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The Segmented Zambezi Sedimentary System from Source to Sink: 2. Geochemistry, Clay Minerals, and Detrital Geochronology

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Abstract :

Elemental geochemistry, Nd isotopes, clay minerals, and U-Pb zircon ages integrated by petrographic and heavy-mineral data offer a multi-proxy panorama of mud and sand composition across the Zambezi sediment-routing system. Detrital-zircon geochronology highlights the four major episodes of crustal growth in southern Africa: Irumide ages predominate over Pan-African, Eburnean, and Neoarchean ages. Smectite, dominant in mud generated from Karoo basalts or in the equatorial/winter-dry climate of Mozambican lowlands, prevails over illite and kaolinite. Elemental geochemistry reflects quartz addition by recycling (Uppermost Zambezi), supply from Karoo basalts (Upper Zambezi), and first-cycle provenance from Precambrian basements (Lower Zambezi). Mildly negative for sediments derived from mafic granulites, gabbros, and basalts, $\delta^{143}\text{Nd}$ values are most negative for sand derived from cratonic gneisses. Intrasample variability among cohesive mud, very coarse silt, and sand is principally caused by the concentration of Nd-rich monazite in the fine tail of the size distribution. The settling-equivalence effect also explains deviations from the theoretical relationship between $\delta^{143}\text{Nd}$ and TNd,DM model ages, suggesting that monazite carries a more negative $\delta^{143}\text{Nd}$ signal than less dense and less durable heavy minerals. Elemental geochemistry and Nd isotopes reveal that the Mazowe-Luenha river system contributes most of the sediment reaching the Zambezi Delta today, with minor supply by the Shire River. Sediment yields and erosion rates are lower by an order of magnitude on the low-relief Kalahari Plateau than in rugged Precambrian terranes. On the Plateau, mineralogical and geochemical indices testify to extensive breakdown of feldspars and garnet unjustified by the presently dry climate. Detrital kaolinite is recycled by incision of Cretaceous- Cenozoic paleosols even in the wetter lower catchment, where inefficient hydrolysis is testified by abundant fresh feldspars and undepleted Ca and Na. Mud geochemistry and surficial corrosion of ferromagnesian minerals indicate that, at present, weathering increases only slightly downstream the Zambezi River.

Keywords : Provenance analysis, Sedimentary geochemistry, δ Al indices, Nd isotopes, U-Pb ages, Detrital zircons, Sediment fluxes, Erosion rates, Kaolinite recycling, Chemical weathering.

“The man who drinks Zambezi waters must always return to drink again”
[Old Matabele saying in *The Leopard Hunts in Darkness*, Wilbur Smith]

1

2

Introduction

3

4 The Zambezi, the fourth longest river in Africa and the largest draining into the Indian Ocean (Fig. 1; Moore et al. 2007), underwent a still incompletely understood multistep evolution through time, 5 influenced by rifting events that punctuated the ~280-Ma-long break-up history of Gondwana (Key 6 et al. 2015). For large tracts the river flows along Permian-Triassic rift troughs and its general 7 eastward slope originated from domal uplift associated with the Early Cretaceous rifting of the South 8 Atlantic in the west (Cox 1989; Moore and Blenkinsop 2002).

10 The modern drainage developed through the Neogene as a consequence of surface uplift of the 11 broad Kalahari Plateau and southwestward propagation of the East African Rift, which created the 12 tectonic depressions occupied by Lake Malawi in the east and by the Okavango inland delta in the 13 west (Ebinger and Scholz 2012). After diverse events of river capture and drainage reversal, a 14 youthful lower course in Mozambique eroded backwards to eventually connect with the upper course 15 on the Kalahari Plateau, forcing it to plunge into Victoria Falls and the basaltic gorges downstream 16 (Wellington 1955; Moore and Larkin 2001). The drainage basin continued to expand in the 17 Quaternary, with the capture of the Angolan Kwando tributary and the presently incipient capture of 18 the large Okavango River as well (Gumbrecht et al. 2001).

19 In the Anthropocene, the course of the Zambezi ceased to be natural and was rigidly segmented 20 by the construction of the great dams that created Lake Kariba (1958) and Lake Cahora Bassa (1974). 21 Because the sediment-routing system (Hinderer 2012; Allen 2017) is strictly partitioned by these two 22 major reservoirs, it is here convenient to distinguish four reaches: the Uppermost Zambezi headwaters 23 as far as the Kwando confluence, the Upper Zambezi that includes Victoria Falls and the gorges as

24 far as Lake Kariba, the Middle Zambezi between the two reservoirs, and the Lower Zambezi
25 downstream of Lake Cahora Bassa (Fig. 1).

26 This study integrates the petrographic and heavy-mineral data illustrated and discussed in the
27 companion paper (Garzanti et al. 2021a) with new data on elemental geochemistry, Nd-isotope
28 geochemistry, clay mineralogy, and detrital-zircon geochronology from the same sample set. The two
29 articles in combination provide a multi-proxy characterization of sediment composition in the diverse
30 tracts of the large Zambezi catchment from the Zambian headwaters to the Mozambican coast. Our
31 main aims are to: a) refine provenance diagnoses based on diverse compositional parameters from
32 both mud and sand fractions of the sediment flux; b) unravel the relative effects of source-rock
33 lithology, recycling, hydraulic sorting, and chemical weathering on the mineralogical and chemical
34 composition of mud and sand generated in humid subequatorial to dry tropical climate; c) infer
35 sediment yields and erosion patterns in diverse parts of the large basin; d) evaluate the relative
36 importance of chemical weathering as occurring in present climatic conditions *versus* mineralogical
37 and chemical signatures of weathering inherited from past climatic conditions *via* recycling of detrital
38 components.

39
40

Geology

41 The Zimbabwe Craton represents the northern part of Archean southern Africa (Fig. 2A). A central
42 terrane flanked by two distinct greenstone belts includes gneisses non-conformably overlain by
43 volcanic rocks and conglomerates. The craton was stabilized between 2.7 and 2.6 Ga and eventually
44 sealed by the Great Dyke swarm at 2575 Ma (Kusky 1998; Jelsma and Dirks 2002; Söderlund et al.
45 2010).

46 Tectonic activity continued through the Paleoproterozoic, when the Proto-Kalahari Craton
47 formed during the major Orosirian episode of crustal growth, and into the Mesoproterozoic, at the
48 end of which the Kalahari Craton was eventually assembled (Hanson et al. 2006; Jacobs et al. 2008).
49 Orosirian orogens include the Ubendian-Usagaran Belts along the southern margin of the Tanzania

50 Craton, and the Magondi Belt, exposing arc-related volcano-sedimentary and plutonic rocks
51 metamorphosed at amphibolite facies along the northwestern margin of the Zimbabwe Craton
52 ([Majaule et al. 2001](#); [Master et al. 2010](#)). The Angola Block far to the west represents instead the
53 southern part of the Congo Craton. It comprises a Central Zone in the east, a Central Eburnean Zone,
54 and the Lubango Zone extending southwards into Namibia, which recorded several distinct magmatic
55 events between 2.0 and 1.77 Ga ([De Carvalho et al. 2000](#); [McCourt et al. 2013](#); [Jelsma et al. 2018](#)).

56 The next major episode of crustal growth is documented by the Irumide Belt extending from
57 southern Zambia to Malawi. The external nappes exposed to the north of the Luangwa Rift include a
58 2.0-1.9 Ga gneissic basement overlain by quartzite, schist, and minor carbonate deposited around
59 1.85 Ga (Muva Supergroup). Granitoid suites were emplaced at 1.65-1.52 Ga, 1.36-1.33 Ga, and 1.05-
60 0.95 Ga ([De Waele et al. 2003](#)). Regional metamorphism increasing southeastwards from greenschist
61 facies to upper amphibolite facies took place at 1.05-1.02 Ga ([De Waele et al. 2006, 2009](#)). The high-
62 grade internal zone is exposed to the north of the Lower Zambezi Valley between the Luangwa and
63 Shire Rivers (Southern Irumide Province; [Alessio et al. 2019](#)) and includes the Tete gabbro-
64 anorthosite Complex (~1.05 Ga; [Westerhof et al. 2008](#)). The Choma–Kalomo Block in southwestern
65 Zambia consists of crystalline basement covered by amphibolite-facies paragneiss and schist yielding
66 zircon grains of Paleoproterozoic age, intruded by two generations of Mesoproterozoic granitoid
67 plutons (1.37 and 1.18 Ga; [Bulambo et al. 2006](#)) and documenting a thermal event between 1.02 and
68 0.98 Ma ([Glynn et al. 2017](#)).

69 The Kalahari Craton of southern Africa was eventually welded to the Congo Craton during the
70 major Neoproterozoic Pan-African orogeny, testified by the Damara–Lufilian–Zambezi Belt
71 stretching from coastal Namibia to Mozambique ([Frimmel et al. 2011](#); [Fritz et al. 2013](#); [Goscombe
72 et al. 2020](#)). The Lufilian Arc consists of Neoproterozoic metasedimentary and metaigneous rocks
73 hosting Cu-Co-U and Pb-Zn mineralizations ([Kampunzu and Cailteux, 1999](#); [John et al. 2004](#);
74 [Eglinger et al. 2016](#)). The Zambezi Belt contains a volcano-sedimentary succession deformed under
75 amphibolite-facies conditions at 0.9-0.8 Ga ([Hanson 2003](#)), whereas eclogite-facies metamorphism

76 dated at 592 Ma constrains the timing of subduction and basin closure ([John et al. 2003](#)) with thrust
77 emplacement dated as 550-530 Ma ([Hargrove et al. 2003](#)).

78 The Gondwana supercontinent, assembled during the Pan-African Orogeny, started to be
79 disrupted towards the close of the Paleozoic, when Karoo sediments began to accumulate in several
80 depocenters across southern Africa. Karoo basins include the elongated Gwembe and Luangwa
81 troughs that control the drainage of the Middle Zambezi and Luangwa Rivers ([Nugent 1990](#)). The
82 several km-thick Karoo Supergroup begins with Upper Carboniferous to lowermost Permian
83 diamictite, followed by Permian to Middle Triassic turbidite and coal-bearing fluvio-deltaic strata.
84 Permian sandstones contain andesitic-dacitic volcanic detritus ([Johnson 1991](#)) and interlayered tuffs
85 yielding ages mainly between 270 and 260 Ma ([Lanci et al. 2013](#); [McKay et al. 2016](#)). Subsidence in
86 the southern retroarc basin was associated with subduction of paleo-Pacific lithosphere, while
87 transtensional stress propagated southwards from the Neotethyan rift in the north ([Catuneanu et al.](#)
88 [2005](#)). The upper part of the succession consists of Upper Triassic to Lower Jurassic braidplain
89 sandstones, floodplain mudrocks, and aeolian sandstones ([Johnson et al. 1996](#)). Karoo sedimentation
90 was terminated by flood-basalt eruptions recorded throughout southern Africa around 183 Ma
91 ([Svensen et al. 2012](#); [Greber et al. 2020](#)).

92 In the Early Cretaceous, opening of the Indian Ocean led to formation of sedimentary basins and
93 extensive volcanism in the Mozambique Channel. In the Cenozoic, fluvial and lacustrine sediments
94 including basal gravel and sand with calcrete were deposited inland in the Kalahari Basin, hosting the
95 largest continuous sand sea on Earth where repeated phases of eolian deposition took place through
96 the Quaternary ([Haddon and McCarthy 2005](#); [Burrough et al. 2019](#)). Southwestward propagation of
97 the East African rift in the late Neogene ([Daly et al. 2020](#)) eventually reached the Kalahari region,
98 where fault-related subsidence in the Okavango Rift is exerting a major control on drainage patterns
99 ([McCarthy et al. 2002](#); [Vainer et al. 2021](#)).

102 *Climate.* Southern Africa, with its largely rural population dependent on rain-fed subsistence
103 agriculture, is particularly vulnerable to climate variability and extreme events. These include severe
104 droughts affecting much of Zambia, Malawi, Zimbabwe, or northern South Africa, alternating with
105 devastating floods such as those periodically striking Mozambique. Cyclones occur in the wet season,
106 and for the first time in 2019 two cyclones caused floods and destruction in the same year (Siwedza
107 et al. 2021). Climate variability has a multitude of forcing factors that interact with each other and
108 wax and wane in their importance through time (Reason et al. 2006; Howard and Washington 2019).
109 Overall, rainfall increases from west to east at subtropical latitudes and from south to north at
110 subequatorial latitudes, the principal sources of humidity being the Indian Ocean in the east and the
111 Atlantic Ocean in the northwest (Fig. 2B).

112 The African continent divides the tropical high-pressure zone into the Indian Ocean and South
113 Atlantic anticyclones, particularly during the austral summer when heating of the landmass reaches
114 maximum. Moisture derived primarily from air masses moving inland from the warm Indian Ocean
115 is reduced by orographic effects along the eastern escarpment and declines progressively westward
116 resulting in increasing aridity. Descending, divergent air masses occur throughout the year along the
117 west coast, where the temperature inversion is reinforced by the northward-flowing Benguela current
118 and upwelling of cold Antarctic waters offshore. Dominant southwesterly winds are dry, and thus
119 contribute to the marked westward decrease in rainfall across the subcontinent. A sharp contrast thus
120 exists between humid coastal Mozambique (maximum annual rainfall 1.5 m) and the Kalahari Plateau
121 inland, where quasi-stationary anticyclonic conditions prevail (Fig. 2C). Mozambique is
122 characterized by tropical climate with a wet season from October to March and a dry season from
123 April to September (average annual rainfall ~0.65 m at Tete).

124 In subequatorial southern Africa, atmospheric circulation is complex. Moist South Atlantic air
125 moved inland by westerly winds converges with Indian Ocean air along the Congo Air Boundary,
126 frequently associated with development of pressure lows and widespread rains across the Kalahari
127 Plateau. Annual rainfall, chiefly associated with the southward shift of the Intertropical Convergence

128 Zone, reaches 1.4 m in Angola during summer (Jury 2010). In winter, when the Intertropical
129 Convergence Zone and Congo Air Boundary migrate northward, interior southern Africa remains
130 generally dry under the influence of the Indian Ocean anticyclone.

131 ***The River System.*** The Zambezi (length 2575 km, basin area $\sim 1.4 \cdot 10^6$ km²) is the largest river of
132 southern Africa. Sourced in northwesternmost Zambia among low ridges of the Kasai Shield (part of
133 the Congo Craton), the river traverses unconsolidated Kalahari sands in the Barotse floodplain, a 30-
134 50 km-wide wetland flooded for several months by Zambezi waters after the rainy season (peak
135 discharge in April).

136 The Uppermost Zambezi starts to flow more swiftly as it first encounters basaltic bedrock at
137 Ngonye Falls, and it is next joined by the Kwando River draining the Kalahari Basin in humid Angola.
138 While entering the Okavango Rift, the Kwando (here named Linyanti and next Chobe) deviates
139 sharply eastward along the tectonic depression hosting large swamps and once large paleolakes
140 (Burrough et al. 2009; Moore et al. 2012). The graben continues eastward into the Machili Flats
141 drained by the low-gradient Kasaya and Ngwezi Rivers, eastern Zambezi tributaries sourced in the
142 Lufilian arc and Choma-Kalomo Block, respectively.

143 Downstream of the Kwando confluence, the Upper Zambezi and its local tributaries — including
144 the Sinde River sourced in the Choma-Kalomo Block — incise into Karoo basaltic lavas and the
145 gradient steepens forming minor rapids upstream of Victoria Falls. Next, after plunging some 100 m
146 into the falls, turbulent Upper Zambezi waters design an astonishing zigzag into steep gorges of black
147 basalt, the result of progressive retreat of the waterfalls during the Quaternary (Derricourt 1976).
148 After receiving tributaries draining Karoo lavas overlain by thin Kalahari dune sand (e.g., Masuie and
149 Matetsi), the Zambezi reaches Lake Kariba shortly downstream of the confluence with the Gwai
150 River. Sourced in the Zimbabwe Craton, the low-gradient senile upper course of the Gwai River is
151 incised — as its east-bank tributaries Umguza and Shangani — in Karoo basalt and sedimentary rocks
152 surrounded by Kalahari dune sand (Thomas and Shaw 1988; Moore et al. 2009). In the youthful lower
153 tract, the Gwai cuts steeply across the Dete-Kamativi Inlier of the Paleoproterozoic Magondi Belt

154 (Glynn et al. 2020) and receives the eastern Tinde tributary mainly draining Pan-African molasse
155 (Goscombe et al. 2020).

156 The Middle Zambezi between Lakes Kariba and Cahora Bassa flows along a Karoo rift trough
157 formed along the Pan-African (Kuunga) suture zone (Goscombe et al. 2020) and hosting a thick infill
158 of basalt-capped Permian-Triassic sedimentary rocks overlying Ordovician to Devonian siliciclastic
159 strata (Nyambe 1999). The major tributaries in this tract are the Kafue and the Luangwa, both from
160 Zambia. The Kafue, the longest Zambezi tributary (length 1576 km, basin area 154,200 km²), is
161 sourced in the Lufilian arc, flows across a 240 km-long swampy flat floodplain, and next drops 550
162 m into a 60-km-long gorge carved in gneiss and metasedimentary rocks of the West Zambezi Belt to
163 join the Zambezi ~75 km downstream of Lake Kariba. The Luangwa (length 770 km, basin area
164 151,400 km²) is sourced in the Ubendian Belt and flows for most of its course along a Karoo rift
165 trough filled with an 8-km-thick Permian-Triassic sedimentary succession (Banks et al. 1995),
166 separating the external nappes of the Irumide Belt in the north from the high-grade Southern Irumide
167 Province in the south. After cutting across Southern Irumide granulites, the Luangwa joins the
168 Zambezi just upstream of Lake Cahora Bassa.

169 The Lower Zambezi in Mozambique receives tributaries from the west (Sangara, Mufa) and north
170 (Luia, Morrunguze) that largely drain high-grade Southern Irumide rocks. The largest tributary is the
171 Shire (*chiri* = steep banks), the outlet of Lake Malawi, which drains largely garnet-free mafic
172 granulites of the Blantyre domain (southern Malawi-Unango Complex) where middle-lower crust at
173 the southern margin of the Congo Craton underwent high-grade metamorphism at ~920 Ma
174 (Goscombe et al. 2020). From the west, the Mazowe and Luenha Rivers drain well into the Archean
175 Zimbabwe Craton in the headwaters. Downstream, the two rivers cut across the polymetamorphic
176 Mudzi migmatitic gneisses remobilized during the Pan-African orogeny, and next across the
177 Neoproterozoic Marginal Gneiss. Lowermost-course tributaries include the Sangadze and Zangue
178 Rivers sourced in the Pan-African Umkondo Belt, comprising greenschist-facies to lower-
179 amphibolite-facies schists thrust onto the margin of the Zimbabwe Craton and upper-amphibolite-

180 facies to granulite-facies gneisses in the core (Stenian Barue Complex). The northern Minjova
181 tributary largely drains the Moatize-Minjova Basin filled by coal-bearing Permian-Triassic Karoo
182 clastic rocks (Fernandes et al. 2015).

183 In the lowermost tract, the Zambezi River flows along the Lower Zambezi graben, originated as
184 a failed arm of the Middle Jurassic Mozambique Basin rift (Butt and Gould 2018). Zambezi sediments
185 have built through time the largest continental shelf along the Indian Ocean coast of Africa (Walford
186 et al. 2005; Ponte et al. 2019). Large sediment volumes, however, are not deposited in front of the
187 Zambezi mouth but dragged northeastward by longshore currents (Schulz et al. 2011; van der Lubbe
188 et al. 2014), forming wide beaches as far as Quelimane and beyond (e.g., Praia da Madal; Fig. 1).

189 **The Zambezi in the Anthropocene.** The course of the Zambezi has been profoundly modified
190 by man since the second half of the last century. The Kariba and Cahora Bassa Dams built on the
191 mainstem, as well as others built on major tributaries, have substantially altered the hydrological
192 regime of the Zambezi River and its delta, and disrupted the natural sediment-routing system
193 efficiently trapping detritus generated upstream (Davies et al. 2000; Beilfuss and dos Santos 2001;
194 Calamita et al. 2019). Lake Kariba (length 223 km, storage capacity 185 km³) is the world's largest
195 artificial reservoir, whereas Lake Cahora Bassa (length 292 km, storage capacity 73 km³) is Africa's
196 fourth-largest (Vörösmarty and Moore 1991).

197 Since Zambia's independence, two big dams have been built also on the Kafue River, at Itezhi-
198 Tezhi (“slippery rock”) and in the Kafue Gorge. The Itezhi-Tezhi Dam (storage capacity 6 km³,
199 completed in 1977) closes the gap through a ridge of ~100-m-high hills where the paleo-Kafue —
200 once flowing southwards towards Lake Makgadikgadi and the Limpopo River — was captured and
201 started to flow eastward as part of the Zambezi drainage (Thomas and Shaw 1991; Moore and Larkin
202 2001). Downstream, the river flows sluggishly in the maze of swampy channels and lagoons of the
203 Kafue Flats and next plunges into the Kafue Gorge, where other dams have been constructed (Upper
204 Kafue Gorge; storage capacity 0.8 km³, operational since 1973) or are under construction (Lower
205 Kafue Gorge). Other dams were built (e.g., Nkhula, Tedzani, Kapichira), or are planned, on the Shire

206 River in southern Malawi. Besides human intervention, the Zambezi sediment-routing system is
207 segmented by natural processes as well, much sediment being retained in large wetlands such as the
208 Barotse floodplain and Chobe swamps on the Uppermost Zambezi, the Kafue Flats, or the Elephant
209 Marsh on the Shire River (Bolton 1984; Moore et al. 2007).

210 *Sediment Fluxes.* Information on sediment loads transported both before and after the
211 construction of the big dams is largely missing throughout the Zambezi drainage basin. In the lack of
212 accurately gauged sediment fluxes, estimates on annual solid transport range widely between 20 and
213 100 million tons (Hay 1998), with a median value around 50 million tons (Milliman and Meade 1983;
214 ESIA 2011; Milliman and Farnsworth 2011). These figures correspond to an average annual sediment
215 yield and erosion rate between 15 and 70 tons/km² and between 0.005 and 0.03 mm (median values
216 ~35 tons/km² and ~0.01 mm). Based on cosmogenic nuclide data, the annual Uppermost Zambezi
217 sediment flux was estimated as 5.5±0.6 million tons (Wittmann et al. 2020), corresponding to a
218 sediment yield and erosion rate of 16±2 tons/km² and 0.006 ±0.001 mm. An annual sediment volume
219 of only 100,000 m³, ascribed to low topographic gradient and presence of vast wetlands on the
220 Kalahari Plateau, was estimated at Victoria Falls, whereas nearly half of the sediment flux (22 out of
221 51 million m³) was considered as generated in the Lower Zambezi catchment (FFEM 2005; van der
222 Lubbe et al. 2016). After closure of the Kariba and Cahora Bassa Dams, annual sediment supply to
223 the Zambezi Delta may have been reduced to as low as 0.8 million m³, between 1% and 7% of which
224 is bedload (Ronco et al. 2010 p.52; ESIA 2011).

225 Uncertain by an order of magnitude are also the estimates of sediment accumulation in Lake
226 Kariba (between 7 and 70 million tons according to Bolton 1984, but only ~4 million tons according
227 to Kunz et al. 2011) and Lake Cahora Bassa (between 20 and 200 million tons according to Bolton
228 1984 and 28.6 million m³ according to Ronco et al. 2010 p.47). Based on sparse data on sediment
229 concentration, annual sediment yields of 40 and 200 tons/km² were estimated for the Gwai and
230 Luangwa catchments, corresponding to average erosion rates of 0.015 and 0.075 mm (Bolton 1984).
231 Similar values were evaluated for minor tributaries in Zambia and Mozambique (200 tons/km²) and

232 for the Luangwa and the rest of the Middle Zambezi (170-250 tons/km², from sediment volumes of
233 14.0 and 14.6 million m³ respectively; [Ronco et al. 2010](#) p.47). Very high annual rates of soil loss (up
234 to 2900 tons/km²) are reported from the Shire catchment in southern Malawi ([Mzuza et al. 2019](#))

235
236

Methods

237 Between 2011 and 2019, 71 sediment samples were collected from active sand bars (57), levees (2)
238 and freshly deposited muds (12) of the Zambezi River and its major tributaries from the source in
239 northwesternmost Zambia to the delta in Mozambique. Full information on sampling sites is provided
240 in [Appendix Table A1](#) and Google Earth™ file [Zambezi2.kmz](#).

241 **Clay and Silt Mineralogy.** The mineralogy of six mud samples from the Middle and Lower
242 Zambezi mainstem and tributaries (Kafue, Sangadze, Shire) was determined on both < 32 µm and <
243 2 µm fractions by X-ray powder-diffraction (XRD) using a PANalytical Aeris equipment with a Cu
244 tube, at 15 kV, 40 mA. The < 32 µm fraction was separated by wet sieving and diffractograms were
245 performed on randomly oriented powder in the range 2-60° (2θ). The < 2 µm fraction, separated by
246 centrifuging according to Stokes' law, was analysed on oriented aggregates after air-drying (2-30°2θ)
247 and solvation with ethylene-glycol and heating at 550°C (2-15°2θ). Mineral proportions were
248 evaluated semi-quantitatively using diagnostic XRD peak areas ([Moore and Reynolds 1997](#); [Kahle et al. 2002](#)).
249 Further technical information is provided in [Dinis et al. \(2020a\)](#). XRD data previously
250 obtained on the < 32 µm fraction of mud samples collected from the Upper Zambezi catchment and
251 main tributaries including the Kwando and the Gwai ([Garzanti et al. 2014a](#); [Setti et al. 2014](#)) were
252 also considered. The mineralogical dataset is provided in [Table 1](#) and [Appendix Table A2](#).

253 **Mud and Sand Geochemistry.** Chemical analyses of 51 sediment samples were carried out at
254 Bureau Veritas Mineral Laboratories (Vancouver, Canada) on quartered aliquots of the < 32 µm (18
255 river muds) and 63-2000 µm fractions (31 river sands) obtained by wet sieving. Two levee silty sands
256 were analysed in bulk. Following a lithium metaborate/tetraborate fusion and nitric acid digestion,
257 major oxides and several minor elements were determined by ICP-ES and trace elements by ICP-MS

258 (see [Appendix A](#) and <http://acmelab.com> for detailed information on adopted procedures, standards
259 used, and precision for elements of group 4A-4B and codes LF200 and LF300).

260 Classic multi-element chemical indices used to estimate weathering and calculated using molar
261 proportions of mobile alkali and alkaline-earth metals include the WIP (Weathering Index; [Parker](#)
262 [1970](#)) and the CIA (Chemical Index of Alteration; [Nesbitt and Young 1982](#)). The WIP, however,
263 merely measures the amount of a set of mobile elements that decreases rapidly wherever quartz is
264 added to the sediment, making it an index of quartz recycling more than an index of weathering.
265 Because correcting the CIA for CaO in carbonates introduces uncertainties ([Garzanti and Resentini](#)
266 [2016](#)), in this study we use the CIA* corrected only for CaO in apatite [$\text{CIA}^* = 100 \cdot \text{Al}_2\text{O}_3 / (\text{Al}_2\text{O}_3$
267 $+ (\text{CaO} - 3.33 \times \text{P}_2\text{O}_5) + \text{Na}_2\text{O} + \text{K}_2\text{O})$]. The weathering effect is best detangled from other controls on
268 geochemical composition if mobile elements (Mg, Ca, Na, K, Sr, and Ba) are considered one by one.
269 This can be done by using $\alpha^{\text{Al}E}$ values, defined as $(\text{Al}/E)_{\text{sample}} / (\text{Al}/E)_{\text{standard}}$ ([Garzanti et al. 2013a](#),
270 [2013b](#)), which compare the concentration of any mobile element E with reference to non-mobile Al
271 in our samples *versus* an appropriately selected standard composition (e.g., the UCC, Upper
272 Continental Crust standard of [Taylor and McLennan 1995](#); [Rudnick and Gao 2003](#)). Aluminium,
273 hosted in a wide range of rock-forming minerals with diverse density, shape, and size including
274 phyllosilicates (concentrated in mud) and feldspars (concentrated in sand) is used as a reference for
275 all elements rather than Ti, Nd, Sm, or Th ([Gaillardet et al. 1999](#)), which are hosted preferentially in
276 ultradense minerals and thus may reach strongly anomalous concentrations owing to hydrodynamic
277 processes. Geochemical data are summarized in [Table 2](#) and provided in full in [Appendix Table A3](#).

278 ***Nd-Isotope Geochemistry.*** Sixteen samples from the Middle and Lower Zambezi catchment
279 were treated using a sequential leaching procedure for quantitative removal of carbonates, Fe-oxide
280 phases, and organic matter ([Bayon et al. 2002](#)). Prior to geochemical analyses, about 80 mg of
281 powdered samples were digested by HF-HCl-HNO₃ mixture (seven < 32 μm fractions, nine 32-63
282 μm classes, and two bulk samples) or alkaline fusion (fourteen 63-2000 μm fractions). Selected major
283 and trace-element abundances were determined at the Pôle Spectrométrie Océan (PSO) with a

284 Thermo Scientific Element XR sector field ICP-MS, using the Tm addition method (Barrat et al.
285 1996). Both the accuracy and precision of measured concentrations were assessed by analyzing three
286 certified reference materials (AN-G, AGV-1, BCR-1).

287 Neodymium isotopes were measured at PSO using a Thermo Scientific Neptune multi-collector
288 ICP-MS, after Nd purification by conventional ion chromatography. Repeated analyses of a JNdi-1
289 standard solution gave $^{143}\text{Nd}/^{144}\text{Nd}$ of 0.512114 ± 0.000005 (2σ , $n=12$), in full agreement with the
290 recommended value of 0.512115 (Tanaka et al. 2000) and corresponding to an external
291 reproducibility of $\pm 0.09 \epsilon$ (2σ). Epsilon Nd values were calculated using the present-day chondritic
292 (CHUR) value of $^{143}\text{Nd}/^{144}\text{Nd} = 0.512630$ (Bouvier et al. 2008). Neodymium depleted mantle model
293 ages ($T_{\text{Nd,DM}}$) were calculated following the approach described in De Paolo (1981), using measured
294 Sm and Nd concentrations ($^{147}\text{Sm}/^{144}\text{Nd} = \text{Sm}/\text{Nd} \times 0.6049$) and present-day depleted mantle values
295 of $^{143}\text{Nd}/^{144}\text{Nd} = 0.51315$ and $^{147}\text{Sm}/^{144}\text{Nd} = 0.2145$. Isotope data, including results previously
296 obtained on muds from the upper part of the Zambezi catchment (Garzanti et al. 2014a), are
297 summarized in Table 3. Geochemical data on all size fractions analysed for isotope geochemistry and
298 isotope-geochemistry data are provided in full in Appendix Tables A4 and A5.

299 ***Detrital-Zircon Geochronology.*** Detrital zircons were identified by Automated Phase Mapping
300 (Vermeesch et al. 2017) with a Renishaw inViaTM Raman microscope on the heavy-mineral separates
301 of 18 samples, concentrated with standard magnetic techniques and directly mounted in epoxy resin
302 without any operator selection by hand picking. U-Pb zircon ages were determined at the London
303 Geochronology Centre using an Agilent 7700x LA-ICP-MS (laser ablation-inductively coupled
304 plasma-mass spectrometry) system, employing a NewWave NWR193 Excimer Laser operated at 10
305 Hz with a 25 μm spot size and $\sim 2.5 \text{ J}/\text{cm}^2$ fluence. No cathodo-luminescence imaging was done, and
306 the laser spot was always placed “blindly” in the center of zircon grains in order to treat all samples
307 equally and avoid bias in intersample comparison (“blind-dating approach” as discussed in Garzanti
308 et al. 2018). The mass spectrometer data were converted to isotopic ratios using GLITTER 4.4.2
309 software (Griffin et al. 2008), employing Plešovice zircon (Sláma et al. 2008) as a primary age

310 standard and GJ-1 ([Jackson et al. 2004](#)) as a secondary age standard. A NIST SRM612 glass was used
311 as a compositional standard for the U and Th concentrations. GLITTER files were post-processed in
312 R using IsoplotR 2.5 ([Vermeesch 2018](#)). Concordia ages were calculated as the maximum likelihood
313 intersection between the concordia line and the error ellipse of $^{207}\text{Pb}/^{235}\text{U}$ and $^{206}\text{Pb}/^{238}\text{U}$ ([Ludwig](#)
314 [1998](#)). The discordance cutoff was set at -5/+15% of the concordia distance ([Vermeesch 2021](#)). The
315 complete geochronological dataset, comprising 2095 concordant ages refined with a ^{208}Pb -based
316 common Pb correction, is provided in [Appendix B](#).

317 ***Statistical methods and graphical displays.*** The relative contribution from each tributary or
318 geological domain to the sediment flux of a trunk river can be quantified with forward mixing models,
319 provided that the compositional signatures of sediment in each potential source are distinct and
320 accurately determined ([Weltje 1997](#); [Garzanti et al. 2012](#)). The forward mixing model calculates a
321 row vector of compositional data as a non-negative linear combination between a matrix of fixed end-
322 member compositions and a row vector of coefficients representing the proportional contribution of
323 each end member to the observation. The accuracy of calculations depends on how distinct and
324 precisely assessed the end-member signatures of each potential source are. Additional information on
325 the method is contained in [Appendix A](#) and [Resentini et al. \(2017\)](#).

326 Statistical techniques used to illustrate our datasets include the compositional biplot ([Gabriel](#)
327 [1971](#); [Aitchison and Greenacre 2002](#)) and multidimensional scaling (MDS; [Vermeesch 2013](#);
328 [Vermeesch and Garzanti 2015](#)). The compositional biplot (drawn using CoDaPack software by
329 [Comas and Thió-Henestrosa 2011](#)) allows discrimination among multivariate observations (points)
330 while shedding light on the mutual relationships among multiple variables (rays). The length of each
331 ray is proportional to the variance of the corresponding variable; if the angle between two rays is 0°
332 or 180° , then the corresponding variables are perfectly correlated or anticorrelated. MDS analysis
333 produces a map of points in which the distance among samples is approximately proportional to the
334 Kolmogorov-Smirnov dissimilarity of their compositional or chronological signatures. The goodness
335 of fit is evaluated using the “stress” value of the configuration (20 = poor; 10 = fair; 5 = good; [Kruskal](#)

336 and Wish 1978). The provenance package of Vermeesch et al. (2016) was used to plot MDS maps
 337 and U-Pb age distributions as kernel density estimates (KDE).

338
 339

Data

340 This section briefly summarizes petrographic and heavy-mineral data (provided in full in Appendix
 341 Tables A6 and A7, and illustrated in detail in the companion paper Garzanti et al. 2021a) and presents
 342 new data on clay mineralogy (Table 1; Fig. 3), elemental geochemistry for mud and sand samples
 343 (Table 2; Fig. 4), REE geochemistry (Fig. 5), Nd-isotope geochemistry (Table 3; Fig. 6), and detrital-
 344 zircon geochronology (Table 4; Fig. 7) from the whole Zambezi River sedimentary system.

345 Five main zircon-age ranges recur among the analysed samples, corresponding to main thermal
 346 events of crustal growth across southern Africa (Hanson 2003; Dirks et al. 2009; Andersen et al. 2016,
 347 2018): A) "Karoo" (Triassic-Permian, 253 ± 21 Ma); B) "Pan-African" (Cambrian-Ediacaran, 571 ± 22
 348 Ma, and Tonian, 792 ± 54 Ma); C) "Irumide" (Stenian, 1036 ± 32 Ma); D) "Eburnean" (Orosirian,
 349 1947 ± 70 Ma); and, E) "Limpopo" (late Neoproterozoic, 2568 ± 47 Ma) (Table 4). A few zircon ages
 350 referred to the Ectasian "Kibaran" (~ 1.35 Ga) or Calymmian/Statherian "Lukamfwa" events (~ 1.55
 351 Ga; De Waele et al., 2003) are recorded mainly in the Upper Zambezi catchment and Luangwa sand.
 352 The youngest zircon ages (103-121 Ma) were obtained from lowermost Zambezi sands.

353 ***The Uppermost Zambezi.*** Uppermost Zambezi and Kwando sands are pure quartzose. Sands of
 354 left Zambian tributaries range from quartz-rich feldspatho-quartzose (Kabombo, Ngwezi) to pure
 355 quartzose (Kasaya). Smectite prevails over kaolinite and mica/illite in mud of the Zambezi mainstem,
 356 and is dominant in Ngwezi and Kwando muds (Fig. 3).

357 In Zambezi and Kwando sands, SiO₂ is overwhelming (> 98%) (Fig. 4A) and other elements very
 358 low, including Zr (62-114 ppm) and REE (Fig. 5). The CIA* is ≥ 77 , the WIP < 1, $\alpha^{Al}Ca \geq 3$, and $\alpha^{Al}Na$
 359 ~ 4 . Ngwezi sand is less SiO₂-rich, with higher Al, Ca, Na, K, Rb, Sr, and Ba but very low Mg
 360 (Fig. 4B). Kasaya sand has intermediate composition. Chemical indices are CIA* 83, $\alpha^{Al}Ca$ 4, $\alpha^{Al}Na$
 361 21 for Zambezi mud and CIA* 77 ± 1 , $\alpha^{Al}Ca$ 2.9 ± 0.5 , and $\alpha^{Al}Na$ 11 ± 4 for Kasaya and Ngwezi muds.

362 Kwando mud has anomalously low $\alpha^{\text{Al}}\text{Ca}$, $\alpha^{\text{Al}}\text{Mg}$, $\alpha^{\text{Al}}\text{Sr}$, and $\alpha^{\text{Al}}\text{Ba}$ (Table 2). The general observed
 363 order of element mobility is $\text{Na} \gg \text{Sr} > \text{Ca} > \text{Mg} \approx \text{K}$ for mud and $\text{Na} \approx \text{Ca} > \text{Sr}$ for sand. In mud
 364 samples, REE patterns normalized to CI carbonaceous chondrites (Barrat et al. 2012) display classical
 365 shapes with higher LREE than HREE fractionation and moderately negative Eu anomaly. Pure
 366 quartzose sands display slightly stronger LREE enrichment, negative Ce anomaly, more strongly
 367 negative Eu anomaly, and low HREE fractionation. The ϵ_{Nd} values vary between -14 and -17 (-15.5
 368 for Uppermost Zambezi mud; Table 3).

369 Ages of zircon grains in Uppermost Zambezi, Kwando, and Kasaya sands display polymodal
 370 spectra, with mostly Cambrian to Stenian ages including a main Irumide and subordinate Pan-African
 371 peaks (Fig. 7). Orosirian ages are common in Uppermost Zambezi and Kwando sands and minor in
 372 Kasaya sand. Kwando sand is distinguished by a Neoproterozoic age cluster, whereas Kasaya sand
 373 yielded several zircon grains with Devonian to Triassic ages.

374 *The Upper Zambezi.* Zambezi sand becomes rapidly enriched in basaltic detritus downstream of
 375 Victoria Falls. Bedload sand and levee silty sand upstream of Lake Kariba are, respectively, quartzose
 376 and plagioclase-rich litho-feldspatho-quartzose with mafic volcanic rock fragments and green augite.
 377 Basaltic detritus increases from west to east also in Zambezi tributaries, from quartzose Sinde sand
 378 to lithic-rich litho-quartzose and quartzo-lithic Masuie and Matetsi sands containing common augite.
 379 Smectite predominates over mica/illite and kaolinite in Zambezi mud and is overwhelming in muds
 380 of the Sinde and Matetsi tributaries, which contain kaolinite (Fig. 3).

381 Silica decreases progressively along the Upper Zambezi, with corresponding increase in most
 382 other elements, including Fe, Mg, Ca, Na, Sr (Fig. 4G, 4H) and REE (Fig. 5C), but not Zr, Hf, and
 383 Nb. Chemical indices upstream of Lake Kariba are $\text{CIA}^* 59 \pm 6$, $\alpha^{\text{Al}}\text{Ca} 0.9 \pm 0.3$, $\alpha^{\text{Al}}\text{Na} 3.9 \pm 0.9$ for
 384 mud, $\text{CIA}^* 45$, $\alpha^{\text{Al}}\text{Ca} 0.5$, $\alpha^{\text{Al}}\text{Na} 1.5$ for silty sand, and $\text{CIA}^* 49$, $\alpha^{\text{Al}}\text{Ca} 0.9$, $\alpha^{\text{Al}}\text{Na} 1.2$ for sand.
 385 Tributaries draining progressively larger portions of Karoo basalts display an even sharper trend from
 386 west to east. Masuie and Matetsi sands have much lower SiO_2 than Sinde sand and higher
 387 concentration of all other elements (Fig. 4A). In these rivers, both mud and sand are markedly

388 enriched in Mg, Ca, Sc, Ti, V, Cr, Fe, Mn, Co, Ni, Cu (Fig. 4B), and display regular chondrite-
 389 normalized REE patterns lacking Eu anomaly (Fig. 5A, 5D, and 5G). The ϵ_{Nd} value is around -12 in
 390 Zambezi mud between Victoria Falls and Lake Kariba, and is much less negative for Matetsi (-4) and
 391 Sinde (-5) muds.

392 Sinde sand yielded a zircon-age spectrum with dominant Irumide peak, common Neoproterozoic
 393 ages, and minor Permian-Triassic, Eburnean, and Neoproterozoic ages. Upper Zambezi sand upstream
 394 of Lake Kariba yielded a polymodal spectrum with main Pan-African and Irumide peaks and minor
 395 Orosirian and Neoproterozoic clusters (Fig. 7).

396 Lower Gwai sand is feldspatho-quartzose with biotite and amphibole. Smectite predominates in
 397 upper Gwai and Umguza muds, whereas mica/illite prevails over kaolinite and smectite in mud of the
 398 lower-course Tinde tributary (Fig. 3). Chemical indices are CIA* 73, $\alpha^{Al}Ca$ 1.9, $\alpha^{Al}Na$ 10 for upper
 399 Gwai mud and CIA* 54, $\alpha^{Al}Ca$ 2.9, $\alpha^{Al}Na$ 1.4 for lower Gwai sand. Umguza mud is similar as upper
 400 Gwai mud, whereas Umguza sand is much richer in Fe, Mg, Ca, Ti, Mn, Sr, V, Co, Ni, and Cu than
 401 lower Gwai sand (Fig. 4C, 4D). Tinde mud is richer in Si, Na, K, Rb, Ba, YREE, Th, U, Zr, Hf, Nb,
 402 Ta, and poorer in Fe, Mg, Ca, P, Mn, Sc, V, Cr, Co, Ni, and Cu (Table 2). The ϵ_{Nd} value ranges
 403 between -12 and -15 in upper Gwai, Umguza and Tinde muds, but it is strongly negative in Gwai
 404 sand upstream of lake Kariba (-25), which yielded a polymodal zircon-age spectrum with major
 405 Neoproterozoic and Eburnean peaks and minor Irumide and Pan-African peaks (Fig. 7).

406 ***The Middle Zambezi.*** Between Lakes Kariba and Cahora Bassa, Zambezi sand has the same
 407 feldspar-rich feldspatho-quartzose composition as Kafue sand, with metamorphic rock fragments,
 408 mica, and amphibole. The Luangwa River carries feldspatho-quartzose sand with granitoid to gneissic
 409 rock fragments and amphibole. Smectite predominates over mica/illite and kaolinite in mud of the
 410 Zambezi mainstem, whereas mica/illite predominates over smectite and kaolinite in Kafue mud (Fig.
 411 3).

412 Chemical indices are CIA* 67 ± 3 , $\alpha^{Al}Ca$ 2.1 ± 0.6 , $\alpha^{Al}Na$ 5.2 ± 0.2 for mud and CIA* 51 ± 3 , $\alpha^{Al}Ca$
 413 2.1 ± 1.3 , $\alpha^{Al}Na$ 1.6 ± 0.2 for sand. The observed order of element mobility is Na > Sr > Ca > K > Ba

414 for mud and $\text{Ca} > \text{Na} > \text{Sr}$ for sand. Luangwa sand is higher in SiO_2 , K, Ba, and lower in most other
 415 elements (especially Mg, Ti, and Sc; Fig. 4C). The finer-grained of the two Kafue sand samples is
 416 notably enriched in Zr, Hf, REE, Th, U, Nb, Ta (Table 2). Kafue and Middle Zambezi muds have
 417 virtually identical chemical composition (Fig. 4D, 4H). All ϵ_{Nd} values range between -14 and -18,
 418 reaching -20 only in the 32-63 μm size class of Luangwa sand.

419 The zircon-age spectrum of Kafue sand displays a dominant Irumide peak with a minor Pan-
 420 African cluster and a few Triassic and Paleoproterozoic to Neoproterozoic ages. Luangwa sand is
 421 characterized by a trimodal spectrum with major Irumide, subordinate Pan-African, and minor
 422 Eburnean peaks, and a few Permian ages (Fig. 7).

423 ***The Lower Zambezi.*** In Mozambique, Zambezi sand ranges from quartzo-feldspathic to feldspar-
 424 rich feldspatho-quartzose with biotite, amphibole, and garnet. Most tributaries carry quartzo-
 425 feldspathic sand with amphibole, garnet, clinopyroxene, hypersthene, and epidote. Feldspar (mostly
 426 plagioclase) is twice as abundant as quartz in Shire sand from Malawi. Metabasite grains are common
 427 in Morrunguze sand. Kaolinite, mica/illite, and smectite occur in subequal amount in Zambezi mud
 428 collected at Tete, whereas smectite predominates over mica/illite and kaolinite is subordinate
 429 upstream of the delta (Fig. 3). Sangadze mud consists almost exclusively of smectite, whereas Shire
 430 mud contains mica/illite and kaolinite in subequal proportions (Table 1).

431 Chemical indices are remarkably constant in sediments of the Zambezi mainstem and most of its
 432 main tributaries ($\text{CIA}^* 70 \pm 3$, $\alpha^{\text{Al}} \text{Ca} 2.0 \pm 0.4$, $\alpha^{\text{Al}} \text{Na} 6.0 \pm 2.6$ for mud and $\text{CIA}^* 50.4 \pm 0.5$, $\alpha^{\text{Al}} \text{Ca}$
 433 1.6 ± 0.5 , $\alpha^{\text{Al}} \text{Na} 1.2 \pm 0.2$ for sand) (Table 2). Plagioclase abundance explains why an Eu anomaly is
 434 not shown (Fig. 5H, 5I). The observed order of element mobility is $\text{Na} > \text{Sr} > \text{Ca} > \text{K} > \text{Rb}$ for mud
 435 and $\text{Ca} > \text{Na} > \text{Sr}$ for sand. In all grain-size fractions of Lower Zambezi sediments, ϵ_{Nd} values become
 436 much more negative from Tete to upstream of the delta, where the very-fine-sand sample yielded a
 437 less negative value than the fine-sand sample (Table 3).

438 Sand of the Morrunguze River draining gabbroic rocks of the Tete Complex is low in SiO_2 (52%),
 439 K, and Rb, notably rich in Fe, Mg, Ca, Ti, Mn, Sc, V, Cr, Co, Ni and Cu (Fig. 4E), and yielded the

440 least negative ϵ_{Nd} value (Fig. 6). Mufa sand, enriched in the same elements but to a much lesser extent,
441 yielded high Zr and Hf concentrations (Table 2) and a more negative ϵ_{Nd} value. Luenha sand is
442 relatively rich in Zr and Hf, U, Nb, Ta and REE, is the richest in Th (Fig. 4E), displays the steepest
443 REE patterns with the largest negative Eu anomaly (Fig. 5H, 5K), and is the only sample with negative
444 loss on ignition (LOI -0.3). Mazowe and Luenha sands have ϵ_{Nd} values of -18 and -19. Shire sand is
445 highest in Al, Na, Sr, and P, and shows a much less negative ϵ_{Nd} value (Fig. 7). Sangadze sand is the
446 richest in K, Rb, and Ba, and displays a strongly positive Eu anomaly (Fig. 5H) — reflecting abundant
447 feldspar and fewer heavy minerals — and a strongly negative ϵ_{Nd} value. Shire mud is low in SiO_2
448 (42%) and highest in Al, Fe, Sr, Ba, P, Sc, V, and Cu (Fig. 4F). Zambezi mud upstream of the delta
449 is high in Zr and Hf, REE, Th, U, Nb, Ta, Cr, Mo, W, Co, and Ni (Fig. 4H), and displays a negative
450 Eu anomaly (Fig. 5I).

451 The U-Pb age spectrum of zircon grains supplied by the Zambezi River to the delta displays a
452 dominant Irumide peak, with common Neoproterozoic, some Orosirian, and a few Neoproterozoic and
453 late Paleozoic ages (Fig. 7). Sangara and Morrunguze zircons show a unimodal Irumide peak, which
454 is associated with minor Neoproterozoic ages in Mufa sand. Luenha and Mazowe sands yielded nearly
455 identical bimodal zircon-age spectra with Neoproterozoic and Neoproterozoic peaks (Table 4). The
456 spectrum of Shire sand is also bimodal but with Irumide and Pan-African peaks. Pan-African ages
457 are more common than Irumide ages in Zangue sand (Fig. 7).

458 ***The Northern Zambezi Delta.*** Estuary and beach sand ~100 km north of the Zambezi mouth is
459 feldspatho-quartzose with amphibole, epidote, and clinopyroxene. The ϵ_{Nd} value of bulk sand ranges
460 between -13 and -18. The U-Pb zircon-age spectrum displays a dominant Irumide peak with common
461 Neoproterozoic, some Orosirian and a few Neoproterozoic and Permian ages, similarly to Lower
462 Zambezi sand (Fig. 7).

463

464

Provenance Insights from Clay Mineralogy and Sediment Geochemistry

465 Clay mineralogy and sediment geochemistry are largely controlled by factors other than provenance.
466 If weathering is intense, then they reflect the lithology of source rocks only poorly, which explains
467 why they have long been used to evaluate weathering rather than provenance (e.g., [Nesbitt and Young](#)
468 [1982](#); [Velde and Meunier 2008](#)). However, despite the complexities associated with multiple controls
469 on sediment composition ([Johnsson 1993](#)), clay minerals and elemental and especially isotope
470 geochemistry do offer complementary information useful to augment the completeness and
471 robustness of provenance analysis for several reasons ([McLennan et al. 1993](#)). Firstly, most other
472 provenance techniques are best suited to tackle sand and, in the case of detrital geochronology, only
473 a millesimal fraction of total sand. Geochemistry, instead, can be applied to bulk-sediment samples
474 of any size fraction from clay to granule. This allows us to investigate almost the entirety of the
475 sediment flux, including clay and silt that constitute the large majority of the detrital mass transported
476 in river systems as suspended load (e.g., [Hay 1998](#); [Milliman and Farnsworth 2011](#)). The aim of this
477 section is thus to complement previous considerations based only on sand with inferences derived
478 independently from the mineralogy and geochemistry of mud.

479 ***Clay Minerals.*** The clay mineralogy of unconsolidated mud depends on weathering processes in
480 soils but reflects provenance as well, especially in dry climates where illite and chlorite are largely
481 derived from phyllosilicate-rich metamorphic bedrock whereas smectite is shed by mafic lava (e.g.,
482 [Chamley 1989](#)). Among the studied samples, smectite is the dominant clay mineral ($\geq 87\%$) in mud
483 transported by the Sinde and Matetsi tributaries of the Upper Zambezi and by the Umguza tributary
484 of the Gwai River, all partly draining Karoo basalt between southern Zambia and western Zimbabwe.
485 Smectite, however, is produced in abundance also in regions lacking significant exposures of mafic
486 rocks (e.g., Kwando catchment) and represents the virtually exclusive clay mineral in mud of the
487 Sangadze River flowing across Mozambican lowlands, indicating incomplete flushing of mobile ions
488 in poorly drained low-relief regions ([Wilson 1999](#)). Illite is the most abundant clay mineral in mud
489 of the Kafue and Shire Rivers chiefly draining Proterozoic crystalline basement, and in mud of the
490 Tinde River draining Neoproterozoic molasse ([Fig. 3](#)).

491 *Sand Geochemistry*. The piece of provenance information most readily obtained from
492 geochemical data is the supply from mafic rocks, revealed by high concentrations of ferromagnesian
493 metals including Mg, Sc, Ti, V, Cr, Mn, Fe, Co, and Ni (Fig. 8; McLennan et al. 1993; von Eynatten
494 et al. 2003). Among the analysed samples, these elements reach the highest values in Masuie and
495 Matetsi sands draining Karoo basalts and in Morrunguze sand draining the Tete gabbro-anorthosite
496 (Fig. 4). Intermediate values for these elements are obtained for the Sinde and Umguza Rivers and
497 for the Middle Zambezi upstream of Lake Kariba, all draining Karoo basalts more marginally (Table
498 2). Other samples in the Upper Zambezi catchment have $\text{SiO}_2 > 90\%$, revealing extensive recycling
499 of pure quartzose Kalahari sand.

500 The virtually identical chemical composition of Kafue and Middle Zambezi muds confirms that
501 the Kafue is by far the most important source of sediment to the Middle Zambezi between Lake
502 Kariba and the Luangwa confluence. Slightly above 80% in Kafue and Middle Zambezi sand, SiO_2
503 raises to nearly 90% in Luangwa sand that contains a greater proportion of detritus recycled from
504 Karoo siliciclastic strata. In sand of Lower Zambezi tributaries, SiO_2 mostly ranges between 70% and
505 80%. Composition is closest to the UCC for sand carried by the Lower Zambezi to the Indian Ocean,
506 confirming its dominantly first-cycle provenance from mid-crustal basement rocks (Garzanti et al.
507 2021a).

508 Chemical indices provide further clues. Because the addition of quartz grains profoundly affects
509 the WIP, but not the CIA*, the CIA*/WIP ratio can be used to detangle the effects of weathering and
510 recycling. This ratio reaches ≥ 100 in Uppermost Zambezi and Kwando sands consisting almost
511 entirely of recycled Kalahari dune sand, decreases to 30 ± 9 in Upper Zambezi sand above and below
512 Victoria Falls, and next drastically to 3.1 ± 0.7 in Upper Zambezi sand and silty sand upstream of Lake
513 Kariba. The CIA*/WIP ratio decreases further to 1.7 ± 0.3 in sand of Middle Zambezi tributaries and
514 mainstem and is lowest (0.9 ± 0.1) in sand of Lower Zambezi tributaries and mainstem, further
515 confirming their mostly first-cycle provenance.

516 **Mud Geochemistry.** The geochemical composition of mud samples is more homogeneous. SiO₂
517 varies between 42% and 54%, being notably higher (68%) only for Tinde mud reflecting recycling
518 of siliciclastic Pan-African molasse. Kwando mud is markedly enriched in Ca, Sr, Mg, and Ba (Fig.
519 4B), revealing contribution from calcrete and dolocrete soils (Shaw 2009; McFarlane et al. 2010). Fe,
520 Ti, Sc, V, Co, and Cu are highest in Sinde mud and Mg in Matetsi mud (Fig. 4A, 4B) largely derived
521 from Karoo basalt. Lower Zambezi mud upstream of the delta is richest in Cr and Ni (Fig. 4H),
522 suggesting significant supply from mafic Proterozoic rocks including the Tete gabbro-anorthosite.

523 **Nd-Isotope Geochemistry.** Although the ¹⁴³Nd/¹⁴⁴Nd composition of sediments is controlled by
524 multiple factors, the provenance signal emerges clearly from data obtained from all analysed size
525 fractions — cohesive mud (< 32 μm), very coarse silt (32-63 μm), and sand (63-2000 μm) —,
526 allowing a sharp distinction between sediments derived from mafic igneous rocks and old granitoid
527 basements (Fig. 9).

528 The least negative ε_{Nd} values characterize Morrunguze sand, largely derived from the upper
529 Stenian Tete gabbro-anorthosite, and Sinde and Matetsi muds, partly derived from Lower Jurassic
530 Karoo basalt (Fig. 6). Mildly negative values were also obtained from Mufa sand, partly derived from
531 Tete mafic rocks, from Shire sediments largely derived from mafic granulites of the Blantyre domain,
532 and from Lower Zambezi silt collected at Tete and mostly derived from the Southern Irumide
533 Province (Table 3).

534 At the other extreme, most negative ε_{Nd} values identify Gwai, Mazowe, and Luenha sands
535 sourced from Archean gneiss of the Zimbabwe Craton. Markedly negative values also characterize
536 mud in the upper catchments of the Zambezi and Gwai Rivers. Epsilon Nd becomes less negative as
537 the Upper Zambezi rushes through the basaltic gorges downstream of Victoria Falls, but much more
538 negative as the Lower Gwai cuts steeply across gneisses of the Dete-Kamativi Inlier. Strongly
539 negative values also characterize Kafue and Luangwa sediments in the middle catchment, Sangadze
540 sediment in the lowermost catchment, and Lower Zambezi sediment upstream of the delta.

541 *Intrasample Variability.* The grain-size-controlled intrasample variability of ϵ_{Nd} values is limited
542 (average standard deviation 1.6 ± 0.9). The sand fraction (63-2000 μm) typically yields a more
543 negative ϵ_{Nd} than the cohesive mud fraction ($< 32 \mu\text{m}$). The most negative ϵ_{Nd} values are obtained
544 from the 32-63 μm size class of sand samples, representing the fine tail of the size distribution where
545 ultradense minerals including LREE-rich monazite concentrate because of the settling-equivalence
546 effect (Rubey, 1933). This is documented by La, Th, and Zr concentrations respectively 7 ± 5 , 9 ± 7 ,
547 and 6 ± 3 times higher in the 32-63 μm size class than in the 63-2000 μm fraction of sand samples,
548 with enrichment factors increasing sharply with sample grain size (corr. coeff. 0.91; data from
549 [Appendix Table A4](#)). The opposite holds for the Lower Zambezi silt collected at Tete ([Fig. 6](#)), where
550 the 32-63 μm size class is part of the coarse tail of the size distribution, which is depleted in denser
551 minerals and yields a less negative ϵ_{Nd} than both $< 32 \mu\text{m}$ and 63-2000 μm fractions ([Table 3](#)).

552 *Intersample Variability.* Samples collected in nearby localities along the Kafue and Lower
553 Zambezi Rivers confirm a tendency towards more negative ϵ_{Nd} values with increasing grain size.
554 Strong grain-size control on ϵ_{Nd} has been recently documented in Congo Fan sediments, where it was
555 ascribed to the more negative ϵ_{Nd} carried by multiply recycled quartz grains originally derived from
556 older terrains on average (figs. 6 and 8 in [Garzanti et al. 2021b](#)). In recycled Congo Fan sediments,
557 however, quartz and monazite account for similar amounts of Nd, quartz being 10^5 times more
558 abundant in weight but monazite containing 10^5 times more Nd. In first-cycle Lower Zambezi
559 sediments, instead, heavy minerals (including monazite) are an order of magnitude more abundant
560 than in Congo sediments, and are thus expected to dominate the REE budget ([Totten et al. 2000](#)).
561 Among minerals frequently found in sediments, only monazite and allanite display very high LREE
562 content, monazite being about three times richer in LREE. In our sample set, monazite resulted to be
563 about five times more abundant than allanite, which implies an order-of-magnitude greater
564 contribution to the LREE sedimentary budget by monazite than by allanite. The tendency of ϵ_{Nd} values
565 to become more negative in sand, and more markedly in the 32-63 μm size class of Middle and Lower
566 Zambezi sand samples, thus suggests that ultradense monazite, strongly concentrated in the fine tail

567 of the size distribution because of the settling-equivalence effect, carries an older (more negative) ϵ_{Nd}
 568 signal than other less dense and less durable minerals (e.g., allanite, titanite, apatite, epidote, and
 569 amphibole).

570 *Model Ages.* Depleted mantle model ages ($t_{Nd,DM}$) for the Zambezi mainstem and tributaries
 571 mainly range between 2.0 and 2.3 Ga (Fig. 6). Mud partly derived from Karoo basalt yielded notably
 572 younger values (1.6 Ga for Sinde and 1.45 Ga for Matetsi), as all size fractions of Shire sediment
 573 largely derived from mafic granulites of the Blantyre domain (1.25-1.45 Ga). Morrunguze sand
 574 largely derived from gabbroic rocks of the Tete domain yielded the youngest value (1.16 Ga), whereas
 575 the oldest values (2.5-2.8 Ga) characterize the three rivers sourced in the Archean Zimbabwe Craton
 576 (Gwai, Mazowe, and Luenha) (Fig. 6).

577 Sm-Nd model ages depend on REE fractionation, being lower in samples containing LREE-rich
 578 minerals such as monazite or allanite and higher in samples containing MREE-rich minerals such as
 579 xenotime, titanite or apatite (fig. 9 in Garzanti et al. 2021b). Because of the settling-equivalence
 580 effect, ultradense monazite grains occur only in the fine tail of the size distribution, which explains
 581 why the 32-63 μm size class of the cohesive Zambezi silt collected at Tete has a higher Sm/Nd ratio
 582 and thus yielded a notably higher Sm-Nd model age than the $<32 \mu m$ fraction despite its slightly
 583 lower ϵ_{Nd} (Fig. 6). In the two Zambezi samples upstream of the delta, the 63-2000 μm fraction has
 584 lower LREE fractionation (higher Sm/Nd) than both 32-63 μm and $< 32 \mu m$ fractions, and thus
 585 yielded older Sm-Nd model ages than the $< 32 \mu m$ fraction despite ϵ_{Nd} values are similar, and almost
 586 the same Sm-Nd model ages than the 32-63 μm size class despite the latter yielded notably more
 587 negative ϵ_{Nd} (Fig. 6).

588

589 *Provenance Insights from Detrital-Zircon Geochronology*

590 Age spectra of detrital zircons enrich provenance information by providing insight into events of
 591 crustal growth in diverse source-rock domains (Fig. 10). In the general case, however, these can only
 592 be considered as “protosources” (Andersen et al. 2016, 2018). They represent the true sediment source

593 only in the specific case of first-cycle detritus supplied directly from igneous or metamorphic
594 basement (Dickinson et al. 2009). This introduces a further major uncertainty in zircon-based
595 provenance analysis, because most sedimentary basins are fed with a mixture of recycled and first-
596 cycle sediments in a proportion that can be evaluated only roughly from independent compositional
597 data.

598 Petrographic, mineralogical, and geochemical data collectively concur to reveal that Uppermost
599 Zambezi sand is dominantly recycled from Kalahari dune fields (Garzanti et al. 2014b). The Kwando
600 River in particular drains entirely within the Kalahari Basin, whereas some first-cycle detritus is
601 supplied by Zambian tributaries sourced in the Lufilian arc or Choma-Kalomo Block. In sharp
602 contrast, Lower Zambezi sand is mostly first-cycle and derived from igneous and metamorphic
603 Precambrian basements rejuvenated during Neogene southward propagation of the East African Rift.

604 **Zircon Ages.** Geochronological analysis indicates the Irumide and Pan-African crustal domains
605 as the main protosources of zircon grains in rivers draining the northern Kalahari Plateau (Fig. 7).
606 Neoproterozoic ages ultimately derived from the Kasai Shield are common only for Kwando sand in the
607 west. Paleoproterozoic grains ultimately derived from the Angola Block are more common in
608 Kwando and Uppermost Zambezi sand than in eastern Zambian tributaries. Among these,
609 Paleoproterozoic zircons are more common in Sinde sand sourced in the Choma-Kalomo Block,
610 whereas Neoproterozoic zircons are much more abundant in Kasaya sand largely derived from the
611 Lufilian Arc. The Irumide peak is invariably prominent. Zircon grains yielding Permian to Triassic
612 Karoo ages constitute a minor population in Sinde sand but are lacking in Upper Zambezi sand
613 downstream of the basaltic gorges, indicating that zircon occurs in tuffs interlayered within Karoo
614 siliciclastic strata but not in the overlying Lower Jurassic basalt.

615 Despite extensive remobilization during the Pan-African orogeny (fig. 7 in Goscombe et al.
616 2020), the analysed center of zircon grains in sand of northern (Kafue, Luangwa, Morrunguze) and
617 western (Sangara, Mufa) tributaries to the Middle and upper Lower Zambezi yielded mostly or even
618 exclusively Irumide ages (Table 4). The dominant Irumide zircon-age peak displayed by Kafue sand

619 suggests that zircons sourced from the Neoproterozoic Lufilian Arc are retained in the Itezhi-Tezhi
620 Reservoir and/or in the Kafue Flats and do not reach the lower gorge, where Irumide-aged zircons
621 are derived largely from the Mpande Gneiss (~1.1. Ga; [Hanson et al. 1994](#); fig. 6 in [Goscombe et al.](#)
622 [2020](#)).

623 Late Neoproterozoic ages are lacking in sand of the Morrunguze River draining entirely within
624 the Southern Irumide Province but more abundant than Irumide ages in sand of the Shire and Zangue
625 tributaries joining the Zambezi upstream of the delta ([Fig. 7](#)). Archean zircons are common ($24\pm 2\%$)
626 in the Gwai, Mazowe, and Luenha Rivers sourced in the Zimbabwe Craton. Gwai sand also carries
627 ~30% of Paleoproterozoic zircons largely derived from the Dete-Kamativi Inlier cut across in the
628 lower course.

629 The multimodal age-spectrum of Zambezi zircons eventually supplied to the delta and dragged
630 by littoral currents to the northern Quelimane region indicates predominance of zircon grains derived
631 directly or indirectly from the Irumide belt, with subordinate late Neoproterozoic zircons mainly
632 supplied by geological domains most severely remobilized during the Pan-African orogeny ([Fig. 7](#)).
633 Orosirian zircons are frequent, Neoproterozoic grains minor, and Permian-Triassic zircons rare ([Table](#)
634 [4](#)).

635 A few Permian-Triassic zircons derived from Karoo volcanic or volcanoclastic layers are
636 identified in Kasaya, Sinde, Gwai Kafue, and Luangwa sands, but were not recorded from any Lower
637 Zambezi tributary. Yet, a few Permian-Triassic zircons occur in Lower Zambezi sand upstream of
638 the delta, suggesting recycling of Karoo strata exposed in the Moatize-Minjova basin. Jurassic or
639 Cretaceous ages are sporadically recorded. Among these, most significant are a couple of grains dated
640 as ~120 Ma in the terminal tract of the Lower Zambezi, indicating provenance from igneous rocks
641 emplaced during the incipient opening of the Mozambique Channel ([König and Jokat 2010](#); [Chanvry](#)
642 [et al. 2018](#)).

643 **Zircon Fertility.** The joint consideration of petrographic, mineralogical, and geochemical
644 datasets does not only offer a panorama of compositional signatures but also useful information to

645 evaluate the zircon fertility of sediment sources (e.g., diverse tributary catchments in the Zambezi
646 drainage system), which is required for a correct use of zircon-age data in the calculation of
647 provenance budgets. Ultradense zircon is invariably segregated in the fine tail of the size distribution
648 of each sample deposited by a tractive current and can be strongly concentrated locally by selective-
649 entrainment processes (Garzanti et al. 2008, 2009). These issues may hamper the accuracy of fertility
650 determinations based on mineralogical or geochemical data (Malusà et al. 2016).

651 In our sample set, sands from the upper part of the Zambezi catchment have a notably lower
652 concentration of zirconium (Zr 232 ppm in one trunk-river sample, but otherwise invariably below
653 the UCC standard and mostly in the 30-80 ppm range) than in the Middle and Lower Zambezi
654 catchment (between 330 and 600 ppm in Kafue and Mufa samples, and above the UCC also for the
655 Middle Zambezi, Luenha, and Shire samples). Zircon concentration, calculated by combining
656 petrographic and heavy-mineral point-counting data (Appendix Tables A6 and A7), is markedly
657 lower in the Uppermost to Upper Zambezi catchment (median of bulk sand in volume 0.02%,
658 maximum 0.2%), than in Middle to Lower Zambezi catchment (median 0.16%, maximum 0.6% for
659 Shire sand).

660 A most useful parameter to detect hydraulically controlled concentration of denser minerals is
661 the weighted average density of terrigenous grains in g/cm^3 (SRD index of Garzanti and Andò 2007),
662 which for each sample should be equal to the weighted average density of source rocks in the ideal
663 absence of environmental bias. The SRD index of most sands in rivers on the Kalahari Plateau ranges
664 between 2.65 and 2.68 (just a little higher than quartz density), increasing to 2.79 and 2.90 for Masuie
665 and Matetsi sands containing 50% and 70% of detritus from dense basaltic rocks. In the Middle to
666 Lower Zambezi catchment, SRD mostly ranges between 2.7 and 2.8, which matches the expected
667 density range for upper to middle crustal basement rocks (Garzanti et al. 2006). The finer-grained
668 Kafue (SRD 2.79) and Luenha (SRD 2.78) sands are those richest in elements preferentially hosted
669 in ultradense minerals (Fig. 4). Luenha sand has high LREE and Th, negative LOI, but only
670 moderately high Zr, indicating concentration of ultradense monazite and magnetite but only

671 moderately high zircon content (0.16% of bulk sand in volume; [Appendix Table A7](#)). The higher
 672 SRD values observed for Shire (2.82) and Morrunguze sands (2.87) reflect contribution from high-
 673 grade and largely mafic basement rocks of the Blantyre and Tete domains (figs. 2 and 7 in [Goscombe
 674 et al. 2020](#)). Ultradense garnet, zircon, monazite, and opaque Fe-Ti-Cr oxides have been markedly
 675 concentrated by selective entrainment processes only in Lower Zambezi sample S5778 (SRD 3.34),
 676 a fluvial garnet placer not analysed for either geochemistry or zircon geochronology.

677 Petrographic, heavy-mineral, and geochemical data converge to indicate that the zircon
 678 concentration in our samples provides a broadly reliable indication of zircon fertility in the
 679 corresponding catchments. Zircon fertilities are estimated to range from 0.02% for Kalahari dune
 680 sands to 0.2% for mid-crustal basement rocks exposed in the Lower Zambezi catchment.

681

682

Provenance Budgets and Erosion Patterns

683 ***Provenance Budgets.*** In this section, independent calculations based on elemental-geochemistry,
 684 Nd-isotope, and geochronological data are used to better constrain the rough provenance budget based
 685 on detrital modes presented in [Garzanti et al. \(2021a\)](#). Integrated compositional data indicate that
 686 Upper Zambezi sand and silty sand delivered to Lake Kariba consist $\geq 80\%$ of recycled quartz-rich
 687 Kalahari dunes and $16\pm 4\%$ of largely basaltic volcanic detritus, the remaining $\leq 5\%$ being derived
 688 from Precambrian basements exposed in Zambia and Zimbabwe. The age spectrum of zircon grains
 689 supplied by the Upper Zambezi to Lake Kariba is intermediate between those of Uppermost Zambezi
 690 and Kwando sands. These two river branches are thus revealed as the most prominent zircon sources,
 691 in a relative proportion that cannot be accurately determined owing to the low and similar zircon
 692 concentration in their sands indicated by mineralogical and geochemical data ([Table 2](#)).

693 Because sand generated in the Upper and Middle Zambezi catchments is trapped in Lakes Kariba
 694 and Cahora Bassa, all sand ultimately delivered to the delta (zircon grains included) is generated
 695 within the Lower Zambezi catchment. Petrographic and heavy-mineral data indicate contribution in
 696 similar proportion (ca. 30-40% each) from the Southern Irumide Province drained upstream of the

697 Luenha confluence and from the Mazowe-Luenha river system sourced in the Zimbabwe Craton. The
698 Umkondo Belt and the Karoo, Cretaceous, and Cenozoic extensional basins drained by the Minjova,
699 Sangadze, and Zangue tributaries contribute much of the rest (~20%), whereas supply from the Tete
700 and Blantyre domains drained respectively by the Morrunguze and Shire tributaries is subordinate
701 ($\leq 10\%$ each; [Garzanti et al. 2021a](#)).

702 New complementary information obtained from elemental geochemistry suggests that as much
703 as 70% of Lower Zambezi sand may be supplied by the Mazowe-Luenha river system, with
704 subordinate contribution from the mainstem upstream of the Zambezi-Luenha confluence (20-25%),
705 and minor supply from the Morrunguze and Shire Rivers ($< 5\%$ each) and other lowermost-course
706 tributaries (~5%). Calculations based on ϵ_{Nd} values of sand (63-2000 μm fraction) confirm that most
707 Lower Zambezi sand is generated in the Mazowe-Luenha catchment (55-65%), with subordinate
708 contribution from the mainstem upstream of the Luenha confluence and other sources. The ϵ_{Nd} values
709 of cohesive mud ($< 32 \mu\text{m}$ fraction) in the Lower Zambezi upstream of the delta are similar or more
710 negative than for sand ([Table 3](#)), indicating that mud contributions from the Mazowe-Luenha river
711 system are not lower, and possibly higher, than for sand.

712 Forward mixing calculations based on zircon-age data suggest that at least half of zircon grains
713 are derived from the Irumide domain, a quarter at most is generated in the Mazowe-Luenha
714 catchment, and a fifth at most in the final part of the Zambezi drainage basin, with minor contribution
715 from the Shire River. Because mineralogical and geochemical data indicate a relatively high zircon
716 fertility for the Shire catchment, all compositional information converges to indicate that the Shire
717 River supplies only a very small part ($< 5\%$) of sediment to the Zambezi Delta. This holds true also
718 for mud, because the smectite/kaolinite ratio increases sharply downstream of the Lower Zambezi,
719 whereas Shire mud contains abundant kaolinite and minor smectite ([Table 1](#)). Minor sediment supply
720 from the Shire River is explained by sequestration in Lake Malawi of all sediment generated in the
721 upper catchment and by further sediment trapping in wetlands and artificial reservoirs downstream
722 ([Mzuza et al. 2019](#)). This inference contrasts with [Just et al. \(2014 p.191\)](#), who reckoned that the

723 Shire River contributes ~28% of total Zambezi sediment load at present (~21% before construction
724 of the Cahora Bassa Dam).

725 *Weighing up Information.* Provenance budgets based on independent datasets and on diverse size
726 fractions are not entirely consistent. Age spectra of detrital zircons point at predominant zircon supply
727 from the Irumide domain exposed in the upper part of the Lower Zambezi catchment, with minor
728 zircon contribution from the Zimbabwe Craton drained by the Mazowe and Luenha Rivers. Instead,
729 elemental-geochemistry and Nd-isotope data suggest that most Lower Zambezi sediment is generated
730 in the Mazowe-Luenha catchment. Although the robustness of diverse sets of calculations is not easily
731 evaluated, it notably increases if end-member signatures are well distinct, precisely determined, and
732 have little variability dependent on grain size, weathering, or hydraulic sorting. Conversely, estimates
733 obtained on a narrow grain-size window or, worse, on a rare mineral within a narrow grain-size
734 window, are least likely to be representative and accurately extrapolated to the entire sediment flux
735 ([Vezzoli et al. 2016](#)).

736 In the case of Lower Zambezi tributaries draining medium/high grade crystalline basements,
737 sand-petrography, heavy-mineral, and elemental-geochemistry signatures show significant overlap
738 and hydrodynamically-controlled variability. Least robust are calculations based on zircon-age
739 spectra, because of the uncertainties involved in zircon-fertility determinations. Nd-isotope
740 geochemistry suffers from a limited number of analysed samples, but the end members are well
741 distinct and precisely defined, all grain sizes have been considered, and grain-size-dependent
742 variability is limited ([Fig. 9](#)). The change towards much more negative ϵ_{Nd} documented in all size
743 fractions of Lower Zambezi sediments downstream of the Luenha confluence cannot be ascribed to
744 Shire sediments — which yielded even less negative ϵ_{Nd} values for all size fractions ([Fig. 6](#)) — but
745 clearly indicates major sediment supply from the Mazowe-Luenha river system. Weighing up all
746 obtained information, we conclude that up to two-thirds of the sediment presently reaching the
747 Zambezi Delta is generated in the Mazowe-Luenha catchment, between a quarter and a third is

748 produced between Lake Cahora Bassa and the Zambezi-Luenha confluence, and the rest downstream,
749 with very limited supply ($\leq 5\%$) from the Shire River.

750 ***Erosion Patterns.*** Because of a general lack of gauged sediment loads, Zambezi sediment fluxes
751 are evaluated with large uncertainties of a full order of magnitude. Erosion patterns across the
752 catchment can thus be only grossly determined. Based on the available sediment-concentration data
753 and sediment-transport models, two end-member domains can be distinguished by their contrasting
754 geomorphological conditions and sediment-generation modality: the low-relief Kalahari Plateau
755 largely covered by eolian sand in headwater regions *versus* rugged igneous and metamorphic terranes
756 extensively exposed between Victoria Falls and Mozambican lowlands.

757 On the plateau, rivers with low channel steepness sluggishly flow for large tracts through
758 wetlands, where sediment is sequestered rather than produced, as in the Barotse floodplain and Chobe
759 marshes on the mainstem or in the Machili and Kafue Flats traversed by the Kasaya, Ngwezi, and
760 Kafue tributaries. Because data on Kwando sediment load are to the best of our knowledge
761 unavailable, information from the Okavango River similarly draining entirely within the Kalahari
762 Basin in Angola ([Shaw and Thomas 1992](#); [McCarthy et al. 2012](#)) allows us to broadly constrain the
763 annual sediment yield and erosion rate in the Kalahari Basin as 2 ± 2 tons/Km² and 0.001 ± 0.001 mm.
764 This is notably lower than estimates based on cosmogenic nuclides for the Uppermost Zambezi
765 catchment including Precambrian terranes in the north (16 ± 2 tons/Km² and 0.006 ± 0.001 mm;
766 [Wittmann et al. 2020](#)). Sediment yield increases by an order of magnitude where channel steepness
767 reaches very high values, as in basaltic gorges downstream of Victoria Falls (40-90 tons/Km²; fig. 3
768 and p.356 in [Garzanti et al. 2021a](#)).

769 A similar sediment-generation pattern characterizes other rivers flowing on the Kalahari Plateau
770 in the headwaters and plunging into bedrock gorges downstream. For the Gwai River, a provenance
771 budget based on petrographic, heavy-mineral, and geochemical data on both fluvial-bar and levee
772 silty sands indicates that sediment yield and erosion rate are between 20 and 50 times higher in the
773 lower course cutting steeply across the Dete-Kamativi Inlier of the Magondi Belt than in the upper

774 course, sourced in the Zimbabwe Craton and draining the Kalahari Basin. The same may hold true
 775 for the Kafue River, where much of the sediment is however trapped in the Kafue Flats and behind
 776 the Itezhi-Tezhi and Kafue Gorge Dams.

777 Annual yields of 150 ± 50 tons/Km², corresponding to erosion rates of 0.06 ± 0.02 mm, were
 778 estimated for Middle and Lower Zambezi tributaries flowing steeply across basement rocks exposed
 779 in the Archean Zimbabwe Craton or in the Proterozoic Irumide, Umkondo, and Zambezi Belts in
 780 southern Zambia, northeastern Zimbabwe, and western Mozambique (Bolton 1984; Ronco et al.
 781 2010). Considering that the Lower Zambezi upstream of the Luenha confluence and the Mazowe-
 782 Luenha river system have similar catchment areas, our provenance budget implies sediment yields
 783 and erosion rates between 1.5 and 2.5 times higher in the latter. Extensive sediment trapping in Lake
 784 Malawi upstream and across wetlands or behind dams downstream (Mzuza et al. 2019) prevents us
 785 to make considerations concerning erosion rates in the Shire catchment.

786
 787

Weathering *versus* Recycling

788 ***Insights from Clay Minerals.*** Clay mineralogy is quite sensitive to weathering conditions. It has
 789 long been observed that kaolinite is abundant in hot humid regions where feldspar hydrolysis is
 790 intense, whereas smectite is common in warm regions with a dry season characterized by intense
 791 evaporation, and illite and chlorite dominate where chemical decomposition is minor (Chamley 1989;
 792 Velde 1995). In modern sediments, the ratio between kaolinite and illite+chlorite [Kao/(Ill+Chl)] may
 793 thus be used as a proxy for weathering intensity (Liu et al. 2007; He et al. 2020). Within our sample
 794 set, kaolinite is significant in all catchments and represents ~40% of the clay-mineral assemblage in
 795 Uppermost Zambezi, Lower Zambezi (Tete sample), and Shire mud (Fig. 3). The Kao/(Ill+Chl) ratio
 796 is > 1 in Uppermost Zambezi, Kwando, Ngwezi, and upper Gwai muds generated on the Kalahari
 797 Plateau, but < 1 in Middle and Lower Zambezi tributaries downstream (Table 1).

798 ***Insights from Mud and Sand Geochemistry.*** Geochemical indices have long been used as
 799 proxies for weathering intensity (e.g., Nesbitt and Young 1982; Price and Velbel 2003), although they

800 may be even predominantly controlled by grain size (von Eynatten et al. 2012, 2016), provenance
801 (Garzanti and Resentini 2016; Dinis et al. 2017), hydraulic sorting, or quartz addition by recycling
802 (Fig. 11). This is especially true for sand, and weathering conditions are thus better reflected in the
803 geochemistry of mud (Dinis et al. 2020b).

804 A most reliable indicator of weathering intensity is α^{AlNa} , which chiefly measures the
805 progressive leaching of Na^+ from the plagioclase lattice. The α^{AlNa} value decreases quite regularly
806 from the Uppermost Zambezi and Kwando Rivers (21-22 for mud, ≥ 4 for sand) to Victoria Falls (4.6
807 for mud, 2.1 ± 0.1 for sand), and to the Middle and Lower Zambezi (4.6 ± 0.4 for mud, 1.2 ± 0.1 for
808 sand). Other α^{Al} values, and consequently the CIA* and its several derivative indices, are more
809 significantly affected by the mineralogy of sediment sources. Most evident is the anomaly of Kwando
810 mud, which is notably enriched in Ca, Sr, Mg, and Ba derived from erosion of calcrete soils (Fig. 11)
811 and consequently yielded very low corresponding α^{Al} indices (≤ 0.4). Masuie, Matetsi, and
812 Morrunguze sediments largely derived from basaltic or gabbroic rocks have high Mg and Ca and
813 consequently low α^{AlMg} and α^{AlCa} (0.4-0.6). Conversely, sediments derived from gneissic basements
814 are enriched in K and Rb largely hosted in K-feldspar and mica, which explains why α^{AlK} and α^{AlRb}
815 are < 1 in several tributaries (e.g., Kasaya, Ngwezi, Gwai, Kafue, Luangwa, Mazowe, Luenha, and
816 Sangadze). Sangadze sediments have the lowest plagioclase/K-feldspar ratio of all analysed sands
817 from the Lower Zambezi catchment, and consequently yielded the highest α^{AlNa} in both mud and
818 sand, and the lowest α^{AlK} and α^{AlRb} in sand.

819 ***Insights from Detrital Minerals.*** Additional indications on weathering intensity can be inferred
820 from the different durability of detrital minerals, although this path is fraught with pitfalls. An
821 example is the unfortunate use of the “Mineralogical Index of Alteration” [$\text{MIA} = \text{Q}/(\text{Q} + \text{F}) \times 100$; Rieu
822 et al. 2007], a misleading parameter not only long demonstrated to markedly increase with grain size
823 (Odom et al. 1976) but also reaching maximum values equally in hyper-humid equatorial and hyper-
824 arid desert conditions (Garzanti et al. 2019; Pastore et al. 2021).

825 The different chemical durability of heavy minerals is more reliably indicative (Bateman and
826 Catt 2007). Particularly useful is the ratio between garnet (G) and other nesosilicates found in
827 amphibole-facies metapelites (SKAS = staurolite + kyanite + andalusite + sillimanite), which in sand
828 of the Lower Zambezi catchment [$G/(G+SKAS) = 72 \pm 21\%$] is the same as in modern first-cycle sand
829 derived from metamorphic basements [$G/(G+SKAS) = 70 \pm 20\%$; Garzanti et al. 2010a]. In contrast,
830 the markedly anomalous low ratios [$G/(G+SKAS) < 5\%$] that characterize sand in the Uppermost
831 Zambezi catchment testify to an almost complete breakdown of garnet, a mineral that proves to be
832 extremely vulnerable in equatorial soils (figs. 9C and 9D in Garzanti et al. 2013a) but very durable
833 in dry tropical climate (Garzanti et al. 2015). In recycled sand of the Kalahari Plateau, even zircon
834 results to be selectively weathered out relative to quartz, as indicated by a zircon concentration lower
835 by an order of magnitude relative to first-cycle sand shed by Precambrian basement rocks in the
836 Middle to Lower Zambezi catchment. Zircon depletion is chiefly ascribed to breakdown of
837 preferentially weathered, strongly metamict old grains (e.g., Balan et al. 2001; Resentini et al. 2020).

838 *Corrosion features.* Direct evidence of chemical attack is provided by surficial dissolution
839 textures on labile ferromagnesian minerals (Velbel 2007). This approach, however, has drawbacks:
840 i) surficial features tell us the state of what is preserved but nothing about how much was destroyed;
841 ii) fresh and strongly weathered grains of the same detrital mineral commonly occur jointly (Van
842 Loon and Mange 2007); iii) slight degrees of corrosion may not be evaluated consistently by different
843 operators. Only semiquantitative hints on the intensity of weathering can thus be obtained.

844 With this aim, the percentage of surficially etched grains and the degree of corrosion was
845 recorded for over 4000 identified transparent heavy minerals following the classification of Andò et
846 al. (2012). In Uppermost Zambezi mainstem and tributaries, pyroxene, amphibole, epidote, staurolite,
847 kyanite, and andalusite are all mainly unweathered, a minority of grains are corroded, and only a few
848 pyroxene grains are deeply etched. In the Upper Zambezi mainstem and tributaries, most grains are
849 unweathered, but the percentage of corroded grains increases and both pyroxene and amphibole may
850 be deeply etched. In the Middle Zambezi mainstem and tributaries, the percentage of corroded heavy

851 minerals increases further, a larger percentage of pyroxene and amphibole grains are deeply etched,
852 and also epidote, garnet, or kyanite may show deep etching. Similar features characterize Lower
853 Zambezi mainstem and tributary sands, where epidote and garnet are even more extensively corroded.

854 ***Recycled Weathering Signatures.*** In the Uppermost Zambezi and Kwando Rivers, sand consists
855 of quartz and durable heavy minerals, with very low $G/(G+SKAS)$ ratio, and $CIA^* > 75$; mud contains
856 abundant kaolinite, CIA^* is > 80 , and $\alpha^{Al}Na$ is > 20 . Such compositional features, typical of sediment
857 produced in hot-humid equatorial climate, are at odds with the semiarid conditions of the Kalahari
858 Plateau today. They testify to an intensity of chemical weathering that cannot occur in the present
859 climatic regime but must have been inherited through multiple recycling from much wetter conditions
860 of the past, most plausibly characterizing the subequatorial belt to the north (Garzanti et al. 2022).
861 Diagenesis between sedimentary cycles contributed too, but cannot account for all features (e.g.,
862 virtually complete selective breakdown of garnet). Weathered detritus from lower latitudes ($\sim 10^\circ S$),
863 for instance, was supplied to the Kalahari Basin by the once connected Chambeshi-Kafue river
864 system, reaching the Uppermost Zambezi via the Machili Flats as supported by fish lineages (Moore
865 and Larkin 2001; Katongo et al. 2005).

866 Clay-mineral assemblages in river muds across southern Africa are not kaolinite-dominated,
867 reflecting the limited efficiency of soil-forming processes and incomplete feldspar leaching under the
868 present climatic conditions (Garzanti et al. 2014a). Kaolinite must thus be largely recycled from
869 widespread relic lateritic paleosols and duricrusts (Partridge and Maud 1987; Dill 2007; Moore et al.
870 2009). Even in southern Malawi — where annual rainfall increases eastwards from ~ 0.8 m to 1.6 m
871 at the foot of Mount Mulanje and up to 2.8 m at high elevation (peak 3002 m a.s.l.) — kaolinite is
872 mostly recycled by fluvial incision of peneplains of Cretaceous to Cenozoic age triggered by base-
873 level lowering of the Shire River (Dill et al. 2005). This is corroborated by the great abundance of
874 fresh feldspar in Shire sand (fig. 5G in Garzanti et al. 2021a), where inefficient plagioclase hydrolysis
875 is testified by undepleted Na and Ca ($\alpha^{Al}Na = \alpha^{Al}Ca = 1.0$).

876 All mineralogical and geochemical parameters, including $Kao/(Ill+Chl)$, CIA^* and $\alpha^{Al}Na$, would
877 consistently indicate that weathering intensity recorded in modern river sediments decreases
878 downstream the Zambezi River. Even when depurated from the physical effect of recycling (i.e.,
879 quartz addition), mud generated in the Uppermost Zambezi catchment appears to be more affected
880 by weathering than Middle and Lower Zambezi mud (Fig. 11). Such evidence, however, by no means
881 implies that weathering on the dry Kalahari Plateau is at present more intense than in the wetter
882 Middle to Lower Zambezi catchment. Rather, this trend reflects mixing of distilled polycyclic detritus
883 originally generated during some wetter stages of the past to the north of the Kalahari dryland with
884 first-cycle detritus shed from Karoo basalts along the Upper Zambezi, followed by abrupt
885 replacement — downstream of Lake Kariba first and of Lake Cahora Bassa next — by largely first-
886 cycle detritus derived from Precambrian basements. Only in the Middle and Lower Zambezi, where
887 detrital minerals are directly derived from basement rocks, their surficial dissolution textures chiefly
888 reflect present conditions of weathering. Together with trends displayed by cohesive mud (Fig. 11B),
889 these are the only features that document a slight increase in weathering intensity from the Middle to
890 the Lower Zambezi catchment.

891

892

Conclusions

893

894 Any compositional parameter is invariably controlled by multiple physical and chemical
895 processes that must be carefully evaluated before provenance and environmental information could
896 be correctly detangled and understood. Diverse datasets obtained by a range of independent methods
897 were thus integrated to constrain the many unknowns, reduce the number of potential alternative
898 solutions, and increase the plausibility of our inferences. Following this rationale, we applied a
899 spectrum of mineralogical, geochemical, and geochronological techniques to shed light on
900 sedimentary processes active in the complex Zambezi big-river system.

901

902 In this study, such an approach allowed us to: 1) characterize the composition of mud and sand
generated in, and transported across, the Zambezi drainage basin; 2) monitor the evolution of

903 compositional signals across a routing system rigidly segmented by both natural (tectonic
904 depressions, lakes, wetlands) and anthropic factors (large reservoirs trapping all sediment generated
905 upstream); 3) make inferences on sediment yields and erosion rates even in the lack of gauged
906 sediment fluxes; 4) assess the intensity of weathering and the origin of weathering signatures (i.e.,
907 present vs. recycled) in diverse parts of the vast catchment.

908 The age spectra of detrital zircons reflect the major episodes of crustal growth in Precambrian
909 southern Africa. Irumide ages are dominant in the Lower Zambezi and in most of its tributaries,
910 excepting the Shire and the Zangue Rivers where Pan-African ages prevail. Neoproterozoic ages
911 characterize the Gwai, Mazowe and Luenha Rivers sourced in the Zimbabwe Craton. Eburnean ages
912 are widely distributed but never prevail. Permian-Triassic (Karoo) ages are minor and Cretaceous
913 ages rare.

914 Smectite is the most widespread clay mineral, dominant in mud from Karoo basalts as in the
915 warm and poorly drained Mozambican lowlands characterized by equatorial/winter-dry climate. Illite
916 is prevalent locally (e.g., Kafue mud) and kaolinite is ubiquitous, reaching maximum abundance in
917 both uppermost and lower parts of the Zambezi catchment. Elemental geochemistry reflects
918 overwhelming quartz addition by recycling of Kalahari dune sand in the Uppermost Zambezi, local
919 supply from Lower Jurassic Karoo basalt in the Upper Zambezi, and chiefly first-cycle provenance
920 from Precambrian basements in the Lower Zambezi.

921 The ϵ_{Nd} values range from mildly negative for sediment derived from Stenian gabbro, Tonian
922 mafic granulite and Jurassic basalt to strongly negative for sand derived from Neoproterozoic cratonic
923 gneiss. The preferential concentration of ultradense monazite in the fine tail of the size distribution
924 owing to the settling-equivalence effect controls the intrasample ϵ_{Nd} variability among cohesive mud
925 (< 32 μm), very coarse silt (32-63 μm), and sand fractions (63-2000 μm) as well as deviations from
926 the theoretical relationships between ϵ_{Nd} and $T_{Nd,DM}$ model ages, suggesting that durable monazite
927 carries a more negative ϵ_{Nd} signal than other REE-bearing minerals (e.g., allanite, titanite, apatite,
928 epidote, and amphibole).

929 Elemental and isotope geochemistry reveal that 55-65% of mud and sand reaching the Zambezi
930 Delta today, after the construction of the Kariba and Cahora Bassa Dams, is generated in the Mazowe-
931 Luenha catchment. Contribution from Irumide terranes exposed upstream of the Luenha confluence
932 is subordinate and supply from the Shire River – the outlet of Lake Malawi – is minor. Although an
933 accurate assessment of sediment yields and erosion rates is hampered by the lack of gauged sediment
934 fluxes, annual estimates are lower by an order of magnitude on the Kalahari Plateau (10-20 tons/Km²
935 and ~0.005 mm) than in rugged terranes exposing Precambrian basements downstream (100-200
936 tons/Km² and ~0.05 mm).

937 All mineralogical [garnet/(staurolite+kyanite+andalusite+sillimanite), kaolinite/(illite+chlorite)]
938 and geochemical parameters (CIA*, α^{AlNa}) consistently point to an intensity of chemical weathering
939 on the Kalahari Plateau that cannot be related to modern dry-climate conditions. Chemical breakdown
940 of virtually all minerals relative to quartz – including feldspars, garnet that is very labile in lateritic
941 soils, and even zircon if strongly metamict – cannot occur in the Kalahari dryland, where kaolinite is
942 recycled. Kaolinite is mostly produced by fluvial incision of relic Cretaceous-Cenozoic paleosols
943 even in the Shire catchment closer to the wetter Mozambican coast, where inefficient plagioclase
944 hydrolysis is testified by the dominance of fresh feldspars and undepleted Ca and Na. Indications of
945 only slightly increasing weathering conditions in the Middle to Lower Zambezi catchment at present
946 times are provided by mud geochemistry and surficial corrosion of pyroxene, amphibole, epidote,
947 kyanite, and garnet.

948

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956

957 SUPPLEMENTARY MATERIAL

958 Supplementary data associated with this article, to be found in the online version at
959 http://dx.doi._____, include information on sampling sites (Table A1) together with clay
960 mineralogy (Table A2), elemental geochemistry (Tables A3 and A4), Nd-isotope geochemistry
961 (Table A5), bulk petrography (Table A6), and heavy-mineral datasets (Table A7). Appendix A
962 contains the caption of appendix tables and further information on the rationale of forward mixing
963 calculations. The complete detrital-zircon geochronology dataset is contained in Appendix B. The
964 Google-EarthTM map of sampling sites [Zambezi2.kmz](#) is also provided.

965

966 DATA AVAILABILITY

967 The mineralogical, geochemical, and geochronological datasets from this study are also available
968 from the senior author upon request.

969 FIGURES

970 **Figure 1.** The Zambezi drainage basin (base map from Google Earth™). White circles indicate
 971 sampling locations (more information in file [Zambezi2.kmz](#)). VF = Victoria falls.

972 **Figure 2.** Geology and climate of southern Africa. **A)** Geological map ([Thiéblemont et al. 2016](#)). **B)**
 973 Precipitation gradients: east/west from warm Indian Ocean to cool Atlantic Ocean; south/north from
 974 Kalahari dryland to humid Congo. **C)** Distribution of climatic zones (Köppen–Geiger classification;
 975 [Kottek et al. 2006](#)): A = equatorial; B = arid; C = warm temperate. Precipitation: W = desert; S =
 976 steppe; f = fully humid; s = summer dry; w = winter dry. Temperature: h = hot arid; k = cold arid; a
 977 = hot summer; and b = warm summer.

978 **Figure 3.** Clay mineralogy. Multiple controls explain erratic trends (dotted arrow) downstream the
 979 Zambezi River (stars). Recycled kaolinite occurs in both Uppermost Zambezi and Lower Zambezi
 980 catchments (e.g., Shire mud). Smectite is derived from Karoo basalt around Victoria Falls (VF) but
 981 also produced on the Kalahari Plateau and Mozambican lowlands (e.g., Sangadze mud). Illite is
 982 derived from metasedimentary and siliciclastic rocks of the Irumide and Pan-African belts (e.g.,
 983 Kafue and Tinde muds).

984 **Figure 4.** Sand and mud geochemistry (in UCC-normalized diagrams chemical elements are arranged
 985 following the periodic table group by group). **A, B)** In Zambezi headwaters, extensive quartz addition
 986 by recycling explains relative depletion of most elements other than Si in sand. Supply from Karoo
 987 basalt leads to marked increase in ferromagnesian metals and lack of Eu anomaly. High Ca, Sr, Mg,
 988 and Ba in Kwando mud reflect reworking of calcrete soils. **C, D)** Kafue samples are slightly enriched
 989 in elements hosted in ultradense minerals. Umguza and Upper Gwai sediments include minor detritus
 990 from Karoo basalt. Luangwa sand is partly recycled from Karoo siliciclastic strata. **E, F)** Most Lower
 991 Zambezi tributaries carry sediment undepleted relative to the UCC. In Morrunguze sand, high
 992 ferromagnesian metals and lack of Eu anomaly reflect supply from the Tete gabbro-anorthosite. High
 993 LREE and Th in Luenha sand suggest presence of monazite. **G, H)** Recycled quartz decreases

994 downstream the Zambezi mainstem. Lower Zambezi sand is only moderately depleted relative to the
 995 UCC, and mud is undepleted.

996 **Figure 5.** Rare Earth Elements. **A, B, C)** Chondrite-normalized REE patterns for sand. HREE trends
 997 are ill defined in pure quartzose sand because of very low concentration of elements with odd atomic
 998 numbers (Tb, Ho, Tm, Lu). **D, E, F)** Chondrite-normalized REE patterns for mud. **G, H, I)** La_N/Yb_N
 999 *versus* europium anomaly. **J, K, L)** LREE versus HREE fractionation. Mafic detritus, conspicuous in
 1000 Matetsi, Masuie and Morrunguze sediments and present in Sinde, Umguza, and Uppert Zambezi
 1001 sediments, has higher REE concentration, lower LREE fractionation, no Eu anomaly, and higher
 1002 HREE fractionation. Absence of Eu anomaly in most Lower Zambezi sands reflects abundance of
 1003 Ca-bearing feldspar. Steepest REE patterns with strongly negative Eu anomaly in Luenha sand
 1004 indicates presence of monazite, whereas strongly positive Eu anomaly in Sangadze sand reflects
 1005 abundant feldspar with lesser quantities of heavy minerals.

1006 **Figure 6.** Relationship between $\epsilon_{Nd(0)}$ and depleted mantle model ages ($T_{Nd,DM}$) for the Zambezi
 1007 mainstem and tributaries. The ϵ_{Nd} and $T_{Nd,DM}$ values are least negative and youngest for detritus from
 1008 Karoo basalt, Tete gabbro, and Blantyre mafic granulite (emplacement ages after [Svensen et al. 2012](#),
 1009 [Westerhof et al. 2008](#), and [Goscombe et al. 2020](#), respectively), and most negative and oldest for
 1010 detritus from Neoproterozoic cratonic gneiss. Most samples have higher Sm/Nd ratio than the UCC and
 1011 thus plot below the red line (based on $Sm/Nd_{UCC} = 0.1735$). The 32-63 μm size class of sand samples,
 1012 representing the fine tail of the size distribution where ultradense monazite is concentrated, has high
 1013 LREE and steeper LREE pattern (lower Sm/Nd) than the 63-2000 μm fraction. Instead, the 32-63 μm
 1014 size class of the Lower Zambezi silt collected at Tete (yellow outline) has low LREE and high Sm/Nd
 1015 because it represents the coarse tail of the size distribution depleted in ultradense minerals.

1016 **Figure 7.** U-Pb age spectra of detrital zircons. Archean ages are common in Gwai, Mazowe and
 1017 Luenha sands sourced in the Zimbabwe Craton. Orosirian ages in Kwando and Uppermost Zambezi
 1018 sands recycling Kalahari dunes and in Gwai sand largely derived from the Paleoproterozoic Magondi

1019 Belt. Irumide ages are widespread, overwhelming in Sangara and Morrunguze sands and dominant in
1020 Lower Zambezi sand. Pan-African zircons are also widespread, but only locally prevalent (Shire and
1021 Zangue sands). Geological domains after [Hanson \(2003\)](#) and [Thiéblemont et al. \(2016\)](#). CK = Choma-
1022 Kalomo block; IB = Irumide Belt; KB = Kibaran Belt; LRZ = Luangwa Rift Zone; MB = Magondi
1023 Belt; MRZ = Malawi Rift Zone; SIP = South Irumide Province; UB = Umkondo Belt; Ub-Usg =
1024 Ubendian-Usagaran Belts.

1025 **Figure 8.** The biplots highlight the relationships among chemical elements in Zambezi sand and
1026 cohesive mud. Provenance control is most evident for sand. Ferromagnesian metals are enriched in
1027 basaltic or gabbroic detritus, Al, Na, Ca, K, Rb, Ba, and Eu hosted in feldspars are enriched in first-
1028 cycle detritus from mid-crustal basements, and Si, Zr, and Hf in sediment recycled from Kalahari
1029 dunes dominated by quartz and durable heavy minerals.

1030 **Figure 9.** Multiple controls on Nd isotope values. Most prominent are the effects of lithology (mafic
1031 detritus being least negative) and average age of source rocks (as highlighted by generally good
1032 correlation with U-Pb age of detrital zircon). Grain-size-controlled intrasample variability is limited.

1033 **Figure 10.** Multidimensional scaling map based on U-Pb zircon-age spectra (green field defined by
1034 two Lower Zambezi samples upstream of the delta and Praia da Madal beach). Irumide ages are
1035 dominant in Sangara and Morrunguze sands. Pan-African ages prevail in Shire and Zangue sands.
1036 Neoproterozoic ages prevail in Gwai sand and are common in Luenha and Mazowe sands. Eburnean ages
1037 are common in Upper Zambezi, Kwando, and Gwai sands but never prevalent. A few Karoo ages
1038 occur in Kasaya and Sinde sands. Closest and second closest neighbours are linked by solid and
1039 dashed lines, respectively.

1040 **Figure 11.** Discriminating the effects of weathering, recycling, and grain size from geochemical data.
1041 Theoretical trends are calculated starting from the UCC standard: quartz-addition trend by
1042 progressively adding SiO₂; weathering trend by progressively subtracting mobile metals relative to Si
1043 and Al. Grain-size trend based on data from Alpine and Himalayan sediments ([Garzanti et al. 2010b](#),

1044 [2011, 2012](#)). In all four panels, sand follows the quartz-addition trend reflecting recycling of Kalahari
 1045 sands (Uppermost and Upper Zambezi catchment) or Karoo and older sandstones and metasandstones
 1046 (Middle and Lower Zambezi catchment) to various degrees. Mud samples broadly follow the
 1047 weathering trend. **A**) Samples plotting far below the regression line ($Al_2O_3 = -0.45 SiO_2 + 45$) include
 1048 Fe-rich Masuie and Matetsi sands from Karoo basalts, and Kwando mud enriched in Ca, Sr, Mg, and
 1049 Ba from calcrete soils. **B**) Uppermost and Upper Zambezi muds plot below the weathering trend (low
 1050 WIP), suggesting weathering signature inherited by recycling. Middle and Lower Zambezi muds
 1051 follow the theoretical trend, hinting at slightly increasing weathering intensity toward the coast.
 1052 Calcrete erosion explains anomalously low CIA* in Kwando mud. **C, D**) Cohesive muds collected
 1053 upstream of Lake Kariba reflect quartz addition from Kalahari dunes (Uppermost Zambezi) or
 1054 Neoproterozoic sandstones (Tinde).

1055 **Table 1.** Silt and clay mineralogy in the Zambezi catchment determined by X-ray powder-diffraction.
 1056 Qz = quartz; KF = K-feldspar; Pl = plagioclase; Carb = carbonate; Amp = amphibole; Hem = hematite;
 1057 Phyll = phyllosilicate; Sme = smectite; Ill = mica/illite; Chl = chlorite (including vermiculite); Kao =
 1058 kaolinite.

1059 **Table 2.** Sand and mud geochemistry in the Zambezi catchment. LOI = loss on ignition. The α values
 1060 are defined as $(Al/E)_{sample}/(Al/E)_{UCC}$. CIA* values corrected only for CaO in apatite.

1061 **Table 3.** Neodymium isotope values and Sm-Nd model ages for cohesive mud (> 32 μm), very coarse
 1062 silt (32-63 μm), and sand (63-2000 μm) fractions of Zambezi sediments (FS = fine sand; VFS = very
 1063 fine sand). * = data from mud samples collected in the upper part of the Zambezi catchment after
 1064 [Garzanti et al. \(2014a\)](#). All other data are from sand samples excepting the Lower Zambezi silt
 1065 collected at Tete.

1066 **Table 4.** U-Pb age peaks of detrital zircons and relative frequencies in modern sands of the Zambezi
 1067 catchment (calculated with Density Plotter; [Vermeesch, 2012](#)). To treat all samples equally and avoid
 1068 bias in intersample comparison the laser spot was always placed blindly in the center of zircon grains.

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To the Editor of
The Journal of Geology

Dear Sir,

please find enclosed the very carefully revised version of our manuscript, entitled “*The Segmented Zambezi Sedimentary System from Source to Sink - 2. Geochemistry, Clay Minerals, and Detrital Geochronology*” and submitted for publication in **The Journal of Geology**. This article integrates with new major and trace element geochemistry, Nd isotope, clay mineral, and U-Pb detrital-zircon age data the recently published study focused on sand petrology and heavy minerals, thus completing our multidisciplinary research project dedicated to the sedimentary system of the Zambezi River.

The manuscript has been thoroughly revised taking into full account the constructive critical comments provided by all three Reviewers, who greatly helped us to check every detail and to amend the flaws that the submitted version contained. The comments provided by Reviewer Basu were given particular attention. We did not eliminate figures but carefully checked, corrected, and improved all of them. As required, we added a new Table 4 providing key numerical information on zircon age-peaks. The account on how we have responded to the concerns expressed by each Reviewer is detailed point-by-point in the separate file “*Zambezi 2 Revision Notes*”.

We thank you heartily for your precious help, attention, and kind interest, hoping that our revised manuscript is now in current order and can thus meet the high requirements of **The Journal of Geology**.

Yours sincerely,

Eduardo Garzanti (on behalf of all co-authors)

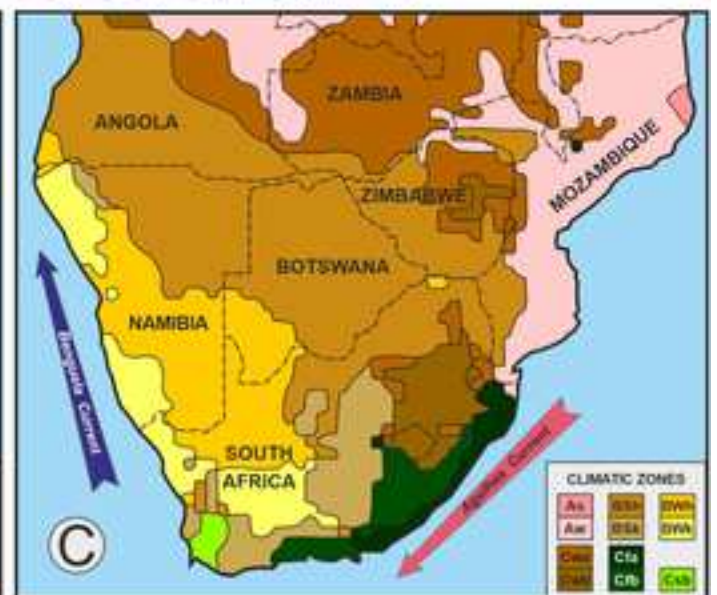
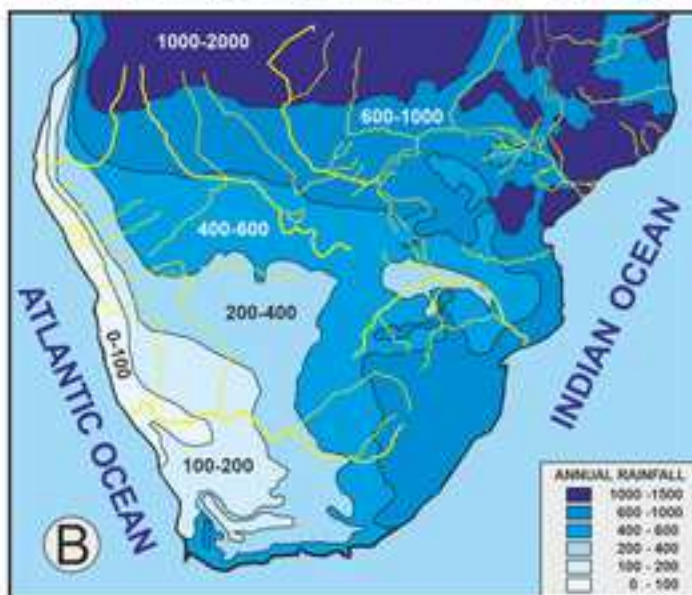
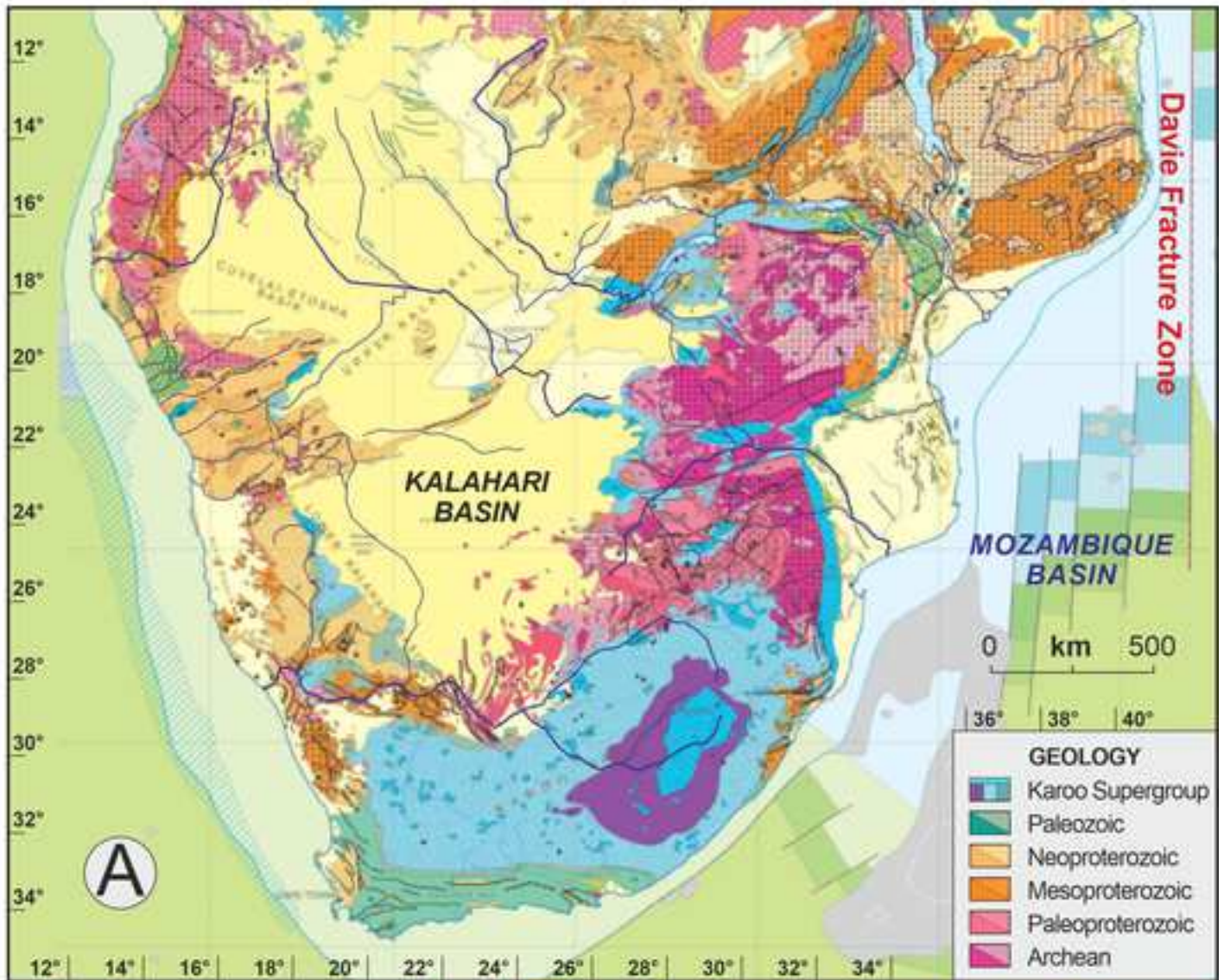
Tel: (39) 02 64482088 • Email: eduardo.garzanti@unimib.it

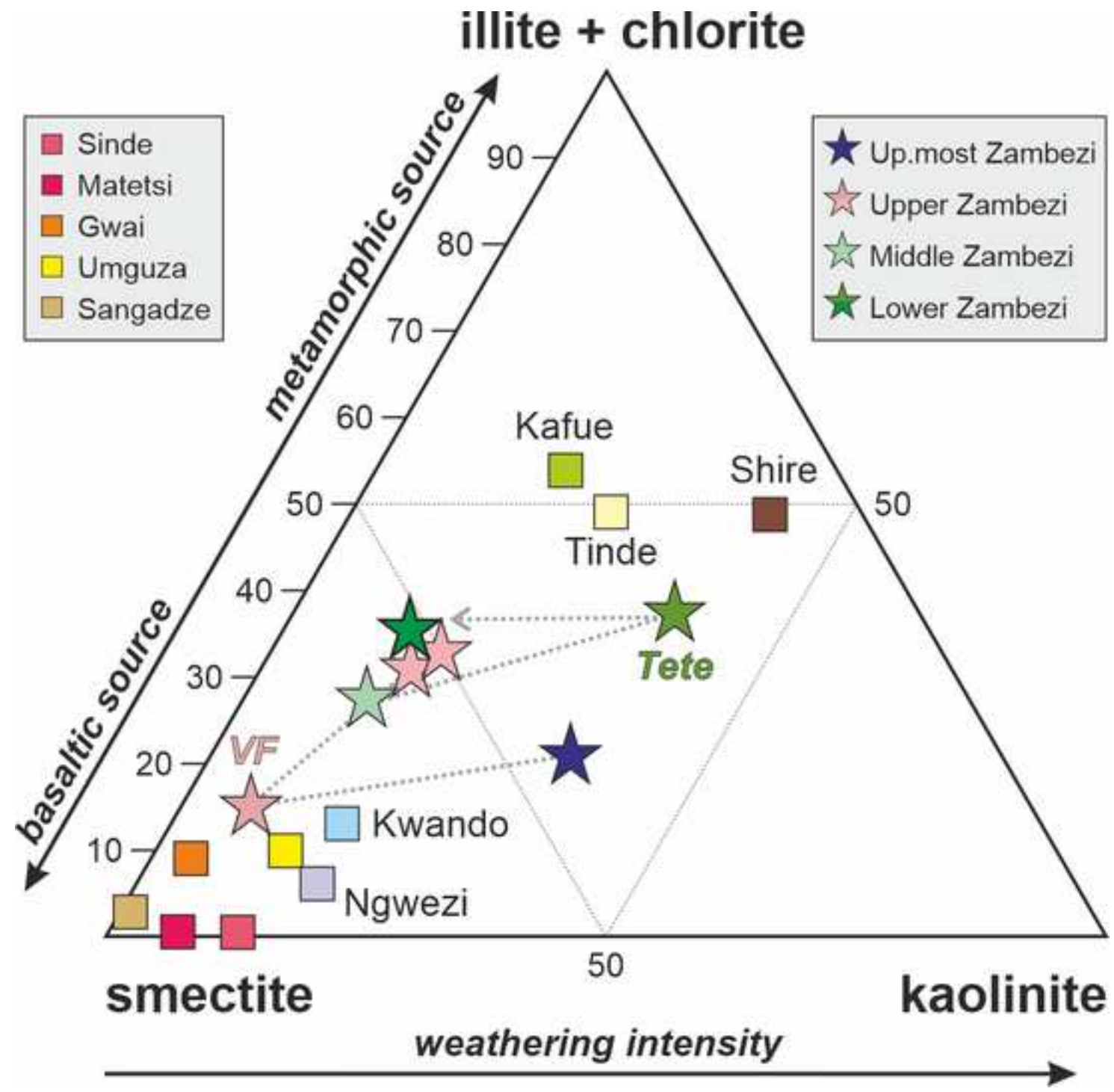
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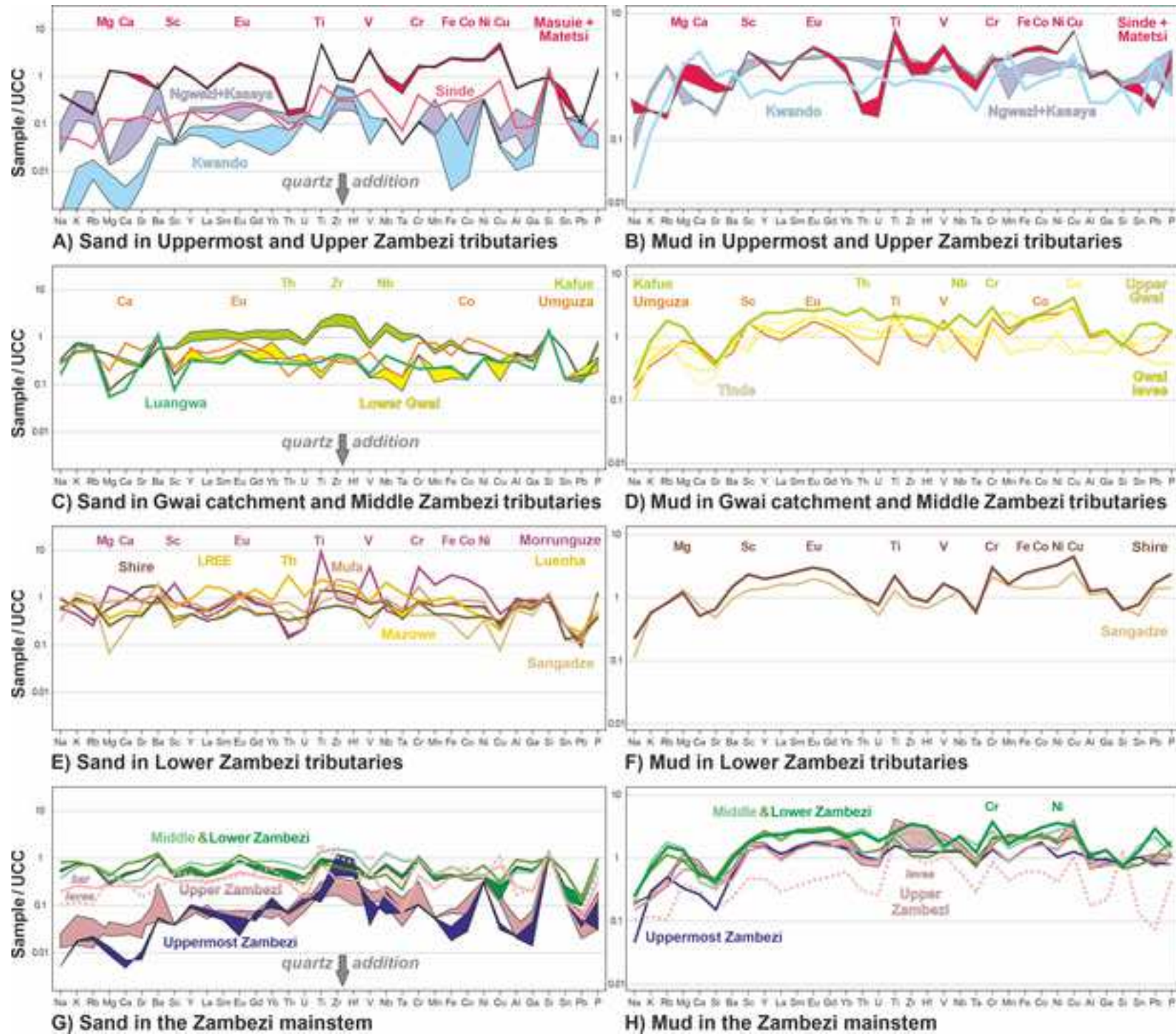
Figure 1

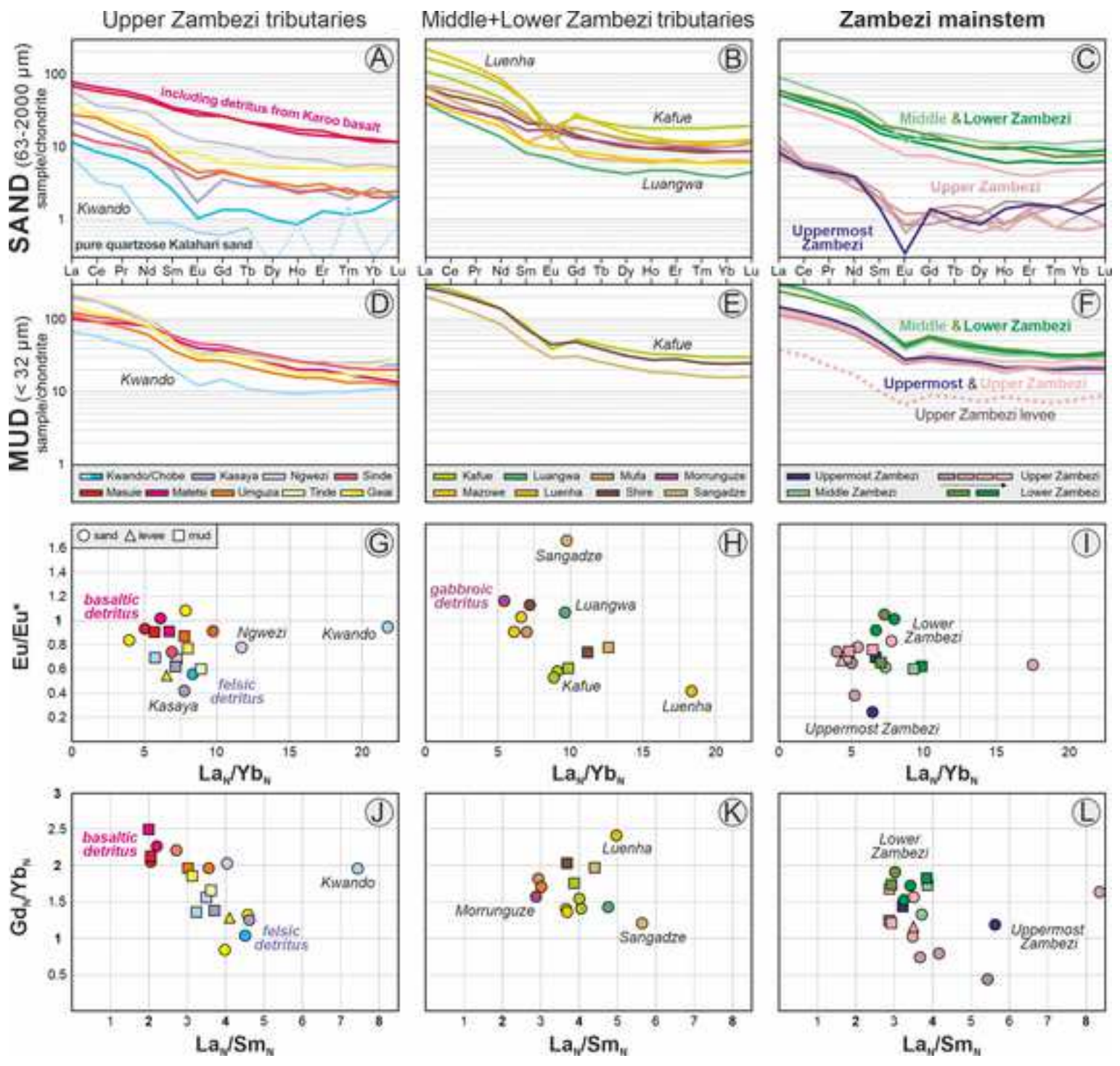
[Click here to access/download;Figure;Figure 1 Zambezi2 Geography.jpg](#)

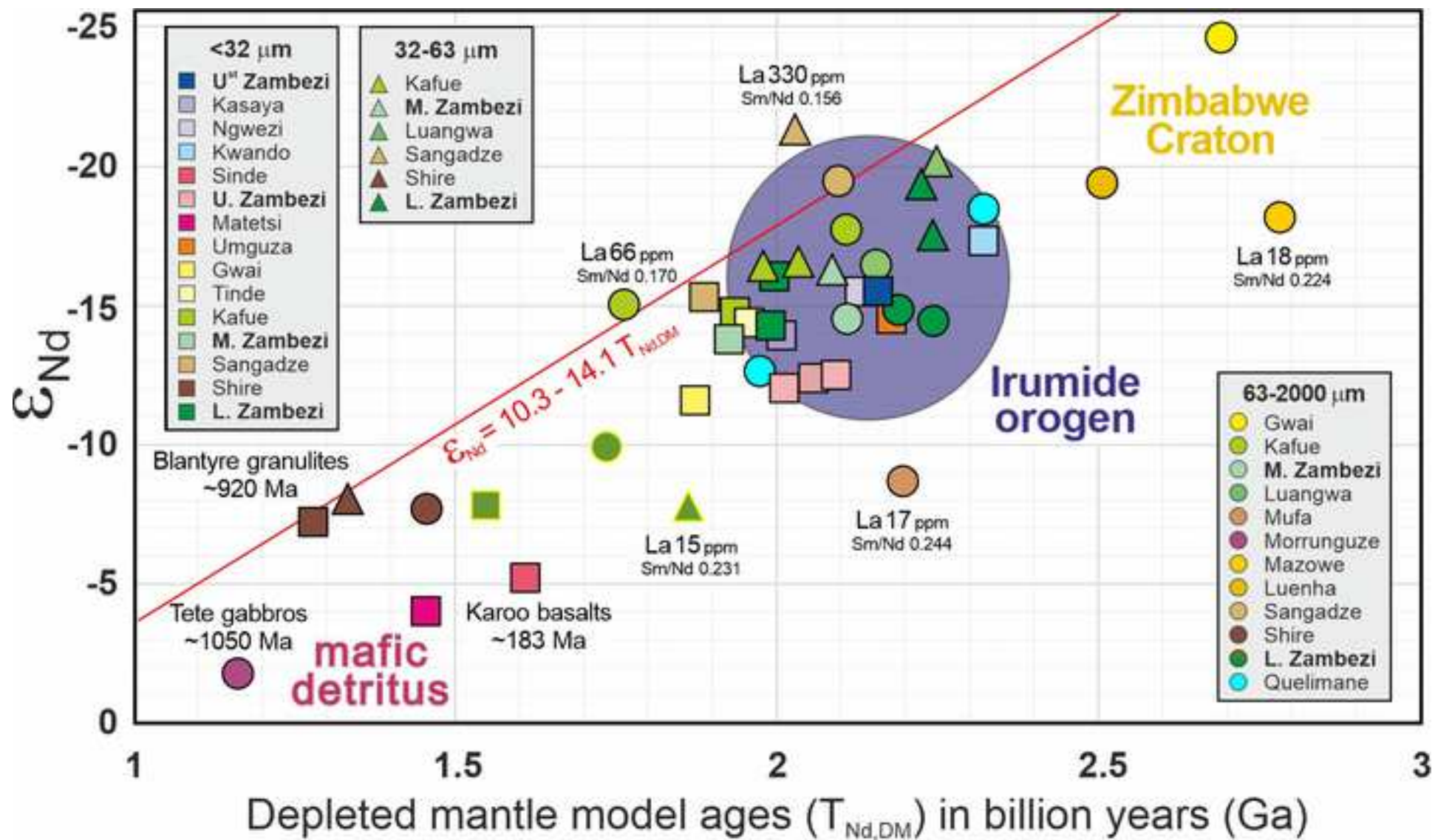












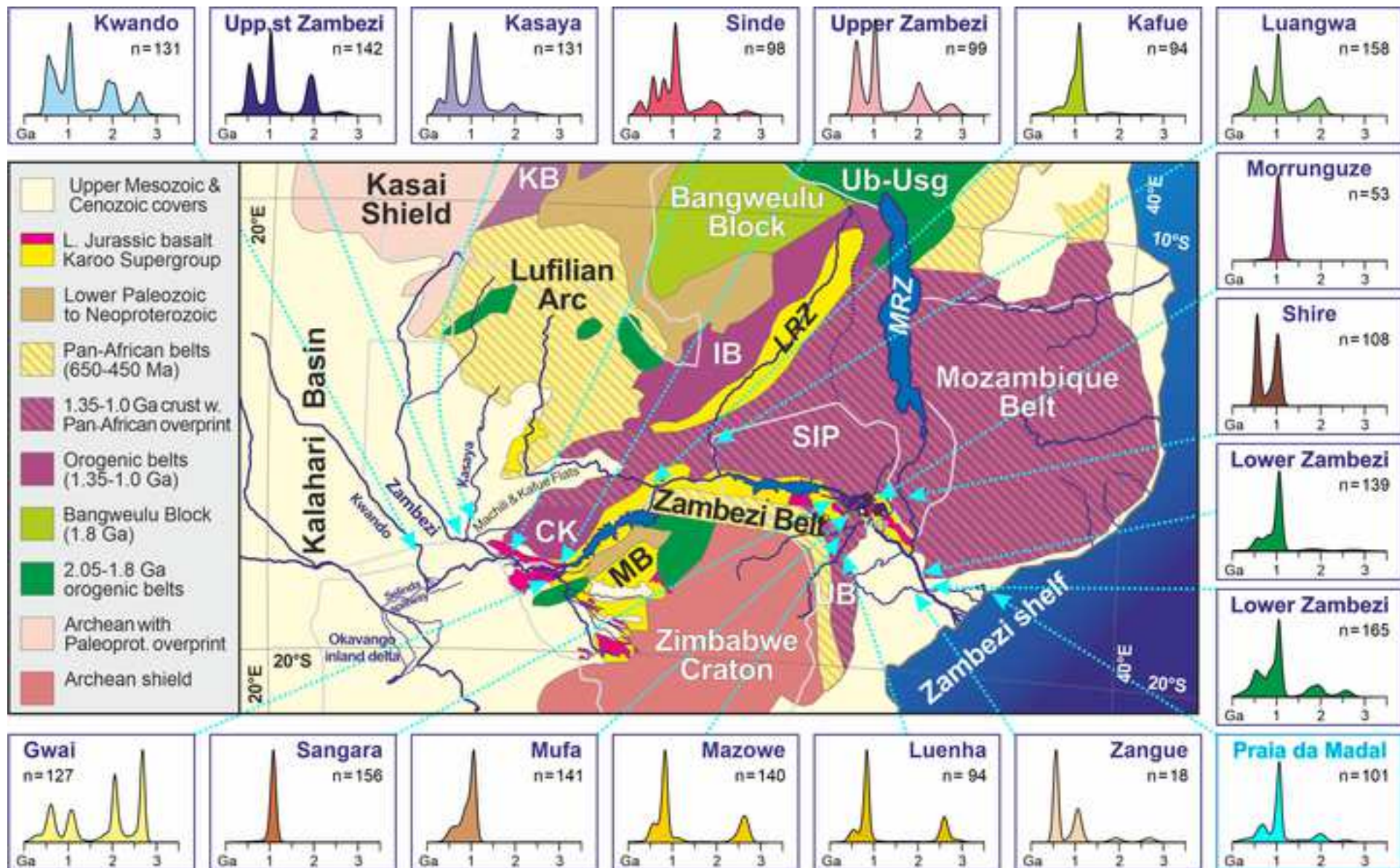


Figure 8

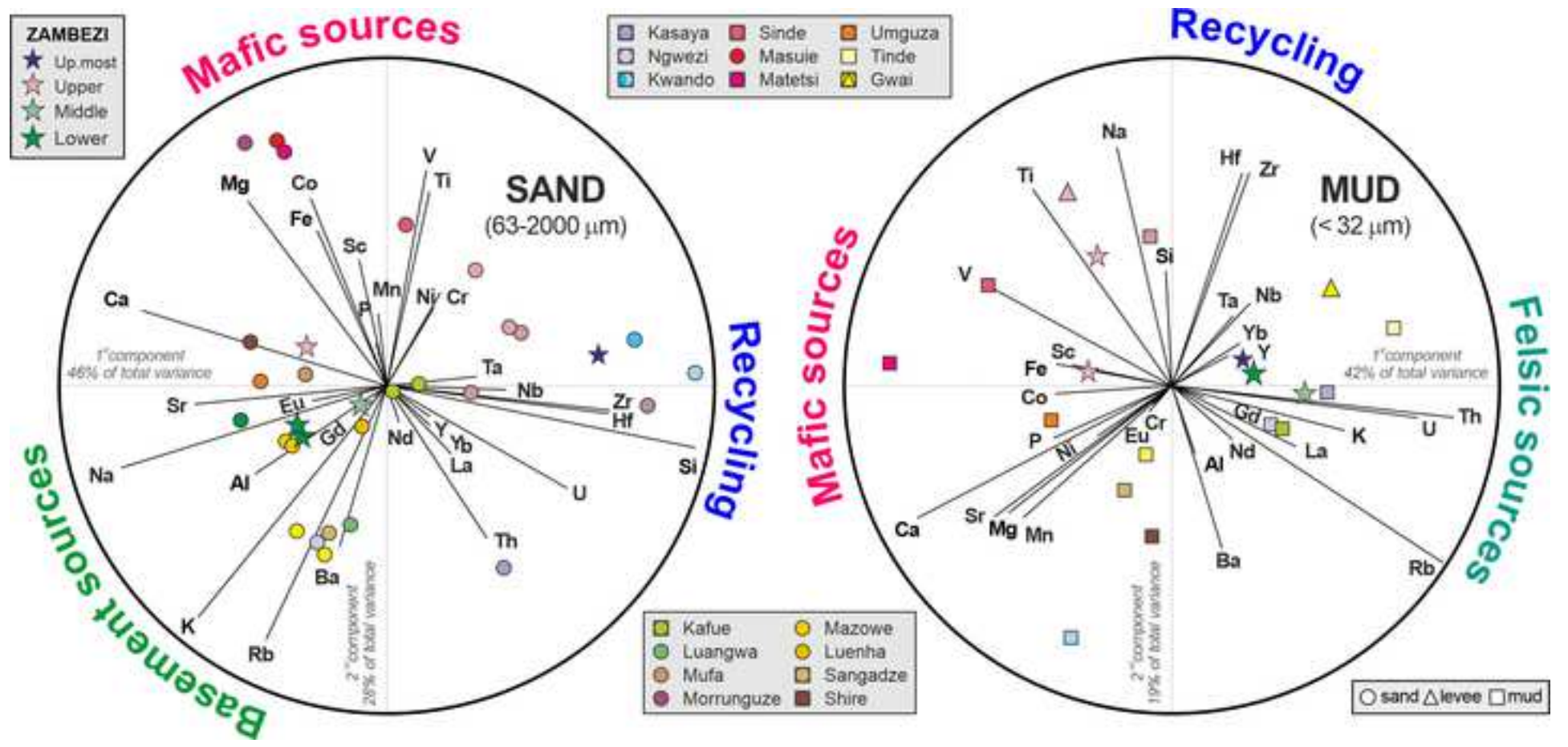


Figure 9

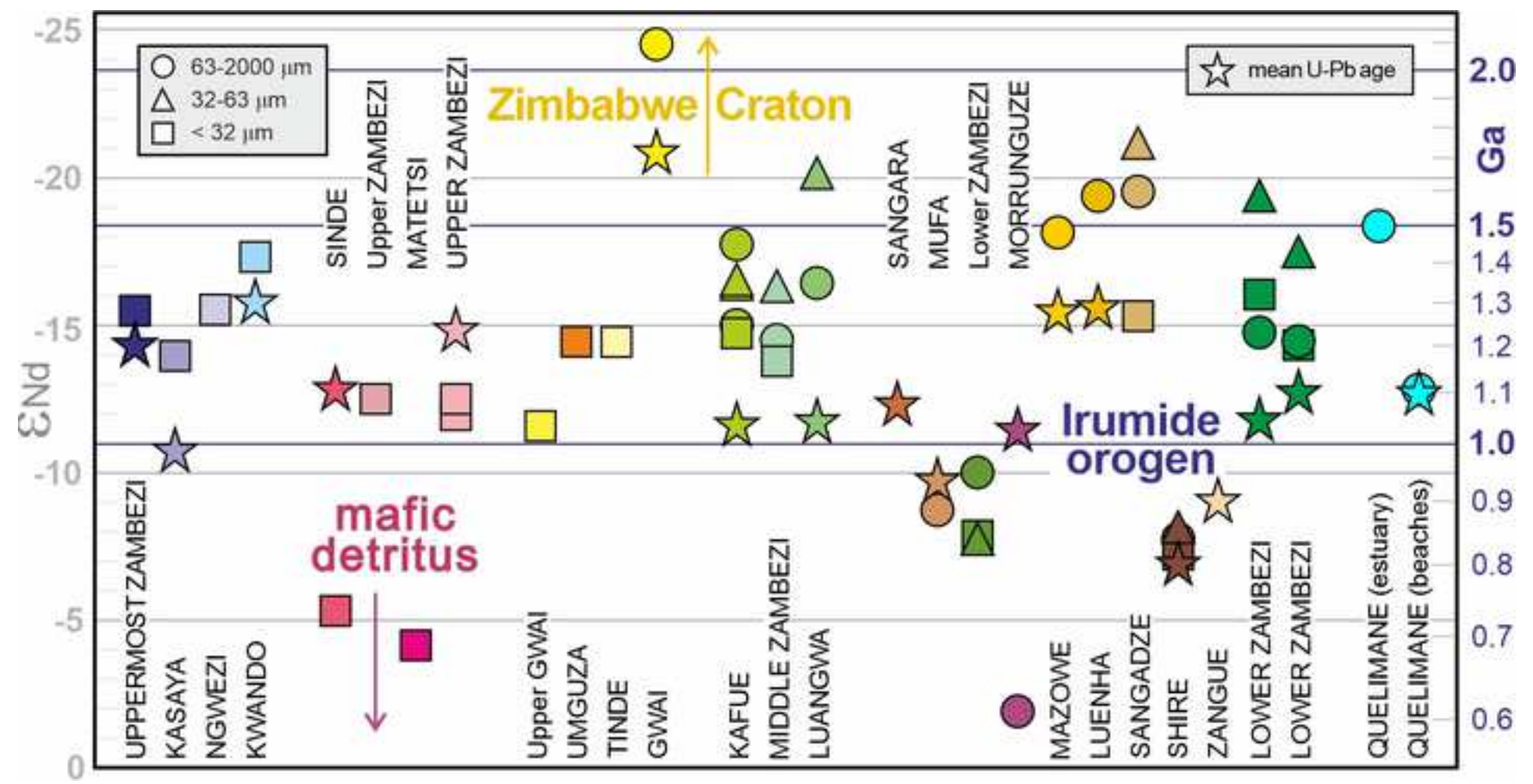
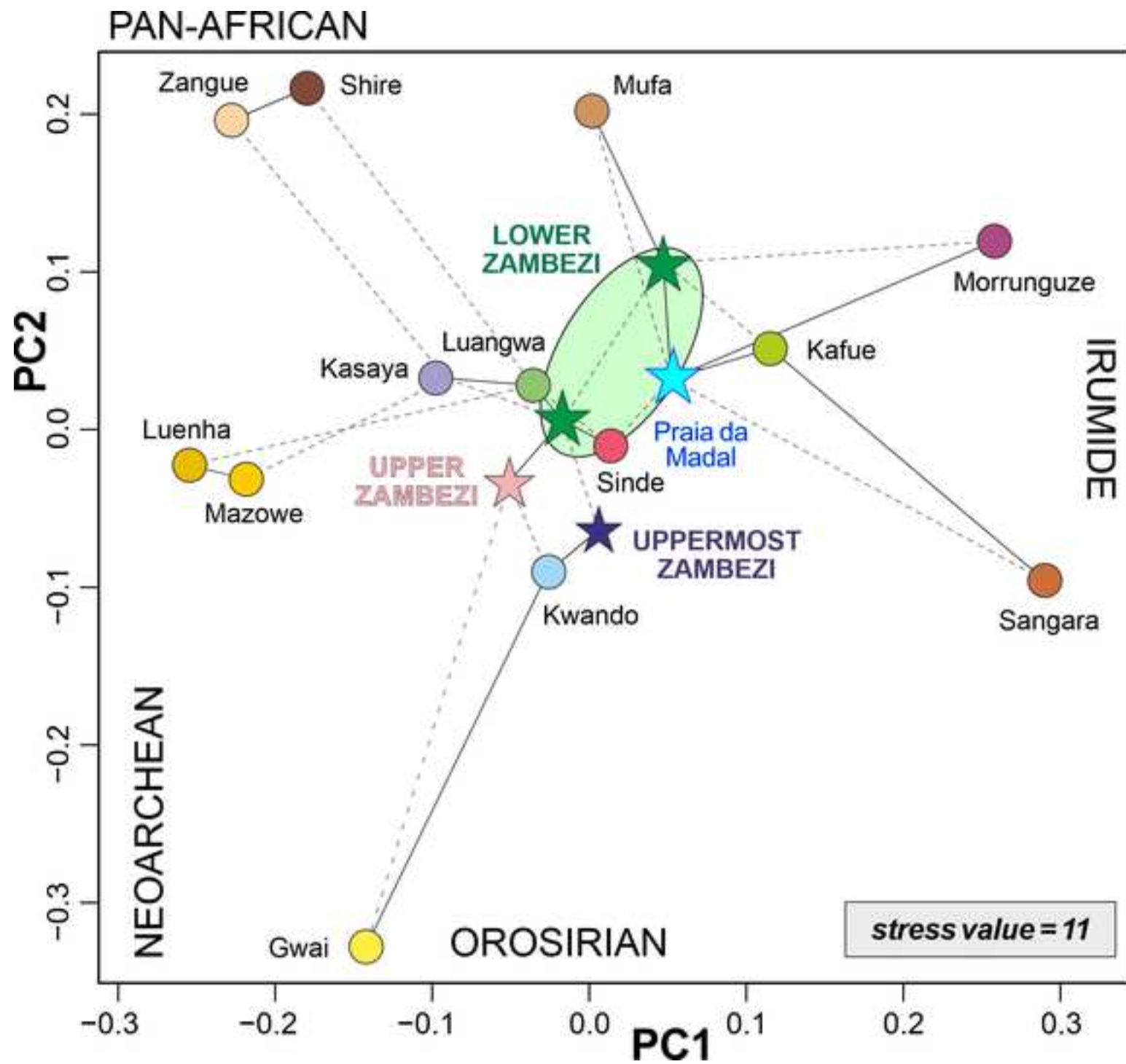


Figure 10



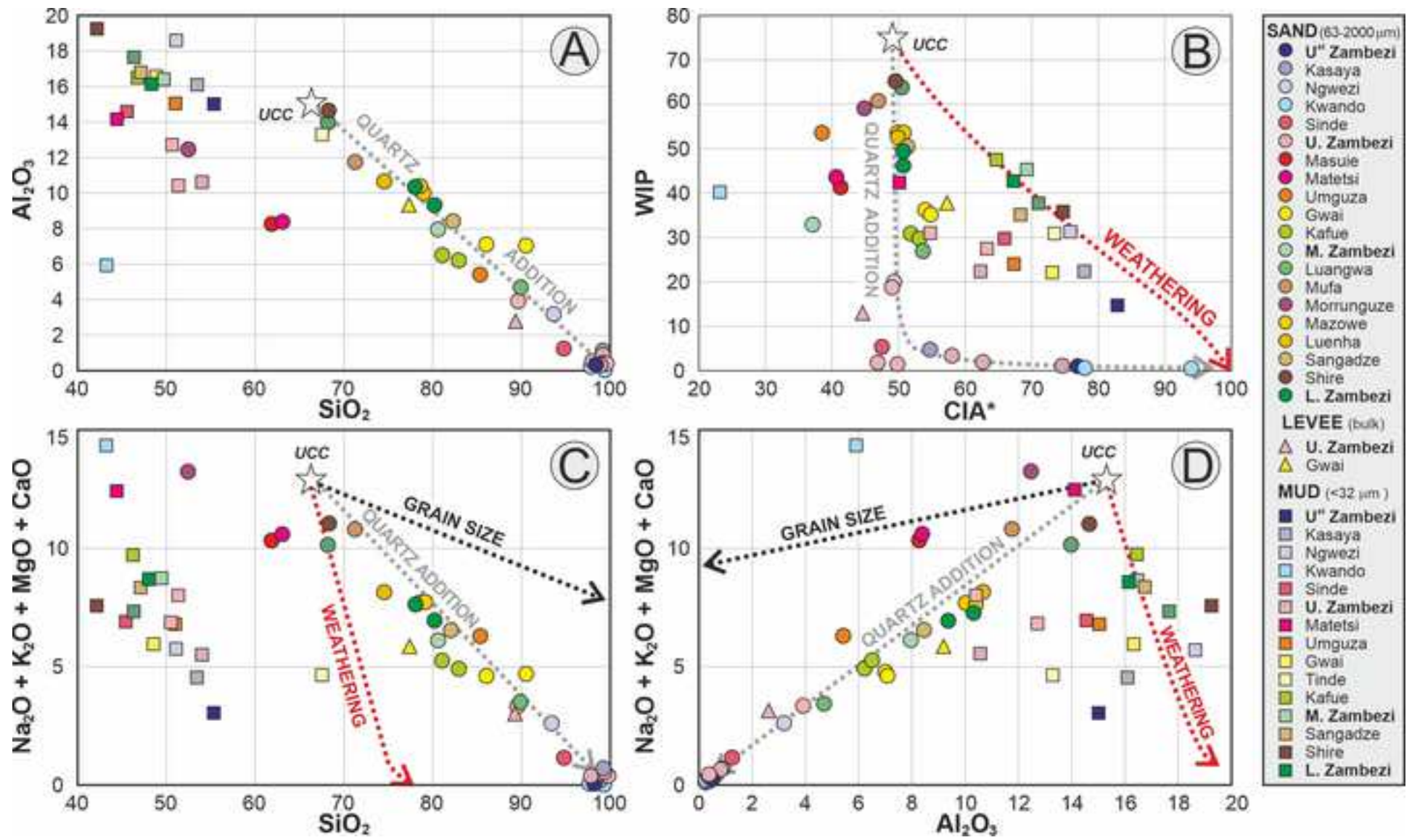


Table 1

River	BULK SAMPLE (< 32 µm fraction, wet sieved)							CLAY MINERALS (< 2 µm)					Kao / (Ill+Chl)	
	Qz	KF	Pl	Carb	Amp	Hem	Phyll	total	Sme	Ill	Chl	Kao		total
Uppermost Zambezi	49	15	1	0	0	0	35	100.0	43	21	0	36	100.0	1.7
Ngwezi	33	11	6	0	0	0	50	100.0	76	6	0	18	100.0	3.0
Kwando	89	0	0	4	0	0	7	100.0	70	13	0	17	100.0	1.3
Sinde	14	6	17	0	0	10	53	100.0	87	0	0	13	100.0	∞
Zambezi @ V.Falls	75	8	2	0	0	0	15	100.0	78	15	0	7	100.0	0.5
Matetsi	7	20	15	4	0	9	45	100.0	93	0	0	7	100.0	∞
Upper Zambezi	41	12	15	0	0	5	27	100.0	54	31	0	15	100.0	0.5
Upper Zambezi	43	11	19	0	0	4	23	100.0	50	33	0	17	100.0	0.5
Unguza	26	8	7	3	0	0	56	100.0	87	4	5	4	100.0	0.4
Upper Gwai	20	4	4	3	0	0	69	100.0	77	5	5	13	100.0	1.3
Tinde	34	24	5	0	0	0	37	100.0	25	40	9	26	100.0	0.5
Kafue	9	7	11	2	2	1	69	100.0	27	54	0.5	19	100.0	0.3
Middle Zambezi	10	7	11	6	1	0.2	64	100.0	60	27	0	12	100.0	0.4
Zambezi @ Tete	19	5	19	0	2	0	54	100.0	25	37	0	38	100.0	1.0
Sangadze	12	8	10	3	0.1	0	67	100.0	96	3	0	1	100.0	0.4
Shire	14	7	16	0	2	0	61	100.0	9	49	0	42	100.0	0.9
Lower Zambezi	17	14	20	0	0	0	49	100.0	52	35	0	12	100.0	0.4

Table 3

River	ϵ_{Nd}			t_{DM} (Ma)		
	<32 μm	32-63 μm	63-2000 μm	<32 μm	32-63 μm	63-2000 μm
Uppermost Zambezi*	-15.5	n.d.	n.d.	2162	n.d.	n.d.
Kasaya*	-14.0	n.d.	n.d.	2006	n.d.	n.d.
Ngwezi*	-15.6	n.d.	n.d.	2128	n.d.	n.d.
Kwando*	-17.3	n.d.	n.d.	2320	n.d.	n.d.
Sinde*	-5.3	n.d.	n.d.	1608	n.d.	n.d.
Zambezi @ Vic.Falls*	-12.5	n.d.	n.d.	2056	n.d.	n.d.
Matetsi*	-4.1	n.d.	n.d.	1454	n.d.	n.d.
Upper Zambezi*	-12.0	n.d.	n.d.	2010	n.d.	n.d.
Upper Zambezi*	-12.5	n.d.	n.d.	2092	n.d.	n.d.
Umguza*	-14.7	n.d.	n.d.	2177	n.d.	n.d.
Upper Gwai*	-11.6	n.d.	n.d.	1872	n.d.	n.d.
Tinde*	-14.4	n.d.	n.d.	1953	n.d.	n.d.
Gwai	n.d.	n.d.	-24.6	n.d.	n.d.	2690
Kafue (FS)	n.d.	-16.5	-17.7	n.d.	2029	2107
Kafue (VFS)	-14.8	-16.4	-15.0	1935	1976	1762
Middle Zambezi	-13.8	-16.2	-14.5	1925	2084	2106
Luangwa	n.d.	-20.1	-16.5	n.d.	2244	2155
Mufa	n.d.	n.d.	-8.8	n.d.	n.d.	2195
L.Zambezi @ Tete°	-7.8	-7.7	-10.0	1552	1864	1735
Morrunguze	n.d.	n.d.	-1.9	n.d.	n.d.	1162
Mazowe	n.d.	n.d.	-18.2	n.d.	n.d.	2781
Luenha	n.d.	n.d.	-19.4	n.d.	n.d.	2504
Sangadze	-15.3	-21.1	-19.5	1887	2028	2094
Shire	-7.3	-8.0	-7.7	1279	1333	1455
Lower Zambezi (FS)	-16.0	-19.3	-14.8	1995	2221	2188
Lower Zambezi (VFS)	-14.3	-17.4	-14.5	1990	2242	2242
Quelimane (estuary)	n.d.	n.d.	-18.3	n.d.	n.d.	2319
Quelimane (beach)	n.d.	n.d.	-12.7	n.d.	n.d.	1968

Table 4

River	Karoo		Pan-African II		Pan-African I		Irumide		Lukamfwa		Eburnean		Limpopo	
Uppermost Zambezi	235 ± 14	2%	561 ± 10	27%			1009 ± 15	39%			1989 ± 31	32%		
Kasaya	281 ± 12	8%	558 ± 10	34%			1100 ± 16	45%			2064 ± 58	13%		
Kwando			575 ± 11	27%			990 ± 17	36%			1891 ± 36	25%	2571 ± 78	12%
Sinde	255 ± 9	7%	570 ± 16	15%	816 ± 30	15%	1079 ± 21	42%			1968 ± 46	21%		
Upper Zambezi			593 ± 11	33%			987 ± 20	34%			1905 ± 46	24%	2578 ± 103	9%
Gwai	243 ± 11	4%	589 ± 12	19%			1064 ± 23	17%			2028 ± 40	29%	2606 ± 52	31%
Kafue	230 ± 13	3%	616 ± 22	11%			1035 ± 12	79%			1866 ± 87	5%	2579 ± 205	2%
Luangwa			550 ± 15	20%	717 ± 39	8%	1066 ± 17	54%	1530 ± 76	8%	1998 ± 82	10%		
Sangara							1061 ± 8	100%						
Mufa			559 ± 19	13%	754 ± 50	12%	1002 ± 14	75%						
Morrunguze							1030 ± 18	100%						
Mazowe			555 ± 14	14%	844 ± 10	57%							2518 ± 41	29%
Luenha			529 ± 18	12%	831 ± 12	58%							2529 ± 49	30%
Shire			564 ± 8	46%			1022 ± 13	54%						
Zangue			560 ± 18	55%			1036 ± 42	33%			1926 ± 197	6%	2676 ± 274	6%
Lower Zambezi			597 ± 15	17%			1013 ± 11	72%			1851 ± 82	6%	2522 ± 91	5%
Lower Zambezi			587 ± 11	21%			1039 ± 12	49%			1878 ± 35	22%	2545 ± 42	8%
Praia da Madal	272 ± 14	4%			686 ± 27	21%	1044 ± 15	56%			1998 ± 53	17%	2558 ± 72	2%