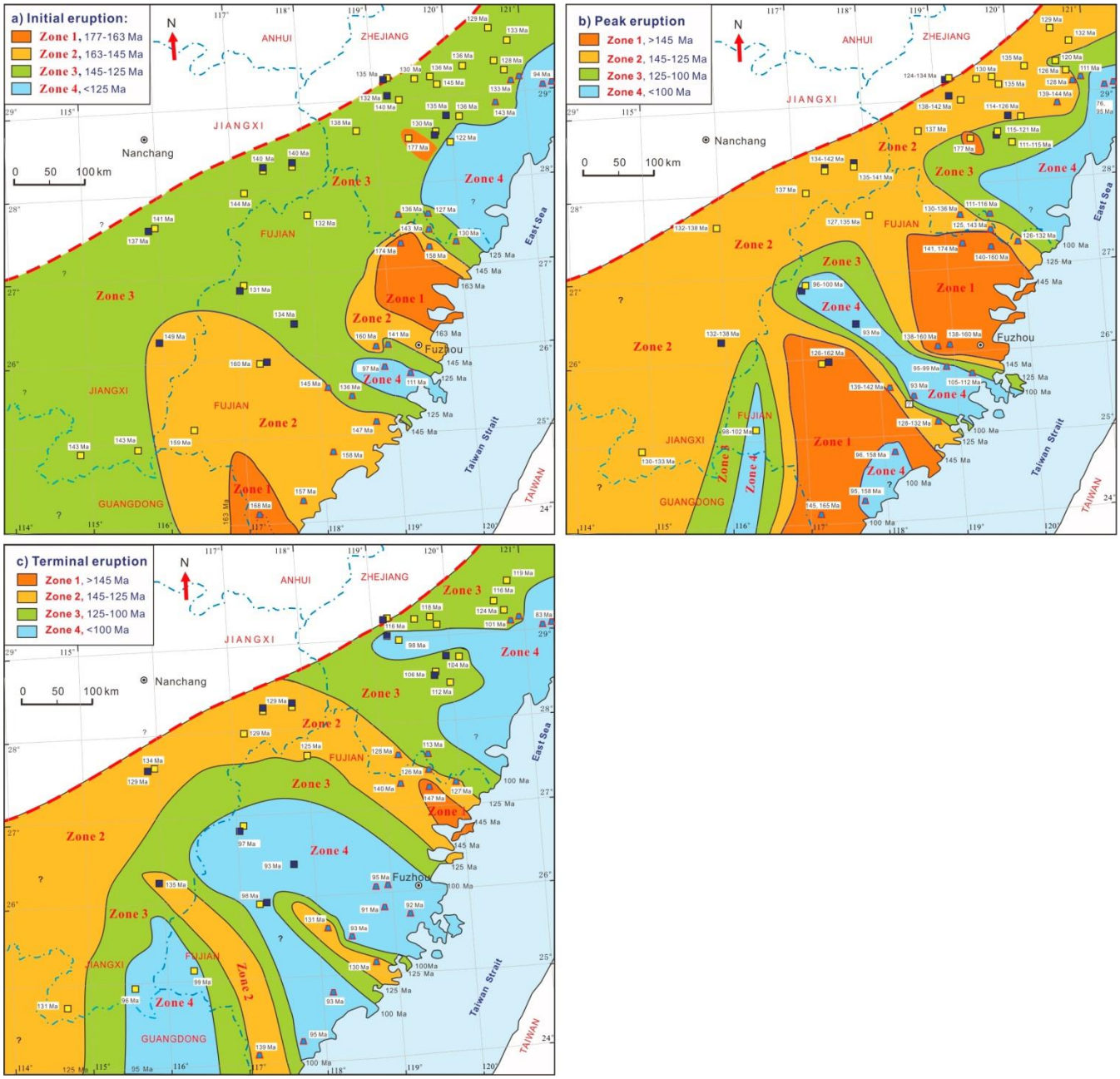


Figure 4 Diagram showing age ranges of volcanism in SE China and correlations with the global Large Igneous Provinces (LIPs) and magmatic flux. a, age range of the Cretaceous LIPs (summary see Coffin & Eldholm, 1994); b, magma flux of the Cretaceous LIPs, mid-ocean ridges, and (except Tethys) global total (Coffin & Eldholm, 1994); c, age range of the volcanism with histogram and relative probability and SE China.



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566 **Figure 5 Sketch maps showing zonations of the late Mesozoic volcanism by age in SE China. Age within white rectangles is the**
567 **eruption time at a location or in a basin/region. Names of color squares and trapezoids refer to Fig. 1. a), zonations of initial**
568 **eruption ages; b) zonations of peak eruption ages; c) zonations of terminal eruption ages.**

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