

1 **Mantle sources of Cenozoic volcanic activities around the South**
2 **China Sea revealed by geochemical and isotopic data using the**
3 **principal component analysis (PCA)**

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24 1. Geological setting

25 Cenozoic basaltic rocks are widespread in the Southeast Asian region and occupy
26 the SCS, Hainan Island, Fujian–Zhejiang, Taiwan, Vietnam, and Thailand (Fig. 1).
27 These regions show evidence of a series of tectonic evolutionary processes, including
28 rifting, subduction, terrane collision, and large-scale continental strike–slip faulting
29 occurring via spatially and temporally complex relations (Cullen et al., 2010; Wang et
30 al., 2012; Yan et al., 2018; Li et al., 2014a, 2014b). The SCS, one of the largest
31 western Pacific marginal basins at the junction of the Eurasian, Indo-Australian, and
32 Philippine–Pacific plates, is surrounded by the South China fold belt in the north, a
33 subduction zone in the east, the Borneo Trough in the south, and the East Vietnam
34 fault in the west (Fig. 1). Hainan Island and the adjacent Leizhou Peninsula are
35 located at the northern edge of the SCS basin and the southeastern margin of the
36 Eurasian plate (Fig. 1). Two volcanic eruptive styles, including massive eruptions
37 from extensional fissures and sporadic eruptions from central volcanoes, led to the
38 formation of massive Cenozoic basaltic flows in Hainan Island (Flower et al., 1992;
39 Ho et al., 2000), which may be attributed to the influences of the Hainan mantle
40 plume (Hoang and Flower, 1998; Wang et al., 2012, 2013; Yan et al., 2018) and the
41 extension of the SCS basin (Cullen et al., 2010; Wang et al., 2012). The
42 Zhejiang–Fujian region of SE China, which is located on the East Asiatic continental
43 margin, shows a wide distribution of abundant Cenozoic basalts belonging to the
44 circum–Pacific volcanic belt, including Xilong, Mingxi, and Nutoushan (Fig. 1; Ho et
45 al., 2003; Chung et al., 1994; Smith, 1998). The Taiwan orogeny was formed by an

46 arc–continent collision in which the buoyant passive margin of Asia underthrust the
47 Manila Trench and collided with the Luzon Arc (Fig. 1; Lin et al., 2019), which
48 mainly consists of five geomorphic units and three NS-trending structural belts from
49 west to east (Tian et al., 2019). The volcanism in Vietnam mostly post-dates the
50 mid-Miocene cessation of the SCS opening (Barr and MacDonald, 1981; Hoang et al.,
51 1996, 2013; An et al., 2017). Large quantities of tholeiites and small amounts of
52 alkaline basalts occupy Vietnam with an area of ca. 23,000 km² (Hoang and Flower,
53 1998; Hoang et al., 1996, 2013; An et al., 2017). Thailand and its surrounding region
54 mainly consist of three tectonostratigraphic units, including a western Sibumasu block,
55 a middle Sukhothai arc terrane, and an eastern Indochina block (Yan et al., 2018; Barr
56 and Macdonald, 1987; Metcalfe, 2011, 2013). The pre-Cambrian basements beneath
57 the Sibumasu and Indochina blocks were formerly part of the eastern Early Paleozoic
58 Gondwanaland (Metcalfe, 2011, 2013; McCabe et al., 1988; Yan et al., 2017). Two
59 major Cenozoic strike–slip faults, including the Mae Ping and Tree Pagodas faults,
60 cut through the western part of Thailand (Fig. 1).

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62 **2. Discussion**

63 **2.1. Effects of the Hainan mantle plume on volcanic activities in different** 64 **periods**

65 The PC1 values of nine 0–12.6 Ma Hainan Island combined samples are
66 consistently positive and range from 0.38 to 4.62; by contrast, the PC1 values of
67 nineteen 4.6–33 Ma Hainan Island samples are negative (-2.66 – -0.44) (Appendix 6;

68 [Wang et al., 2012; Liu et al., 2015](#)). This result indicates than an enriched OIB-type
69 mantle plume plays a more significant role in Hainan volcanic activities during the
70 period 0–12.6 Ma than in those during the period 4.6–33 Ma. A similar phenomenon
71 occurs in the seamounts of the SCS, the expansion center of the SCS, Zhejiang–Fujian,
72 Thailand, and Vietnam ([Appendix 6](#)). In detail: (1) in the seamounts of the SCS, eight
73 younger combined volcanic rocks (3–8 Ma) have high positive PC1 values in the
74 range of 1.38–7.63, whereas, three older volcanic rocks (16.5 Ma) have relatively low
75 positive PC1 values (0.75 –1.27) ([Appendix 6; Yan et al., 2008, 2015](#)). (2) In the
76 expansion center of the SCS, the 3.8–12.8Ma combined volcanic samples are
77 characterized with a high PC1 value (1.29–7.63), whereas the three older volcanic
78 rocks (15–17 Ma) have relatively low negative PC1 values (-2.90 – -2.01) ([Appendix](#)
79 [6; Zhang et al., 2017, 2018](#)). (3) In the Zhejiang–Fujian region, younger 0–16.2 Ma
80 volcanic rocks have significantly higher PC1 values (1.27–7.08) compared with three
81 older volcanic rocks (>9 Ma), which have low positive or even negative values (-2.86
82 – -2.60) ([Appendix 6; Huang et al., 2017](#)). (4) In Thailand and its surrounding areas,
83 older volcanic rocks (8.8–11 Ma) have negative PC1 values (-0.18), while younger
84 volcanic rocks (0.6–3.3 Ma) have high positive PC1 values ranging from 0.44 to 4.62
85 ([Appendix 6; Yan et al., 2018](#)). (5) In Vietnam, some older volcanic rocks (7.0–16.5
86 Ma) have low PC1 values ranging from -1.81 to -0.51, whereas some younger
87 volcanic rocks (0.2–9.6 Ma) have significantly higher positive PC1 values (1.07–4.93)
88 ([Appendix 6; Hoang et al., 2018](#)).

89 The above observations reveal that younger combined samples collected in the
90 same area have higher PC1 values than older samples. Similar phenomena have been
91 observed in the SCS and its surrounding areas ([Appendix 6](#)), thus indicating that the
92 phenomenon is not accidental but, instead, quite common in our study area. We
93 speculate that the influence of the Hainan mantle plume on the Cenozoic volcanic
94 activity in this area gradually strengthened. Specifically, the influence of the Hainan
95 mantle plume on nearby young volcanic activity during the period of <13 Ma is much
96 stronger than that during the period of >13 Ma.

97 We used the PCA method to analyze Sr, Nd, and Pb isotopic ratios and present the
98 PC values of volcanic rocks from the SCS and its surrounding areas ([Appendix 7](#)). (1)
99 For the volcanic rocks from the expansion center of the SCS, 3.8–12.8 Ma SCS
100 samples have higher PC1 compositions relative to those of 15–17 Ma SCS samples
101 ([Appendix 7](#); [Yan et al., 2008](#); [Zhang et al., 2017, 2018](#)), thus suggesting that younger
102 volcanic activities (3.8–12.8 Ma) are more affected by enriched OIB- and EM1-type
103 mantle plumes than older volcanism (15–17 Ma). (2) The same pattern could be
104 observed in Thailand, where relatively younger volcanic activity (0.6–3.3 Ma) is more
105 influenced by OIB- and EM1-type mantle plumes compared with older volcanism
106 (8.8–11 Ma; [Appendix 7](#); [Yan et al., 2018](#)). (3) Although seamounts in the SCS and
107 Hainan Island have positive PC1 values, they do not show obvious differences at
108 different ages ([Appendix 7](#); [Tu et al., 1991](#); [Li et al., 2013](#); [Yan et al., 2008](#)). This
109 result may be attributed to the fact that the PC1 value calculated by isotopic ratios
110 represents a variety of enriched isotope components (e.g., OIB- and EM1-type

111 components); thus, distinguishing whether the volcanic activity of different ages is
112 affected by the mantle plume is difficult. Even though relatively older volcanic rocks
113 are not significantly affected by the Hainan mantle plume compared with younger
114 rocks, they may still be affected by other enriched isotopic compositions.

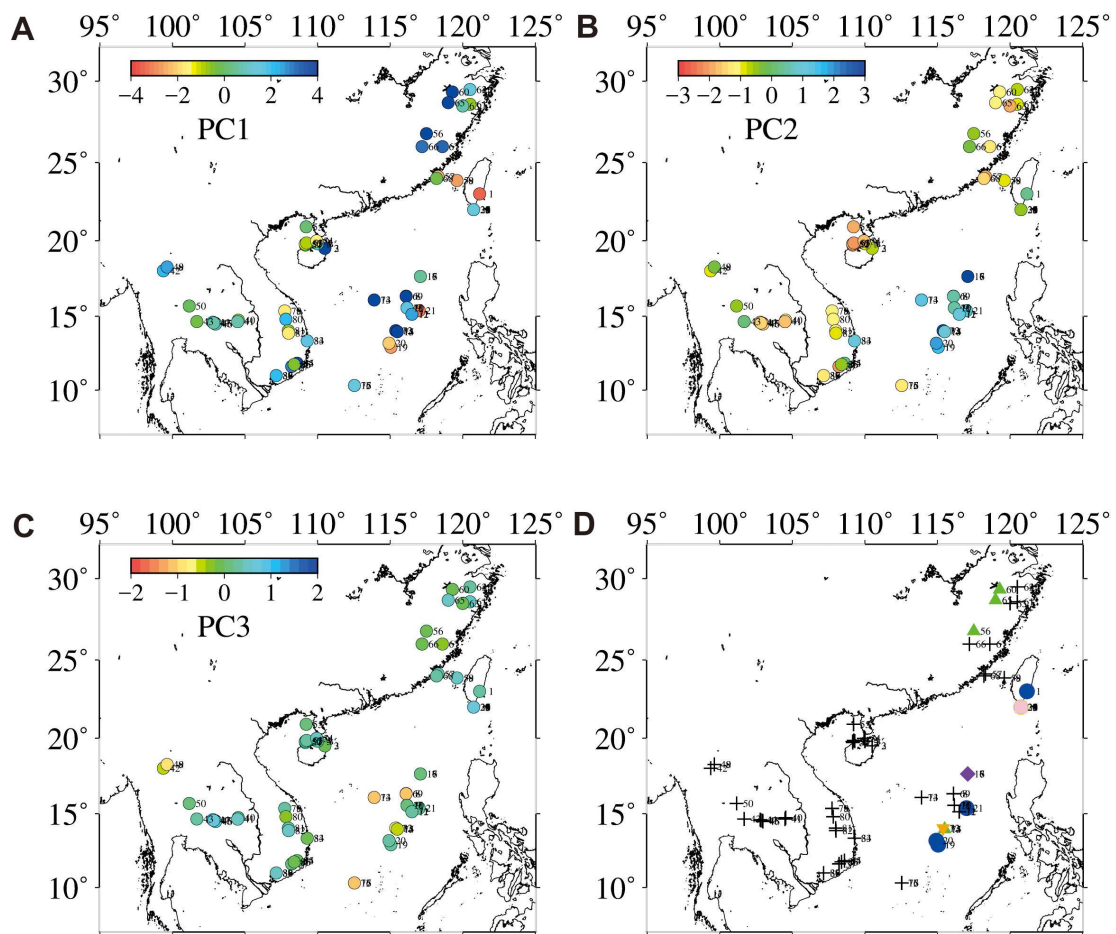
115 The results of isotope PCA calculations indicate that volcanic rocks from
116 Thailand and the expansion center of the SCS show significant age variation patterns.
117 Relatively older volcanic rocks are subject to a smaller degree of PC1 composition
118 (i.e., enriched OIB- and EM1-type mantle sources) compared with younger samples
119 ([Appendix 7](#)). This conclusion is consistent with the results of trace element PCA
120 calculations described earlier.

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122 **2.2. Spatial scope of the influence of the Hainan mantle plume on Southeast Asia**

123 The latest geochemical studies show that the influence range of the Hainan
124 mantle plume includes a wide region, such as Hainan Island, the SCS, Thailand, and
125 Vietnam ([Wang et al., 2012](#); [Liu et al., 2015](#); [Yan et al., 2008, 2015, 2018](#); [Zhang et](#)
126 [al., 2017, 2018](#); [An et al., 2017](#); [Hoang et al., 2018](#)). This study collected trace
127 element data of Cenozoic volcanic rocks from the above regions as well as Fujian,
128 Zhejiang, and Taiwan. PCA and cluster analysis of the trace element data revealed
129 that over 75% samples clustered into a sample that is highly similar to the OIB
130 standard sample ([Fig. 3](#)). This cluster sample extends from Zhejiang–Fujian in the
131 north to Vietnam–SCS in the south and from Thailand in the west to Southern Taiwan
132 in the east (black cross in the [Fig. S1D](#)). The PC1 value of this cluster sample is much

133 higher than its PC2 and PC3 values ([Appendix 3](#)). PC1 may reflect the origin of the
134 OIB-type mantle plume as a result of the above analyses, thus, the Hainan mantle
135 plume may be assumed to play a significant role in the volcanism of these areas and
136 affect the geochemical characteristics of their volcanic rocks. Similar findings (black
137 cross in the [Fig. S2C](#)) can be obtained from the PCA and cluster analysis of isotopic
138 ratios ([Figs. 4, 5](#)).

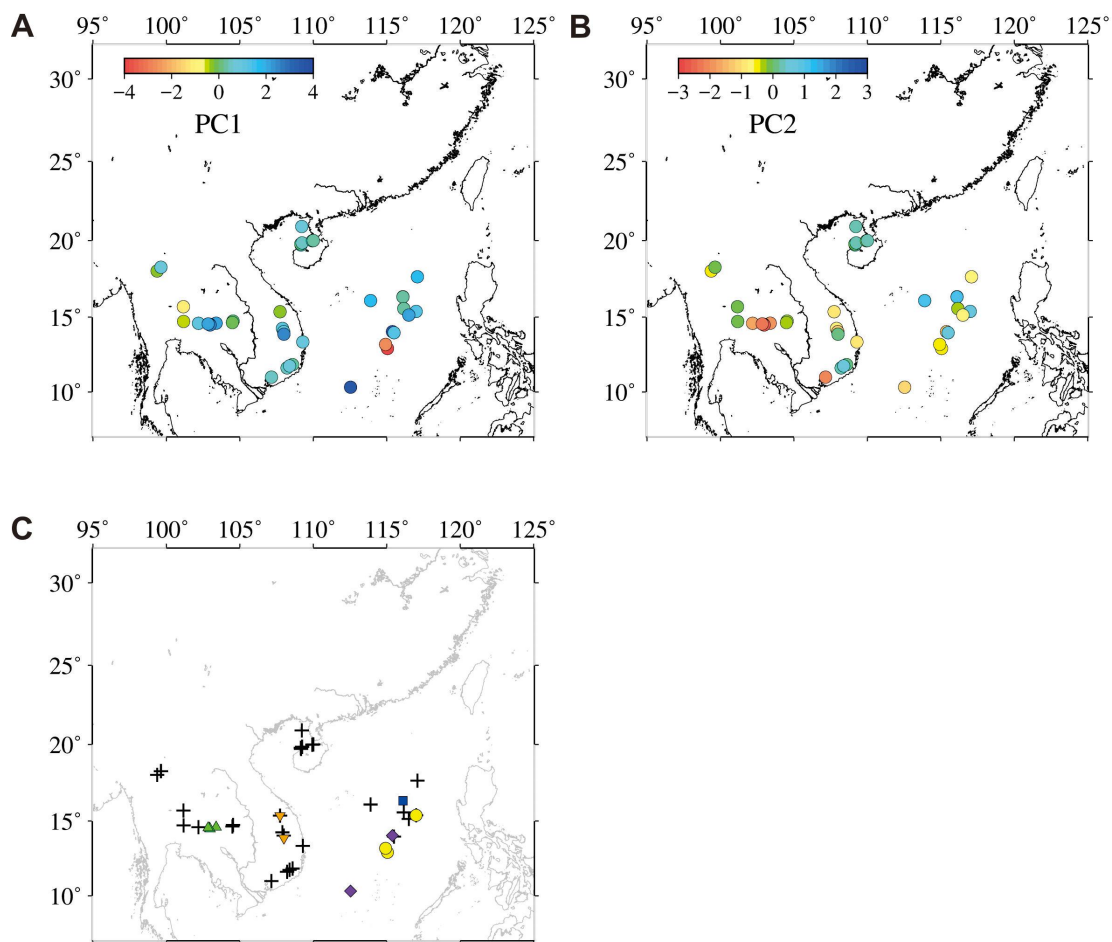


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141 **Fig. S1** The spatial distribution of <33Ma volcanic rocks from the Southeast Asian
142 based on principal component values calculated by trace elements.

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146 **Fig. S2** The spatial distribution of <33Ma volcanic rocks from the Southeast Asian
 147 based on principal component values calculated by Sr, Nd, Pb isotopic ratios.

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150 **References**

151 An, A. R., Choi, S. H., Yu, Y. & Lee, D. C. Petrogenesis of Late Cenozoic basaltic
 152 rocks from southern Vietnam. *Lithos.* **272–273**, 192–204 (2017).

153 Barr, S. M. & MacDonald, A. S. Geochemistry and geochronology of late Cenozoic
 154 basalts of southeast Asia. *Geological Society of America Bulletin (Part II)*. **92**,

155 1069–1142 (1981).

- 156 Barr, S. M. & Macdonald, A. S. Nan River suture zone, northern Tailand. *Geology*. **15**,
157 907–910 (1987).
- 158 Chung, S. L., Sun, S. S., Tu, K., Chen, C. H. & Lee, C. Y. Late Cenozoic basaltic
159 volcanism around the Taiwan Strait, SE China: product of
160 lithosphere-asthenosphere interaction during continental extension. *Chemical
161 Geology*. **112**, 1–20 (1994).
- 162 Cullen, A., Reemst, P., Henstra, G., Gozzard, S. & Ray, A. Rifting of the South China
163 Sea: new perspectives. *Petroleum Geoscience*. **16**, 273–282 (2010).
- 164 Flower, M. F. J., Zhang, M., Chen, C. Y., Tu, K. & Xie, G. Magmatism in the South
165 China Basin: Post-spreading Quaternary basalts from Hainan Island, south China.
166 *Chemical Geology*. **97**, 65–87 (1992).
- 167 Ho, K. S., Chen, J. C. & Juang, W. S. Geochronology and geochemistry of late
168 Cenozoic basalts from the Leiqiong area, Southern China. *Journal of Asian
169 Earth Sciences*. **18**, 307–324 (2000).
- 170 Ho, K. S., Chen, J. C., Lo, C. H. & Zhao, H. L. ⁴⁰Ar-³⁹Ar dating and geochemical
171 characteristics of late Cenozoic basaltic rocks from the Zhejiang-Fujian region,
172 SE China: eruption ages, magma evolution and petrogenesis. *Chemical Geology*.
173 **197(1–4)**, 287–318 (2003).
- 174 Hoang, N., Flower, M. F. J. & Carlson, R. W. Major, trace element, and isotopic
175 compositions of Vietnamese basalts: interaction of hydrous EM1-rich
176 asthenosphere with thinned Eurasian lithosphere. *Geochimica et Cosmochimica
177 Acta*, **60**, 4329–4351 (1996).

178 Hoang, N. & Flower, M. F. J. Petrogenesis of Cenozoic basalts from Vietnam:
179 implication for origins of a 'diffuse igneous province'. *Journal of Petrology*. **39**,
180 369–395 (1998).

181 Hoang, N., Flower, M. F. J., Chi, C. T., Xuan, P. T., Quy, H. V. & Son, T. T.
182 Collision-induced basalt eruptions at Pleiku and Buon Me Thuot, south-central
183 Viet Nam. *Journal of Geodynamics*. **69**, 65–83 (2013).

184 Hoang, T., Choi, S. H., Yu, Y., Pham, T. H., Nguyen, K. H. & Ryu, J. S. Geochemical
185 constraints on the spatial distribution of recycled oceanic crust in the mantle
186 source of late Cenozoic basalts, Vietnam. *Lithos*. **296–299**, 382–395 (2018).

187 Huang, X. W., Su, B. X., Zhou, M. F., Gao, J. F. & Qi, L. Cenozoic basalts in SE
188 China: Chalcophile element geochemistry, sulfide saturation history, and source
189 heterogeneity. *Lithos*. **282–283**, 215–227 (2017).

190 Li, N., Yan, Q., Chen, Z. & Shi, X. Geochemistry and petrogenesis of Quaternary
191 volcanism from the islets in the eastern Beibu Gulf: evidence for Hainan plume.
192 *Acta Oceanologica Sinica*. **32 (12)**, 40–49 (2013).

193 Li, C. F., Lin, J. & Kulhanek, D. K. South China Sea tectonics: Opening of the South
194 China Sea and its implications for Southeast Asian tectonics, climates, and deep
195 mantle processes since the late Mesozoic. *Int. Ocean Discovery Program Sci.*
196 *Prospectus*. **349**, 1–111 (2014a).

197 Li, C. F., Xu, X., Lin, J., Sun, Z. & Zhang, G. L. Ages and magnetic structures of the
198 South China Sea constrained by deep-tow magnetic surveys and IODP
199 Expedition 349. *Geochemistry. Geophysics. Geosystems*. **15(12)**, 4958–4983

200 (2014b).

201 Lin, C. T., Harris, R., Sun, W. D. & Zhang, G. L. Geochemical and geochronological
202 constraints on the origin and emplacement of the East Taiwan Ophiolite.
203 *Geochemistry. Geophysics. Geosystems.* **20**, 2110–2133 (2019).

204 Liu, J. Q., Ren, Z. Y., Nichols, A., Song, M. S., Qian, S. P., Zhang, Y. & Zhao, P. P.
205 Petrogenesis of Late Cenozoic basalts from North Hainan Island: Constraints
206 from melt inclusions and their host olivines. *Geochimica et Cosmochimica Acta.*
207 **152(1)**, 89–121 (2015).

208 McCabe, R., Celaya, M., Cole, J., Han, H. C., Ohnstad, T., Pajitprapapon, V. &
209 Thitipawarn, V. Extension tectonics: the Neogene opening of the north-south
210 trending basins of central Thailand. *Journal of Geophysical Research.* **93**,
211 11899–11910 (1988).

212 Metcalfe, I. Palaeozoic-Mesozoic history of SE Asia. *Geological Society, London,*
213 *Special Publications.* 355, 7–35 (2011).

214 Metcalfe, I. Gondwana dispersion and Asian accretion: tectonic and palaeogeographic
215 evolution of eastern Tethys. *Journal of Asian Earth Sciences.* **66**, 1–33 (2013).

216 Smith, A. D. The geodynamic significance of the DUPAL anomaly in Asia. In: Flower,
217 M. F. J., Chung, S. L., Lo, C. H. & Lee, T. Y. (Eds.), *Mantle Dynamics and Plate*
218 *Interactions in East Asia. Geodynamics Series.* vol. 27. American Geophysical
219 Union Press, Washington, D.C., pp. 89–105 (1998).

220 Tian, Z. X., Yan, Y., Huang, C. Y., Zhang, X. C., Liu, H. Q., Yu, M. M., Yao, D. &
221 Dilek, Y. Geochemistry and geochronology of the accreted mafic rocks from the

222 Hengchun peninsula, Southern Taiwan: origin and tectonic implications. *Journal*
223 *of Geophysical Research: Solid Earth*. **124**, 2469–2491 (2019).

224 Tu, K., Flower, M. F. J., Carlson, R. W., Zhang, M. & Xie, G. Sr, Nd, and Pb isotopic
225 compositions of Hainan basalts (south China): implications for a subcontinental
226 lithosphere Dupal source. *Geology*. **19**, 567–569 (1991).

227 Wang, X. C., Li, Z. X., Li, X. H., Li, J., Liu, Y., Long, W. G., Zhou, J. B. & Wang, F.
228 Temperature, pressure, and composition of the mantle source region of Late
229 Cenozoic basalts in Hainan Island, SE Asia: a consequence of a young thermal
230 mantle plume close to subduction zones?. *Journal of Petrology*. **53(1)**, 177–233
231 (2012).

232 Wang, X. C., Li, Z. X., Li, X. H., Li, J., Xu, Y. G. & Li, X. H. Identification of an
233 ancient mantle reservoir and young recycled materials in the source region of a
234 young mantle plume: implications for potential linkages between plume and
235 plate tectonics. *Earth and Planetary Science Letters*. **377**, 248–259 (2013).

236 Yan, Q. S., Shi, X. F., Wang, K. S., Bu, W. R. & Xiao, L. Major element, trace
237 element, and Sr, Nd and Pb isotope studies of Cenozoic basalts from the South
238 China Sea. *Science in China Series D: Earth Sciences*. **51(4)**, 550–565 (2008).

239 Yan, Q. S., Castillo, P., Shi, X. F, Wang, L. L., Liao, L. & Ren, J. Geochemistry and
240 petrogenesis of volcanic rocks from Daimao Seamount (South China Sea) and
241 their tectonic implications. *Lithos*. **218–219**, 117–126 (2015) .

242 Yan, Q., Metcalfe, I. & Shi, X. U-Pb isotope geochronology and geochemistry of
243 granites from Hainan Island (northern South China Sea margin): Constraints on

244 late Paleozoic-Mesozoic tectonic evolution. *Gondwana Research*. **49**, 333–349
245 (2017).

246 Yan, Q., Shi, X., Metcalfe, I., Liu, S., Xu, T., Kornkanitnan, N. & Zhang, H. Hainan
247 mantle plume produced late Cenozoic basaltic rocks in Thailand, Southeast Asia.
248 *Scientific reports*. **8(1)**, 2640 (2018).

249 Zhang, G. L., Chen, L. H., Jackson, M. G. & Hofmann, A. W. Evolution of carbonated
250 melt to alkali basalt in the south china sea. *Nature Geoscience*. **10(3)**, 229–235
251 (2017).

252 Zhang, G. L., Luo, Q., Zhao, J., Jackson, M. G., Guo, L. S. & Zhong, L.F.
253 Geochemical nature of sub-ridge mantle and opening dynamics of the South
254 China Sea. *Earth and Planetary Science Letters*. **489 (1)**, 145–155 (2018).
255