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1	Highly heterogeneous mantle caused by recycling of oceanic
2	lithosphere from the mantle transition zone
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20 Abstract

21 Geochemical heterogeneities observed in the mantle are usually attributed to 22 recycling of oceanic lithosphere through subduction. However, it remains hotly 23 debated where recycled material stagnates, and how quickly it can be liberated back to 24 surface. This knowledge gap hinders our understanding of mantle circulation and the 25 chemical evolution of the Earth. Here we address these questions using a combination 26 of geochronology and geochemistry from South China Sea (SCS) seamounts. The 27 Shixingbei seamount lavas formed during active seafloor spreading at c. 19.1 Ma 28 show limited geochemical variability, whereas the Zhenbei-Huangyan seamount chain formed during the post-spreading stage at c. 7.8 Ma and displays a wide range of 29 30 compositions. However, melt inclusions in olivine and plagioclase from the 31 Zhenbei-Huangyan basalts show considerably greater isotopic variability than seen in 32 the whole rock compositions of both the SCS syn- and post-spreading lavas. A 33 previously unidentified third mantle source component (FOZO) revealed by 34 olivine-hosted melt inclusions along with both depleted (DMM) and enriched (EMII) 35 mantle components is required in the source region to explain the observed isotopic 36 and chemical variability. On the basis of our results, the age of the recycled ocean crust and sediments in this region are estimated to be c. 120 - 350 Ma. We infer that 37 38 these enriched components in the SCS lavas come from the mantle transition zone. 39 Variations in mantle source heterogeneity coupled with melting process control 40 spatial-temporal (spreading vs. post-spreading stage) geochemical variations of lavas

from the SCS and surrounding areas. Together with the results from published studies, we propose that marginal basins are one of the major locations on Earth where oceanic and/or continental lithosphere are transferred into the upper mantle and transition zone, representing an important source of upper mantle heterogeneity. We provide a simple conceptual model linking plate subduction and upper mantle heterogeneity and the volcanism in the SCS and surrounding areas.

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48 Keywords

South China Sea; mantle transition zone; recycled oceanic crust; EMII and FOZO;
mantle heterogeneity

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#### 52 **1. Introduction**

In general, mid-ocean ridge basalts (MORBs) have relatively uniform and 53 54 overall depleted compositions, while oceanic island basalts (OIBs) display 55 considerable chemical heterogeneity (e.g., Gale et al., 2013; Hoernle et al., 2011; 56 Hofmann, 1997; Jackson and Hart, 2006; Salters and Stracke, 2004; Stracke et al., 2005). Five endmembers are commonly proposed to explain the isotopic 57 heterogeneity observed in oceanic rocks: depleted MORB-source mantle (DMM), 58 enriched mantle one (EMI) and two (EMII), high time-integrated  $\mu$  (<sup>238</sup>U/<sup>204</sup>Pb) 59 60 mantle (HIMU), and a focal zone (FOZO) component (e.g., Hanan and Graham, 1996; Hickey-Vargas, 1992; Stracke et al., 2005; Workman and Hart, 2005; Zindler 61

and Hart, 1986). The cause of the geochemical differences between MORBs and OIBs
and the chemical heterogeneity of Earth's mantle is mainly attributed to mantle
convection, but the understanding of where recycled material stagnates and how it is
mingled and arrives back at the surface, is still debated.

MORBs originate from a large-scale homogeneous, incompatible element-poor 66 67 shallow and degassed mantle, whereas OIBs are related to mantle plumes sampling 68 both ancient undegassed domains and recycled crustal reservoirs (e.g., Castillo et al., 69 2010; Hofmann, 1997; Jackson and Hart, 2006; McCoy-West et al., 2016; Salters and 70 Stracke, 2004; Stracke et al., 2005; Workman and Hart, 2005; Zindler and Hart, 71 1986). Nevertheless, the two-layer mantle convection regime proposed based on 72 geochemical data (e.g., Hofmann, 1997) is hard to reconcile with geophysical 73 observations for whole-mantle convection (van der Hilst et al., 1997). Alternatively, a 74 ubiquitous distribution of small- to moderate-scale heterogeneity in the upper mantle 75 is considered to be responsible for geochemical variability of MORBs and OIBs 76 (Castillo et al., 2010; Meibom and Anderson, 2004).

The South China Sea (SCS), one of the marginal basins in the western Pacific, formed by seafloor spreading between 33 – 15 Ma (Li et al., 2014). During the period of SCS seafloor spreading, magmatic activities primarily occurred in the SCS basin and southeast China, then, after cessation of SCS seafloor spreading, intraplate volcanism was widely distributed in the SCS basin and adjacent areas (such as southeast (SE) China, the Pearl River basin, and Indochina; **Fig. 1**) (Wang et al., 2013; Zou and Fan, 2010). Previous studies show remarkable differences between the
geochemical compositions of the spreading stage MORBs and post-spreading
seamount lavas (Qian et al., 2021). However, the causes of these remain unclear. In
addition, it remains unclear why volcanism after cessation of spreading is widespread
in the SCS and surrounding areas.

88 Although bulk-rock geochemistry of mafic volcanic rocks is commonly used to 89 evaluate the geochemical heterogeneity of the mantle, most magmas that reach the 90 surface represent complex mixtures of melts, which through their homogenization and 91 re-equilibration with the surrounding mantle at shallower depths effectively erase 92 much of their original source heterogeneity (Kamenetsky et al., 1997; Kent, 2008; 93 Peate et al., 2012). Detailed studies of individual rock samples, especially of the 94 isotopic composition of their various constituents, including crystal cargo and melt 95 inclusions trapped in the crystals, can be used to assess the heterogeneity of the 96 underlying mantle source (e.g., Hanyu et al., 2019; Jackson and Hart, 2006; 97 Kamenetsky et al., 1997; Kent, 2008; Qian et al., 2017; Ramos and Tepley Iii, 2008; 98 Rose-Koga et al., 2017; Sobolev et al., 2011). In this study we examine the age and 99 geochemistry (whole rock; matrix glass; olivine (ol), clinopyroxene (cpx) and 100 plagioclase (plag) crystal; and ol and plag-hosted melt inclusion compositions) of the 101 from the syn-spreading Shixingbei lavas seamount. and post-spreading 102 Zhenbei-Huangyan seamount chain in the SCS (Fig. 1). Our results display considerably larger isotopic variability than observed previously from whole rocks 103

analyses, pointing to mantle source heterogeneity beneath the SCS resulting from recycling of oceanic crust and sediments. The enriched mantle reservoirs are most likely introduced from the mantle transition zone (MTZ) into the upper mantle. Based on the temporal-spatial evolution of volcanism and our new geochemical and geophysical evidence, we propose that the Cenozoic magmatism in SE Asia is generated by active upwelling of mantle material linked to curved subduction systems.

- 111
- 112 **2.** Geological setting and samples

113 The SCS is one of the largest marginal seas in the western Pacific (~3,500,000 km<sup>2</sup>), with the Eurasian plate to the north, the Philippine Sea plate to the east, the 114 115 Australian plate to the south, and the Indian plate (or Indochina block) to the west 116 (Fig. 1). The SCS developed as a result of multiple episodes of continental rifting and 117 seafloor spreading during the Cenozoic (Briais et al., 1993; Li et al., 2014; Sibuet et 118 al., 2016). The deep-water basin of the SCS comprises three parts, the East Sub-basin 119 (ESB), Southwest Sub-basin (SWSB) and Northwest Sub-basin (NWSB) (Fig. 1). Constrained by mapping magnetic anomalies and cores recovered from the 120 121 International Ocean Discovery Program (IODP) Expedition 349 (microfossil age 122 constraints), seafloor spreading started 33 Ma near the northern continental margin of 123 the SCS (corresponding to the oldest seafloor spreading magnetic anomaly C12), and 124 ceased at c. 15 Ma in the ESB and c.16 Ma in the SWSB (Li et al., 2014). Late 125 Cenozoic volcanism is widespread and a prominent feature in SE Asia (including SCS, SE China, and Indochina Peninsula), with the majority of magmatism younger 126 127 than sea-floor spreading in the SCS (between c. 33 and 16 Ma) (e.g., Hoang & 128 Flower, 1998; Wang et al., 2013; Yan et al., 2018; Zou and Fan, 2010). The SCS and 129 nearby areas are surrounded by several subduction zones (Fig. 1), indicating that the 130 volcanism in SE Asia took place in the same tectonic setting. In addition, the SCS 131 post-spreading seamount lavas and synchronous intraplate lavas in SE China and 132 Indochina Peninsula share similar chemical and isotopic characteristics with typical 133 E-MORB to OIB-type incompatible element distributions, and distinctive Pb isotope 134 compositions similar to EMII-type OIB (e.g., Hoang & Flower, 1998; Qian et al., 2021; Wang et al., 2013; Yan et al., 2018; Zou and Fan, 2010), suggesting a common 135 136 mantle source.

137 The SCS preserves both seafloor spreading and post-spreading volcanic features. 138 Seafloor spreading stage seamounts are mostly distributed in the northern SCS; 139 post-spreading stage volcanism is more extensive in the SCS basin and predominantly 140 occurs in ESB and SW SCS basin, such as two north-south trending seamount chains 141 (Longbei and Longnan seamounts; Nanyue and Beiyue seamounts) in the SWSB, and an east-west trending Zhenbei-Huangyan seamount chain in ESB. The samples for 142 143 this study were collected from Shixingbei seamount, which is not in a chain, and the 144 Zhenbei-Huangyan seamount chain in the SCS (Fig. 1) with the ROPOS remotely 145 operated subsea vehicle operated by the Canadian Scientific Submersible Facility 146 (Fig. S1). Shixingbei seamount lies in the middle of the SCS Basin (Fig. 1). The 147 Zhenbei and Huangyan seamounts formed after seafloor spreading ceased in the ESB 148 and are aligned northeastwards from the fossil spreading ridge (Li et al., 2014) (Fig. 149 1). Most lavas from these seamounts display pillow structures with columnar jointing 150 (Fig. S1c and d). The basalts from the SCS seamounts are typically variably vesicular 151 and porphyritic (3-20 %) with phenocrysts dominated by olivine (ol), clinopyroxene 152 (cpx) and plagioclase (plag) (Fig. S2a, b, c, d, e). In some samples, ol crystals contain 153 spinels (Fig. S2a, b), and ol and plag crystals also contain melt inclusions up to 140 154 μm in size (Fig. S2c, e). Biotite is found in rare evolved samples (such as trachytes), 155 in which alkali-feldspar and cpx dominate the phenocryst assemblage (Fig. S2f).

156

#### 157 **3. Analytical methods**

158 Complete description of the analytical techniques can be found in the 159 supplementary material and are briefly discussed here. Plag separates from two 160 samples (2061-R7 from Shixingbei seamount and 2055-R12 from Huangyan 161 seamount) were selected for Ar-Ar age determinations by an ARGUS-VI 162 multi-collector instrument at the Argon geochronology laboratory of the Oregon State 163 University.

A U-Pb age on zircon from trachyte 2059-R16 from Zhenbei seamount was dated using a Thermo Element XR instrument connected to a RESOlution M-50 laser at Guangzhou Institute of Geochemistry, Chinese Academy of Science (GIG-CAS).

167	Major and trace elements of the SCS seamount whole-rock samples were analyzed
168	using a PANalytical AXIOS-Advanced X-ray fluorescence spectrometer at the State
169	Key Laboratory of Marine Geology (SKLMG) in Tongji University, and an Agilent
170	7900 inductively coupled plasma-mass spectrometer (ICP-MS) at GIG-CAS,
171	respectively. The whole-rock Sr-Nd-Pb isotopes of volcanic samples were analyzed
172	using a Neptune Plus multi-collector (MC-)ICP-MS at GIG-CAS. Major and trace
173	elements of minerals and glasses (matrix glass and melt inclusion) were analyzed
174	using a JEOL JXA-8230 electron probe micro-analyzer at SKLMG in Tongji
175	University and an excimer 193 nm laser ablation system (RESOlution M-50) coupled
176	to a Thermo Element ICP-MS instrument at GIG-CAS, respectively. In situ Sr-Pb
177	isotopes of feldspar, matrix glass and melt inclusion were analyzed using a Neptune
178	Plus MC-ICP-MS with a RESOlution M-50 laser at GIG-CAS.

179

180 **4. Results** 

181 4.1 Geochronology

Some samples described here were collected from Shixingbei seamount (**Fig. 1**), and the age of volcanism at these sites was unknown prior to this study. Plag in volcanic rock sample 2061-R7 from the Shixingbei seamount yield a  ${}^{40}$ Ar/ ${}^{39}$ Ar plateau age of 19.10 ± 0.78 Ma (**Fig. 2A**), indicating the off-axis Shixingbei seamount formed during the syn-spreading stage (33 – 16 Ma) of the SCS. The two lavas from the Huangyan and Zhenbei seamounts in the SCS ESB yielded significantly younger

188	ages. Sample 2055-R12 from the Huangyan seamount gave a <sup>40</sup> Ar/ <sup>39</sup> Ar plateau age of
189	7.78 $\pm$ 0.21 Ma ( <b>Fig. 2B</b> ), which affirms a previously published K-Ar age data (7.77 $\pm$
190	0.49 Ma) for samples from the same seamount (Wang et al., 2009). U-Pb dating of
191	zircon grains from the Zhenbei seamount trachyte 2059-R16 yields a weighted mean
192	$^{206}$ Pb/ $^{238}$ U age of 7.76 ± 0.13 Ma (n = 31; <b>Fig. 2C</b> ). The Zhenbei-Huangyan seamount
193	chain samples are nearly age-synchronous and do not show clear age progression.
194	Thus, the available data indicate that the Shixingbei seamount formed while seafloor
195	spreading was still active in the SCS, whereas the Zhenbei-Huangyan seamount lavas
196	formed after spreading ceased.

197

#### 198 *4.2 Geochemistry*

The studied Shixingbei seamount and Zhenbei-Huangyan seamount chain lavas show temporal variations in geochemical compositions. The Shixingbei seamount lavas formed during the SCS spreading stage are tholeiitic basalts (alkalis (Na<sub>2</sub>O +  $K_2O$ ) = 3.03 – 3.59; SiO<sub>2</sub> = 49.59 – 51.32; MgO = 4.90 – 5.93; all oxide contents in weight % normalized to 100% on volatile-free basis), and possess similar major element compositions to spreading stage MORBs sampled by IODP Expedition 349 within the SCS basin (**Fig. 3**).

The post-spreading Zhenbei-Huangyan seamount lavas range from alkali basalt through trachyte (SiO<sub>2</sub> = 45.2 - 62.7; MgO = 9.6 - 0.7) to basanite (SiO<sub>2</sub> = 43.4 -44.4; MgO = 10.4 - 11.8) (**Fig. 3**). MgO is highest in ol-hosted melt inclusions (7.2 - 209 11.9), intermediate in plag-hosted melt inclusions (4.5 - 6.3) and lowest in matrix glass (2.2 - 4.6) (Fig. 3B). As MgO decreases in ol-hosted melt inclusions, whole 210 211 rocks and matrix glass with MgO > 4.5, both SiO<sub>2</sub>, and Al<sub>2</sub>O<sub>3</sub> increase and FeO<sup>t</sup> (t = 212 total) decrease overall (Fig. 3), reflecting an assemblage dominated by ol, cpx and 213 Fe-oxide fractionation. Plag melt inclusions and matrix glass with MgO < 4.5 (Fig. 3), 214 however, are shifted to lower Al<sub>2</sub>O<sub>3</sub> and Na<sub>2</sub>O and higher FeO<sup>t</sup>, consistent with plag joining the fractionating phases. Ol have relatively high CaO (0.16 - 0.35 wt.%)215 216 (Table S5), favoring crystallization from mafic melts rather than being xenocrysts 217 from mantle peridotite (Qian et al., 2021). Cpx have diopsidic to augitic compositions  $(En_{35-44}Fs_{11-20}Wo_{41-48})$  with relatively high Mg#  $(100Mg/(Mg+Fe^{+2}) = 63 - 80;$  Table 218 219 **S3**). Plag range from bytownite to andesine ( $An_{57-75}$ ; **Table S4**).

220 The Zhenbei-Huangyan seamount whole rock lavas, matrix glass and plag-hosted 221 melt inclusions have incompatible element contents characteristic of intraplate 222 volcanic rocks, showing overall enrichment in highly to moderately incompatible 223 elements, slight relative Nb enrichment and relative K and Pb depletion (Fig. 4A, B). 224 Notably, the SCS Shixingbei seamount tholeiitic basalts have significantly lower 225 incompatible trace element concentrations than the Zhenbei-Huangyan seamount 226 lavas (Fig. 4A), but slightly higher incompatible trace element concentrations than 227 MORBs erupted during the SCS spreading stage (Fig. 4A). They also have lower 228 La/Sm, Sm/Yb, and Nb/Zr than the Zhenbei-Huangyan seamount lavas (Fig. 4C, D).

229	The Shixingbei tholeiitic basalts $(n = 4)$ formed during the SCS spreading stage
230	have less radiogenic ${}^{87}$ Sr/ ${}^{86}$ Sr (0.7033 – 0.7034) and ${}^{206}$ Pb/ ${}^{204}$ Pb (18.537 – 18.571),
231	and more radiogenic $\epsilon_{Nd}(5.2-5.4)$ than the Zhenbei-Huangyan lavas $({}^{87}\text{Sr}/{}^{86}\text{Sr}$ =
232	$0.7035 - 0.7043$ , ${}^{206}\text{Pb}/{}^{204}\text{Pb} = 18.591 - 18.809$ , and $\epsilon_{Nd} = 3.2 - 4.8$ ) (Fig. 5). In
233	general, matrix glass ( ${}^{87}$ Sr/ ${}^{86}$ Sr = 0.7036 - 0.7039; ${}^{207}$ Pb/ ${}^{206}$ Pb = 0.832 - 0.838;
234	$^{208}$ Pb/ $^{206}$ Pb = 2.092 - 2.098) and plag ( $^{87}$ Sr/ $^{86}$ Sr = 0.7035 - 0.7041; $^{207}$ Pb/ $^{206}$ Pb =
235	0.828 - 0.843; <sup>208</sup> Pb/ <sup>206</sup> Pb = 2.054 - 2.111) within the Zhenbei-Huangyan seamounts
236	show a similar range of Sr-Pb isotope compositions as those exhibited by the whole
237	rocks ( ${}^{87}$ Sr/ ${}^{86}$ Sr = 0.7035 - 0.7043; ${}^{207}$ Pb/ ${}^{206}$ Pb = 0.831 - 0.838; ${}^{208}$ Pb/ ${}^{206}$ Pb = 2.079 -
238	2.090) (Fig. 6; Table S1, S2 and S4). Plag-hosted melt inclusions extend to much
239	more radiogenic Sr ( ${}^{87}$ Sr/ ${}^{86}$ Sr = 0.7037 – 0.7067) ( <b>Fig. 6; Table S2</b> ). Ol-hosted melt
240	inclusions extend to considerably more radiogenic Sr ( ${}^{87}$ Sr/ ${}^{86}$ Sr = 0.7028 - 0.7062)
241	and Pb ( $^{207}$ Pb/ $^{206}$ Pb = 0.778 - 0.856; $^{208}$ Pb/ $^{206}$ Pb = 1.959 - 2.099) isotopes ( <b>Fig. 6</b> ;
242	Table S2). The melt inclusions in the most forsteritic of $(Fo_{88 - 89})$ have more
243	radiogenic Pb isotope ratios ( $^{207}$ Pb/ $^{206}$ Pb = 0.778 - 0.807; $^{208}$ Pb/ $^{206}$ Pb = 1.958 - 2.035)
244	than melt inclusions in Fo <sub>82-87</sub> ol ( $^{207}$ Pb/ $^{206}$ Pb = 0.816 – 0.856; $^{208}$ Pb/ $^{206}$ Pb = 2.050 –
245	2.099) ( <b>Table S2</b> ). Two melt inclusions in the same ol crystal have different ${}^{87}$ Sr/ ${}^{86}$ Sr
246	(0.70581 for 2055-R7-28MI1, 0.70658 for 2055-R7-28MI2) ( <b>Tables S2</b> ).

### **5.** Discussion

249 5.1 A three component mixing model for SCS seamount lava genesis

250	The SCS seamount lavas have a large range in Pb, Sr and Nd isotope
251	compositions (Fig. 5). It is critical to determine if this isotopic variability reflects a
252	heterogeneous source, or whether crustal contamination during magma ascent and
253	emplacement has modified the isotopic signal. Since ${}^{87}$ Sr/ ${}^{86}$ Sr ratios in 6 – 15 Ma
254	oceanic crust (e.g. IODP sites 504 and 1256) (Höfig et al., 2014) generally do not
255	exceed 0.7040 (only ~5%), assimilation of in-situ altered oceanic crust is unlikely to
256	generate the radiogenic <sup>87</sup> Sr/ <sup>86</sup> Sr (extending to 0.7067) observed for lavas from the
257	SCS and surrounding areas. Sediment assimilation is also unlikely due to the paucity
258	of sediment (< 10 m) on the seafloor at the time the seamounts formed (Li et al.,
259	2014). It is noteworthy that the isotope systematics of most of these lavas do not
260	appear to be significantly affected by crustal assimilation, as suggested by isotope
261	ratios being mostly independent of the degree of differentiation (e.g. MgO, Mg/Si,
262	Mg# and/or Fo content) (Fig. S6). In addition, the SCS seamount lava and most
263	intraplate lava samples with high- $\varepsilon_{Nd}$ (> 2.7) from the SE Asia have a large variation
264	in Nb/U with no correlations with $\epsilon_{Nd},$ indicating insignificant crustal contamination
265	(Fig. S6). By contrast, the low- $\epsilon_{Nd}$ (< 2.7) lavas (mainly from the Indochina
266	Peninsula; Wang et al., 2013) show strong covariation between Mg/Si, Nb/U, and $\epsilon_{\rm Nd}$
267	(Fig. S6), indicating that they are most likely affected by crustal contamination.

Lava whole rock compositions are commonly assumed to represent melt compositions (possibly, except for melt volatile element compositions), implying that minerals crystallized from that melt should inherit the whole rock composition. This 271 assumption, however, is not valid for the Zhenbei-Huangyan lavas. The most mafic 272 (MgO = 10.4 - 11.8) basanitic lavas represent complex mixtures of evolved basaltic melt (matrix glass with Mg# (100 Mg/(Mg + Fe<sup>2+</sup>)) = 32 - 44) and a complex crystal 273 274 cargo, primarily mafic ol (Fo<sub>82 - 89</sub>) and cpx (Mg# = 63 - 80) but also Fe oxides and 275 plag. Plag have major element and Sr-Pb isotopic compositions compatible with being 276 phenocrysts because their isotopic compositions are similar to those of their host 277 whole rocks. The plag- and ol-hosted melt inclusions, on the other hand, show a large 278 variation in Sr isotopes and record interaction with an EMII-like component with radiogenic  ${}^{87}$ Sr/ ${}^{86}$ Sr ( $\geq 0.7067$ ) (Fig. 6). In addition, the plag- and ol- melt inclusions 279 280 have higher MgO contents and show larger variations in isotopic compositions compared to the matrix glasses (Fig. 3, 6). Ol, thus, must have crystallized from more 281 282 mafic melts than the matrix glasses (Fig. 3, 6).

283 Whole rock Sr-Nd-Pb isotope compositions of post-spreading lavas from the 284 SCS and nearby areas (SE China and Indochina) (Fig. 5) have previously been 285 explained by mixing of at least two major components: depleted upper mantle or the 286 SCS spreading lavas and an EMII-like component (Wang et al., 2013; Zou and Fan, 287 2010). However, this two-component mixing model does not adequately explain the 288 isotope compositions of ol and plag-hosted melt inclusions from Zhenbei-Huangyan 289 seamounts (Fig. 6). Instead, they require mixing of at least three distinct components 290 (Fig. 6):

1) **DMM**: Zhenbei ol-hosted (Fo<sub>83 - 84</sub>) melt inclusions and IODP Site U1431 MORB lavas (Zhang et al., 2018) have non-radiogenic Sr ( ${}^{87}$ Sr/ ${}^{86}$ Sr  $\leq$  0.7029) and moderate radiogenic Pb ( ${}^{207}$ Pb/ ${}^{206}$ Pb > 0.86;  ${}^{208}$ Pb/ ${}^{206}$ Pb  $\geq$  2.10) isotopes, characteristic of a depleted upper mantle MORB type source;

2) **FOZO**: Huangyan melt inclusions hosted in magnesium-rich ol (Fo<sub>88 - 89</sub>) in equilibrium with mantle peridotite have non-radiogenic Sr ( ${}^{87}$ Sr/ ${}^{86}$ Sr = 0.7036 – 0.7037) and radiogenic Pb ( ${}^{207}$ Pb/ ${}^{206}$ Pb = 0.78 – 0.81;  ${}^{208}$ Pb/ ${}^{206}$ Pb = 1.96 – 2.03) isotopes. They are similar to the FOZO component ubiquitous in mid-ocean-ridge and intraplate basalts (Hanan and Graham, 1996; Stracke et al., 2005);

300 3) **EMII**: Huangyan melt inclusions hosted in plag and some ol (at least three ol 301 crystals) require a component with the most radiogenic Sr ( ${}^{87}$ Sr/ ${}^{86}$ Sr  $\ge 0.7067$ ) and 302 moderately radiogenic Pb ( ${}^{207}$ Pb/ ${}^{206}$ Pb = 0.826 - 0.845;  ${}^{208}$ Pb/ ${}^{206}$ Pb = 2.06 - 2.10) 303 isotopes.

304 Clearly, plag and ol crystals crystallizing from the same magma batch should possess identical isotopic compositions, therefore the large range of plag- and 305 306 ol-hosted melt inclusions compositions seen in the SCS seamount lavas (i.e. Fig. 6) 307 requires trapping melts of isotopically distinct parental magmas (Qian et al., 2017; 308 Ramos and Tepley Iii, 2008; Rose-Koga et al., 2017). The FOZO-like signature in 309 particular is only clearly identified in a few ol-hosted melt inclusions, implying that 310 the proportion of the FOZO component contributing to erupted lavas must be small, 311 and/or that the FOZO-like fragments in the mantle are so small that they are rarely 312 sampled in melt inclusions. They are probably present in the whole rock, but they are 313 so diluted that the FOZO "flavor" cannot be distinguished. It is apparent that 314 shallow-level storage and magma mixing resulted in dilution of the FOZO component 315 in the SCS seamount lavas, before eruption.

316

#### 317 5.2 Rapid recycling of slab-derived source components

318 We now discuss the possible origin of the enriched components (EMII and 319 FOZO) necessary to explain the isotopic variations of the crystals and their MIs in the 320 SCS (Fig. 6). Some plag- and ol-hosted melt inclusions have more radiogenic <sup>87</sup>Sr/<sup>86</sup>Sr than matrix glasses and plag crystals, and form mixing arrays between 321 322 SCS-spreading or Pacific MORBs and EMII (Fig. 6). A potential source component 323 for EMII is delaminated lower crustal material, but the H<sub>2</sub>O/Ce ratios of the SCS 324 seamount lavas are much higher than those of lower continental crust (LCC) granulite xenoliths from SE Asia (Qian et al., 2021). In addition, the LCC has higher <sup>87</sup>Sr/<sup>86</sup>Sr, 325  $^{143}$ Nd/ $^{144}$ Nd and lower  $^{206}$ Pb/ $^{204}$ Pb (< 18) (Zhang et al., 2018). Therefore, it is unlikely 326 327 to be the source of the SE Asian (including the SCS seamount) volcanism.

Subducting marine sediment, seawater-altered ocean crust and EMII have similar Pb isotope compositions but can extend to more radiogenic <sup>87</sup>Sr/<sup>86</sup>Sr and could serve as this end member (**Fig. 6**). Compared to matrix glass, plag-hosted melt inclusions extend to lower (Ce, Nd)/Pb but higher Ba/La ratios (**Table S2**), also favoring sediments as the EMII component. Geophysical studies provide additional support for recycled oceanic crust components beneath the SCS and Indochina. They could
originate from subducted Neo-Tethyan slabs from the southwest (230 – 90 Ma)
(Chung et al., 2005), or paleo-Pacific and Pacific slabs from the east (300 – 120 Ma)
(Li and Li, 2007). Given that the SE China region was far from the Neo-Tethyan
tectonic realm during the Mesozoic, it is likely that recycled Neo-Tethyan slabs are
not the major source of the FOZO-like signature observed in Cenozoic lavas.

339 The observed Sr and Pb isotopic heterogeneity in young (130-170 Ma) Pacific 340 oceanic crust and marine sediments from ODP sites 801 and 1149, east of the Mariana 341 and Izu-Bonin arcs (Hauff et al., 2003), respectively, covers the entire range exhibited by SCS spreading MORB, post-spreading volcanism and their crystal cargoes and 342 343 melt inclusions, and even extends to more extreme compositions (Fig. 6). For the 344 purpose of our modelling, we considered Pacific MORB from site 801C with high µ  $(^{238}\text{U}/^{204}\text{Pb} = 51)$  and radiogenic Pb  $(^{206}\text{Pb}/^{204}\text{Pb} = 20.03; ^{207}\text{Pb}/^{206}\text{Pb} = 0.777;$ 345  ${}^{208}$ Pb/ ${}^{206}$ Pb = 1.881; Hauff et al., 2003) as the FOZO component (Hanan and Graham, 346 347 1996; Stracke et al., 2005). Marine sediments (clay, chert and nannofossil marl) from Sites 801 and 1149 in Pacific Ocean with very radiogenic Sr ( ${}^{87}$ Sr/ ${}^{86}$ Sr = 0.70954) and 348 relatively radiogenic Pb ( $^{206}$ Pb/ $^{204}$ Pb = 18.61,  $^{207}$ Pb/ $^{206}$ Pb = 0.845, and  $^{208}$ Pb/ $^{206}$ Pb = 349 350 2.089) (Hauff et al., 2003) as the EMII component. Our isotope modeling results (Fig. 351 5 and 6) show that the isotopic heterogeneity seen in the SCS spreading- and 352 post-spreading lavas could represent mixtures of depleted upper mantle with recycled 353 oceanic crust and sediment.

354	The FOZO component ubiquitous in mid-ocean-ridge and intraplate basalts was
355	proposed to represent young ocean crust recycled through the transition zone and/or
356	upper mantle (Hanan and Graham, 1996; Stracke et al., 2005). Since the SCS lavas
357	have ${}^{3}\text{He}/{}^{4}\text{He}$ (7.4 – 8.0 R/R <sub>A</sub> ) similar to or lower than MORB (Qian et al., 2021), the
358	MTZ is the most likely storage location for ocean crust evolving to FOZO (Hanan and
359	Graham, 1996). Here we use a Monte Carlo simulation (see Fig. 7, S7, and Table
360	S11; see supplementary material for further details) to constrain the possible
361	combinations of source maturation times (in Ma) and $\mu(^{238}\text{U}/^{204}\text{Pb})$ values required to
362	reproduce the FOZO-like Pb isotope signature seen in the SCS. The Monte Carlo
363	simulation (Fig. 7, S7 and Table S11) randomly chooses an initial Pb isotope
364	compositions and a $\boldsymbol{\mu}$ value, which falls within the range seen in typical unradiogenic
365	paleo-Pacific oceanic crust, then calculates the time required to evolve to the FOZO
366	endmember. Pb isotope compositions always take longer to evolve with a lower $\mu$ , but
367	all scenarios are consistent with the relatively short timescales (< 500 Myr) needed to
368	generate the required FOZO-like isotope signature (Fig. 7). Based on the average $\mu$ of
369	42 measured in Pacific oceanic crust all the potential initial compositions can
370	reproduce the FOZO-like component in less than 120 – 350 Myr (Fig. 7). Thus, on the
371	basis of the unique geochemical constraints and geological setting, it seems most
372	plausible that the MTZ beneath SE Asia was most likely enriched by subducted
373	materials from past paleo-Pacific plate subduction. Given that the subduction of the
374	paleo-Pacific plate beneath SE Asia took place between c. 250 and 150 Ma (Li and Li,

375 2007), the timescales for oceanic crust recycling in marginal sea basin (e.g., < c. 250 376 Ma) are significantly shorter than those associated with the plate deep subduction and 377 mantle plume upwelling (possibly > 1.0 Gyr) (Chase, 1981; Hanyu et al., 2019; 378 Stracke et al., 2003). It has previously been shown that the Pb isotopic composition of 379 FOZO can be generated from 300 Ma-age oceanic crust, if some Pb from a continental source (e.g. pelagic sediments) was added during hydrothermal alteration 380 381 shortly after crust formation (Hanan and Graham, 1996). In particular, given that the 382 abundant seamount lavas or OIBs formed during the seafloor spreading and 383 post-spreading stage are highly variable in isotopic composition, their subduction and 384 recycling could be a plausible process to produce enriched MORB or intraplate lavas 385 isotopic signatures over relatively short timescales. The MTZ has been proposed as 386 the most likely storage location for such young ocean crust evolving to these 387 FOZO-like intermediate compositions (Hanan and Graham, 1996; Stracke et al., 388 2005). Thus, recycling of ocean crust through the upper mantle and transition zone 389 can occur on relatively short time scales. This rapid reworking is consistent with other 390 studies that have argued for short ( $\leq 600$  Myr) recycling times based on evidence 391 from mid-ocean ridge and intraplate volcanism (e.g. Hoernle et al., 2011; Mazza et 392 al., 2019; McCoy-West et al., 2016; Sobolev et al., 2011). Ultimately, recycling of 393 enriched oceanic lithosphere through the transition zone over those short time scales 394 (< 250 Myr) seen in the SCS has the potential to significantly enhance upper mantle 395 heterogeneity.

397 5.3 Source heterogeneity and melting process control on geochemical variations of
398 late Cenozoic volcanism

Our new Ar-Ar dating results show that the Shixingbei seamount is  $19.10 \pm 0.78$ Ma (**Fig. 2A**), and is 3 – 5 Myr younger than the underlying MORBs, which formed around 23 – 24 Ma with a spreading rate of c. 25 km/Myr based on magnetic anomaly data (Li et al., 2014). The final age of seafloor spreading in the ESB of the SCS is estimated to be c. 15 Ma (Li et al., 2014). The Shixingbei seamount, is  $\leq$  150 km away from the present-day fossil ridge axis in the ESB (**Fig. 1**), indicating that it formed off-axis on the flank of the fossil ridge.

406 Off-axis seamounts can form near a ridge axis if an excessive magma supply is 407 available from residual mantle upwelling feeding volcanic activity on the ridge axis. 408 Following abandonment of a spreading ridge, the average degree of mantle melting 409 under the abandoned ridge progressively decreases with time due to the decrease in 410 mantle upwelling rate (seafloor spreading rate) and the increase in overlying 411 lithosphere thickness by conductive cooling as it moves further off-axis (Brandl et al., 412 2012; Haase et al., 2011). This is observed in the off-axis Shixingbei seamount where 413 the seafloor spreading rate in the SCS decreased rapidly due to a ridge jump at 25 Ma 414 (Li et al., 2014). As off-axis seamount lavas are generated by lower degrees of 415 melting of heterogeneous mantle they would contain a larger contribution from more 416 fusible and enriched components, which are expected to have relatively enriched 417 geochemical flavor (e.g., Brandl et al., 2012; Castillo et al., 2010; Haase et al., 2011).
418 Compared to the SCS spreading stage MORBs, the off-axis Shixingbei seamount
419 basalts have more enriched trace element (elevated La/Sm and Nb/Zr, Fig. 4) and
420 isotopic compositions (more radiogenic Sr and less radiogenic Nd isotope signatures,
421 Fig. 5). Thus, the off-axis Shixingbei seamounts lavas were generated by lower
422 degrees of melting of a heterogeneous magma source relative to SCS MORBs
423 generated at the spreading center.

The Zhengbei and Huangyan seamounts have been dated at c. 7.8 Ma (Fig. 2), 424 425 which is much younger than the underlying fossil spreading ridge in the ESB of the SCS that was active at 15 Ma (Li et al., 2014). Previous studies have shown that 426 427 buoyancy-induced upwelling by melt-depleted residual mantle and associated lower 428 degree melting relative to spreading stage can be responsible for continued 429 magmatism on fossil ridges for several Myr after active spreading ceased (Brandl et 430 al., 2012; Castillo et al., 2010; Haase et al., 2011). A similar mechanism can explain 431 to some extent the enriched geochemical signatures observed in some seamounts on 432 the SCS fossil ridges that erupted several Myr after spreading ceased. The post-spreading stage Huangyan and Zhenbei seamount lavas are characterized by 433 434 E-MORB- to OIB-type incompatible element abundances, and compared to the 435 spreading stage MORBs and off-axis Shixingbei seamount lavas, show a significantly 436 higher La/Sm, fractionated HREE element patterns (Sm/Yb  $\gg$  1), and more enriched Sr and Nd isotope compositions (Fig. 4, 5). This suggests that the post-spreading 437

438 seamount lavas were most likely generated by lower degrees of melting with residual 439 garnet in their mantle source. The geochemical compositions of SCS seamount lavas 440 together with the new age data indicate that the variable degrees of melting of 441 heterogeneous mantle can reproduce the trace element and isotope variations in the 442 lavas.

443 Interestingly, similar features are observed in intraplate volcanism throughout SE 444 Asia, there is a large contrast in the compositions of magma generated during the SCS 445 spreading (33 – 16 Ma) and post-spreading (post-16 Ma) stages (Fig. 4, 5, 8C and D). 446 Spatially, compared to the post-spreading seamount lavas in the SCS basin, there are significantly larger chemical and isotopic variations (e.g., Sm/Yb and <sup>87</sup>Sr/<sup>86</sup>Sr) in the 447 448 intraplate basalts from neighboring areas (western- and northern-group), although this 449 could simply reflect much less data from the SCS basin (Fig. 8E, F). The 450 post-spreading basalts in the SCS basin erupted on thinner lithosphere (45 - 50 km)451 have geochemical characteristics consistent with higher degrees (F) and shallower 452 depths (P) of melting (e.g., lower Sm/Yb; Fig. 8E), whereas intraplate basalts erupted 453 on thicker lithosphere (60 - 90 km) (Wu et al., 2004) (e.g., south China and 454 Indochina, north and west of the SCS) show the inverse (Fig. 8E). In accordance with 455 these observations, we propose that there was a common mantle source beneath SE 456 Asia during the spreading and post-spreading stages, but the enriched (OIB-type) 457 mantle was generally more diluted in the SCS MORBs and seamount lavas due to 458 greater degrees of melting at shallower depths. Our new results support a model where variable degrees of melting of a heterogeneous mantle are responsible forgeochemical variability of MORBs and OIBs (Meibom and Anderson, 2004).

461

#### 462 5.4 Volcanism in SE Asia linked to subduction-induced mantle upwelling

463 The cause of intraplate volcanism and dynamics of mantle upwelling in SE Asia 464 are subject to debate, and have been variously attributed to the Hainan mantle plume 465 (e.g. Wang et al., 2013; Yan et al., 2018; Zou & Fan, 2010), plate subduction induced mantle flow (e.g. Lin et al., 2019; Qian et al., 2021), and eastward mantle flow escape 466 467 in response to Indo-Eurasian collision (e.g., Hoang & Flower, 1998). The volcanism in SE Asia is not easily explained by the model of a mantle plume rooted at the core-468 469 mantle boundary under Hainan island because: (1) multiple volcanic episodes of 470 distinct chemistry and age are observed in SE Asia (Hoang and Flower, 1998; Fig. 1); 471 (2) the volcanism around Hainan island is much younger and obviously weaker than that in nearby areas (SE China, SCS and Indochina) (Fig. 1); (3) <sup>3</sup>He/<sup>4</sup>He of SCS 472 seamount glasses (7.36 to 8.03  $R/R_A$ ) are significantly lower than classical mantle 473 474 plume-derived basalts (Hawaii and Iceland) but typical of normal MORB (Qian et al., 2021); (4) there is no well-defined 'hotspot' track associated with age-progressive 475 476 volcanism in SE Asia (Li et al., 2014; this study); and, (5) broad (i.e. non plume-shaped), ambiguous P-wave tomographic anomalies above the transition zone 477 478 together with weaker, diminishing slow anomalies below the transition zone beneath

479 SE Asia, challenge the view of the 'Hainan plume' as a classical plume structure (Lin480 et al., 2019).

481 The SE Asia region is surrounded by several sub-duction zones (Fig. 1), 482 suggesting a potential association between plate subduction and magmatic activity 483 (Lin et al., 2019). During the seafloor spreading stage of the SCS, in addition to 484 formation of new oceanic crust and a small number of seamounts near the spreading 485 center, igneous activity (with ages between 26 - 16 Ma) occurred predominantly along coastal SE China and the SCS northern margin (see Fig. 1, 8A). Its spatial 486 487 extent could indicate that the spreading stage volcanism might be induced by westward subduction of the Pacific and Philippine Sea plates beneath Eurasia, since 488 489 these were located along eastern SE Asia. Intraplate magmatism within Indochina 490 (peaking < 8 Ma) is generally younger than along coastal SE China (generally 10 - 16Ma) (Hoang and Flower, 1998; Fig. 1, 8A and B). Seismic tomography studies show 491 492 evidence for the subduction of the Indo-Australian plate beneath SE Asia along the 493 Sunda trench with slab segments deflected horizontally in the MTZ towards the SCS 494 and Indochina (Li and van der Hilst, 2010). Therefore, it seems likely that Cenozoic 495 volcanism in Indochina could be related to subduction-induced mantle upwelling along the Sunda trench. Hainan island is far from any subduction zones and shows 496 497 intraplate volcanism peaking < 5 Ma, much younger than the age of the volcanism in 498 the SCS and coastal areas of SE China (Zou and Fan, 2010; Fig. 1, 8B). Thus, the 499 spatiotemporal distribution and evolution of the intraplate volcanism could plausibly 500 be active upwelling of mantle material linked to circular subduction systems of the 501 Pacific, Philippine Sea, and Indo-Australian plates (**Fig. 9**). Indeed, geodynamic 502 models that tested a variety of input SE Asian plate reconstructions demonstrated 503 robust, localized upper mantle upwellings within SE Asia during the Cenozoic that 504 were induced by the regional subduction (Lin et al., 2019).

505 Here, we propose a simple conceptual model linking the flux from multiple 506 subducting plates to upper mantle heterogeneity, and volcanism in SE Asia (Fig. 9). 507 The induced mantle upwelling associated with local subduction can be generated 508 ahead of a deep subducting slab and adjacent to edges of the stagnant slab segments in 509 the transition zone, despite being far from arcs (Faccenna, 2010). Recent geodynamic 510 models showed that this slab flux would stimulate upwelling of the enriched MTZ 511 reservoirs and eventually lead to volcanic eruptions (Mather et al., 2020). We suggest 512 that this subduction-induced upwelling originating from the MTZ and surrounding 513 area as revealed by seismic tomography, and associated decompression melting feed 514 the diffuse and non-age-progressive volcanism in SE Asia. A similar mantle return 515 flow, driven by long-lived subduction, has also been proposed to cause the intraplate volcanism across eastern Australia and Zealandia, as well as northern and 516 517 northeastern China (Mather et al., 2020; Zhao et al., 2011). Slab-triggered mantle 518 return flow beneath SE Asia is significantly more complex than under eastern 519 Australia-Zealandia and, northern and northeastern China, as SE Asia is surrounded 520 by complicated convergent subduction systems on three sides. Therefore, it remains 521 challenging at present to reconstruct and pinpoint the exact geochemical signature and 522 mantle flows associated with a specific subducted slab flux. Further geodynamic 523 modeling, high-resolution seismological studies and geochemical and age constraints 524 on magmatism are required to quantify the link between cycling of 525 subduction-induced mantle flows and time-space evolution of volcanism.

526

#### 527 **6.** Conclusions

We present new geochemical and geochronological data for lavas from the South China Sea (SCS) seamounts. Through comparison with existing geochemical data we investigate mantle source compositions and the mantle evolution, and infer a possible mechanism to generate mantle heterogeneity and the volcanism in the SCS and surrounding areas. The major results of this work are as follows:

533 1. The Shixingbei seamount lavas formed during the syn-spreading stage (19.1 534 Ma; 33-16 Ma) are limited to tholeiitic basalts. The Zhenbei-Huangyan seamount 535 lavas formed during the post-spreading stage (7.8 Ma) show wide variations in 536 chemical compositions, ranging from basanite through alkali basalt to trachyte. The SCS Shixingbei seamount tholeiitic basalts are more depleted in incompatible trace 537 element (e.g. La/Sm and Sm/Yb), and have less radiogenic <sup>87</sup>Sr/<sup>86</sup>Sr and more 538 radiogenic <sup>143</sup>Nd/<sup>144</sup>Nd relative to most post-spreading lavas. Mantle source 539 heterogeneity together with melting process control spatial-temporal (spreading and 540

541 post-spreading stage) geochemical variations of lavas from the SCS and surrounding542 areas.

543 2. Zhenbei-Huangyan seamount whole rocks, matrix glass and plag show 544 limited isotopic variation, while ol-hosted and plag-hosted melt inclusions show large 545 variations in radiogenic Sr and Pb isotope ratios. A three-component mixing model 546 (DMM-EMII-FOZO) is required to adequately explain the variability in isotopic 547 compositions of the ol-hosted and plag-hosted melt inclusions.

548 3. The combined trace element and isotopic data of the SCS seamount melt 549 inclusions, minerals and lavas demonstrate that the mantle source in the region is influenced by a package of young recycled oceanic crust with variable amounts of 550 551 sediment. Combined with geophysical evidence, this demonstrates that recycling of 552 ocean crust and marine sediments through the upper mantle from the MTZ can occur 553 on relatively short time scales (< 250 Myr), much faster than those associated with the 554 plate deep subduction and mantle plume upwelling (possibly > 1 Gyr). This new 555 evidence shows that the production of mantle reservoirs enriched by the storage of 556 past subducted slabs in the mantle transition zone is an important mechanism in the 557 generation of distinctive isotopic domains that were generally thought to be related to 558 deep recycling.

4. Our results show that marginal basins are important sites for introducing enriched oceanic and continental lithospheric material, i.e., chemical heterogeneity, into the upper mantle and transition zone. Subduction-induced upwelling from the 562 mantle transition zone and nearby areas can explain the late Cenozoic volcanism563 observed in SE Asia.

564

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576

#### 577 Figure captions

**Fig. 1**. Simplified geological map of the South China Sea (SCS) and adjacent regions, and sampling locations. (A) Bathymetric map of the present-day SCS and adjacent regions. Distribution of late Cenozoic intraplate volcanism in the SCS basin (red circles) and surrounding area (e.g., western group including Indochina block and Hainan (grey circles); and northern group including Southeast China and Taiwan 583 (blue circles)) are shown with ages labeled in red. Spreading stage volcanism (pink dots) is also shown with ages labeled in black (Age data for both are from the 584 585 GEOROC database (http://georoc.mpchmainz.gwdg.de/georoc)). Major magnetic 586 anomalies (blue lines) are also shown and marked with anomaly numbers after Li et 587 al. (2014). The red dashed lines show the final fossil ridges and the pink dashed line 588 marks the location of the ridge jump with the arrow indicating the direction of ridge 589 migration. Bathymetric taken from GEBCO\_2014 data were Grid 590 (http://www.gebco.net). SXB, Shixingbei; ZB, Zhenbei; HY, Huangyan. ESB, East 591 Sub-basin; SWSB, Southwest Sub-basin; NWSB, Northwest Sub-basin. (B) The 592 major subduction zones surrounding the SCS and the Indochina Peninsula. Inset map 593 showing location of tomographic profile (A1-A2) presented in Fig. 9.

594

595 Fig. 2. The ages of the South China Sea seamount lava samples. (A-B) <sup>40</sup>Ar/<sup>39</sup>Ar age
596 spectrum for plagioclase from the Shixingbei and Huangyan seamount lavas. (C)
597 U-Pb zircon concordia plot for sample 2059-R16 from Zhebei seamount. Inset is the
598 weighted average of zircon <sup>206</sup>Pb/<sup>238</sup>U ages.

599

Fig. 3. Variation of major elements in whole rocks, melt inclusions and matrix glasses for South China Sea (SCS) seamount lavas. (A) SiO<sub>2</sub> versus (vs.) total alkali (Na<sub>2</sub>O+K<sub>2</sub>O) content, MgO vs. (B) FeO<sup>t</sup>, (C) Al<sub>2</sub>O<sub>3</sub>, and (D) CaO for whole rocks (WR), matrix glasses (mgl), and olivine (ol)-hosted and plagioclase (plag)-hosted melt

inclusions (MI) from Shixingbei, Huangyan and Zhenbei seamounts. Data of
spreading stage MORB and post-spreading lavas in the SCS and intraplate volcanism
in surrounding area (e.g. Southeast China, Taiwan and Indochina) are from the
GEOROC (http://georoc.mpchmainz.gwdg.de/georoc). Mineral abbreviations cpx and
ilm denote clinopyroxene and ilmenite, respectively.

609

610 Fig. 4. Variation of trace elements in whole rocks, melt inclusions and matrix glasses 611 for South China Sea (SCS) seamount lavas. (A-B) Incompatible multi-element 612 diagrams, (C) Nb/Zr vs. La/Sm, (D) Sm/Yb vs. La/Sm for whole rocks (WR) from 613 Shixingbei, Huangyan and Zhenbei seamounts, and matrix glass and melt inclusion 614 from Huangyan lavas. Trace element data of primitive mantle, N-MORB, E-MORB 615 and OIB are from Sun and McDonough (1989). The SCS spreading stage MORBs 616 (Zhang et al., 2018), and Pacific and Indian MORBs (Gale et al., 2013) are shown for 617 comparisons. Data source of intraplate volcanism in southeast (SE) Asia is the same 618 as in Fig. 3. To minimize the effect of crustal contamination, intraplate lavas in SE 619 Asia with  $\varepsilon_{Nd} < 2.7$  have been excluded.

620

Fig. 5. Geochemical characteristics of the South China Sea (SCS) seamount samples.
(A) <sup>87</sup>Sr/<sup>86</sup>Sr vs. <sup>143</sup>Nd/<sup>143</sup>Nd, (B) <sup>143</sup>Nd/<sup>143</sup>Nd vs. <sup>206</sup>Pb/<sup>204</sup>Pb, (C) <sup>87</sup>Sr/<sup>86</sup>Sr vs.
<sup>206</sup>Pb/<sup>204</sup>Pb, (D) <sup>207</sup>Pb/<sup>204</sup>Pb vs. <sup>206</sup>Pb/<sup>204</sup>Pb for whole rocks (WR) from Shixingbei,
Huangyan and Zhenbei seamounts. Data sources: the SCS spreading stage MORBs

625 (Zhang et al., 2018); Pacific MORBs, EMI-type and EMII-type oceanic island basalts 626 (Stracke et al., 2003), and intraplate lavas in the SCS and SE Asia from the GEOROC database (http://georoc.mpchmainz.gwdg.de/georoc). The modeling results show 627 628 mixing trends (solid lines) between depleted SCS MORB mantle end-component 629 (DMM) and recycled oceanic crust and sediments. Dashed lines show mixing between 630 averaged Pacific MORB, and typical EMII mantle derived partial melt. To minimize the effect of crustal contamination, intraplate lavas in southeast Asia with  $\varepsilon_{Nd} < 2.7$ 631 632 have been excluded. The main parameters in the mixing model are given in 633 Supplementary Table S9.

634

Fig. 6. Isotopic variations in melt inclusions, whole rocks and matrix glasses for 635 South China Sea (SCS) seamount lavas. (A) <sup>87</sup>Sr/<sup>86</sup>Sr vs. <sup>207</sup>Pb/<sup>206</sup>Pb and (B) 636 <sup>207</sup>Pb/<sup>206</sup>Pb vs. <sup>208</sup>Pb/<sup>206</sup>Pb isotope diagram for whole rocks from Shixingbei seamount 637 638 lavas, and whole rocks (WR), matrix glass (mgl), olivine (ol)-hosted and plagioclase 639 (plag)-hosted melt inclusions (MI) and feldspars from Huangyan and Zhenbei 640 seamount lavas, illustrating that at least three components are required to explain the observed variations in isotopic composition: DMM-depleted SCS MORB-source 641 upper mantle, represented by the most depleted SCS MORB (Zhang et al., 2018), 642 643 FOZO-type component, interpreted as reflecting the young recycled material in the 644 mantle transition zone (Hanan and Graham, 1996; Stracke et al., 2005), well represented by young recycled Mesozoic Pacific MORB (Hauff et al., 2003), and an 645

646 EMII-type component - represented by Mesozoic Pacific Ocean sediment (Hauff et al., 2003). The main parameters in the three-component-mixing model between DMM 647 648 and recycled oceanic crust and sediments are the same as in Fig. 5. Data sources: 649 unpublished data from olivine-hosted melt inclusions from Hainan island lavas, to the 650 north of the SCS, from Ren (personal communication 2021); the SCS post-spreading 651 lavas, and Pacific and Indian MORB (http://georoc.mpch-mainz.gwdg.de/georoc/); 652 U1431 SCS spreading stage MORBs (Zhang et al., 2018); Mesozoic Pacific ocean 653 crust (Hauff et al., 2003); EMII (typical EM-2 OIBs), and HIMU (typical HIMU 654 OIBs) end members (Stracke et al., 2003); FOZO (Stracke et al., 2005) and C (Hanan and Graham, 1996). To minimize the effect of crustal contamination, intraplate lavas 655 656 in southeast Asia with  $\varepsilon_{Nd} < 2.7$  have been excluded.

657

658 Fig. 7. Results of Monte Carlo simulations for Pb model evolution of the recycled 659 component in FOZO mantle source. (A) Monte Carlo results derived from Equation (1) in the supplementary material for R1 values.  $\mu = {}^{238}U/{}^{204}Pb$ , the green area shows 660 661 the range for  $\mu$  values for Pacific oceanic crust and the dashed blue line defines the 662 average  $\mu$  value; (B) Modelling  $\mu$  in the mantle source. Modelled  $\mu$  compositions that are necessary to produce radiogenic  ${}^{206}\text{Pb}/{}^{204}\text{Pb}$  of typical FOZO ( ${}^{206}\text{Pb}/{}^{204}\text{Pb} = 20.03$ ) 663 signature starting from an un-radiogenic sample ( $^{206}Pb/^{204}Pb = 18.5$ ) of the 664 paleo-Pacific oceanic crust (Hauff et al., 2003). Each line corresponds to possible 665

source ages and the necessary  $\mu$  for that source to generate the radiogenic <sup>206</sup>Pb/<sup>204</sup>Pb of typical FOZO lavas.

668

Fig. 8. Spatial and temporal evolution of Cenozoic lavas in the South China Sea 669 670 (SCS) and surrounding areas. (A-B) Histogram showing age ranges for Cenozoic 671 (spreading- and post-spreading stage) lavas in the SCS and surrounding areas. (C-D) 672 Temporal variation of geochemical composition of spreading- and post-spreading stage lavas in the SCS and surrounding areas. (E-F) Spatial variation of geochemical 673 674 composition of intraplate lavas from the SCS, western group (Indochina and Leigiong, to the west of the SCS) and northern group (including south China and 675 Taiwan, to the north of the SCS) shown in Fig. 1. Data for ages and compositions of 676 677 Cenozoic Southeast **GEOROC** lavas in the Asia from the 678 (http://georoc.mpchmainz.gwdg.de/georoc). Intraplate lavas in southeast Asia affected 679 by crustal contamination have been excluded.

680

Fig. 9. Schematic model illustrating the formation mechanism of the Cenozoic
volcanism in southeast Asia based on integration of seismic and geochemical data.
Mantle structure beneath the SCS shown by FWEA18 full waveform tomography
P-wave perturbations (Tao et al., 2018) and earthquakes (red circles). Tomographic
fast anomalies indicate a swath of sub-horizontal subducted oceanic lithosphere
beneath the SCS at 500 – 700 km depths from previous subduction events. Although a

687 small amount of stagnant material may be older recycled slabs from past subduction, most of the visible stagnant slab segments stored in the transition zone imaged by 688 seismic tomography are likely to be younger subducted slabs from nearby 689 690 circum-SCS subduction zones (i.e. Indo-Australian, Pacific, Philippine Sea plates, or 691 Proto-SCS slabs). Even after SCS spreading ceased at 15 Ma, the more buoyant 692 metasomatized mantle continues to upwell and melt to form the SCS seamounts and possibly intraplate basalts in South China and Indochina with EMII type 693 694 compositions.

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