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Provenance, routing and weathering history of heavy minerals from coastal
 placer deposits of southern Vietnam

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14

15 Abstract

16 Heavy mineral rich sands along the coastal margin of southern Vietnam often 17 contain commercial deposits of ilmenite and zircon but their origin is unknown. 18 A multi-method approach based on petrology, geochemistry and detrital 19 zircon geochronology was used to define the provenance and transport 20 history of these mainly Quaternary sands. A trend of progressive enrichment 21 of ilmenite TiO₂ content, from north to south, was observed. This reflects 22 increased levels of weathering attributed to a wider coastal margin and shelf 23 in the south combined with a succession of erosion and reburial events 24 associated with interstadial and interglacial sea-level changes. Weathering

25 took place during lowstands. Detrital zircon U-Pb age signatures collected 26 from 25 major river outlets along the coast of Vietnam helped to locate potential sand sources. Prominent age groups spanning 90-120 Ma and 220-27 28 250 Ma with a minor group at 400-500 Ma are present in all of the detrital 29 zircon U-Pb age distributions of contemporary beach sands and Quaternary 30 coastal dune placer deposits. Proterozoic grains are also present but 31 constitute < 10% of dated grains. The main source terrain for the placer 32 sands is southern Vietnam where there are widespread outcrops of Mesozoic 33 magmatic rocks. Detrital zircon U-Pb age signatures from river sands that 34 drain this area are identical to zircon age distributions in placer sands. River 35 sands from northern Vietnam, the Mekong and its delta contain abundant 36 Paleozoic and Proterozoic zircons, which are largely absent from the placer 37 sands, and so are ruled out as primary sources.

38

Keywords: Ilmenite, zircon U-Pb, provenance, Vietnam, weathering, seallevel change

41

42 **1. Introduction**

Beach and dune placer deposits occur along the 3260 km coastline of Vietnam, as well as offshore in water depths up to 30 m or more, are economically important sources of ilmenite, rutile and zircon (Fig. 1). The heavy mineral rich sands are mainly found in beach dunes, beach ridge, washover and backshore deposits associated with Holocene to Pleistocene sealevel changes. Onshore deposits occur as bands, typically 1-4 m thick,

49 that extend 1-3 km inland from the coast, and are up to 10 km in length. Most 50 of the high-value deposits are found south of latitude 16°N particularly in the central SE Vietnam provinces Ninh Thuan and Binh Thuan (Fig. 1), to the 51 52 northeast of Ho Chi Minh City. Surveys made in 2011 by the Department of 53 Geology and Mineral Resources of Vietnam estimated that there are at least 54 650 million tons of ore reserves along the coastal margins between northeastern Vietnam and Vung Tau in the south. Sand ilmenite content 55 typically varies from 10 to 100 kg/m³ although some locations have 56 concentrations well above this. Rutile contents are usually less than 1 kg/m³, 57 although in some places it can reach up to 3-4 kg/m³ (e.g., coastal areas 58 59 north of Da Nang). Zircon abundances also vary; the highest average content 60 (up to 12 kg/m³) can be found in the coastal sections of Ham Tan in Binh 61 Thuan Province (Fig. 1). Mineral grain sizes are typically in the range of 0.16-62 0.25 mm. Understanding the origin of these minerals and the processes by 63 which they became concentrated is the primary motivation of this study.

64 Ilmenite is an important source of titanium oxide. Fresh unaltered ilmenite has TiO₂ wt% values up to the stoichmetric value of 52.6 wt%. Chemical 65 66 weathering and alteration, especially in oxidising and/or acidic environments, 67 can change ilmenite chemistry by reducing Fe and Mn, increasing Ti and adding AI, Si, Th, P, V and Cr (Pownceby, 2010). The distribution and 68 69 proportions of the different types of altered grains in the heavy mineral sands 70 influences their commercial value as a source of Ti, and therefore it is 71 important to understand the distribution and proportions of the different types of altered grains in the deposits which requires identifying where the alteration 72 73 occurred and defining grain transport history.

74 Coastal sands along the southeast-central coastline of Vietnam typically 75 comprise an outer and inner sand barrier. The former consists of loose white sand that sometimes form tombolos (e.g., Ho Gom Peninsula). The inner 76 77 sand barrier located up to 20 km inland consists of light yellow to reddish 78 yellow sands that include dunes found at elevations over 100 m above sea 79 level, such as in the area north of Vung Tau or Ham Thuan Bac district (Fig. 80 1). Whether these sands are locally derived is unclear. The aim of this study is 81 to better understand the environmental processes that led to the alteration 82 and concentration of the heavy minerals and to define where the sand came 83 from. It is known that the sands are closely linked to sea-level oscillations 84 during the Quaternary, especially Holocene glacioeustatic changes between 8 85 and 5 ka (Stattegger et al., 2013). Falling sea level causes remobilisation of 86 coastal sands deposited during highstands and increases bedrock erosion 87 inland. Larger volumes of sediment would have been more intensely 88 weathered during glacial periods (Wan et al., 2017) and the subaerial 89 exposure of unconsolidated shelf sediments during associated lowstands 90 would have affected ilmenite chemistry by causing enrichment in TiO₂. Wave 91 action and longshore drift would also have contributed to the winnowing 92 process, sorting grains according to size and density, hence it is entirely 93 possible that sand grains are far removed from their original source areas.

94

95 **2. Regional geology and geomorphology**

96 The source and volume of beach sands depend on wind, wave and tide
97 regimes as well as local erosion and fluvial transport rates. Detailed study of a

98 river catchment in northern coastal Vietnam has indicated that greatest 99 erosion reflected by river bedload and chemistry occurs within the mountainous regions where precipitation rates are highest, and that both 100 101 weathering and erosion rates are linked to monsoon intensity (Jonell et al., 102 2016). Transport of sediment to the coast is dominated by discharge from the 103 Mekong River in the south and by the Song Ma and Song Hong (Red River) in 104 the north. Between these large rivers, that have their headwaters in Tibet and 105 southwest China, the central areas of Vietnam are more locally drained by 106 relatively small river catchments (Fig. 2) that have their headwaters in the 107 nearby steep mountain ranges of the central highlands. Despite their small 108 size, these rivers have been important sources of sediment to the coast and 109 shelf as there is a relatively short distance between the wet highlands and the 110 coastal plain, evidenced by high Quaternary sedimentation rates (from 0.5 to 111 1.2 m/ka) on the local continental shelf (Schimanski and Stattegger, 2005).

112 The trend of the coastline and shelf areas of Central Vietnam tend to follow 113 the NNW- and NW-striking faults formed during the Triassic or earlier. 114 Ordovician to Permo-Triassic granulite and amphibolite facies metamorphic 115 rocks of the elevated Kontum Massif, which broadly lies between latitudes 116 14°N and 15°N, form the northern margin of the study area. Whilst zircon U-117 Pb geochronology has recorded Proterozoic ages between 1480-1350 and 900-600 Ma for local orthogneiss (Nguyen, et al., 2001; Tran, et al., 2003), 118 119 charnokites, biotite-sillimanite-cordierite-garnet gneiss, schists, amphibolites, 120 and granitoids originally mapped as Archean and Proterozoic have since been 121 dated as Silurian and Triassic (Indosinian) (Carter et al., 2001; Nam et al., 122 2001; Hiet et al., 2015, 2016). These rock types are not seen south of latitude

123 13°N where Mesozoic granitoids dominate. West of Nha Trang are late 124 Carboniferous-early Permian rocks of the Dac Lin Formation. These comprise terrigenous sediments interbedded with intermediate volcanics, mainly 125 126 andesitic basalts and tuffs. During the Triassic, closure of Tethys and final welding between Indochina and South China blocks caused significant 127 128 deformation across much of northern Vietnam. This event is known as the 129 Indosinian orogeny. Stratigraphy and radiometric ages of magmatic and 130 metamorphic rocks support a Middle Triassic age for final closure of the 131 Paleo-Tethys ocean (Faure et al., 2014). The study area was relatively unaffected by deformation related to this event. Triassic rocks are mainly 132 133 confined to the northern part of the study area where the Mang Yang 134 Formation includes rhyolites and tuffs associated with intracontinental rifts 135 (Tran et al., 2011). Jurassic rocks are more widespread and occur as 136 andesites, dacite and tuffaceous sandstones (Deo Bao Loc Formation). They 137 are especially widespread in the western area between latitudes 13°30'N and 138 12°N. By contrast, the eastern region is dominated by Cretaceous magmatic 139 rocks related to a former active continental margin. The widespread 140 occurrence of arc-related magmatic rocks across the study area, including 141 granitoids and rhyolites, is linked to subduction of the Palaeo-Pacific oceanic 142 crust beneath southern China, Vietnam and southern Borneo (Shellnut et al., 2013; Hall and Breitfeld, 2017). 143

Within the study area there are three main suites of Cretaceous magmatic rocks. The Dinhquan and Deoca complexes are found along the South Vietnamese coast. Petrological characteristics of the Dinhquan complex comprise hornblende-biotite diorites, granodiorites and minor granites. The

148 Deoca complex consists of granodiorite, hornblende-biotite granite (phase I), 149 biotite-hornblende granite, granosyenite and biotite syenite (phase II), and granite porphyry, granular aplite and pegmatite (dike phase). U-Pb zircon 150 151 ages range from 88±1.5 - 109±7.0 Ma (Thuy et al., 2004) to 115.4±1.2 -152 118.2±1.4 Ma (Shellnutt et al., 2013). The Ankroet Complex is smaller than 153 the Dinhquan and Deoca complexes and is located further inland, at higher 154 elevations. Rock types include medium to coarse grained porphyroid biotite 155 granite. Published zircon U-Pb ages are 93.4±2.0 – 96.1±1.1 Ma (Thuy et al., 156 2004) and 86.8±1.6 Ma (Shellnutt et al., 2013). Geochemical work by Shellnut et al (2013) show the upper Lower Cretaceous granitic batholiths are I-type 157 158 (partial melting of dehydrated middle/lower crust) and the Upper Cretaceous 159 (i.e., ~90 Ma) granitic rocks have compositions similar to A-type (differentiated 160 mafic parental magmas) associated with an extensional tectonic regime, most 161 probably trench retreat caused by slab rollback. Ankroet rocks are associated 162 with this extensional setting.

163 Cenozoic fluvial-shallow marine clastic sedimentary rocks in the study area 164 are the Oligo-Miocene Di Linh Formation, the early Pliocene to Pleistocene 165 Song Luy Formation, and the late Pliocene to Pleistocene Ba Mieu Formation. 166 Study of detrital zircon U-Pb ages from these units recorded abundant 167 Cretaceous ages, as well as Permian-Triassic and Ordovician-Silurian 168 sources. The youngest unit also records a significant increase in Precambrian (Hennig et al., 2018). Also found across the study area are 169 zircons 170 widespread late Cenozoic basaltic lava flows up to several hundred metres 171 thick (Hoang and Flower, 1998). Alkali basaltic magmatism began in the 172 middle Miocene and has a geochemistry that fits with sources of recycled

eclogitic oceanic crust from the Hainan plume (An et al., 2017). Eruptions and
lava flows often appear to have exploited local fault zones re-activated by
South China Sea opening.

176 Patterns of sediment accumulation and concentration of heavy mineral sands along the coastal shelf and margins appear to track past sealevel changes. 177 Direct evidence to support this can be found in optically stimulated 178 179 luminescence (OSL) dating studies of stratigraphically oldest barrier sands 180 exposed at Suoi Tien (10°57'16"N - 108°15'30"E) and Hon Gom (12°41.64'N - 109°45.27'E) (Fig. 1) (Quang-Minh et al., 2010) that include layers enriched 181 182 in ilmenite and zircon. These gave deposition ages ranging from 8.3±0.6 to 183 6.2±0.3 ka BP, contemporaneous with the local postglacial maximum sealevel 184 highstand. Much older red shallow marine sands at Suoi Tien were dated to 185 101±16 ka whilst white sand at the bottom of the sequence could be as old as 186 276±17 ka and correspond to an earlier sea-level highstand. Detailed 187 reconstructions of mid to late Holocene sealevel for Southeast Vietnam can 188 be found in Stattegger et al. (2013).

189 Although sealevel fluctuations are important, the concentration of heavy 190 minerals likely involved a combination of factors that included sediment 191 transport history along the shelf and coastline (influenced by sediment supply) 192 and hydrodynamic conditions. The latter is dominated by the East Asian 193 monsoon system that blows from the northeast in winter and southwest in the 194 summer. The northeast monsoon has most impact on northern Vietnam and 195 the southwest monsoon on central and southern regions (Pham, 2003). 196 Although seasonal reversal of the monsoon system also switches longshore 197 currents from southerly to northerly, the long-term trend of sediment transport

can also be affected by local coastal geomorphology. This makes it difficult to
 predict long-term trends in coastal sediment transport, as demonstrated by
 modeling studies of longshore transport to define impacts of sealevel changes
 associated with climate change (Dastgheib et al., 2016).

202

203 3. Methods and Approach

204 The study area covers the section of Vietnamese coastline where most heavy 205 mineral sands are found, which is between 15°N and 10°N (Fig. 1). Since 206 placer deposits represent biased sand composition we used a multi-method 207 approach and defined the geochronological, geochemical and mineralogical 208 signatures of representative placer deposits to locate sand source areas and 209 define the extent of alteration and transport. Results are then compared 210 against data collected from each of the main rivers along the Vietnamese 211 coastline including 2 samples (X and Y) from the upper Mekong within (Laos 212 (Fig. 2). This approach will enable a model of locally derived vs longshore 213 transport derived to be tested.

214 Sampling of placer deposits included nearby contemporary beach sands as 215 these might preserve geographic links to source areas compared to older 216 sands that are likely to have seen more extensive reworking and mixing, 217 although reworking of older sediments would negate this assumption. 218 Recognising that during transport selective entrainment based on variations in 219 grain density produces a compositional bias we sampled sands with a typical 220 grain size range between 65-500 µm. River sands were collected as close to 221 river mouths as possible from active channel beds and point bars where

heavy minerals tend to be concentrated. Beach sands were sampled (Fig. 3) in areas documented as rich in heavy minerals and taken from dark sand layers in the upper shoreface following removal of the lighter coloured top few centimeters. Also included are sand samples from onshore shallow boreholes drilled in prospective mining areas. In all cases efforts were made to avoid areas subject to obvious anthropogenic disturbance. In total 25 river and 18 onshore placer sand samples (typically between 1 to 2 kg) were collected.

229 Quantification of mineral types and abundances was made using automated energy-dispersive X-ray spectroscopy (SEM-EDS) coupled with expert 230 software analysis on a QEMSCAN[®] platform which allows micron-scale 231 232 mapping and mineral identification of samples (Pirrie and Rollinson, 2011). 233 Polished grain mounts of untreated sands were scanned at a resolution of 10 234 µm yielding c. 5000 to 12000 grain counts per slide. The acquired EDS 235 spectra were interpreted automatically by reference to a database of mineral 236 compositions.

237 Detrital zircon U-Pb geochronology is used to help define ilmenite provenance 238 since both Ilmenite and zircon are normally found in similar source rock types 239 and would be expected to behave similarly during transport as they have 240 similar specific gravities (4.5-4.7). Detrital zircon geochronology is widely used 241 in provenance studies due to stability of the mineral and U-Pb system (e.g., 242 Jonnell et al., 2017; Singh et al., 2017). Detrital zircon grains were separated 243 by standard heavy liquid techniques. Grains for dating were selected 244 randomly from polished grain mounts and analysed by laser ablation 245 inductively coupled plasma mass spectrometry at the London Geochronology 246 Centre based in University College London using a New Wave 193 nm laser

247 ablation system coupled to an Agilent 7700 quadrupole-based ICP-MS. 248 Typical ablation parameters used 25 µm spots with a 10 Hz repetition rate and an energy fluence of ca. 2:5 J/cm². Instrumental mass bias and depth-249 250 dependent inter-element fractionation of Pb, Th and U were corrected for 251 using Plesovice as an external zircon standard (Sláma et al., 2008). Time-252 resolved signals that record evolving isotopic ratios with depth in each crystal were processed using Glitter 4.4 data reduction software. This removed 253 254 spurious signals caused by inclusions, mixing of growth zones or fractures. Calculated ²⁰⁶Pb/²³⁸U ages were used for grains younger than 1000 Ma, and 255 the ²⁰⁷Pb/²⁰⁶Pb age for older grains. Grains with a complex growth history or 256 257 disturbed isotopic ratios, with > +5/-15% discordance, were rejected.

To characterize ilmenite chemistry and to test for ilmenite alteration by weathering grains (circa 100 per sample) from representative river and placer sands were selected for electron microprobe analysis. A JEOL JXA-8100 Electron Probe Microanalyzer Scanning Electron Microprobe fitted with an Oxford Instruments X-act PentaFET Precision detector was used to carry out the analyses on polished grain mounts. Qemscan mineral maps helped with grain identification.

265

266 4. Results and Interpretation

4.1 Petrology

Table 1 summarises mineral abundances of representative river and
Quaternary sands. Despite a wide presence of basaltic rocks olivines are

270 rarely found in river sands and none were detected in the Qemscan analyses 271 of untreated sand (Table 1). Pyroxenes are present in river sands but are missing from the coastal placer sands suggesting that there has been loss 272 273 due to weathering. Minerals diagnostic of heavy to medium grade metamorphic rocks are common. Similar abundances of high-grade 274 275 metamorphic minerals siliminite, kyanite and andalusite are present in both 276 river and beach sands, although they are more abundant in the area between 277 latitudes 14-16°N where outcrops of high-grade Proterozoic metamorphic 278 rocks are more widespread. Amphiboles are especially common in the river 279 sands between 12 and 16°N but abundances systematically decrease to the 280 south (Fig. 4). By contrast, amphiboles are sparse in the contemporary beach 281 sands (Table 1) suggesting either removal by weathering and physical 282 abrasion, helped by its cleavage (Garzanti et al., 2015), or density sorting 283 during transport. The latter is unlikely given that the ultrastable high-grade 284 metamorphic minerals, siliminite and kyanite, which are only slightly denser 285 than amphibole, are present in both river and beach sands (typically 0.1 to 286 0.4% of grains, Table 1). Aside from loss by weathering and abrasion it is 287 also possible that the absence of amphiboles reflects minimal south-directed 288 longshore transport, i.e., rivers sands are not dispersed very far along the 289 coast. The latter seems more likely as the denser minerals garnet, rutile, 290 ilmenite and zircon that do not breakdown as easily as amphibole during 291 transport, also show decreasing abundances between northern and southern 292 rivers and that levels in the beach sands always have a lower content than 293 river sands. By contrast, levels of feldspars increase southwards in river 294 samples but remain low in most heavy mineral sand samples. This provides

clear evidence that some density separation is taking place in the marineenvironment.

297

298 **4. 2. Ilmenite geochemistry**

299 Results of a subset of samples selected for ilmenite microprobe analyses (Fig. 300 5) show that although some fresh unaltered ilmenite grains are present most 301 ilmenites have been altered and this increased grain titanium contents to above stoichiometric levels (i.e., > 52.6 wt%). Plot 5A, of river sands, shows 302 303 that there are some regional differences whereby the proportion of altered 304 grains increases to the south. This implies that rivers in the north of the study 305 area deliver fresher ilmenite to the coast and offshore. Plot 5B compares 306 ilmenite from Holocene sands (Quang-Minh et al., 2010) along the coast. These data also show a trend of increased levels of weathering to the south. 307 Comparison between river sands and nearby Holocene sands (Plot 5C) show 308 309 dissimilar distributions supporting alteration after river deposition. Plot 5D 310 compares modern beach sands along the coast and again the greatest 311 amount of alteration is seen in the south.

312

4.3. Detrital zircon U-Pb river sand results

Data from each of the main river outlets along the coast of Vietnam provide a simple way of capturing signatures of the local geology against which coastal sand data may be compared (full analytical results are provided in the supplementary section). A summary of age distributions of individual river

318 samples (Fig. 6), displayed as Kernel density (KDE) plots (Vermeesch, 2012), 319 show rivers from northern and central Vietnam drain older rocks than rivers in southern Vietnam (Fig. 3). Both the Song Hong (Red River) and Mekong have 320 321 age distributions dominated by a wide range of Proterozoic ages that reflect 322 source rocks in the catchments beyond Vietnam, e.g. Mekong samples X and 323 Y from Laos (Fig. 2). The proportion of 400-500 Ma zircons is seen to 324 increase southwards at the expense of Proterozoic grains (Fig. 6). South of 325 14°N, river (sample L onwards) zircon age distributions are dominated by 326 either Permo-Triassic, Cretaceous or Ordovician-Silurian peaks (Fig. 6). The 327 Permo-Triassic ages are likely to be volcanic rather than granitic as the main 328 rocks types in the study area are rhyolites and tuffs belonging to the Mang 329 Yang Formation although Triassic granulites are known in the Kontum area 330 (Carter et al., 2001). The majority of age spectra contained a few Proterozoic 331 ages, some of which are clearly related to inherited cores (Supplementary 332 Figure 1). This observation is consistent with Shellnut et al., (2013) who noted 333 magma mixing with older basement was required to explain the composition 334 and inherited ages of the Cretaceous granites.

335 As visual comparison of KDE plots is subjective the data, were also plotted as 336 Multidimensional Scaling (MDS) maps (Vermeesch, 2013). The MDS 337 approach, based on Kolmogorov-Smirnov effect size, group samples with 338 similar age spectra, and pull apart samples with different spectra. The MDS 339 map (Fig. 6) clearly shows two groups of samples. The left group comprises 340 rivers from northern Vietnam plus the Mekong that have abundant Proterozoic 341 ages. The right-hand group comprises river samples from central and 342 southern Vietnam which are dominated by Permo-Triassic and Cretaceous

343 age peaks. These two groups reflect changes in regional geology whereby 344 northern Vietnam is dominated by Proterozoic and Paleozoic metamorphic basement, compared to the south where Mesozoic granitoids and Cenozoic 345 346 basalts dominate. A transition between these groups occurs around the 347 Kontum massif, which marks the northern limit of the main study area. The 348 catchment of river K (Da Rang) spans this junction and therefore plots 349 between the two main clusters. Based on these results it will be possible to 350 identify if any of the heavy mineral sands originated from northern Vietnam.

351

352 4.4. Detrital zircon U-Pb coastal sand results

353 KDE plots of detrital zircon ages from coastal Quaternary (Q) and modern 354 beach (MB) sands (Fig. 7) show prominent age groups spanning 90-120 Ma 355 and 220-250 Ma plus a minor group at 400-500 Ma. The age distributions are 356 remarkably similar across the whole study area, differing only in the 357 proportions of zircons within each age group. The accompanying MDS plot suggests samples Q1 and Q2, from north of Nha Trang (Fig. 1) are different 358 359 from the rest but this is simply due to fewer Cretaceous ages compared to the 360 other samples, despite being located less than 50 km from the Cretaceous 361 Deo Ca magmatic Complex. The Da Rang (river K) is local to samples Q1 and 362 Q2 and shows a similar age distribution (Fig. 7) although there are fewer 363 Cretaceous and Ordovician-Silurian zircons. South of the Da Rang, all other 364 rivers, apart from the Mekong, are dominated by Cretaceous zircons.

365

366 **5. Discussion**

367 Heavy mineral sand mineralogy data support derivation from a mixture of 368 magmatic and high-grade metamorphic lithologies. Many sands contain trace 369 amounts of the high-grade metamorphic minerals sillimanite and kyanite 370 (present in both river and coastal sands) but olivine and pyroxenes are 371 missing despite the widespread occurrence of Neogene basalts throughout 372 southern Vietnam (see Table 1). Likely sources of sillimanites are outcrops of 373 biotite-sillimanite-cordierite-garnet gneiss in the Kontum district. This is 374 supported by the higher amounts of sillimanite in rivers G and H (Fig. 2) that drain this area. However, sillimanite is also present farther south in the Song 375 376 Cai (L in Fig. 2) and in Holocene heavy mineral sands near Phan Rang (Q4 in 377 Fig. 1), a region dominated by Cretaceous granites. Rocks west of Nha Trang 378 have been mapped as Proterozoic amphibole gneiss and schists so it is 379 conceivable that sillimanite rocks may also exist in this area.

380 Feldspar contents in river sands, which are typically between 10 and 40%, 381 have been reduced to < 2% in most heavy mineral sands indicating 382 considerable density separation (and/or weathering) within the marine 383 environment. Whilst none of these observations enable specific source areas 384 to be identified, several common trends have been recognized amongst the 385 petrological, geochemical and geochronological datasets that reflect the 386 sediment routing system. Amphibole abundances decrease from north to 387 south and ilmenite TiO₂ content increases southwards. Relatively fresh 388 ilmenite is delivered to the oceans by rivers in central Vietnam (e.g., Song 389 Cau) compared to rivers in the south (e.g., Sai Gon), where alteration due to 390 weathering is more developed (Fig. 5A). However, ilmenite TiO₂ content in 391 river sands do not match local heavy mineral deposits (Fig. 5C). Collectively,

this evidence shows most of the alteration must have taken place after deposition by rivers. One possibility is that the wider coastal plains found in the south are more conducive to intermediate storage (and weathering) before remobilisation and final deposition (Fig. 8).

396 Southward widening of the SE Vietnam Shelf area has not only increased the 397 distance between sediment sources to the middle and outer shelf but also 398 created a wide plain that would have been exposed to weathering during the 399 late Pleistocene and Holocene lowstands. With rising sealevel some of this sand would have been remobilised and transported inland, especially during 400 401 the Holocene highstand between 6-7 ka. Sand was subsequently reworked by 402 wave activity and redeposited during interstadial and interglacial 403 transgressions. The narrow continental shelf farther north and the proximity of 404 the mountainous terrain to the coast limit the amount of surface area exposed 405 weathering in the northern and central coastal areas.

406 Studies of modern and late Pleistocene to Holocene stratigraphy of the shelf 407 areas of central and southern Vietnam (Dung et al., 2013, 2014; Stattegger et 408 al., 2013; Tan et al., 2014) have identified at least five major seismic units and 409 three bounding surfaces that can be linked to known sealevel adjustments 410 including relict beach-ridge deposits at water depths of about ~130 m below 411 present that are associated with the last glacial lowstand. More importantly, in 412 relation to understanding the processes by which sands became weathered 413 and enriched in heavy minerals, studies (Bui et al., 2013; 2014) have noted an 414 absence of falling stage systems tract deposits. This can be explained as the 415 result of inner and middle shelf deposits being subjected to erosion and 416 reworking during successive sea-level falls following highstands and

417 reworking again during the following transgression. Repeating cycles of 418 reworking would also have been influenced by strong monsoon driven bottom 419 currents evidenced by numerous NE–SW oriented sand waves that today are 420 found at modern water depths of 20-40 m (Bui et al., 2013). Figure 8 shows 421 former coastlines associated with past lowstands and their relationship to 422 onshore and offshore placer sands (Quang-Minh et al., 2010; Stattegger et 423 al., 2013).

424 Detrital zircon data help to define where placer sands came from. Results from rivers along the coast of Vietnam show clear differences in zircon age 425 426 distributions between northern Vietnam and central to southern Vietnam that 427 directly reflect changes in the local geology (Fig. 6). Differences between river 428 and placer zircon age distributions (Fig. 9) rule out sources from northern and 429 central Vietnam, which are dominated by older rocks. Exceptions are Mekong 430 river samples that yielded significant numbers of Precambrian zircon ages. 431 Similar old ages are also found in the late Pliocene to early Pleistocene Ba 432 Mieu Formation (proto-Mekong) found east of Ho Chi Minh City (Hennig et al., 433 2018). Much of this formation has been eroded away and therefore if these 434 rocks (and paleo Mekong deposits in general) were an important source there 435 should be significant numbers of Precambrian zircon ages present in the 436 coastal sands. That this is not the case shows that Mekong river sands 437 (modern or ancient) could not have been the main source of the placer sands. 438 Geochronological and geochemical characteristic of placer and contemporary 439 sands support a local origin defined by river catchments that are dominated 440 by Cretaceous magmatism associated with an active continental margin, i.e., 441 the Da Lat zone and areas to the south. Apart from Quaternary samples Q1

442 and Q2 that contain a larger proportion of ages between 220-250 Ma and 443 400-500 Ma, there is no significant difference between modern and older sand 444 deposits (Fig. 7). This is likely due to mixing associated with changes in 445 sealevel. Lack of Precambrian grains in the coastal placer deposits and 446 beach sands rule out significant longshore transport from the north or 447 reworking of paleo-Mekong sands in the south.

448

449 **6.** Conclusions

450 Placer sands along the coastal margins of central and southern Vietnam have been enriched in heavy minerals by cycles of deposition, weathering and 451 452 erosion, and reburial associated with interstadial and interglacial sealevel 453 changes. Weathering took place during lowstands. Geochemical and 454 geochronological data show sands were derived from river catchments that 455 contain outcrops of Cretaceous magmatic rocks. Results do not support 456 significant longshore transport from northern Vietnam or from the Mekong delta in the south. Had there been significant transport from the north, placer 457 458 sands would contain large numbers of zircons with Proterozoic and Paleozoic 459 ages that typify the geology of these areas, including the large catchment 460 area of the Red River that extends into South China. Mekong sources can be ruled out for similar reasons. Ilmenite sources were observed in all of the main 461 462 river outlets along the southern to central Vietnamese coastline although fresh 463 unaltered grains were mainly found in the central region. A progressive 464 enrichment of ilmenite TiO₂ content was observed from north to south due to 465 more intense weathering related to a widening of the shelf area. This would

have increased surface area exposure of unconsolidated shelf sediments to
weathering during glacial sea-level lowstands and remobilisation and mixing
during subsequent transgressions.

469

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477

478 **References**

479 An, A-R., Choi, S.H., Yu, Y., Lee, D-C., 2017. Petrogenesis of Late Cenozoic

480 basaltic rocks from southern Vietnam. Lithos 272, 192-204.

- 481 Carter, A., Roques, D., Bristow, C., Kinny, P., 2001. Understanding Mesozoic
- 482 accretion in Southeast Asia: significance of Triassic thermotectonism

483 (Indosinian orogeny) in Vietnam. Geology 29, 211–214.

- 484 Dastgheib, A., Reyns, J., Thammasittirong, S., Weesakul, S., Thatcher, M.,
- 485 Ranasinghe, R., 2016. Variations in the Wave Climate and Sediment
- 486 Transport Due to Climate Change along the Coast of Vietnam. Journal of
- 487 Marine Science and Engineering 4, 86, doi:10.3390/jmse4040086.
- 488 Dung, B.V., Stattegger, K., Unverricht, D., Phung, V.P., Nguyen, T.T., 2013.
- 489 Late Pleistocene–Holocene seismic stratigraphy of the Southeast Vietnam.
- 490 Shelf. Global and Planetary Change 110, 156-169.

491 Dung, B.V., Nguyen, T.T., Stattegger, K., Phung, V.P., Tran, T.D., Bui, X.D.,

492 2014. Late Pleistocene-Holocene seismic stratigraphy of Nha Trang Shelf,

493 Central Vietnam. Marine and Petroleum Geology 58, 789-800.

494 Garzanti, E., Resentini, A., Andò, S., Vezzoli, G., Pereira, A., Vermeesch, P.,
495 Lancaster, N., 2015. Physical controls on sand composition and relative

496 durability of detrital minerals during ultra-long distance littoral and aeolian

497 transport (Namibia and southern Angola). Sedimentology 62, 971-996.

Grey, I.E., Li, C., 2001. Low temperature roasting of ilmenite—phase
chemistry and applications. Proceedings of the Australasian Institute of
Mining and Metallurgy 306, 35–42.

501 Hall, R., 2012. Late Jurassic–Cenozoic reconstructions of the Indonesian 502 region and the Indian Ocean. Tectonophysics 570–571, 1–41.

Hall, R., Breitfeld, H.T., 2017. Nature and Demise of the Proto-South China
Sea. Bulletin of the Geological Society of Malaysia 63, 61-76.

Hieu, P.T., Dung, N.T., Nguyen, T.B.T., Minh, N.T., Minh, P., 2016. U–Pb
ages and Hf isotopic composition of zircon and bulk rock geochemistry of
the Dai Loc granitoid complex in Kontum massif: Implications for early
Paleozoic crustal evolution in Central Vietnam. Journal of Mineralogical and
Petrological Sciences 111, 326-336.

Hieu, P.T., Yang, Y.Z., Binh, D.Q., Nguyen, T.B.T., Dung, L.T., Chen, F.,
2015. Late Permian to Early Triassic crustal evolution of the Kontum
massif, central Vietnam: zircon U–Pb ages and geochemical and Nd–Hf
isotopic composition of the Hai Van granitoid complex. International
Geology Review 57, 1877-1888.

- Hoang, N., Flower, M.F.J., 1998. Petrogenesis of Cenozoic basalts from
 Vietnam: implication for origins of a 'diffuse igneous province'. Journal of
 Petrology 39, 369-395.
- Hoang, N., Flower, M.F.J., Xuan. P.T., 1996. Petrology of Vietnam basalt in
 the late Cainozoic. Institute of Geology and Mineral Resources Hanoi, 1,
 142-155.
- Jonell, T.N., Clift, P.D. Hoang, L.V., Hoang, T., Carter, A., Wittmann, H.,
 Böning, P., Rittenour, T., 2017. Controls on Erosion Patterns and Sediment
 Transport in a Monsoonal, Tectonically Quiescent Drainage, Song Gianh,
 Central Vietnam. Basin Research 29, 659–683.
- Lam, N.T., 2009. Hydrodynamics and Morphodynamics of a Seasonally Force
 Tidal Inlet System. Ph.D. Thesis, Delft University of Technology, Delft, The
 Netherlands.
- Metcalfe, I., 2009. Late Palaeozoic and Mesozoic tectonic and
 palaeogeographic evolution of SE Asia. In Buffetaut, E., Cuny, G., Le
 Loeuff, J., Suteethorn, V. (Eds), Late Palaeozoic and Mesozoic
 Ecosystems in SE Asia. Geological Society of London, Special
 Publications, pp. 315, 7 22.
- Nam, T.N., Sano, Y., Terada, K., Toriumi, M., Quynh, P.V. and Dung, L.T.,
 2001. First Shrimp U–Pb zircon dating of granulites from the Kontum
 massif (Vietnam) and tetonothermal implications. Journal of Asian Earth
 Sciences 19, 77–84.
- 537 Pham, V.N., 2003. Bien Dong Monograph, Vol. II—Meteorology. Hanoi
 538 National University Publisher, Hanoi, 565 pp. (in Vietnamese).

- 539 Pirrie, D., Rollinson, G.K., 2011. Unlocking the applications of automated 540 mineral analysis. Geology Today 27, 235-244.
- Pownceby, M., 2010. Alteration and associated impurity element enrichment
 in detrital ilmenites from the Murray Basin, southeast Australia: a product of
 multistage alteration. Australian Journal of Earth Sciences 57, 243-258.
- 544 Pubellier, M., Morley, C.K., 2014. The basins of Sundaland (SE Asia):
- 545 Evolution and boundary conditions. Marine and Petroleum Geology 58, 546 555-578.
- Quang-Minh. D., Frechen, M., Nghi, T., Harff J., 2010. Timing of Holocene
 sand accumulation along the coast of central and SE Vietnam. International
 Journal of Earth Science 99, 1731–1740.
- 550 Sláma, J., Košler, J., Condon, D.J., Crowley, J.L., Gerdes, A., Hanchar, J.M.,
- Horstwood, M.S., Morris, G.A., Nasdala, L., Norberg, N., 2008. Plešovice
 zircon—a new natural reference material for U–Pb and Hf isotopic
 microanalysis. Chemical Geology 249, 1-35.
- 554 Shellnutt, J.G., Lan, C-Y., Long, T. V., Usuki, T., Yang, H-J., Mertzman, S.A.,
- Lizuka, Y., Chung, S-L., Wang, K-L., Huse, W-Y., 2013. Formation of Cretaceous Cordilleran and post-orogenic granites and their microgranular enclaves from the Dalat zone, southern Vietnam: Tectonic implications for
- the evolution of Southeast Asia. Lithos 182-183, 229-241.
- 559 Singh, A., Thomsen, K.J., Sinha, R., Buylaert, J-P., Carter, A., Mark, D-F.,
- 560 Mason, Densmore, A.L., Murray, A.S., Jain, M., Paul, J., and Gupta, S.,
- 561 2017. Counter-intuitive influence of Himalayan river morphodynamics on
- 562 Indus Civilisation urban settlements. Nature Communications 8, 1617,
- 563 doi:10.1038/s41467-017-01643-9.

Stattegger, K., Tjallingii, R., Saito, Y., Michelli, M., Nguyen, T.T., Wetzel, A.,
2013. Mid to Late Holocene sea-level reconstruction of Southeast Vietnam
using beachrock and beach-ridge deposits. Global and Planetary Change,
110, 214-222.

Suggate, S., Hall, R., 2013. Using detrital garnet compositions to determine
provenance: a new compositional database and procedure. In Scott, R.A.,
Smyth, H.R., Morton, A.C, Richardson, N. (Eds.), Sediment Provenance
Studies in Hydrocarbon Exploration and Production, Geological Society of
London Special Publication, 386, pp. 373-393.

574 2014. Pliocene–Quaternary evolution of the continental shelf of central 575 Vietnam based on high resolution seismic data. Journal of Asian Earth 576 Sciences 79, 529–539.

Tan, M.T., Dung, L.V., Bach, L.D., Bieu, N, Nghi, T., Long, H.V., Huong, P.T.,

- 577 Thuy, N.T.B., Muharren, S., Wolfgang, S., Fukun, C, 2004. Granitoids in the 578 Dalat zone, Southern Vietnam: age constraints on magmatism and regional 579 geological implications. International Journal of Earth Science 93, 329-340.
- 580 Tran V.T., and Vu, K., (Eds.), 2011. Geology and Earth Resources of
- 581 Vietnam: General Department. of Geology, and Minerals of Vietnam.
- 582 Hanoi, Publishing House for Science and Technology, 634 pp.
- 583 Vermeesch, P., 2012. On the visualisation of detrital age distributions.
- 584 Chemical Geology 312, 190-194.

573

- 585 Vermeesch, P., 2013. Multi-sample comparison of detrital age distributions.
- 586 Chemical Geology 341, 140-146.
- 587 Wan, S., Clift, P.D., Zhao, D., Hovius, N., Munhoven, G., France-Lanord, C.,
- 588 Wang, Y., Xiong, Z., Huang, J., Yu, Z., Zhang, J., Ma, W., Zhang, G., Li, A.,

- 589 Li, T., 2017. Enhanced silicate weathering of tropical shelf sediments 590 exposed during glacial lowstands: a sink for atmospheric CO2. Geochimica 591 et Cosmochimica Acta 200, 123-144.

- **FIGURE CAPTIONS**
- **Table 1**. QEMSCAN mineral percentages (by volume) for untreated river and
 598 Quaternary beach sands from central and southern Vietnam.

Figure 1. Locations of placer and beach sands samples and main commercial
 extraction sites in southern Vietnam. Samples with illmenite composition
 data, and mineralogical data reported in Table 1, are also indicated.

Figure 2. Locations of river sand samples collected from each of the main
river outlets along the coast of Vietnam. Sample prefixes are given in
brackets.

Figure 3. Map of study area geology showing locations of sand samples

Figure 4. Abundance of amphiboles in river sands from central Vietnam as afraction of total grains scanned on the Qemscan slide.

613 **Figure 5.** Ti and Fe contents of ilmenite grains from river and coastal sand 614 samples.

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Figure 6. Kernel density and Multidimensional Scaling plots of the detrital
 zircon U-Pb results from the river samples shown in Figure 2.

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Figure 7. Kernel density and Multidimensional Scaling plots of detrital zircon
U-Pb results from coastal sands. Prefix Q indicates a Quaternary sand and
MB modern beach sands.

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Figure 8. Relationship between late Pleistocene to Holocene sealevel
change, shorelines and locations of the heavy mineral sands. The lower
plot shows the link between OSL dated sands and Holocene sea level
based on data from Quang-Minh et al., (2010) and Stattegger et al., (2013).

Figure 9. Multidimensional Scaling plot combining all detrital zircon samples
 apart from the Mekong and Red rivers that have been excluded due to their
 markedly different age spectra that rule out these rivers as sand sources.

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