1	REVISION 2
2	Fluvial transport of impact evidence from cratonic interior to passive margin:
3	Vredefort-derived shocked zircon on the Atlantic coast of South Africa
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13	Abstract
14	Meteorite impacts produce shocked minerals in target rocks that record diagnostic high
15	pressure deformation microstructures unique to hypervelocity processes. When impact craters
16	erode, detrital shocked minerals can be transported by fluvial processes, as has been
17	demonstrated through studies of modern alluvium at some of the largest known impact
18	structures. In South Africa, detrital shocked minerals from the 2020 Ma Vredefort impact
19	structure have been documented in the Vaal River basin, downriver from the structure. However,
20	the ultimate fate of distally transported detrital shocked minerals in fluvial systems is not well
21	understood, and is an important parameter for constraining the location of a source crater. Here,
22	we report results of an extensive microstructural survey of detrital zircon from the Orange River
23	basin and the Atlantic coast of South Africa to search for the presence of far-travelled Vredefort-

24	derived detrital shocked zircon grains in different modern sedimentary environments. Three
25	shocked grains were found out of 11,168 grains surveyed (0.03%) by scanning electron
26	microscopy, including two in beach sand on the Atlantic coast and one from an Orange River
27	sandbar 15 km upstream from the river mouth. Shock-produced {112} twins documented by
28	electron backscatter diffraction in each of the three grains confirm their impact provenance, and
29	U-Pb ages from 3130 to 3040 Ma are consistent with derivation from bedrock at the Vredefort
30	impact structure. These results demonstrate the transport of Vredefort-derived shocked zircon to
31	the coast via the Vaal-Orange river system, which requires 1940 km of fluvial transport from
32	their point source on the Kaapvaal craton to the Atlantic coast passive margin. These results
33	further demonstrate that shocked zircon grains can be detected in detrital populations at
34	abundances < 1%, and can ultimately be transported outside their basin of origin when they
35	arrive at continental margins. Detrital shocked zircon thus constitutes long-lived evidence of
36	former impacts, as they retain microstructural evidence of shock deformation, as well as
37	geochemical (U-Th-Pb) fingerprints of their source terrain. The study of detrital shocked
38	minerals uniquely merges impact cratering with sedimentology, as identification of detrital
39	grains with diagnostic shock microstructures in siliciclastic sediments can be applied to search
40	the sedimentary record for evidence of eroded impact structures of any age, from the
41	Phanerozoic to the Hadean, which can aid in reconstructing the impact record of Earth.
42	Keywords: shocked zircon; Vredefort impact structure; electron backscatter diffraction, Orange
43	River; fluvial transport; shock metamorphism
44	Introduction
45	The terrestrial impact record has been largely removed due to geologic processes that
46	obscure or destroy impact craters. Only 190 confirmed impact structures are currently recognized

47	on Earth (Spray, 2001), while ~8700 impact structures >0.01 km in diameter have been
48	identified on the Moon (Losiak et al., 2008). During meteorite impact, pressures and
49	temperatures in target rocks can reach 100s of GPa and 1,000s of °C, greatly exceeding values of
50	endogenic crustal metamorphism (Melosh, 1989). Under these conditions, certain minerals in
51	target rocks record unique microstructures that form as a consequence of shock metamorphism
52	(French, 1998; Leroux et al., 1999; Gibson and Reimold, 2008). Some shocked minerals eroded
53	from impact structures, such as zircon, are resilient enough to survive as detrital grains.
54	Identification of shock microstructures in detrital minerals can thus be used to detect evidence of
55	impact structures that have been eroded or are otherwise unknown (Cavosie et al., 2010;
56	Erickson et al., 2013a; Thomson et al., 2014; Reddy et al., 2015).
57	Shock features in zircon generally manifest as different types of planar microstructures
58	that form from 20 to 40 GPa (Leroux et al., 1999) and include planar fractures (PFs), planar
59	deformation features (PDFs), planar deformation bands (PDB), {112} twins, and the high
60	pressure polymorph reidite (Wittmann et al., 2006; Timms et al., 2012; Erickson et al., 2013b;
61	Cavosie et al., 2015a). Shocked zircon grains that experience high temperature can recrystallize
62	to a granular texture consisting of neoblasts (Bohor et al., 1993; Cavosie et al., 2016), and
63	ultimately dissociate. At present, {112} twins (Moser et al., 2011; Timms et al., 2012; Erickson
64	et al., 2013a,b; Thomson et al., 2014; Cavosie et al., 2015b; Erickson et al., 2016) and the
65	polymorph reidite (Glass and Liu, 2001; Wittmann et al., 2006; Cavosie et al., 2015a; Reddy et
66	al., 2015) are considered the only diagnostic microstructural evidence for hypervelocity impact
67	deformation of zircon.

Located southwest of Johannesburg in South Africa, the Vredefort Dome is a ~90 km
wide erosional remnant that exposes the central uplift of one of the largest and oldest precisely

70	dated impact structures on Earth (Fig. 1) (Wieland et al., 2005; Gibson and Reimold, 2008). It
71	has been interpreted as a multi-ring structure originally ~300 km wide (Grieve and Therriault,
72	2000), and geothermobarometry studies of exposed rocks indicate that the upper 8 to 11 km have
73	eroded since its formation at 2020 Ma (Kamo et al., 1996; Therriault et al., 1997; Gibson et al.,
74	1998). Exposed components of the Vredefort impact structure include a ~40 km core of Archean
75	granitoids and gneisses (Kamo et al., 1996; Hart et al., 1999; Moser et al., 2001; Flowers et al.,
76	2003; Armstrong et al., 2006), surrounded by a ~20-25 km collar of late Archean to
77	Paleoproterozoic metavolcanic and metasedimentary rocks (Wieland et al., 2005; Gibson and
78	Reimold, 2008). Shocked zircon grains have been reported from a wide range of Vredefort
79	bedrocks (e.g., Kamo et al., 1996; Gibson et al., 1997; Moser, 1997; Moser et al., 2011).
80	The Orange River is the largest river in South Africa, occupying a \sim 900,000 km ² basin
81	that drains roughly half of the Kaapvaal Craton (Fig. 1) (Bremner et al., 1990; Compton and
82	Maake, 2007). The Vaal River is a right-bank tributary of the Orange River that flows through
83	the Vredefort Dome in a generally southwest direction (Fig. 1). The combined Vaal-Orange
84	fluvial system flows approximately 1940 km downstream from the Vredefort Dome to the
85	Atlantic coast, where the Orange River has been discharging since the Cretaceous (de Wit et al.,
86	2000; Gibbon et al., 2009; Garzanti et al., 2012). The location of the Orange River mouth has
87	migrated between 31°S and its current location at 28°S over time (Dingle and Hendey, 1984).
88	Currently, most Orange River discharge originates from the upper Orange, with $< 2\%$
89	contributed from below the Vaal-Orange confluence (Benade, 1988). At the outflow of the
90	Orange River on the Atlantic Coast, longshore drift transport is from the south to the north,
91	driven by the Benguela current (Fig. 1) (Garzanti et al., 2012). Once discharged, sediments from

the Orange River are transported as far as the Namib Sand Sea on the Atlantic coast of Namibia
by coastal and eolian processes (e.g., Garzanti et al., 2012).

Vredefort-derived detrital shocked minerals have previously been documented in the 94 Vaal River, including zircon, monazite, and quartz, and are thus predicted to occur downstream 95 in the Orange River. Cavosie et al. (2010) reported shocked zircon abundances of 36 - 64% in 96 alluvium within the Vredefort Dome. Erickson et al. (2013a) documented detrital shocked zircon 97 in the Vaal River outside the Vredefort Dome; at 103 km downstream from the impact structure, 98 99 they found shocked zircon abundances of 18% in alluvium (81/452 grains). The most distal alluvium sample analyzed in that study was 759 km downstream from the impact structure, only 100 101 \sim 3 km upriver from the Vaal-Orange confluence, which had a shocked zircon abundance of 2% 102 (5/253 grains).

Given the ubiquitous occurrence of detrital shocked minerals in the Vaal River, the goal 103 of this study is to evaluate if Vredefort-derived shocked zircon can be detected beyond the 104 confluence of the Vaal and Orange rivers, at distal locations downstream from the Vredefort 105 106 Dome in the Orange River. Samples of modern alluvium from the middle and lower Orange River, beach sand on the Atlantic coast of South Africa, and alluvium near the only other known 107 impact structure in the Orange River basin were collected to test the extreme limits of distal 108 transport, in terms of distance and detectability, for how far detrital shocked minerals can travel 109 110 in a fluvial system, be identified, and linked to their source terrane. Given the time-intensive aspect of surveying large numbers of individual grains, our study focused exclusively on zircon. 111 Methodology 112 Fourteen sites were sampled to search for distally-transported detrital shocked zircon in 113 modern sediment (Fig. 1, Table 1). These include seven alluvium samples from the middle and 114

115 lower stretches of the Orange River below the Vaal-Orange confluence, and two heavy mineral beach sand samples at the Orange River mouth on the Atlantic coast. In addition to sampling 116 117 Orange River alluvium and beach sand at the river mouth, five other sediment samples were 118 collected to evaluate additional environments where detrital shocked zircon might reside. These 119 include three samples of modern beach sand from south of the Orange River mouth, as far south as the mouth of the Oliphants River, to evaluate if shocked zircon grains are being transported 120 121 northward along the Atlantic margin from previous locations where the Orange River discharged 122 since the Cretaceous (Dingle and Hendey, 1984). Additionally, two modern alluvium samples near the Morokweng impact structure, the only other known impact structure in the Orange River 123 124 basin, where collected to evaluate if detrital shocked zircon from this Jurassic-age structure are 125 present in local fluvial systems that could potentially be transported by tributary streams to the Orange River. 126

Seven of the samples were collected in 2009 (09VDxx series), and seven in 2014 127 (14VDxx series); one of the 2009 sites on the Atlantic coast (09VD48) was resampled in 2014 128 129 (14VD80). Distances listed for samples in the Orange River are reported relative to the highway Route 53 bridge over the Vaal River at Parys, which is the closest point of the Vaal River to the 130 131 center of the Vredefort impact structure. Distances listed for samples on the Atlantic coast and 132 near the Morokweng impact structure are relative to the Orange River mouth. Samples were 133 washed, dried and sieved to $< 0.5 \text{ mm} (1\phi)$. Zircon grains were further concentrated using heavy 134 liquids and a Frantz magnetic separator. Extreme care was taken to eliminate cross-sample 135 contamination, as shocked zircon abundances were assumed to be low. Zircon grains were handpicked and placed on aluminum scanning electron microscope 136

137 (SEM) stubs. Documentation of shock microstructures on grain exteriors was done using

138	backscattered electron imaging (BSE) with a Hitachi S-3400N SEM at the Eugene Cameron
139	Electron Microprobe Lab at the University of Wisconsin-Madison. Further SEM analysis was
140	conducted using a Tescan MIRA3 field emission gun (FEG) SEM at the Microscopy and
141	Microanalysis Facility in the John de Laetre Centre at Curtin University. The FEG-SEM was
142	used for BSE and panchromatic cathodoluminescence (CL) imaging, and electron backscatter
143	diffraction (EBSD) analysis. Automated EBSD maps of regions of interest were generated by
144	indexing electron backscatter diffraction patterns (EBSPs) on user-defined grids. Mean angular
145	deviation values for each map ranged from 0.25 to 0.61°; the only post-collection filtering used
146	was the wildspike correction. Maps of whole grains and smaller regions of interest were
147	collected using step sizes ranging from 100 to 500 nm. EBSD analyses were collected with a 20
148	kV accelerating voltage, 70° sample tilt, 20.5 mm working distance, and 18 nA beam current.
149	EBSPs were collected with a Nordlys Nano high resolution detector and Oxford Instruments
150	Aztec system using routine data acquisition settings for zircon (e.g., Reddy et al., 2007). EBSD
151	maps and pole figures were processed using the Tango and Mambo modules in the Oxford
152	Instruments/HKL Channel 5 software package.
153	EBSD maps were used to illustrate different types of microstructures (figures 3, 4, 5).
154	Band contrast (BC) maps display electron backscatter pattern quality, and were used as a
155	background image to facilitate description of microstructures along with other EBSD maps.
156	Texture component (TC) maps display variations in crystal orientation relative to a user-selected
157	reference point (indicated by the red cross in figures 3c, 4c, 5c). Inverse pole figure (IPF) maps
158	are color-coded according to Miller index to identify variations in crystallographic orientation.
159	The grain boundary (GB) function inserts a colored line (red) along the boundary of adjacent
160	pixels that form a specific misorientation about a specific axis. In figures 3, 4, and 5, the GB

161 function was used to identify {112} twins, which are defined by a 65° misorientation about
162 <110>.

163	U-Th-Pb geochronology data were collected for three shocked zircon grains. The grain
164	from sample 09VD48 was analyzed using SHRIMP-RG in the SUMAC laboratory at Stanford
165	University in 2010. Four analyses of grain 09VD48-2 were performed using a 25 μ m primary
166	beam. The ${}^{238}\text{U}/{}^{206}\text{Pb}$ ages obtained were calibrated with zircon standard R33 (${}^{238}\text{U}/{}^{206}\text{Pb}$ age =
167	419 Ma) (Black et al., 2004), and U concentration was calibrated with zircon standard MAD
168	(4196 ppm U) (Barth and Wooden, 2010). Grains from samples 14VD77 and 14VD80 were
169	analyzed using SHRIMP II at Curtin University in 2015. SHRIMP II analyses were collected
170	using a 20 μ m diameter spot with a primary beam of 1.9 nA. A total of eight analyses were
171	conducted on the two zircon grains, including five analyses of zircon 14VD80-373 and three
172	analyses of zircon 14VD77-1224. The 238 U/ 206 Pb ages were calibrated with zircon standard R33,
173	while U concentration was calibrated using zircon standard M257 (840 ppm U) (Nasdala et al.,
174	2008). Data reduction was done using SQUID (Ludwig, 2001a) and ages calculated and plotted
175	using Isoplot (Ludwig, 2001b).
176	Results
177	A total of 11,168 detrital zircon grains from the three disparate geographic areas of South

A total of 11,168 detrital zircon grains from the three disparate geographic areas of South Africa were surveyed to search for shock features. Shocked grains were only identified in three samples near the Orange River mouth on the Atlantic coast (Fig. 1b). A summary of the imaging and geochronological results is presented below.

181 Middle Orange River alluvium (5 sites, 2187 zircon grains)

182 From the middle Orange River a total of 2187 detrital zircon grains from five modern
183 alluvium samples were surveyed. Samples 14VD69, 09VD38, 09VD44, 09VD45 and 09VD46

represent alluvium at increasing distances of 761, 776, 1054, 1202, and 1219 km, respectively,

downriver from the Vredefort Dome. Sample 14VD69 was collected from a sandbar 2 km

186 downstream from the Vaal-Orange river confluence. No shocked zircon grains were found in the

populations investigated, which ranged from 41 (09VD45) to 1153 (14VD69) grains per sample

188 (Table 1).

189 Atlantic coast, near Orange River mouth (4 sites, 7326 zircon grains)

A total of 7326 zircon grains were surveyed from two modern alluvium samples collected 190 191 from sandbars (14VD76, 14VD77), and two modern beach sand samples (09VD48, 14VD80) at and near the mouth of the Orange River, approximately 1940 km downriver from the Vredefort 192 Dome. Three grains were identified that preserve conspicuous, closely-spaced (~5 µm) planar 193 194 microstructures in multiple orientations on exterior surfaces (Fig. 2, Table 1). One of the grains was found in a sandbar 15 km upriver from the Atlantic coast (14VD77-1224) (Fig. 2a-c), and 195 the other two grains were found in beach sand on the Atlantic coast at the mouth of the Orange 196 River (09VD48-2 and 14VD80-373) (Fig. 2d-i). Each of the three sediment samples yielded a 197 198 single detrital shocked zircon, which equates to abundances of 0.09% (1/1055, 09VD48), 0.04% 199 (1/2745, 14VD80), and 0.03% (1/3174, 14VD77) per sample.

200 On polished surfaces, the PFs are readily visible with BSE and CL imaging. In zircon

201 14VD77-1224 three orientations of PFs, some of which offset the margin of the grain, are visible

in the BSE image (Fig. 3a). Shock features are not obvious in the CL image of this grain, which

shows both oscillatory and sector zoning (Fig. 3b). EBSD mapping revealed three sets of {112}

shock-twins misoriented $65^{\circ} < 110$ relative to the host grain, and $< 10^{\circ}$ of cumulative

- 205 misorientation across the grain accommodated by low-angle boundaries (Fig. 3c-e). Zircon
- 14VD80-373 shows one orientation of PFs in the BSE image (Fig. 4a). PFs are faintly visible in

207	CL (Fig. 4b), which shows a light rim around a dark core that appears disturbed. EBSD mapping
208	revealed a set of {112} shock-twins in the same orientation as one of the PF sets (Fig. 4c-e). Up
209	to 20° of cumulative misorientation across the grain was detected (Fig. 4c), however with the
210	exception of grain margins, the majority of cumulative misorientation across the grain is $<10^{\circ}$,
211	and accommodated by low-angle boundaries. Zircon 09VD48-2 contains two sets of PFs visible
212	in BSE and CL images (Fig. 5a-b). EBSD mapping revealed a single orientation of {112} shock-
213	twins, and a set of planar deformation bands (PDBs) parallel to the c-axis (Fig. 5c-d). Up to 15°
214	of cumulative misorientation across the grain was detected (Fig. 5c); the majority of cumulative
215	misorientation across the grain is <10°, and accommodated by low-angle boundaries and PDBs.
216	Atlantic coast, south of the Orange River mouth (3 sites, 1502 zircon grains)
217	A total of 1502 detrital zircon grains were surveyed from three modern beach sand
218	samples collected south of the Orange River mouth on the Atlantic coast. Samples 14VD79,
219	14VD86, and 14VD89 were collected at increasing distances of 87, 339, and 401 km,
220	respectively, south of the Orange River mouth (Table 1) in order to detect if shocked zircon
221	grains were being transported northward from former sites of the Orange River mouth via
222	longshore drift. Sample 14VD89, located 401 km south of the Orange River mouth, was sampled
223	at the mouth of the Oliphants River, which represents the southern-most location of the paleo-
224	Orange River mouth since the Cretaceous (Dingle and Hedey, 1984). However, no shocked
225	zircon grains were identified in the three populations surveyed, which ranged from 170
226	(14VD79) to 783 (14VD89) grains per sample.
227	Morokweng area, Northwest Province (2 sites, 153 zircon grains)
228	A total of 153 detrital zircon grains from two samples of modern alluvium near the
229	Morokweng impact structure were surveyed. Sample 09VD51 was collected from the Phepane

230 River, and sample 09VD52 was collected from the Mashowing River; these two rivers, both dry, drain the northern and southern regions of the Morokweng structure. Both fluvial systems are 231 232 tributaries of the Molopo River, which is a right-bank tributary of the Orange River; the two 233 samples are located 1248 and 1165 km upriver from the Orange River mouth, on the southern 234 margin of the Kalahari desert (Fig. 1). Both samples yielded few zircon grains, resulting in a relatively small number of grains surveyed. No shocked zircon grains were found in either 235 population, which ranged from 86 (09VD51) to 67 (09VD52) grains per sample (Table 1). 236 **U-Th-Pb** geochronology 237 A total of 12 SHRIMP analyses were made on the three shocked zircon grains from the 238 239 Orange River mouth area (Table 2). Five analyses of zircon 14VD80-373 are variably discordant 240 and co-linear, and yield a concordia upper intercept age of 3092 ± 18 Ma (2σ , MSWD=0.65, n=5) (Fig. 6a). Four analyses of grain 09VD48-2 are also variably discordant and co-linear, and yield 241 242 a concordia upper intercept age of 3040 ± 16 Ma (2σ , MSWD=0.63, n=4) (Fig. 6b and Table 2). 243 Three analyses of grain 14VD77-1224 are not co-linear, but overlap in a cluster on the concordia 244 curve, with one spot showing slight reverse discordance; together they yield a weighted mean 207 Pb/ 206 Pb age of 3130±16 Ma (2 σ , MSWD=0.91, n=3) (Fig. 6c). The data from grains 245 09VD48-2 and 14VD80-373 show variable discordance; discordia regressions define lower 246 247 intercept ages of 268±490 Ma and 809±380 Ma, respectively, which indicate that Pb-loss 248 occurred long after the 2020 Ma Vredefort impact (Fig. 6a-b). Discussion 249 **Shock microstructures** 250 The three detrital zircon grains (09VD48-2, 14VD77-1224 and 14VD80-373) discovered 251 near the mouth of the Orange River at the Atlantic coast preserve unambiguous shock 252

253 microstructures. The grains contain multiple orientations of PFs visible on external surfaces,

which correlate to features visible in polished section. Each grain contains between one and three

- orientations of {112} twins as measured by EBSD, as well as evidence of crystal-plastic
- deformation accommodated by low-angle boundaries. The formation of planar microstructures
- and {112} shock-twins requires pressures of 20 GPa (Leroux et al., 1999; Morozova, 2015), thus
- confirming an unequivocal impact-provenance origin for these grains.
- 259 Provenance of the detrital shocked zircon grains

260 An origin from the Vredefort Dome. The microstructures described above provide clear evidence of shock deformation, but cannot alone be used to determine the provenance of the 261 262 grains. The three zircon grains yield Archean ages of 3040 ± 16 , 3092 ± 18 , and 3130 ± 16 Ma, 263 which are interpreted as bedrock crystallization ages, given that shock-twinning has not been shown to reset U-Pb ages at the scale of a 20 µm SHRIMP spot (Cavosie et al., 2015b). The 264 265 above ages correlate well with ages of bedrock exposed in the core of the Vredefort impact structure, which range from 3500 to 3010 Ma (Kamo et al., 1996; Hart et al., 1999; Moser et al., 266 267 2001; Flowers et al., 2003; Armstrong et al., 2006). Pb-loss within the analyzed zircon grains 268 does not record the 2020 Ma Vredefort impact, and was likely caused by either the Neoproterozoic Pan-African orogeny or the 1.11 - 1.02 Ga Kibaran orogeny (Jacobs et al., 1993; 269 Kamo et al., 1996; Reimold et al., 2000). Similar Pb-loss ages have been reported in studies of 270 271 shocked zircon from Vredefort bedrock (Flowers et al., 2003; Armstrong et al., 2006; Moser et 272 al., 2011) and in detrital shocked zircon suites in the Vaal River (Erickson et al., 2013a), which 273 lends further support to our interpretation that the grains described here originated from the Vredefort impact structure. 274

275 The Morokweng impact structure? The possibility that the three detrital shocked 276 zircon grains described here did not originate from the Vredefort structure was also considered. Only one other impact crater has been confirmed in the Orange River basin. The 145 Ma 277 278 Morokweng impact structure, located in the Northwest Province of South Africa, is a ~70 km 279 wide crater (Hart et al., 1997). The Morokweng structure is within the Molopo River basin, a 280 northern right-bank tributary whose confluence with the Orange River is 635 km upriver from the Atlantic coast. The location of Morokweng inside the Orange basin allows for the possibility 281 282 that detrital shocked zircon at the Orange River mouth may have originated from this structure. However an origin from Morokweng is considered unlikely for the following reasons. 283 284 The Morokweng structure is buried, and shocked bedrock is not exposed. Archean granitoids ranging from 3.0 to 2.9 Ga occur regionally but are unshocked; most local bedrock 285 286 belongs to the 2.25-2.5 Ga Transvaal and Griquatown groups, which are also unshocked (Corner 287 et al., 1997), and are too young to have yielded the ca. 3.0-3.1 Ga shocked zircon grains described here. Ephemeral streams crosscut the Morokweng structure, two of which, the Phepane 288 and Mashowing dry rivers, were sampled for this study. Corner et al. (1997) reported shocked 289 290 quartz in two of 82 cobbles sampled from the bed of the Mashowing River. However, our survey 291 of 86 detrital zircon grains from the Mashowing River and 67 detrital zircon grains from the 292 Phepane River did not identify any shocked zircon grains (Table 1). For the past 1000 years, a 293 barchan dune has blocked the Molopo River 15 km upstream from its confluence with the Orange River; today the Molopo is a dry ephemeral river that only flows during infrequent 294 295 floods (Bremner et al., 1990). The Morokweng impact structure likely contributed detrital shocked minerals to local fluvial systems in the Kalahari region after its formation at 145 Ma, 296 297 and before subsequent burial. However, we cite the absence (or at very least, low abundance) of

detrital shocked zircon in modern sediment at the site of the buried structure, the absence of
exposed ca. 3.0-3.1 Ga shocked-zircon bearing bedrock, and the minimal discharge of the dry
regional fluvial systems together to indicate that it is highly unlikely that detrital shocked zircon
grains from the Morokweng structure are currently being transported ~1200 km downstream to
the mouth of the Orange River in modern alluvium.

An unidentified impact structure? An origin of the shocked grains from an unidentified 303 impact structure located in southern Africa was also considered, as it is possible that the grains 304 305 originated from an impact crater within the Orange River basin that has yet to be identified. The Orange River basin covers nearly a million square kilometers (Bremner, 1990), and drains the 306 western half of the Kaapvaal craton (Fig. 1). The ca. 3.0-3.1 Ga Archean crystallization ages 307 308 require that the shocked zircon grains originated on the Kaapvaal craton (unless they were shocked as detrital grains in a sedimentary rock deposited pre-impact, a highly speculative 309 scenario which we do not consider further). The three shocked zircon grains each contain {112} 310 311 twins, and are thus interpreted to have originated from the central uplift of an impact structure 312 that exposes shocked ca. 3.0-3.1 Ga Archean crust, analogous to the Vredefort Dome (e.g. Moser et al., 2011). While this explanation is speculative and not viewed as likely, the discovery of a 313 new impact structure in southern Africa would not be unusual given the overall low density of 314 impact craters confirmed in Africa (Spray, 2001; Reimold and Koeberl, 2014). Three new impact 315 316 structures have been confirmed in Africa since 2010 (Folco et al., 2010; Ferriere et al., 2011; 317 Chennaoui Aoudhehane et al., 2016). If a new impact structure in the Orange River basin is later 318 discovered, the provenance characteristics of the detrital shocked grains described here can be revisited to further evaluate their potential source crater. 319

Taken together, the diagnostic shock microstructures, narrow range of U-Pb ages from 320 321 3040 to 3130 Ma, ubiquitous presence of shocked zircon in modern Vaal River alluvium, the 322 definitive presence of shocked zircon in lower Orange River alluvium, the paucity of shocked 323 zircon in modern sediment south of the Orange River mouth and near the Morokweng impact 324 structure all support our preferred interpretation, whereby the detrital shocked zircon grains discovered on and near the Atlantic coast originated from the Vredefort impact structure, and 325 326 were transported downstream by fluvial processes in the combined Vaal-Orange rivers nearly 327 1940 km downstream (Fig. 1).

328 Sedimentology of detrital shocked zircon in the Orange River

Detrital shocked zircon abundance. The abundance of detrital shocked zircon in the 329 Vaal-Orange river system decreases with distance from their inferred point source, the Vredefort 330 impact structure. Cavosie et al. (2010) documented detrital shocked zircon in the Vaal River 331 within the Vredefort Dome, reporting abundances ranging from 36 - 64%. The abundance 332 decreases downstream in the Vaal River, from 18% at 100 km to 2% at 759 km near the Vaal-333 334 Orange confluence (Erickson et al., 2013a). Results from the four samples in this study located at 335 or near the mouth of the Orange River, summed together, yield an abundance of 0.04% (3/7326) at nearly 2000 km downriver. The abundance values cited here, based on identification of single 336 shocked grains, are variable, and dependent on the total number of grains analyzed, which also 337 338 varied among samples. While none of the samples from the Orange River mouth area yielded 339 more than a single shocked zircon, we note the reproducibility of this result between samples 340 09VD48 and 14VD80, which were collected from the same location 5 years apart. These two samples were collected, processed, and analyzed by different persons, and each yielded a single 341 shocked zircon in the population surveyed, which ranged from 1055 to 2745 grains per sample. 342

343 Paucity of shocked zircon in the middle Orange River. One puzzling result of this 344 study is that no detrital shocked zircon grains were found in samples from the middle Orange River. The middle Orange River sample suite includes five modern alluvium samples located 345 346 from 761 to 1219 km downstream from the Vredefort Dome, including sample 14VD69, located 347 only 2 km below the Vaal-Orange confluence. The number of grains investigated per sample from the middle Orange River (samples 09VD38, 09VD44, 09VD45, and 09VD46) is somewhat 348 lower (<500 grains per sample) in comparison with the number of grains investigated in samples 349 350 near the Orange River mouth (352-3174 grains per sample). However, >1000 grains were surveyed from the sample 2 km below the Vaal-Orange confluence that did not yield any 351 352 shocked grains (14VD69, 0/1153). This sample is located only ~5 km further downstream from a known detrital shocked zircon site in the Vaal River (sample 09VD41) that yielded 5 shocked 353 grains from a population of 253 grains investigated (2%, Erickson et al., 2013a). We predicted 354 identifying up to 20 detrital shocked zircon grains in the population of 1153 grains surveyed 355 from sample 14VD69 based on the prior results from sample 09VD41. One possible explanation 356 for not finding shocked zircon in the middle Orange River is that the high sediment production 357 and high runoff from the upper Orange may dramatically dilute sediment input from the Vaal 358 River near and below the confluence. 359

Mode of detrital transport. The mode of transport that resulted in detrital zircon from the Vredefort Dome being present nearly 2000 km downriver merits further discussion. Each of the three detrital shocked grains described here were collected as modern sediment (beach sand and alluvium) in environments uniquely dominated by medium-to-coarse grained sand, with no obvious coarser material locally present (e.g., pebbles or larger particles). Based on field observations, the zircon grains appear to have been transported to and deposited at the sites

where they were collected as loose grains. However, the three shocked zircon grains have 366 remarkably well preserved prisms and pyramids, and show only minor evidence of abrasion and 367 rounding. In this regard, their exterior surfaces are similar to those of detrital shocked zircon 368 grains described from the Vaal River, where the planar microstructures are pervasively etched, 369 but the grains otherwise show little evidence of sedimentary abrasion (Cavosie et al., 2010; 370 Erickson et al., 2013a). While we cannot rule out the possibility that these grains were 371 incorporated in larger particles of rock for part of their transport history in the Vaal-Orange 372 373 rivers, we have no evidence to support this mode of transport, as clasts of Vredefort-derived bedrock have thus far not been reported downriver of the impact structure. The absence of 374 pronounced sedimentary abrasion (and subsequent rounding) in grains that have experienced 375 376 distal transport in fluvial systems might instead reflect the lack of an eolian transport history. Detailed studies of texturally mature Precambrian quartz arenite occurrences have identified the 377 critical role of eolian transport in both the abrasion and rounding of detrital quartz grains and 378 379 associated accessory minerals (e.g., Dott, 2003). Given that the Vaal River cross-cuts and is entrenched in shocked bedrock at the Vredefort Dome, it is reasonable to conclude that shocked 380 zircon grains transported in the Vaal-Orange rivers experienced little, if any, history of eolian 381 transport. We thus attribute the well-preserved forms of the shocked zircon grains on and near 382 the Atlantic coast to the dominance of fluvial processes as the main mechanism that transported 383 384 the grains in modern alluvium nearly 2000 km downriver from the Vredefort structure. 385 Our results from southern Africa, together with previous studies, suggest that SEM imaging surveys for detrital shocked minerals in alluvium eroded from known impact structures 386 should be able to identify shocked grains in a sample as small as ~250 zircon grains if they are 387

located <750 km from the source crater (Cavosie et al., 2010; Erickson et al., 2013a). However,

389	the surveyed population should be expanded to >1000 zircon grains when searching for distally
390	transported detrital shocked grains (>750 km), or if the location of the source crater is unknown
391	or only suspected (Reddy et al., 2015). In our experience, surveying populations of 1000 (or
392	more) grains per sample by SEM is practical from an analytical point of view, and more
393	significantly, allows an evaluation of the presence of shocked zircon at a minimum abundance
394	level of 0.1% (1/1000). The size of the impact structure, level of erosion, extent of exposed
395	shocked bedrock, and abundance of zircon-bearing target rock will all influence the sedimentary
396	record of detrital shocked zircon. Additional studies are required at other sites to evaluate if the
397	erosional record of the Vredefort impact structure is a general result, or a consequence of the
398	unique geologic history of the region (c.f, Thomson et al., 2014).
399	Implications
400	These results demonstrate that shocked zircon from ancient and deeply eroded impact
401	structures can survive fluvial transport to distal localities, and preserve impact evidence in the

for detrital shocked zircon previously documented within the Vredefort Dome and the Vaal
River (Cavosie et al., 2010; Erickson et al., 2013a).

form of diagnostic shock microstructures. Our results greatly expand known transport distances

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One implication of the discovery of detrital shocked zircon on the Atlantic coast of southern Africa is that shocked grains are evidently resilient enough to survive distal fluvial transport, and can end up in areas beyond the drainage basin they originated in. Sediment, including detrital zircon grains, from the Orange River basin has been found along the Atlantic coast of Namibia (Stone, 2013), as far as 700 km north of the Orange River mouth, at Walvis Bay (Lancaster and Ollier, 1983; Bluck et al., 2007; Vermeesch et al., 2010). The above observations clearly imply that Vredefort-derived shocked zircon grains are likely to occur in

412 detrital populations at these locations along the coast of Namibia, albeit at predictably low

413 abundance levels (<0.1%).

414	Considering the process of distal transport of shocked grains further back in time, detrital
415	shocked zircon grains from Phanerozoic and Precambrian impact structures are predicted to
416	reside in younger sedimentary rocks. Detrital grains as old as 4.4 Ga (Cavosie et al., 2005;
417	Cavosie et al., 2007; Valley et al., 2014) provide clear evidence that populations of ancient
418	detrital zircon are preserved, which may contain shocked grains at detectable abundance levels
419	(e.g., <0.1%). Discovery of such grains would provide the first ground-truth evidence for the
420	earliest meteorite bombardment history of Earth, and allow the testing of models for
421	establishment of habitable conditions on the early Earth.
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609	Figure Captions
610	Figure 1. Simplified map of southern Africa. (a) Significant elements include the Kaapvaal
611	craton, Vredefort Dome, Vaal and Orange river basins (shaded), and the subdivisions of the

612 Orange River. Stars show locations of sediment samples. (b) Sample locations near and at the

- 613 Orange River mouth on the Atlantic coast.
- **Figure 2.** Exterior images of three detrital shocked zircon grains using backscattered electrons.
- (a) Image of zircon 14VD77-1224, found in an Orange River sandbar 15 km upriver from the
- Atlantic coast. (b) Close-up image showing 2 sets of PFs. (c) Close-up image showing 3 sets of
- 617 PFs. (d) Image of zircon 14VD80-373, found in beach sand on the Atlantic coast. (e) Close-up
- 618 image showing 4 sets of PFs (f) Close-up image showing 3 sets of PFs. (g) Image of zircon
- 619 09VD48-2, found in beach sand on the Atlantic coast. (h) Close-up image showing 1 set of PFs.
- 620 (i) Close-up image showing 2 sets of PFs. Arrows indicate orientations of planar fractures.
- **Figure 3.** Interior images of grain 14VD77-1224. (a) BSE image showing three PF orientations
- 622 (arrows). (b) CL image showing oscillatory zoning and location of geochronology analyses with

623 ²⁰⁷Pb/²⁰⁶Pb ages (in Ma). (c) Combined band contrast (BC), grain boundary (GB) and texture

- 624 component (TC) map showing twins (red lines) and crystal-plastic deformation. Red cross
- 625 indicates reference location. (d) Orientation map showing three sets of twins, labeled (t1-t3).
- 626 This map combines BC, GB, and is colored with an inverse pole figure (IPF-Z) scheme. (e)
- 627 Stereonets (lower hemisphere) of orientation data from (d) showing crystallographic relationship
- between twins and host zircon. The twins are sets of lamellae orientated 65° about $\{110\}$; each
- 629 twins shares a $\{112\}$ axis with the host grain.
- **Figure 4.** Images of grain 14VD80-373. (a) BSE image showing one PF set (arrow). (b) CL
- 631 image showing disturbed zoning and location of geochronology analyses with ²⁰⁷Pb/²⁰⁶Pb ages
- 632 (in Ma). (c) Combined band contrast (BC), grain boundary (GB) and texture component (TC)
- map showing twins (red lines) and crystal-plastic deformation. Red cross indicates reference
- 634 location. (d) Orientation map showing one sets of twins (labeled t1). This map combines BC,

635	GB, and is colored with an inverse pole figure (IPF-Z) scheme. (e) Stereonets (lower
636	hemisphere) of orientation data from (d) showing crystallographic relationship between twins
637	and host zircon.
638	Figure 5. Interior images of grain 09VD48-2. (a) BSE image showing two PF sets (arrows). (b)
639	CL image showing sector zoning and location of geochronology analyses with 207 Pb/ 206 Pb ages
640	(in Ma). (c) Orientation map showing one set of twins (red lines), planar deformation bands
641	(PDB), and crystal-plastic deformation. Red cross indicates reference location. (d) Stereonets
642	(lower hemisphere) of orientation data from (c) showing crystallographic relationship between
643	twins and host zircon.
644	Figure 6. U-Pb concordia diagrams. Results of the SHRIMP analyses on zircon grains (a)
645	14VD80-373, (b) 09VD48-2, and (c) 14VD77-1224. Regressions in the top two panels are not
646	anchored to the concordia curve.
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Sample	Latitude	Longitude	Zrn ^a , grains surveyed	Zrn, shocked	Zrn, % shocked	Distance (km)	
Middle Ora	Middle Orange River ^b						
14VD69	S29°04.626'	E23°37.358'	1153	0	0	761	
09VD38	S29°13.087'	E23°21.045'	481	0	0	776	
09VD44	S28°52.803'	E21°59.145'	379	0	0	1054	
09VD45	S28°43.740'	E20°59.142'	41	0	0	1202	
09VD46	S28°38.121'	E21°05.377'	133	0	0	1219	
		subtotal	2187				
Atlantic coa	st and Lower Or	ange River, near	Orange River m	outh ^b			
14VD77	S28°33.344'	E16°32.903'	3174	1	0.03	1925	
14VD76	S28°34.015'	E16°30.439'	352	0	0	1929	
14VD80 ^c	S28°38.59'	E16°28.32'	2745	1	0.04	1939	
09VD48 ^c	S28°38.59'	E16°28.32'	1055	1	0.09	1939	
		subtotal	7326				
Atlantic coa	st, south of Oran	ge River mouth ^d					
14VD79	S29°15.180'	E16°52.055'	170	0	0	87	
14VD86	S31°15.357'	E17°51.726'	549	0	0	339	
14VD89	S31°42.321'	E18°11.559'	783	0	0	401	
		subtotal	1502				
Near the Mo	Near the Morokweng impact structure ^d						
09VD52	S26°35.153'	E22°40.559'	67	0	0	1165	
09VD51	S25°47.282'	E22°54.882'	86	0	0	1248	
		subtotal	153				
Total			11168	3	0.03		

Table 1. Sample location information and summary of shocked zircon search.

^a Zrn = zircon.

^bDistance cited is from the Route 53 bridge in Parys to the sample location.

^c Collected from a 2 km long sand spit at the Orange River mouth on the Atlantic coast;

the location cited is in the middle of the sand spit.

^d Distance cited is from the sample location to the mouth of the Orange River.

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Spot	U (ppm)	Th (ppm)	Th/ U	% comm ²⁰⁶ Pb ^a	²⁰⁴ Pb/ ²⁰⁶ Pb	²⁰⁸ Pb ^a / ²³² Th	²⁰⁷ Pb ^a / ²³⁵ U	²⁰⁶ Pb ^a / ²³⁸ U	$^{207}{Pb^{a}}/^{206}{Pb^{a}}$	²⁰⁶ Pb ^a / ²³⁸ U Age (Ma)	²⁰⁷ Pb ^a / ²⁰⁶ Pb ^a Age (Ma)	% conc
14VD77-1224												
1224-1	96	88	0.94	0.18	0.0001	0.139 ± 0.003	20.32 ± 0.3912	0.6055 ± 0.0106	0.24341 ± 0.00198	3052 ± 85	3142 ± 26	97
1224-2	348	468	1.39	0.17	0.0001	0.153 ± 0.002	19.92 ± 0.2911	0.6000 ± 0.0074	0.24077 ± 0.00187	3030 ± 60	3125 ± 25	97
1224-3	207	234	1.17	0.17	0.0001	0.161 ± 0.003	20.99 ± 0.3964	0.6367 ± 0.0095	0.23912 ± 0.00278	3176 ± 75	3114 ± 37	102
14VD80-373												
373-1	183	61	0.35	1.03	0.00057	0.126 ± 0.003	16.74 ± 0.2413	0.5303 ± 0.0070	0.22893 ± 0.00133	2743 ± 59	3045 ± 19	90
373-2	107	46	0.45	0.80	0.00044	0.126 ± 0.005	17.64 ± 0.2903	0.5563 ± 0.0083	0.22994 ± 0.00156	2851 ± 69	3052 ± 22	93
373-3	340	175	0.53	0.39	0.00021	0.159 ± 0.003	19.06 ± 0.2787	0.5893 ± 0.0084	0.23463 ± 0.00081	2987 ± 68	3084 ± 11	97
373-4	263	112	0.44	0.26	0.00014	0.148 ± 0.005	18.85 ± 0.2405	0.5875 ± 0.0072	0.23265 ± 0.00087	2979 ± 58	3070 ± 12	97
373-5	283	94	0.35	0.43	0.00024	0.149 ± 0.002	18.46 ± 0.2378	0.5753 ± 0.0070	0.23269 ± 0.00095	2930 ± 58	3071 ± 13	95
09VD48-2												
48-1	130	150	1.19	0.04	0.00002	0.5379 ± 0.0110	16.767 ± 0.356	0.5379 ± 0.0110	0.22608 ± 0.00122	2775 ± 92	3024 ± 17	92
48-2	85	56	0.68	0.27	0.00015	0.5935 ± 0.0127	18.595 ± 0.414	0.5935 ± 0.0127	0.22724 ± 0.00143	3004 ± 102	3033 ± 20	99
48-3	101	91	0.93	0.59	0.00033	0.5656 ± 0.0119	17.885 ± 0.423	0.5656 ± 0.0119	0.22933 ± 0.00250	2890 ± 98	3047 ± 35	95
48-4	112	94	0.87	0.02	0.00001	0.5693 ± 0.0119	17.910 ± 0.384	0.5693 ± 0.0119	0.22815 ± 0.00110	2905 ± 98	3039 ± 15	96

Table 2. Detrital shocked zircon U-Th-Pb isotopic data from SHRIMP.

comm = common; conc = concordance.

^a Indicates a ²⁰⁴Pb corrected Pb value after Ludwig, 2001a.

Uncertainty in isotopic ratios and ages are listed at 2σ .

The % concordance is calculated as follows: $[(^{206}Pb^a/^{238}U age)/(^{207}Pb^a/^{206}Pb^a age)] \ge 100\%$.



Figure 1



Figure 2



Figure 3



Figure 4



Figure 5

