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- 1 Shallow gas and gas hydrate accumulations influenced by magmatic complexes
- 2 in the Pearl River Mouth Basin, South China Sea
- 3
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16 Abstract

We analyse occurrences of gas hydrates and free gases above a 1200 km² complex of 17 igneous volcanoes and sills using newly acquired three-dimensional (3D) seismic data 18 19 in the Pearl River Mouth Basin of the South China Sea. In strata above volcano summits, we recognize three enhanced reflection layers at different stratigraphic 20 21 levels, including discontinuous Bottom Simulating Reflectors (BSRs) and pluming 22 BSRs. We also show that the lateral tips of eight sills, which at times appear to terminate against supra-sill faults, underlie gas chimneys linked to gas hydrate and 23 free gas in shallower strata. These supra-sill faults and gas chimneys, as well as 24 25 supra-volcano faults, provided pathways for upward fluid (gas) migration from deep sources. Given the spatial relationship between volcanoes, sills, free gas, and gas 26

hydrates, we suggest that the igneous structures focused fluid migration into overlying
faults and gas chimneys. We also explore whether higher geothermal gradient at the
summits of the volcanoes caused the base of gas hydrate stability zone to be shallower,
forming a pluming BSR. Our study highlights how volcanic systems can influence the
formation and distribution of later gas hydrate and free gas.

Keywords: Gas hydrate; Free gas; Magmatism; Fluid migration; Pearl River Mouth
Basin

34

35 1. Introduction

Gas hydrates are crystalline solids that can form in low-temperature and 36 high-pressure conditions, and are found often within sedimentary basins along 37 38 continental margins, in permafrost, and potentially under ice sheets (Shipley et al., 1979; Sloan and Koh, 2007; Hornbach et al., 2008). The generation of highly 39 saturated gas hydrate is controlled by the availability of hydrocarbon gas released 40 41 from a source and the presence of migration pathways that enable gas to flow to a suitable reservoir where it can become trapped (e.g., Collett et al., 2009). However, as 42 43 the temperature and/or pressure of such reservoirs changes, any present gas hydrates may destabilize and disaggregate, causing gas to migrate into surrounding strata (e.g., 44 Hornbach et al., 2008; Collett et al., 2009). The evolution of hydrate-bearing reservoir 45 temperature and/or pressure conditions may thus affect gas energy supply, seafloor 46 failure, ocean acidification, and the global redistribution of carbon (e.g., Biastoch et 47 al., 2011; Rupple and Kesler, 2017; Wu et al., 2022). 48

49	In addition to containing gas hydrates, many sedimentary basins across the
50	continental margins contain evidence of significant magmatic events, some of which
51	were associated with the release of substantial volumes of hydrocarbon and carbon
52	dioxide-rich hydrothermal fluids (e.g., Planke et al., 2005; Jamtveit et al., 2004;
53	Svensen et al., 2006; Grove, 2013; Omosanya et al., 2018; Phillips and Magee, 2020).
54	Such magmatism can also drive host rock deformation (e.g., compaction and uplift)
55	and locally elevate temperatures (e.g., Schutter, 2003; Jamtveit et al., 2004; Sun et al.,
56	2020b, 2022a). Magmatism can thus influence the structural, thermal, and
57	geodynamic evolution of sedimentary basins and petroleum systems (e.g., Schutter,
58	2003; Planke et al., 2005; Infante-Paez & Marfurt, 2017; Sun et al., 2020b, 2022a),
59	and could therefore impact concurrent or later gas hydrate evolution. For example, in
60	the Joetsu basin, Japan Sea, massive gas hydrates have been formed by a mixture
61	gases related to magmatic-mantle fluids that migrated upwards through gas chimneys
62	(Snyder et al., 2020). Gas hydrate and fluid migration pathways forming from carbon
63	dioxide-rich fluids in the mid-Okinawa trough, Japan, have also been related to a
64	magmatic source (Sakai et al., 1990; Hou et al., 2005).

In this study, we use reprocessed and newly acquired three dimensional (3D) seismic reflection data to explore potential links between sills, volcanoes, and gas hydrates. In particular, we examine how normal faults and gas chimneys connect gas-related enhanced reflections and BSRs to buried volcanoes and sub-volcanic sills in parts of the Pearl River Mouth Basin, South China Sea. The Pearl River Mouth Basin is a key exploration area for oil, gas, and gas hydrate in the South China Sea (e.g.,

Zhang et al., 2007; Pang et al., 2008; Yang et al., 2015, 2017). Gas hydrates and 71 accompanying free gas have been confirmed in the area by logging while drilling 72 73 (LWD) during the drilling expeditions GMGS1, 3, 4, and 5 (e.g., Zhang et al., 2007; Yang et al., 2015, 2017a). There is also evidence for intense, ancient magmatic 74 activity within the Pearl River Mouth Basin (e.g., Zhao et al., 2016, 2019; Kong et al., 75 2018), which appears to have influenced fluid migration pathways (Sun et al., 2019, 76 2020b, c, d). We explore the age of magmatic activity in our study area and how 77 78 volcanic systems potentially influence the formation of fluid migration pathways.

79

80 **2. Geological Setting**

81 *2.1 Regional geology*

The South China Sea developed through multiple phases of continental rifting 82 between 75-40 Ma (Tg-T80) and 40-32 Ma (T80-T70), and records a rift-to-drift 83 transition (Hayes and Nissen, 2005; Ding et al., 2018, 2020). Rifting was controlled 84 by subduction of the Palaeo-Pacific Plate below the Eurasian Plate (Deng et al., 2019; 85 Ding et al., 2019, 2020; Wang et al., 2021). The Pearl River Mouth Basin is located on 86 the northern slope of the South China Sea (Fig. 1a) and its formation involved a 87 syn-rift stage in the Eocene (~75 Ma to 32 Ma), a phase of continental breakup in the 88 Oligocene (32 Ma to 23.8 Ma), and a final post-rift subsidence stage since the 89 Miocene (23.8 Ma to recent) (Fig. 2; e.g., Clift and Lin, 2001; Pang et al., 2008). 90 Three main tectonic events have since affected the evolution of the Pearl Mouth River 91 92 Basin, including the Zhuqiong Event (~23.8 Ma), the Baiyun Event (23.8–10.5 Ma), and the Dongsha Event (beginning at 10.5 Ma, and peaking at 5.5 Ma) (Lüdmann and 93

Wong, 1999; Zhao et al., 2016, 2019; Sun et al., 2020c, d). Magmatic activity in the
Pearl River Mouth Basin mainly occurred in two stages at 64-32 Ma and 23.8-15.5
Ma (e.g., Yan et al., 2006; Li et al., 2014; Zhao et al., 2016, 2019; Sun et al., 2019,
2020b, c, d; Zeng et al., 2019; Zhang et al., 2021).

The study area is specifically situated in the Baiyun Sag, which comprises numerous submarine canyons on the continental slope, and the Yunli Low Uplift (Fig. 1). The Yunli Low Uplift separates the Baiyun Sag from the Liwan Sag, and hosts a volcano exposed on the present day seafloor (Fig. 1b, the white dashed line). Deep-water gas-fields (e.g., LW3-1-1), as well as a gas hydrate occurrence zone, occur within the Baiyan Sag (Fig. 1).

104 *2.2 Stratigraphy and petroleum system*

105 Continental, lacustrine facies mudstones and coals in the Late Cretaceous-Early Oligocene Wenchang and Enping formations represent the regional hydrocarbon 106 source rocks in the Baiyun Sag, supplying sufficient thermogenic gas for gas fields 107 and free gas accumulations (Fig. 2; e.g., Pang et al., 2008; Zhu et al., 2009). Marginal 108 marine sandstones were deposited during the Late Oligocene-Early Miocene (Zhuhai 109 and Zhujiang formations), which form the most important hydrocarbon reservoirs and 110 carrier beds in the Baiyun Sag (Fig. 2; Zhu et al., 2009). Above these, deep-water 111 depositional systems of the Hanjiang, Yuehai and Wanshan formations developed, as 112 well as bathyal-abyssal mudstones and silty mudstones deposited in the submarine 113 canyon-ridge system on the continental slope of Pearl River Mouth Basin (e.g., Zhou 114 et al., 2015). The Dongsha event (10.5 Ma to present) induced limited regional uplift, 115

erosion, and faulting within the Pearl River Mouth Basin (e.g., Su et al., 1989; Zeng et
al., 2019), inducing the release of overpressured fluid in the Baiyun Sag (Kong et al.,
2018); it has been suggested that this overpressure release provided the hydrocarbon
gas required for development of the concentrated gas hydrates found in the area
(Wang et al., 2011; Sun et al., 2020a).

121 2.3 Magmatic complexes (VCI, VCII and VCIII) in the Baiyun Sag

Recent studies have shown post-rift magmatism was widely distributed in the 122 northern South China Sea (e.g., Zhao et al., 2016, 2019; Song et al., 2017; Yang et al., 123 124 2017b; Wang et al., 2019b; Zeng et al., 2019; Sun et al., 2020b, c, 2022a; Zhang et al., 2021), producing magmatic complexes in the west (VCI) and centre (VCII) of the 125 Baiyun Sag (Fig. 3). Drilling of the VCI and VCII reveals the volcanoes comprise 126 127 hydrothermally altered basalt lavas and tuffs, intercalated with thin-bedded limestone and clastic beds (Li and Rao, 1994; Zhao et al., 2016; Sun et al., 2020c). Igneous 128 activity at VCI occurred between the start of the Miocene (23.8 Ma) and the end of 129 130 the Early Miocene (17.6 Ma), as suggested by the K-Ar dating and seismic-stratigraphic relationships (Qin et al., 2000; Sun et al., 2014; Zhao et al., 131 2016). Many submarine volcanic mounds, associated lava flows, and underlying 132 igneous sills are associated with VCI; they can be identified by their high amplitude 133 and positive polarity reflections in seismic data (Sun et al., 2014; Zhao et al., 2016, 134 2019; Ma et al., 2018). Shallow gas accumulations with negative-polarity reflections 135 are commonly located directly above volcanoes and linked to them by normal faults 136 (Sun et al., 2014; Zhao et al., 2016; Yang et al., 2017b). 137

In this study, we focus on the VCIII magmatic complex, which spans the southeastern Baiyun Sag (referred to as the Shenhu area) and the Yunli Low Uplift (Figs. 3 and 4). Previous studies have recognized from seismic reflection data that a large seamount dominates the center of VCIII (Fig. 1b, the white broken line; Zhou et al., 2018; Zhao et al., 2019; Sun et al., 2022b), but its architecture and relationship with the hydrocarbon and gas hydrate system is poorly known.

144 *2.4 Geological controls on gas hydrate occurrence*

In the Shenhu gas hydrate-drilling area, high-amplitude reflections just above 145 146 bottom-simulating reflectors (BSRs) have been interpreted as accumulations of gas hydrate with elevated saturations (e.g., Yang et al., 2015, 2017a; Wang et al., 2016, 147 2021). These BSRs primarily occur along the crest of four ridges (Wang et al., 2014, 148 149 2016; Yang et al., 2015, 2017a; Zhang et al., 2017, 2020; Qian et al., 2018; Jin et al., 2020a). High concentrations of gas hydrate accompanying free gas and structure II 150 gas hydrate (i.e. composed of heavy-hydrocarbon gases) have been confirmed at 151 152 borehole sites W18, W19, W11 and W17 (Fig. 1A) (e.g., Yang et al., 2015, 2017a; Qian et al., 2018; Wei et al., 2018); quantitative modelling indicates that gas hydrates 153 formed ~29,000-19,000 years ago at Sites W18 and W19 (Fang et al., 2019; Jin et al., 154 2020a). The gas compositions in the core samples at Sites W18, W19 and W17 are 155 mainly composed of methane (C1), heavy hydrocarbon gases (C2), and a small 156 amount of carbon dioxide (CO₂, 100-300 ppm) (Zhang et al., 2007; Yang et al., 2015, 157 2017a; Qian et al., 2018). Double BSRs or II-BGHSZ (base of gas hydrate stability 158 zone) have been found due to the influence of thermogenic gas and complex 159

sedimentation and erosion of the migrating canyons (e.g., Qian et al., 2018; Zhang et 160 al., 2020a). Overall, localized high-temperature and mixed hydrocarbon gases 161 (thermogenic and biogenic gases) are likely responsible for the occurrence of the 162 recent active hydrate system and structure II gas hydrate in the Baiyun Sag (Jin et al., 163 2020a; Sun et al., 2020a). Normal faults and gas chimneys appear to have provided 164 the pathways for fluid migration from deeper sedimentary thermogenic and biogenic 165 gas sources (e.g., Wu et al., 2009; Wang et al., 2011; Zhang et al., 2017, 2020a, b; 166 Cheng et al., 2020; Jin et al., 2020a). 167

168

169 **3. Data and Methods**

170 *3.1 Seismic data and interpretations*

We use high-resolution, 3D seismic reflection data of the Yunli Low Uplift that 171 were acquired and processed by the China National Offshore Oil Corporation 172 (CNOOC) in 2018. The 3D seismic data in the study area covers \sim 1300 km² (Fig. 1). 173 Acquisition involved 390 channel streamers and 6 cables >4950 m in length. The bin 174 spacings are 12.5 m and 25 m in the in-line and cross-line directions, respectively, and 175 the sampling interval is 2 ms. The dominant frequency gradually decreases downward 176 from the seafloor (~50 Hz) to where the igneous features of interest are located (~35 177 Hz). Using P-wave velocities of 1.5 km/s to 2.7 km/s for the interval of interest strata 178 and 4.5 km/s for the volcanic complexes, determined from the Site BY7-1-1 borehole 179 by Sun et al., (2020d), we calculate that the vertical seismic resolution ($\lambda/4$) decreases 180 181 from \sim 7.5 m at the seafloor to \sim 20 m in the sedimentary strata hosting the volcanic complexes. Seismic attributes, such as variance, maximum and minimum amplitudes, 182

and root mean square (RMS) were extracted from the 3D seismic data to help reveal
discontinuous structures and the distributions of gas hydrate and free gas in the
Baiyun Sag and Yunli Low Uplift.

We mapped horizons corresponding to prominent, positive polarity reflections 186 marking the top of volcanoes (TM). In addition to a positive polarity, we defined sills 187 based on their high-amplitude reflections and lateral discontinuity (Planke et al., 188 2005). Eleven regional strata sequence boundaries were mapped and tied to the 189 nomenclature proposed by CNOOC (Figs. 2 and 3), specifically: Seafloor, T20 (2.49 190 191 Ma), T30 (5.5 Ma), T32 (10.5 Ma), T35 (13.5 Ma), T40 (15.5 Ma), T50 (17.5 Ma), T60 (23.8 Ma), T70 (32.0 Ma), T80 (40 Ma), Tg (65 Ma) (e.g., Pang et al., 2008; 192 Dong et al., 2009; Ma et al., 2018). The ages of strata reflections onlapped onto 193 194 volcanoes were used to constrain the relative time of igneous activity (Trude et al., 2003; Planke et al., 2005, 2017). 195

196 *3.2 BSR classifications*

197 In the seismic data, bottom simulating reflectors (BSRs) are interpreted to be the result of a negative impedance caused by the transition from gas hydrate-bearing and 198 water-bearing sediments above to free gas-bearing sediments below, and 199 approximately represent the base of gas hydrate stability zone (BGHSZ) (e.g., Shedd 200 et al., 2012; Wang et al., 2014). Three types of BSR can be identified in the seismic 201 profiles, namely continuous BSRs, discontinuous BSRs and pluming BSRs. The 202 continuous and discontinuous BSRs have been identified in previous studies as they 203 cross-cut strata, are marked by a polarity reversal, and are subparallel the seafloor 204

(Wang et al., 2016; Zhang et al., 2020a). We refer to pluming BSRs as areas where a
BSR is locally elevated and shows an approximate convex toward the seafloor shape,
perhaps indicative of a shallower BGHSZ related to the presence of hot fluids
(Holbrook et al., 1996; Shedd et al., 2012; Daigle and Dugan, 2014; Le et al., 2015).

High amplitude and positive polarity reflections above the BSR(s) have been interpreted as gas hydrate in seismic profiles in the area (Wang et al., 2016). Similarly, polarity reversals and enhanced reflections associated with underlying velocity pull-down reflections below the BSR(s) have been interpreted as free gas and gas migration pathways (e.g., Collett et al., 2009; Boswell et al., 2012; Yoo et al., 2013).

214 *3.3 Calculating the base of gas hydrate stability zone (BGHSZ)*

The BGHSZ marks the bottom boundary where gas hydrate is stable and its 215 216 position is controlled by the temperature, pressure conditions, pore water salinity, and gas compositions of the near-seabed sediments (Xu and Ruppel, 1999; Sloan and 217 Koh, 2007). In general, gas hydrate occurs above this basal boundary and 218 219 hydrocarbon gas (mainly methane) is trapped below the BGHSZ. Mostly, the depth of BGHSZ corresponds to the BSR found from the seismic data in the Pearl River 220 221 Mouth Basin (e.g., Wang et al., 2014; Qian et al., 2018; Jin et al., 2020a). Due to a lack of well log data in the Yunli Low Uplift, we simulated the BGHSZ to help 222 identify the boundary between gas hydrate and free gas. Specifically, parameters of 223 inferred seafloor temperature, geothermal gradient from Sites SH2, SH3 and SH5 224 (Zhang et al., 2007) were used to calculate the likely depth of the BGHSZ for 225 methane using the CSMHYD program (Sloan and Koh, 2007). The modelled depths 226

of the BGHSZ were changed into time domain using an average velocity for the 227 near-seabed sediment of is 1650 m/s, which was obtained from Site W25 (Zhou et al., 228 229 2022). We added the modelled and time-converted BGHSZ as a horizon within the seismic reflection data and compared its location to the interpreted BSRs and 230 pluming BSRs. Based on the modelled location of the BGHSZ, we extracted 231 coincident maximum, average positive, and minimum amplitude attributes from the 232 seismic reflection data to show the distributions of gas hydrate and free gas. 233 Mmaximum and average positive amplitude attributes are widely used to display the 234 235 distribution of hydrates as they highlight positive waveforms related to gas hydrate above the BGHSZ (e.g. Wang et al., 2016; Portnov et al., 2021). In contrast, the 236 minimum amplitude attribute extracted below the BGHSZ shows negative waveform 237 238 information indicating the distribution of free gas (e.g. Wang et al., 2016).

239

240 4. **Results**

241 *4.1. Gas accumulations*

Seismically anomalous responses of free gas characterized by low frequency, negative polarity, laterally restricted, and enhanced reflections have been widely identified in the study area. In the gas hydrate-drilling area of Baiyun Sag, the minimum amplitude attribute extracted along the modelled BGHSZ within the four submarine canyons of the Baiyun Sag shows that enhanced reflection zones along both the ridges and the troughs (Fig. 5a, b). The enhanced reflections occur as either isolated seismic features distributed along a discrete stratigraphic level, or are marked by stacked reflection pairs that span several stratigraphic layers below the BGHSZ
(Figs. 6 and 7). Gas trapped below the BSRs (or BGHSZ) but above T35 (15.5 Ma) is
underlain by faults and/or 'chimneys' where background seismic reflections are
locally dim and disturbed (Figs. 6a, b and 7).

253 In the Yunli Low Uplift, south of the gas hydrate-drilling zone, the distributions of enhanced reflections occur broadly in three zones to the north of the VCIII central 254 volcanic edifice (Fig. 8a). These enhanced reflections are recognized above the 255 summits of three buried volcanoes (M1, M2 and M3; see section 4.3) at three different 256 257 stratigraphic levels (Fig. 9). The deepest enhanced reflection occurs above M1, ~600–650 ms two-way travel time (TWT) below the seafloor and coincides with T32 258 (10.5 Ma) (Fig. 9a, e). The second deepest layer broadly occurs along T30 (5.5 Ma) 259 260 above M2 and can reach up to the BGHSZ where it develops into a discontinuous or pluming BSR, ~250-330 ms TWT below the seafloor (Fig. 9b, e). The shallowest 261 enhanced reflection occurs above M3, ~180-200 ms TWT below the seafloor and just 262 263 beneath T20 (2.49 Ma), and is locally marked by a pluming BSR (Fig. 9c, d, e).

264 *4.2. Gas hydrate occurrence*

4.2.1 Gas hydrate in the Baiyun sag

The modelled BGHSZ, which partly coincides with the BSRs, occurs ~250–300 ms TWT below the seafloor on the seismic sections (Figs. 6 and 7). We extract the maximum amplitude and average positive amplitude attribute along the BGHSZ with a time window that extends 50 ms TWT upward to show the distribution of gas hydrate-bearing layers (Fig. 5c, d). In plan-view, there is a NW-SE linear but patchy distribution of high amplitude with positive polarity reflectivity along the four crests of submarine ridges in the Baiyun Sag (Fig. 5c, d). The continuous BSRs occur at Sites SH2 and W17 (Figs. 6a, b, c and 7), while a discontinuous BSR is developed at Site W18 (Fig. 6d and 7a). In places, instances of single or multiple high amplitude reflections occur above the BSR (Figs. 6 and 7); these have been confirmed as corresponding to gas hydrates by drilling (e.g., Zhang et al., 2007; Yang et al., 2015, 2017a).

4.2.2 Gas hydrate in the Yunli Low Uplift

279 In the Yunli Low Uplift, potential gas hydrate-bearing layers are revealed by the maximum amplitude attribute map and occur along a broad zone that terminate north 280 of the VCIII (Fig. 8b). Discontinuous BSRs and pluming BSRs are observed in 281 282 shallow sediments above the M2 and M3 volcanoes (Fig. 9e-f). The discontinuous BSR is developed in shallow sediments (325 ms TWT below the seafloor) to the north 283 of M2, above three or four small volcanoes (Fig. 9b). Similar discontinuous, 284 285 moderate-high amplitude, layer reflections occur in this area above the BSR, perhaps indicating the potential local occurrence of gas hydrate-bearing sediments (Fig. 9b). 286 Pluming BSRs occur above the summits of the M2 and M3 volcanoes; these Pluming 287 comprise high-amplitude, convex-upwards reflections that cross-cut 288 BSRs background stratigraphic reflections and are shallower than the modelled BGHSZ (Fig. 289 9b, c, the blue broken line). The pluming BSR is shallower above M3 (189 ms TWT 290 below seafloor) than that above M2 (245 ms TWT below seafloor). Localized high 291 amplitude reflections above the pluming BSRs potentially indicate the occurrence of 292

293 gas hydrates (Fig. 9d).

294 *4.3. Volcanoes*

295 All identified volcanoes have a common top surface (TM), which is typically characterized by a high-amplitude, positive polarity reflection (Fig. 9). We recognize 296 a large central volcanic edifice (VCIII) in the Yunli Low Uplift, which has a ~15 km 297 diameter, is ~1100 ms TWT high (~2640 m assuming an interval velocity of 4.8 km/s), 298 breaches the seafloor, and is surrounded by a series of buried, smaller volcanoes with 299 diameters <1 km (Fig. 4). Of these smaller volcanoes, we focus on those we name M1, 300 301 M2 and M3 as they appear to underlie free gas, gas hydrate, and BSR features in shallower strata (Fig. 9). M1 has a diameter of ~2 km, a height of ~250 ms TWT 302 (~290 m), and its summit is situated ~900 ms TWT (~1000 m) below the seafloor (Fig. 303 304 9a). M2 comprises two summits situated ~700 ms TWT (~800 m) below the seafloor, and has a diameter of ~2.5 km and a height of ~400 ms TWT (~460 m) (Fig. 9b). M3 305 is closest (~4 km) to the large central volcano, has a summit ~500 ms (~570 m) below 306 307 the seafloor, a diameter of ~2 km, and a height of ~270 ms (~310 m) (Fig. 9c). The oldest horizon that onlaps onto the volcanoes is T40 (15.5 Ma) (Fig. 9a, b). Above the 308 volcanoes in places, strata display minor convex-upward folds and associated normal 309 faults that offset the fold crests (Fig. 9). 310

311 *4.4. Sills*

Positive polarity, laterally discontinuous, and moderate-to-high amplitude reflections that are either concordant with or cross-cut surrounding reflections are observed between T80 and T50 (Fig. 6b). We interpret these as igneous sills, and eight

sills were found in this area, named S1-8. The geometries of three types of sills are 315 recognized: sub-horizontal, inclined, and saucer-shaped (Fig. 6e-h). Eight sills were 316 317 mapped, mainly located within the Baiyun Sag in the northern margin away from the large central volcano. These sills have widths ranging from 0.7-8 km and lengths of 318 2-13 km (Figs. 6 and 7). Many normal faults appear to intersect the edges of sills in 319 the deeper strata, and connect upwards to the shallow free gas and gas hydrate-bearing 320 sedimentary rocks (Figs. 6 and 7c). Sills recognized within the volcanoes are poorly 321 imaged and difficult to map clearly. 322

323 *4.5. Faults and gas chimneys*

Normal faults and gas chimneys are widely developed. A variance slice extracted 324 near T32 shows a few E-W striking tectonic faults, which occur between T20 (2.49 325 326 Ma) and Tg (Figs. 3 and 10). Normal faults also occur above the sills across the gas hydrate-drilling area and these strike NW-SE (Fig. 10); these supra-sill faults extend 327 down deepest from T80 (40 Ma) and commonly intersect lateral sill tips, but poor 328 imaging often hinders their recognition at deeper levels (Figs. 6 and 7). Supra-sill 329 faults are usually ~1000-3000 ms TWT and 6-8 km in length (Figs. 6b, 7c and 10); 330 these are taller than those faults found above volcanoes, which are developed between 331 T40 (16.5 Ma) and T20 (2.6 Ma), and thus ~500 ms TWT (<600 m) high (Fig. 9a, b, 332 c). Supra-volcano faults have lengths of <3 km in the Yunli Low Uplift (Fig. 10), and 333 are often radially distributed (Fig. 11a-c). 334

Features interpreted as gas chimneys are characterized by acoustic blanking, chaotic reflections, and/or velocity pull-downs and are the most common in the gas hydrate drilling area (e.g., Wu et al., 2009; Sun et al., 2012; Wang et al., 2014; Cheng
et al., 2021). These chimneys occur above the sills and volcanoes and are 200-3500
ms TWT (230-4100 m) high (Figs. 6, 7 and 9). For example, in the Yunli Low Uplift,
gas chimneys occur within folded strata above volcano summits (Fig. 9). Variance
slices show gas chimneys have high variance values and, if isolated, are sub-circular
in plan-view at horizon T32 (Fig. 10). Most of the gas chimneys are linearly
distributed along the crest of four submarine canyons (Fig. 10).

344 *4.6. Summary of spatial correlation between gas accumulations and igneous features*

345 We overlaid maps to show the relationships between the various geological elements we identify and the occurrences of free gas and potential gas hydrate (Fig. 346 12). The supra-sill faults are developed within the potential gas hydrate and free gas 347 348 zones of the Baiyun Sag and appear to connect the sills with shallow enhanced-reflections and high-amplitude layers (Figs. 6 and 7). For example, the 349 continuous BSR appear connected to sill S8, which occurs at ~6.5 s TWT (~7.5 km), 350 351 by three normal faults at Site SH2 (Fig. 6a, b). Similarly, normal faults appear to connect multiple layers of enhanced reflections to sills S5 and S7 near Sites W11 and 352 W17 (Fig. 7c). In the Yunli Low Uplift, free gas- or potential hydrate-bearing layers 353 occur above the summit of buried volcanoes M1, M2, and M3 (Figs. 10 and 12). 354 There is no obvious gas hydrate distribution at the top of M1, as the free gas-bearing 355 layer is much deeper than the modelled BGHSZ (Figs. 9a, e and 12a). Both 356 supra-volcanic faults and gas chimneys occur above the summit of M2 and M3, and 357 appear to connect the volcanoes to the shallower potential gas hydrate- and free 358

359 gas-bearing sediments (Figs. 9, 10, and 12).

360

361 5. Discussion

362 5.1 Age of magmatism and associated structures

The central volcanic edifice (VCIII) and surrounding volcanoes (e.g., M1-M3) 363 occur at the same stratigraphic level and are onlapped by T40, implying they extruded 364 just before ~15.5 Ma. The age of VCIII is thus similar to VCI and VCII in the Baiyun 365 Sag. In the west of the Baiyun Sag, VCI is 17.1±2.5Ma (Early Miocene), and the 366 oldest volcanic material intersected by BY7-1-1 yields a K-Ar date of 35.5±2.78 Ma 367 (Middle Eocene) (Qin, 2000). In the centre of the Baiyun Sag, seismic-stratigraphic 368 onlap relationships suggest the volcanic plumbing system of VCII was mainly 369 370 emplaced after 17.5 Ma, with activity ceasing before 15.5 Ma (Zhao et al., 2016, 2019). 371

There is no borehole data from the sills and no clear seismic-stratigraphic relationships (e.g., onlap onto intrusion-induced forced folds) that we can use to constrain their age of emplacement (e.g., Trude et al., 2003). However, the sills occur within strata between ~T80-T50, suggesting some if not all were therefore emplaced after 23.8 Ma (Figs. 6 and 10). Due to their close spatial association with the volcanoes, we suggest that the sills were likely emplaced at the same time as and fed volcanism (Sun et al., 2022b).

379 Sedimentary layers directly above the volcanoes (such as M1) show a four-way 380 dip closures, i.e. dome-shaped folds (Fig. 9a, c). These folds occur in strata deposited

long-after after the volcanoes were formed (~15.5 Ma) and are thus not related to 381 magmatism; e.g., T32 (10.5 Ma) is often folded within these structures, indicating the 382 383 folds formed over ~5 Myrs after volcanism. The shallowest folded strata observed is probably the horizon T30 (i.e. 5.5 Ma) (Fig. 9b). We suggest fold formation was likely 384 due to differential compaction, whereby the igneous material comprising the 385 volcanoes compacted less than the adjacent sedimentary column meaning strata 386 directly above the volcanoes does not subside as much as elsewhere (e.g., Phillips and 387 Magee, 2020; Wang et al., 2022). Associated with this differential compaction folding 388 389 was the development of crestal normal faults and probably micro-fractures in response to outer-arc stretching (e.g., Cosgrove and Hillier, 1999). 390

391 5.2 Gas migration and gas hydrate accumulation influenced by magmatic activity

392 Fluid migration from deeply buried sedimentary rocks along the faults, gas chimneys, and permeable inclined strata have been widely reported in many basins to 393 influence gas hydrate accumulation and distribution (e.g., Kinoshita et al., 2011; 394 395 Boswell et al., 2012; Collett et al., 2019; Saito et al., 2019; Cheng et al., 2020; Portnov et al., 2020, 2021). In the Gulf of Mexico, for example, the location of gas 396 hydrate reservoirs is often governed by salt diapirism, which causes overlying 397 anticlinal structures favorable for accumulation of gas and gas hydrate, and an 398 associated fault array that provides potential fluid flow pathways (Boswell et al., 2012; 399 Portnov et al., 2020). Several studies have also found that intrusive and extrusive 400 magmatic features can focus syn- and post-emplacement fluid migration in 401 sedimentary basins, such as offshore southeastern Australia (Holford et al., 2017), in 402

the central Atlantic magmatic province (Capriolo et al., 2021), and in the Møre basin 403 of the northeast Atlantic continental (Svensen et al., 2004). In these works, it has been 404 405 argued that emplacement and cooling fracture networks in igneous intrusions improve their permeability, such that when sills are interconnected (i.e. a sill-complex) they 406 can provide extensive migration pathways (Rateau et al., 2013). Similarly, within 407 volcanoes, the complex intercalation of lavas and volcaniclastics, which can have 408 high porosities and permeabilities, and presence of igneous intrusions can facilitate 409 fluid flow (Holford et al., 2017). However, as of yet there have been few studies 410 411 examining the structural control magmatic systems play on gas hydrate occurrence.

In the Baiyun Sag, normal faults and gas chimneys extending upwards from 412 different stratigraphic levels seem to provide important pathways for fluid migration, 413 414 which has led to the formation of extensive free gas and gas hydrate (Figs. 6 and 7, e.g., Wu et al., 2009; Wang et al., 2011; Zhang et al., 2017; Cheng et al., 2020; Jin et 415 al., 2020a). We show that some of the free gas accumulations are located directly 416 417 above and vertically connected to volcanoes and sills by faults and gas chimneys, and are trapped in differential compaction folds above the volcanic complex (Fig. 9, Zhao 418 et al., 2016; Ma et al., 2018; Sun et al., 2020b). These seismic-stratigraphic 419 relationships indicate fluid migration and gas accumulation occurred long-after 420 magmatism ceased, but that the intrusion network and volcanoes were porous and/or 421 permeable enough that fluids exploited them as pathways. Upon reaching the lateral 422 tips of sills or summits of volcanoes, these fluids then migrated along supra-sill and 423 supra-volcano faults to shallower reservoirs and traps. 424

425 5.3 The thermodynamic influences on the BGHSZ

The pluming BSRs, characterized by localized, bright spots, discontinuous 426 427 reflections (Fig. 9b, c), may be caused by locally elevated geothermal gradient compared to the surrounding area (Shedd et al., 2012), or diverse gas composition 428 effects due to salt-driven evolution on gas hydrate system (Portnov et al., 2021). We 429 just calculate the changes of BGHSZ by the influence of temperature due to lack of 430 gas compositions in this area. By comparing the modelled BGHSZ to the observed 431 BSR locations, the thermodynamic impact of volcanoes on the gas hydrate occurrence 432 433 can be evaluated. We calculated the depth of the BGHSZ assuming the gas was pure methane, but used various geothermal gradients and seafloor temperatures to interpret 434 the anomalous changes at three sites above M2 and M3 (Fig. 13a, b, c, locations 1-3 435 436 shown in Fig. 9b and c). Location 1 occurs above the volcanic basement, but not above a volcano, and is marked by a discontinuous BSR reflection at 325 ms TWT, 437 which corresponds to a depth of 259 m using the P-wave velocity of 1650m/s at Site 438 W25. Here, the modelled depth of the BGHSZ, comprising 100% methane, is 274 m 439 using a seafloor temperature of 5.53 °C and a geothermal gradient of 49.31 °C/km 440 (based on data from Site SH3), as well as a water depth of 1766 m (Fig. 13a); this 441 makes the BGHSZ slightly deeper than the observed BSR depth. Both of Location 2 442 and Location 3 show pluming BSRs above M2 and M3 at depths of 245 ms and 189 443 ms TWT, which broadly equate to depths of 195 m and 159 m, respectively (Table 1). 444 The modelled depths of the BGHSZ at locations 2 & 3 are 208 m and 204 m, given a 445 high geothermal gradient of 67.60 °C/km and a seafloor temperature of 4.72 °C 446

(based on data from Site SH5), as well as water depths of 1830 m and 1789 m (Fig. 447 13b, c and Table 1). The pluming BSRs above M2 and M3 occur at shallower levels 448 449 than the BGHSZ modelled using the geothermal gradient measured at Site SH5. The calculated BGHSZ for thermogenic gas mixing will be deeper than that for pure 450 methane, according to the hydrate phase equilibrium curve (Sloan and Koh, 2007). 451 There may be an interplay between temperature and gas composition effects. For 452 example, an increase in temperature would drive BSR formation upwards, whereas a 453 gas mix containing heavier hydrocarbons would drive BSR formation downwards. 454 455 Similar to seafloor cold seeps that occur above the southern summit of the Hydrate Ridge, Cascadia subduction zone (e.g., Torres et al., 2004), the abnormally shallow 456 BSRs we observe may be related to locally active hot fluids. Such anomalously 457 458 shallow BSRs can also be formed due to overheating of seafloor sediments and the geothermal field not reaching equilibrium due to seafloor erosion (Bang et al., 2005; 459 Jin et al., 2020b). However, we observe no evidence of seafloor erosion above or near 460 461 the buried volcanoes (Fig. 9). Moreover, the spatial overlap between the pluming BSRs, free gas, and volcanoes (Fig. 12) suggests that the pluming BSRs could be 462 influenced by upward flow of warm/hot fluids from the buried volcanoes. Salt bodies 463 in the Gulf of Mexico have similarly been shown to focus warm/hot fluid flow, locally 464 elevating the geothermal gradient (e.g., Portnov et al., 2020). 465

466

Table 1 Comparison of calculated BGHSZ and identified BSR depths at threelocations.

BGHSZ	Water	Geothermal	Seafloor	Observed BSR	Observed	Calculated
	depth (m)	gradients (°C	temperature	in TWT (ms)	BSR in depths	BGHSZ (m)
		/km)	(°C)		(m)	
Location 1	1766	49.31	5.53	325	259	274
Location 2	1830	67.60	4.72	245	195	208
Location 3	1789	67.60	4.72	189	159	204

469

470 *5.4 Controlling factors on the free gas and gas hydrate accumulation*

471 Gas hydrate drilling expeditions indicate that the migration of deep thermogenic and biogenic gas from the Wenchang and Enping formations (i.e. below T70) and 472 shallow formations (i.e. T40 to present) in the Baiyan Sag occurred between 14-6.5 473 Ma and 5-0 Ma, after igneous activity at ~17.5-15.5 Ma (e.g., Yang et al., 2015, 2017a; 474 Zhao et al., 2016; Qian et al., 2018; Su et al., 2018; Sun et al., 2020a, c, d; Zhang et 475 476 al., 2020b). A small amount of CO₂ (100-300 ppm) was found in the gas hydrate-bearing samples at Sites W18, W19 and W17 (Zhang et al., 2007, 2020a), 477 478 which may be related with volcanic-sourced fluids, similar to those found in the Japan 479 Sea (Snyder et al., 2020). Alternatively, the CO₂ may be sourced from microbial degradation of petroleum (i.e. organic matter) (e.g., Rajbongshi and Gogoi, 2020). 480 Regardless, both overpressured and hydrate occurrence simulations show that heavy 481 482 hydrocarbon gases, methane and CO₂ require migration pathways to transport fluids from deeper to shallower strata (Kong et al., 2018; Sun et al., 2020a). 483

In the Baiyun Sag, although the sedimentary succession is thick, igneous sills are widely distributed and their lateral terminations often coincide with normal faults or gas chimneys (Figs. 6, 7 and 9). We suggest that the interconnected network of sills, and perhaps dykes not visible in the seismic reflection data, enabled gas migration

from deep sedimentary rocks to the shallow strata (see Rateau et al., 2013). In the 488 Yunli Low Uplift, gas-rich strata are found within the differential compaction folds 489 490 above volcanoes (e.g., M1, M2 and M3), which thus act as traps (Fig. 9). We suggest that faulting and fracturing generated by differential compaction folding provided gas 491 492 flow pathways, aiding hydrocarbon migration that was already focused into and through the volcanoes (see Holford et al., 2017). Overall, we show free gas migrated 493 through sills, volcanoes, normal faults and chimneys and accumulated: (i) in folded 494 sedimentary strata below the BGHSZ (i.e. first enhanced reflections, in Figs. 9a and 495 496 14); or (ii) where only thin stratal packages overlie the buried volcanoes, the gas was able to migrate above the BGHSZ and form hydrates (Figs. 9b, c and 14). The paleo 497 channels/turbidites of the migrating canyons also provide coarse-grained sediments at 498 499 the BGHSZ, which provide favorable reservoirs for gas hydrate accumulations (Jin et al., 2020a; Zhang et al., 2020b). High geothermal gradients developed locally in the 500 vicinity of the volcanoes, caused the BGHSZ to be locally shallower, thus allowing 501 pluming BSRs to develop (Fig. 14). Overall, we present evidence suggesting that 502 volcanic systems may influence gas hydrate and free gas occurrence and 503 accumulation in two aspects: (i) promoting fluid migration pathways; (ii) temperature 504 anomalies. 505

506

507 6. Conclusions

Newly acquired three-dimensional (3D) seismic reflection data from the Baiyun
Sag and Yunli Low Uplift reveal that volcanic systems influence the occurrence and

accumulation of gas hydrate and free gas in the Pearl River Mouth Basin, South China 510 Sea. The buried volcanic complexes and igneous sills were formed between 23.8-15.5 511 512 Ma, followed by the development of fluid migration pathways consisting of overlying faults and gas chimneys, partly in response to differential compaction. The 513 accumulation zones of gas hydrate and free gas widely overlap with the mapped 514 distribution of volcanoes and sills. This free gas and gas hydrate are commonly 515 associated with fluid migration pathways, and are occasionally trapped by differential 516 compaction folds formed above the volcanoes or in local coarse-grained sedimentary 517 518 rocks in the submarine canyon system above the sills. Volcanoes and sills likely facilitated cross-strata migration of hydrocarbon gas from deep sources and into 519 supra-sill faults, supra-volcano faults, and gas chimneys that connected to to 520 521 shallower reservoirs. Pluming bottom simulating reflectors (BSRs) occur in shallow strata above the summit of buried volcanoes, and may be formed by the local high 522 geothermal gradients related to the release of hydrothermal fluids from the volcanoes. 523 This study supports a growing body of work that shows volcanic systems may 524 influence fluid migration and accumulation in sedimentary basins long after magmatic 525 activity has ceased. 526

527

528 **Declaration of Competing Interest**

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

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- 544 <u>https://drive.google.com/file/d/1aHTs_fmMAU7WZwETEA10H3G-LnMW0G9E/vie</u>
- 545 <u>w?usp=sharing.</u>

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881 Figure captions

Fig. 1. (a) The regional map shows the location of the Pearl River Mouth Basin, northern South China Sea, including the Baiyun sag and the Liwan sag; (b) The enlarged map is the seafloor time structure map showing the distributions of submarine canyons and volcano complex (white broken line) in the Baiyun Sag and Yunli Low Uplift. The rectangle shows to the location of Figs. 4b, 10 and 12.

887

Fig. 2. The Cenozoic geological column, lithology and major tectonic events for the Pearl River
Mouth Basin (Modified from Pang et al., 2008; Dong et al., 2009; Ma et al., 2018).

890

Fig. 3. Regional seismic line (a) and the interpreted profile (b) across Sites BY6-1-1 and BY7-1-1
(Location is in Fig. 1a) show the strata horizons and distribution of the west (VCI), centre (VCII)
and east (VCIII) volcanic complexes in the Baiyun Sag and Yunli Low Uplift. The VC I and VC II
have been found in Zhao et al., 2016, 2019; Sun et al., 2020b.

895

Fig. 4. (a) 3D seismic views and (b) plane map of the interpreted time map of the top of volcano complexes III (horizon TM) show the geomorphology characteristics of buried volcanic complexes (VCIII) and gas hydrate-drilling sites. Regional normal faults presented as linear structures with sharp boundaries are also observed. The rectangles and black line show the locations of Figs. 5, 8 and Fig. 6a, b respectively.

901

902 Fig. 5. (a) Minimum amplitude attribute extracted along BGHSZ with the time windows
903 downward 50 ms shows the free gas distribution near BGHSZ. (b) Minimum amplitude attribute

904 extracted from the time windows between BGHSZ and T32 shows the free gas distribution in the
905 deeper sediments. (c) Maximum amplitude attribute and (d) average positive amplitude extracted
906 along BGHSZ with the time windows upward 50 ms shows the gas hydrate distribution. The
907 distribution area of free gas is larger than that of gas hydrate. The black lines show the locations of
908 Fig. 7a,b.

909

Fig. 6. Regional seismic line (a) and the interpreted profile (b) show the seismic characteristics of 910 911 igneous sills, free gas and gas hydrate in the Shenhu area of the Baiyun Sag. Gas hydrate-bearing 912 sediments have high amplitude just above the BGHSZ/BSR, while free gas is shown by the enhanced reflection below the BGHSZ/BSR. The accompanying relationships can be observed 913 914 between sills and free gas/gas hydrate, which are connected to supra-sill faults and chimneys. The 915 location of seismic line is shown in Fig. 4b. (c, d) Enlarged seismic profiles show the occurrence 916 of continuous BSR (CBSR), discontinuous BSR (DBSR), gas hydrate and free gas at Sites SH2 917 and W18; (e-h) Enlarged seismic profiles shows the various architectures of sills in different deep 918 strata. The BGHSZ represents the base of gas hydrate stability zone.

919

920 Fig. 7. 3D seismic views of the magmatic sills, BSRs, gas hydrates, and corresponding high 921 amplitude zones. (a) Perspective view of arbitrary seismic section through Sites SH2, W18, and 922 W17 shows the line distributions of sills (red arrow); (b) Seismic profile through Sites W11 and 923 W17 shows that supra-sill faults connect gas hydrate and free gas accumulations to the sills with 924 high-amplitude and pull up reflections. The seismic profiles locations are shown in Fig. 5.

925

Fig. 8. (a) Minimum amplitude attribute extracted with the time window between BGHSZ and T40 shows the free gas distribution in the Yunli Low Uplift. The spatial distribution of three enhanced reflection layers surround the summits of three volcanoes (i.e. M1, M2 and M3), respectively. (b) Maximum amplitude attribute extracted along BGHSZ with the time window upward 50 ms shows the gas hydrate distribution in the Yunli Low Uplift. The black lines shows the locations of Fig. 9a-e.

932

933 Fig. 9. The configurations of buried volcanoes, free gas, gas hydrate and normal faults (Locations

934 shown in Fig. 8). in the Yunli Low Uplift. (a) The supra-volcano normal faults and 935 first layer enhanced reflection occurred within compaction fold above the magmatic volcano 936 (M1); (b) Blanking, supra-volcano faults, corresponding DBSR, pluming BSR and second layer enhanced reflection occurred above the magmatic volcano (M2), highlighting their closely 937 relationships. (c) 3D visualization of horizon TM shows the distribution of magmatic volcano (M3) 938 939 and corresponding pluming BSR, blanking and the third enhanced reflection occurring above the summit of M3. (d) Enlarged seismic profile shows the corresponding pluming BSR in the third 940 941 enhanced reflection above M3; (e) 3D visualization of minimum amplitude attributes extracted 942 along horizons T32, T30 and T20 show the distributions of the first, second and third enhanced reflection layers above the M1, M2 and M3, respectively. The red dash lines referring to normal 943 faults. (f, g) Waveforms of the seafloor, high amplitude, and BSR reflections with polarity 944 945 indicated.

946

947 Fig. 10. (a) Variance slices along horizon T32 show the low correlations between faults, gas 948 chimneys, and volcanoes positions respectively, and the statistical rose diagram of faults shows 949 NE-SW striking. (b) Geological interpretation map of the distribution of non-magmatic faults, 950 supra-sill fault and supra-volcano faults and gas chimneys corresponding to sills and buried 951 volcanoes. The rectangle and black line show locations of Figs. 11a and 14, respectively.

952

Fig. 11. (a) Variance slice of 3250 ms and (b) corresponding geological interpretation map show the radial faults distribution in the overlying sediments above the volcano (M1). The location of Fig. 11a is shown in Fig. 10a. The seismic and geological interpretation profiles (c-f) across the volcano (M1) show the development of radial faults above the summit of volcano. The red dash lines referring to supra-volcano faults.

958

Fig. 12. The overlapped distribution map of geological elements shows magmatic sills, volcanoes,
non-magmatic faults, supra-sill fault and supra-volcano faults, free gas and potential gas hydrates
in the Baiyun Sag and Yunli Low Uplift.

962

963 Fig. 13. (a-c) The calculated BGHSZ at Location 1-3 compared to the BSR indicaties the

964 influences of high geothermal gradient (GD) for BGHSZ above the buried volcanoes. The965 location1-3 have been shown in Fig. 9b and c.

966

967 Fig. 14. (a) The seismic section and the schematic model (b) across gas hydrate-drilling Site SH2 968 and buried submarine volcanoes showing the closely relationship between gas hydrate occurrence 969 and magmatic complexes, comprehensive geological elements in the study area, including 970 magmatic sills, volcano complexes (VCIII), gas migration, free gas and gas hydrate occurrence. 971 Different types of fluids can be transported along faults and gas chimneys and accumulate in the 972 shallow sediments. Faults and gas chimneys directly occurred within the strata above the sills and buried volcanoes. The occurrence of pluming BSR above indicates the thermodynamic influences 973 974 of magmatic volcanoes for gas hydrate system. 975



Age (Ma)	Chono	logy	Strata	Lithology	Seismic reflector	Petroleum system		Tectonic event	Basin evolution	Depositional environment
0	Quaternary		Wanshan		-T20	▼				
5 —	Pliocene				-T30	drate		Dongsha		
10 -		Late	Yuehai		T22	as or hy reserve		event	nce	<i></i>
13.5- 15-	fiocene	Middle	Hanjiang		-T35	Ŭ ▼	Seal		Subside	Slope bathyal
17.5-	~	~			-T40					
20-		Early	Zhujiang		-130 0000	voir		Baiyun event		
25-	0				-T60	Reser		_	d	
30-	ligocene	Late	Zhuhai						Breaku	Netritic
	Ō	н.			-T70					
35-		te	Enping				~	1nd Zhuqiong		
40-		La	mm		—Т80		e roc	event		
40	ne				100		ourc		I	
45 –	Eoce	Middle	Wenchang				S	2nd Zhuqiong	ntinenta rifting	Lacustrine & Fluvial
50-		-		~~~~				event	Co	
75				 مرير	-Tg	2	Ľ	_ Shenhu event		
15-	Cretaceous			سر Mu سر	dstone					
80-				Sandstone Magmatism	Basement					



























