1	Deciphering the enigmatic origin of Guyana's diamonds. (Revision 2)
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3	Key Words: diamond, Guyana, Guiana Shield, Roraima Supergroup.
4	Abstract
5	Diamonds have long been mined from alluvial terrace deposits within the rainforest of Guyana,
6	South America. No primary kimberlite deposits have been discovered in Guyana, nor has there
7	been previous studies on the mineralogy and origin of the diamonds. Paleoproterozoic terranes in
8	Guyana are prospective to diamond occurrences because the most productive deposits are
9	associated spatially with the eastern escarpment of the Paleoproterozoic Roraima Supergroup.
10	Geographic proximity suggests that the diamonds are detrital grains eroding from the <1.98 Ga
11	conglomerates, metamorphosed to zeolite and greenschist facies. The provenance and
12	paragenesis of the alluvial diamonds are described using a suite of placer diamonds from
13	different locations across the Guiana Shield. Guyanese diamonds are typically small, and those
14	in our collection range from 0.3 to 2.7 mm in diameter; octahedral and dodecahedral, with lesser
15	cubic and minor macle forms. The diamonds are further subdivided into those with abraded and
16	non-abraded surfaces. Abraded diamonds show various colors in cathodoluminescence whereas
17	most non-abraded diamonds appear blue. In all populations, diamonds are predominantly
18	colorless, with lesser brown to yellow and very rare white. Diamonds are predominantly Type
19	IaAB and preserve moderate nitrogen aggregation and total nitrogen concentrations ranging from
20	trace to ~1971 ppm. The kinetics of nitrogen aggregation indicate mantle-derived residence
21	temperatures of $1124 \pm 100$ °C, assuming residence times of 1.3 Ga and 2.6 Ga for abraded and
22	non-abraded diamonds respectively. The diamonds are largely sourced from the peridotitic to

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eclogitic lithospheric upper mantle based on both  $\delta^{13}$ C values of -5.82 ± 2.45‰ (VPDB-LSVEC) and inclusion suites predominantly comprised of forsterite, enstatite, Cr-pyrope, chromite, rutile, clinopyroxene, coesite, and almandine garnet. Detrital, accessory minerals are non-kimberlitic. Detrital zircon geochronology indicates diamondiferous deposits are predominantly sourced from Paleoproterozoic rocks of 2079 ± 88 Ma.

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## Introduction

Diamonds have been mined for the past century from alluvial gravels along the rivers and 29 creeks deep within Guyana's Amazon rainforest. The diamonds are found as placers in paleo-to-30 modern channels, terraces, and streambeds. Guyana's diamonds are found in headless placers, 31 with the most productive gravels associated spatially with the eastern escarpment of the Roraima 32 Supergroup, which suggests that the diamonds originate from this detrital source (Fig. 1). The 33 humid tropical climate and ancient weathering profile of the Guiana Shield makes addressing 34 diamond provenance a difficult task. Exploration has been driven by artisanal miners who 35 prospect using detrital indicator minerals. No primary kimberlites are known in the region, and 36 there is a lack of prior research on the nature and origin of diamonds in Guyana (e.g., Gibbs and 37 Barron 1993, Shields and Letendre 1999, Persaud 2010). 38

Several hypotheses exist on the origin of Guyana's diamonds. Diamonds may be sourced from yet undiscovered primary kimberlites or igneous intrusions (Shields and Letendre 1999; Persaud 2010). The most likely location would be associated spatially with the highly magnesian ultramafic intrusives of the  $1.7 \pm 0.2$  Ga, Badidku suite (Olszewski et al. 1977). These rocks intruded during a tectonically quiet period of the late Trans-Amazonian orogeny (Gibbs and Barron 1999) and might be associated with other ultramafic intrusions such as kimberlites. The only confirmed examples of primary diamond deposits in the Guiana Shield are in Guaniamo,

Venezuela, and Dachine, French Guiana (Fig. 1), where diamonds are found in Neoproterozoic 46 kimberlite sills and metamorphosed Paleoproterozoic ultramafic and pyroclastic shoshonites or 47 lamprophyres, respectively (Capdevila et al. 1999; Magee and Taylor 1999; Kaminsky et al. 48 2000; Channer et al. 2001; Kaminsky et al. 2004; Wyman et al. 2008; Smith et al. 2016). 49 Kimberlites exist in Roraima, Brazil, but these are not diamondiferous (Svisero et al. 2017, 50 Cabral et al. 2017). Known diamondiferous kimberlites from West Central Africa are an 51 additional potential source for Guvana's alluvial deposits (Reid 1974 and Briceno 1984). These 52 regions were immediately adjacent prior to the Jurassic rifting and opening of the Atlantic 53 Ocean; separated by 200 to 400 km (Fig. 1). If diamonds were derived from Africa then they 54 would be more prevalent in the northeastern Guiana Shield where diamonds are not found. 55 Furthermore, West African diamonds have been traced to the host diamondiferous kimberlites 56 which range in age from 92 - 846 Ma (Bardet and Vachette 1966; Andrews-Jones 1968; Hall 57 1972; Janse 1996; Fourie et al. 1998; Fourie et al. 2000; Kiviets 2003; Skinner et al. 2004). More 58 locally, the Avanavero mafic dykes and sills (<1.79 Ga) have been suggested to be 59 diamondiferous, but these dykes are tholeiitic norites and gabbros and thus unlikely to be 60 diamond-bearing (Da Silva Rodrigues 1991; Reis et al. 2000; Heesterman et al. 2005). Finally, 61 the diamonds may be derived as detrital grains weathering directly from diamondiferous 62 conglomerates of the Paleoproterozoic Roraima Supergroup (e.g., Gibbs and Barron 1993; 63 Meyer and McCallum 1993). This may be plausible because alluvial diamonds are sometimes 64 65 found in gravels with abundant, rounded quartz, red jasper, and quartzite clasts; also observed in Roraima Supergroup conglomerates. Placer diamonds in Mutum, Brazil are mined from alluvial 66 terraces shed possibly from conglomerates of the Tepequém Formation, which is equivalent to 67 the Arai Formation pebbly sandstones and volcaniclastics of the Roraima Supergroup (Santos et 68

al. 2003; Reis et al. 2017). Similarly, in Suriname, diamonds are found in alluvium shed from
metamorphosed Proterozoic conglomerates or ultramafic volcaniclastics of the Rosebel group,
but their provenance remains unknown (van Kooten 1954; Schönberger and de Roeve 1974;
Schönberger 1975; Bosma et al. 1983; Ramlal 2018; Naipal et al. 2019). The Roraima
Supergroup and similar sedimentary sequences may have been a sink for diamonds erupted by
unknown Paleoproterozoic kimberlites, and may be detrital evidence for some of Earth's earliest
kimberlites.

Guyana's diamonds are a known commodity in the global diamond trade, referred to as 76 77 "British diamonds," stemming from a colonial past. Economically valuable diamonds in Guyana are small, most commonly ranging from 0.1 to 0.4 carats although larger stones are found. 78 Guyanese diamonds are predominantly colorless (G-J), yellow (K-M), and brown. Pink and 79 green body colors are rare. Exploration activities conducted by Golden Star resources recovered 80 alluvial diamonds and accessory minerals from Amatuk (Fig. 1) and are described as octahedral 81 with moderate resorption textures. Green and brown spotting and skins are attributed to radiation 82 damage from accessory zircon, monazite, uraninite, micas, and K-feldspar (Breeding et al. 2018), 83 when residing in a placer environment. Surface abrasion and breakage post-dating resorption 84 along crystal edges were interpreted to be derived from either a high energy fluvial environment 85 or agitation during recovery (Shields and Letendre 1999). 86

We present the morphology, abrasion and dissolution textures, nitrogen concentration, carbon isotopic composition, and inclusion mineralogy of Guyana's alluvial diamonds to learn about their provenance, host magma characteristics, residence temperatures, and mantle source beneath the Guiana Shield. We characterize Guyana's diamonds and their accessory minerals, thereby facilitating meaningful comparisons with other diamond deposits in the Guiana Shield

and elsewhere. Diamonds were obtained from the Ekereku, Jawalla, Konawaruk, Kamarang, 92 Kurupung, Maikwak, and Monkey Mountain alluvial deposits in Guyana (Fig. 1). Most 93 diamonds crystallized from peridotitic lithosphere, although there is a lesser population of 94 eclogitic diamonds. These diamonds can be divided into two basic groups; those with and 95 without sedimentological abrasive textures. Importantly, non-abraded and abraded diamonds 96 have statistically different cathodoluminescence, ultraviolet blue light luminescence, and Fourier 97 Transform Infrared (FTIR) responses. Abraded diamonds are evidence for a detrital, polycyclic 98 source weathering from the Roraima Supergroup. Non-abraded diamonds may be derived from 99 the Roraima Supergroup, but have experienced less transport and recycling. 100 **Regional Geology** 101 Diamonds occur across the Guiana Shield and lesser portions of the Guaporé Shield 102 within the Amazonian craton of South America (Fig. 1). The Guiana Shield is exposed for over 103 900,000 km<sup>2</sup> along the northern margin of the Amazonian Craton. Chemical weathering prevails 104 throughout much of the Guiana Shield, resulting in thick sequences of saprolite capped by "tor" 105 formations of large boulders of resistive and residual host rock, with the saprolite regolith 106 stripped away by weathering (Kroonenberg and Gersie 2019). Although exposures are limited by 107 accessibility and extreme tropical weathering, there is broad consensus that the Paleoproterozoic 108 evolution of this craton was dominated by episodes of accretionary orogenic events around an 109 Archean core complex during the Main Trans-Amazonian (2.26-2.08 Ga) to Late Trans-110 Amazonian (2.07-1.93 Ga) orogenies (Tassinari 1997; Vanderhaeghe et al. 1998; Reis et al. 111 2000; Delor et al. 2003; Cordani and Teixeira 2007; Fraga et al. 2009; Daoust et al. 2011; 112 Kroonenberg et al. 2016). 113

114	The Guiana Shield is divided into four major Paleoproterozoic terranes (Daoust et al.
115	2011), one Mesoproterozoic terrane, and two isolated Archean terranes (Norcross 1997;
116	Kroonenberg et al. 2016). Crustal development in the Guiana Shield created a series of
117	greenstone belts, associated gneisses, and amphibolites of the Maroni-Itacaiunas Belt (Tassinari
118	et al. 1997). The greenstone belts underwent several episodes of deformation, intrusion, and
119	metamorphism between 2.26 Ga and 2.08 Ga, followed by cooling between 2.08 Ga and 1.93
120	Ga. These events are interpreted to reflect the expression of the Trans-Amazonian Orogeny in
121	the Guiana Shield (Cordani and De Brito Neves 1982; Gibbs and Barron 1993; Norcross 1997;
122	Daoust et al. 2011). Much of the region was then unconformably overlain by the Roraima
123	Supergroup, which is a stratigraphic succession dominated by interbedded 1.98 - 1.78 Ga
124	sandstones and conglomerates deposited from rocks eroding from earlier greenstone terranes
125	(Priem et al. 1973; Santos et al. 2003). Finally, coeval mafic dykes of the Avanavero Suite
126	intruded the entire sequence at ~1.79 Ga (Reis et al. 2000).
127	The region has been a stable craton, only modified by erosional processes, throughout
128	much of the late Phanerozoic. The upper Proterozoic was marked by a period of prolonged uplift
129	with no evidence of sedimentation (Gibbs and Barron 1993). Changing climate and repeated
130	uplift cycles since the late Triassic have resulted in continuing surface evolution of the Guiana
131	Shield. Rifting of South America from Africa beginning in the early Jurassic was associated with
132	accelerated weathering and fluvial activity (McConnell 1968). The regional drainage patterns
133	and depositional systems have evolved in response to millions of years of river capture,
134	rejuvenation, degradation, and aggradation initiated by faulting and rifting since the opening of
135	the Atlantic Ocean. Today the region preserves a complex network of high alluvial, terrace,
136	alluvial flat, river bed, buried channel, and plateau deposits. Repeated cycles of erosion and

deposition have led to complex diamond placer deposits with detrital assemblages reflecting 137 variable provenance and timing. The diamonds are found alongside accessory phases that include 138 quartz, topaz, jasper, rutile, anatase, zircon, ilmenite, gold, corundum, and tourmaline, with 139 minor garnet and chromite. Olivine and perovskite are notably absent. Diamond is the only 140 residual mineral that certainly eroded from kimberlite rock. Despite the possibility for primary 141 kimberlite sources and the complex geologic history of the Guiana Shield, placer deposits are the 142 only diamond sources that have been discovered in Guyana. 143 **Methods** 144 We acquired a collection of diamonds from miners and a local diamond merchant (Kay's 145 Diamond Enterprise Ltd.). We first cataloged each stone by measuring its dimensions and mass. 146 The overall morphology of the diamonds was then assessed using a Zeiss Scope AX IO 147 petrographic microscope. A focus was delineating crystal shape, dissolution textures, and 148 abrasion. Some features were documented using high resolution scanning electron microscopy 149 energy dispersive X-ray (SEM-EDS) spectroscopy at the Baylor University Center for 150 Microscopy and Imaging. 151 The nitrogen content of 415 diamonds were measured, and the aggregation state 152 calculated for the majority of samples using a Thermoscientific Nicolet iN10 FTIR spectrometer. 153 Analyses were performed across  $675-4000 \text{ cm}^{-1}$  in cooled transmission mode using a 200 x 200 154 µm aperture size, 64 scans, and spectral resolution of 4 cm<sup>-1</sup>. Nitrogen concentration was 155 calculated from individual spectra by applying the Beer-Lambert law and absorption values of 156 nitrogen bands at 1365, 1284, and 1175 cm<sup>-1</sup>, using the least-squares fitting approach (e.g., 157 Howell et al. 2012). 158

159	Effective thickness (x) was calculated by normalizing the absorption coefficient ( $\mu$ ) of
160	diamond lattice bands at 2443, 2158, and 2026 cm <sup>-1</sup> , where $x = \mu_{2443}/5$ ; $x = \mu_{2158}/12$ ; $x = \mu_{2026}/12$
161	and the average effective thickness was then calculated (Kaminsky and Khachatryan 2012).
162	Stable carbon isotope analyses ( $\delta^{13}$ C, in ‰ <sub>VPBD-LSVEC</sub> ) were performed at the Baylor University
163	Stable Isotope Laboratory using a Costech Elemental Combustion System 4010 connected to a
164	Thermo-Electron Delta V Advantage continuous flow Isotope Ratio Mass Spectrometer (CF-
165	IRMS) through a Thermo Conflo IV interface. Fifty-seven crushed and powdered diamond
166	samples (<0.5mm) which range from $0.05 - 0.90$ mg (mean - $0.26 \pm 0.15$ mg) were loaded into
167	silver capsules, flash combusted at 1000°C to convert the diamond to CO <sub>2</sub> , which was carried to
168	the CF-IRMS by a constant helium gas flow.
169	Optical cathodoluminescence (CL) of 556 diamonds was observed using a Nikon-Japan
170	LV UEPI microscope equipped with a low vacuum Reliotron III that operated at 7.5-9 kV and
171	0.3-0.5 amps. The CL data was supplemented with ultraviolet blue light (UV) luminescence
172	(425-495 nm) observed in 558 diamonds using an Olympus BX51 petrographic microscope
173	equipped with an EXFO X-cite 120 fluorescence illumination system, with an exposure time of
174	approximately 10 s for most samples. Each crystal's color, or lack thereof, was documented for
175	both CL and UV observations.
176	Inclusions within 91 diamonds were identified using Raman spectra collected with a
177	Thermoscientific DXR Raman microscope equipped with a 532 nm laser operating at 8 mW, a
178	~2- $\mu$ m spot, and a high resolution 1800 lines mm <sup>-1</sup> grating. Inclusion species were identified by
179	matching diagnostic peak positions and heights of unknown spectra with those in the RRUFF

180 spectral database (Lafuente et al. 2016).

Accessory minerals were extracted from heavy mineral separates that we collected from 181 buried alluvial and colluvial deposits in the Kurupung region (UTM WGS 84 Zone 20N, 182 804678E, 675615N), approximately 4km from the base of the Roraima Supergroup escarpment. 183 Rare diamonds were also recovered from these heavy mineral separates. Samples were sieved, 184 separated by mass in a shaker table, and cleaned in a sonic bath. Hand-picked accessory minerals 185 of garnet, chromite, and ilmenite were mounted in epoxy, polished at Baylor University and 186 analyzed at the University of Texas at Austin, using a JEOL JXA-8200 Electron Microprobe 187 (EPMA). Zircon crystals were also handpicked and analyzed for U-Pb crystallization ages using 188 laser-ablation-inductively coupled plasma-mass spectrometry (LA-ICPMS) at the University of 189 Arkansas Trace Element and Radiogenic Isotope Laboratory. For each sample, 150 detrital 190 zircon grains were mounted on a glass slide using double-sided tape. Grain surfaces were ablated 191 using an ESI 193 nm Excimer laser ablation system and a Thermo iCap quadropole ICP-MS. 192 Data acquisition parameters include a 35 µm spot size, laser repetition rate of 10Hz, helium flow 193 rate of 0.8 L min<sup>-1</sup>, and a fluence of ~4.5 J cm<sup>-2</sup>. Zircon from Plesovice (337.13 Ma; Slama et al., 194 2008) was used as a primary standard, with R33 (419 Ma; Black et al., 2004) as a secondary 195 standard. The weighted mean average of R33 analyses was within 1% of the accepted age. Data 196 were reduced in Iolite v. 3.71 (Paton et al., 2011), and individual analyses were manually 197 trimmed to avoid zones of high discordance and <sup>204</sup>Pb bearing inclusions. Twenty-four analyses 198 were excluded because they were not zircon or were contaminated by inclusions. An additional 199 17 zircon grains (5.6% of the total) were excluded on the basis of discordance ( $^{207}$ Pb/ $^{206}$ Pb versus 200 <sup>206</sup>Pb/<sup>238</sup>U) exceeded 10% or 5% reverse discordance. In total, 259 concordant zircon U-Pb 201 analyses were retained (Fig. A1; Table A5). 202

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#### **Results**

# 204 Morphology

205	Diamonds in our collection range in size from 0.3 to 2.7 mm in diameter; with a mean of
206	$1.1 \pm 0.2$ mm. Masses range from 0.01 g to 0.95 g. Diamonds are predominantly colorless (93%)
207	with lesser amounts of brown (4%) to yellow (3%) and very rare white (1%) variants (Fig. 2).
208	Approximately half of the diamonds show no radiation spotting (55%). The remainder has green
209	to green-blue skins or green spotting that covers up to $\sim 100\%$ of the surface area. A small
210	population (5%) show combined overprinting of brown and green spotting (Table 1).
211	Diamonds across all regions are octahedral (30%) and dodecahedral (30%), with lesser
212	combination (16%), flattened or elongate (15%), and cubic (3%) forms (Fig. 3) (Table 1). Those
213	from Ekereku present more dodecahedrons (46%) than octahedrons (31%). Kamarang has the
214	most flattened or elongated diamonds (26%), with the remainder being octahedral, dodecahedral,
215	and various combinations. Twinned diamonds and aggregates are rare from any location.
216	Fragments account for 9% of diamonds analyzed and resorbed fracture or fragmented diamond
217	surfaces were not observed.
218	Fine stepwise, lamellar trigonal faces are common, whereas flat faces and sharp edges are
219	uncommon (Fig. 3). Resorption textures, including terraces, tear drop hillocks, and dissolution
220	pits are common in 95% of diamonds. Most octahedral diamonds have resorbed edges, and in the
221	case of Ekereku, more diamonds are fully resorbed to dodecahedrons. Tear drop hillocks are the
222	most common dissolution texture (Fig. 3f). The most common pit dissolution textures are flat
223	bottom (37%) and point bottom (12%) trigons and tetragons (19%) (Table 2). Very fine (8%) to
224	fine (3%) point bottom trigons are more abundant than coarse to very coarse point bottom
225	trigons. Very fine (5%) to fine (14%) flat bottom trigons are slightly more abundant than coarse
226	(12%) to very coarse (1%) flat bottom trigons. Point bottom hexagons, and trapezoids are

notably rare. Late stage etching features such as corrosion sculptures, shallow depressions, ruts, 227

and glossy surfaces are observed in two-thirds of the diamonds. Less than 5% of diamonds show 228

Edge abrasion is found in 44% of abraded diamonds and is the most common

- fine to medium microdisc textures, but when present occur in swarms. 229
- sedimentologic-induced surface texture (Fig. 3i). Percussive marks along crystal edges, 231
- scratches, and crescentiform fissures are less common (Fig. 3j). Most diamonds have some 232
- degree of surface abrasion from minor scratches and edge abrasion and some have no apparent 233
- surface abrasion. This distinction in surface texture is the basis to subdivide Guyana's diamonds 234
- into abraded (85%) and non-abraded (15%) populations. Abraded diamonds may reflect an older 235
- population with extensive fluvial communition, whereas, the non-abraded diamond population 236
- may reveal a shorter distance and/or time of fluvial transport. Ekereku has the highest relative 237
- proportion of non-abraded diamonds followed by Kamarang and Kurupung, whereas Jawalla, 238
- Konawaruk, Monkey Mt., and Maikwak, have no abraded diamonds. Abraded diamonds average 239
- mass  $(9.3 \pm 21.5 \text{ mg})$  is larger than that of non-abraded diamonds  $(5.5 \pm 4.2 \text{ mg})$ . Non-abraded 240
- diamonds are more dodecahedral in form (45% compared to 25%) (Table A1). Resorbed abraded 241
- diamonds present more flat-bottomed dissolution textures (85.3%) than point-bottom dissolution
- textures (18.7%). Non-abraded diamonds also present greater flat-bottomed dissolution textures 243
- (56.5%) than point-bottom dissolution textures (43.5%), but to a lesser degree (Table A1). 244
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## **Cathodoluminescence and UV luminescence**

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Guyana's diamonds cathodoluminesce green (35%), blue (25%), and turquoise (18%)

- with lesser orange, red, and yellow (Table 3). Abraded and non-abraded diamonds demonstrate 247
- different responses to CL. Guyana's abraded diamonds dominantly show green CL (39%), 248
- 249 turquoise (19%), and blue (19%), with minor orange, red, and yellow (~10%), with 11% non-

250	luminescent (Fig. 4a). In contrast, non-abraded diamonds cathodoluminesce blue (59%), minor
251	green (13%), turquoise (10%), and orange (4%), with 13% showing no CL. No yellow or
252	combination CL was observed. Statistical analyses ( $\chi^2$ tests) demonstrate with >99% certainty
253	that the frequencies of CL response between abraded and non-abraded diamonds are independent
254	(Table 4).
255	Ultraviolet luminescence also yields a clear distinction between abraded and non-abraded
256	diamonds. Abraded diamonds UV luminesce green (84%) with very minor orange (1%), red
257	(2%), and yellow (3%) variants, and 10% with no apparent luminescence. In contrast, non-
258	abraded diamonds show a smaller population of green-luminescent grains (52%), and minor
259	populations of red- (1%) and yellow-luminescent (1%) grains. A large proportion of non-abraded
260	diamonds yielded no luminescence response (45%) (Fig. 4b). We note that similar CL and UV
261	response distributions are observed for all diamonds regardless of geographic location.
262	Nitrogen and Carbon isotope composition
263	The nitrogen concentration of diamonds range between trace ( $< 5 \text{ ppm} \sim \text{Type IIa}$ ) and
264	1971 ppm, with a mean of $315 \pm 24$ ppm and rare outliers from 1175 to 1971 ppm. Diamonds are
265	predominantly Type IaAB, accounting for 53%, with Type 1aA at 22%, and Type IaB at 9%.
266	17% of the diamonds are Type IIa and thus show no nitrogen aggregation (Fig. 5). No Type IIb
267	diamonds are observed.
268	Marked differences in nitrogen concentration exist between abraded and non-abraded
269	diamonds (Table 4). Non-abraded diamonds have higher average nitrogen concentrations
270	compared to abraded diamonds with means of $621 \pm 47$ ppm and $271 \pm 21$ ppm, respectively
271	(Fig. 5; Table 4). The nitrogen concentrations of non-abraded diamonds preserve a more uniform
272	distribution with lower skewness and kurtosis coefficients of 0.26 and 1.91, whereas the abraded

273	diamonds display a pronounced positive skewness (1.82) and a higher kurtosis (6.62) (Table 4;
274	Fig. A1). Multivariate density Finite Mixture Modeling of nitrogen concentrations was applied to
275	determine if subpopulations could be readily identified within the greater abraded and non-
276	abraded diamond populations, based on the Bayesian Information Criteria which demonstrates
277	that a 21 component mixture is the most appropriate choice (Fig. A2) (e.g., Schwartz, 1978;
278	Galbraith and Green 1990; Liang and Forman 2019; McLachlan et al. 2019). The Finite Mixture
279	Modeling identified ten distinct subpopulations within the abraded diamonds and seven
280	subpopulations within the non-abraded diamonds (Table 4). The abraded and non-abraded
281	diamonds may share six subpopulations, with nitrogen concentrations of $604 \pm 48$ , $390 \pm 48$ , $180$
282	$\pm$ 16, 47 $\pm$ 3, 23 $\pm$ 1, and 4 $\pm$ 0.3 ppm. The non-abraded population may have one additional
283	group centered at $968 \pm 79$ ppm, whereas the abraded diamonds may have additional populations
284	at $13 \pm 1$ , $84 \pm 8$ , $1234 \pm 99$ , and $1911 \pm 150$ ppm. Nitrogen concentrations reported here are
285	average and bulk compositions intended to characterize the diamond populations as a whole.
286	Heterogeneity within each individual diamond is expected but not elaborated further.
287	Guyanese diamonds have an average $\delta^{13}$ C value of -5.82 ± 2.45‰ with a range from -
288	16.11 to -2.88‰. These $\delta^{13}$ C values are slightly more depleted than diamonds from Arenápolis
289	and Boa Vista, Brazil, and are significantly more enriched in <sup>13</sup> C than those of Guaniamo,
290	Venezuela (-15 $\pm$ 2.78‰), and Dachine, French Guiana (-24.65 $\pm$ 4.12‰) (Fig. 6). Diamonds
291	from West and Central Africa preserve a mode of -3.5‰, making them more enriched than
292	Guyana's diamonds (Cartigny et al. 2014). Abraded diamonds have an average $\delta^{13}C$ value of -
293	$6.11 \pm 2.71\%$ and non-abraded diamonds have an average $\delta^{13}C$ value of $-5.06 \pm 1.36\%$ . For
294	comparative spectroscopy, luminescence, $\delta^{13}$ C, morphology, and inclusion paragenesis of
295	Guyana's diamonds please see supplementary Tables A1-A4.

#### 296 Inclusions

297	Diamond-hosted inclusions in order of decreasing abundance include forsteritic olivine,
298	enstatitic orthopyroxene, rutile, chromite, garnet, clinopyroxene, and coesite (Table 5).
299	Epigenetic inclusions consist of graphite, which are restricted to internal fractures within the
300	diamond or along relaxed inclusion margins. Sulfide inclusions are rare. Most primary inclusions
301	of olivine and orthopyroxene are cubo-octahedral, whereas clinopyroxene are prismatic to
302	elongate cubo-octahedral. Forsteritic olivine is the most common inclusion, found in 6.3% of all
303	diamonds examined. When found, olivine occurs as single inclusions or in clusters. Olivine
304	inclusions are colorless and range in size from $\sim 5$ to 500 $\mu m.$ Orthopyroxene is found in 2.3% of
305	diamonds. It is colorless and inclusions are $\sim 5$ to 550 $\mu m$ across. Garnet inclusions occur in
306	0.6% of diamonds. Garnets range in size from $\sim 25$ to 200 $\mu m.$ Three garnets are purple and one
307	is red-brown.
308	Accessory mineral geochemistry

Guyana's alluvial diamond deposits are frequently found with non-309 kimberlite water worn quartz, topaz, jasper, rutile, anatase, zircon, ilmenite, gold, corundum, and 310 tourmaline, with minor garnet and chromite. Traditional kimberlite indicator minerals are 311 uncommon but chromite, garnet, and ilmenite occur in our samples (Table A4). Two populations 312 of mantle derived and crustal derived garnets are observed based on discrimination diagrams of 313 Schulze et al. 2003 (Fig. 7). Crustal garnets are Mn-rich spessartines ( $Alm_{27\pm19}Py_{2\pm5}Sp_{70\pm23}$ ). The 314 315 mantle derived garnets (Alm<sub>3±3</sub>Py<sub>12±2</sub>Sp<sub>84±3</sub>) are also low Cr and have moderately low TiO<sub>2</sub> (~ 0.19 wt%). SEM-EDS profiles of both garnet populations show no obvious compositional zoning 316 or evidence of metamorphic overprint. Accessory ilmenite grains are MgO poor  $(0.17 \pm 0.4)$ 317 318 wt%) and TiO<sub>2</sub> rich ulvospinels (72.2  $\pm$  7.6 wt%) (Wu<sub>72 $\pm$ 16</sub>Ru<sub>28 $\pm$ 16</sub>). We analyzed chromite in our

samples and compared them to previous analyses of accessory chromite conducted by Goldenstar resources from diamondiferous alluvial deposits in Amatuk (Shields and Letendre 1999). Most chromites have moderate to low  $TiO_2$  (0.5 ± 0.8 wt%) and  $Al_2O_3$  (14.2 ± 5.6 wt%) demonstrating magmatic arc (ARC) and mid ocean ridge basalt (MORB) affinities.

323 Detrital zircon geochronology

Two representative diamond-bearing heavy mineral separates from the Kurupung region 324 were targeted for reconnaissance detrital zircon geochronology. The first was from colluvium in 325 a raised terrace, approximately 3 m above the modern river (KU002). The second was collected 326 from an alluvial gravel horizon within a buried fluvial channel. Most zircons are well rounded by 327 abrasion. A minor proportion ( $\sim 8\%$ ) of zircons occur as slightly abraded to non-abraded 328 euhedral, doubly terminated prisms. Detrital zircon age distributions of both buried alluvium and 329 terrace colluvium samples are dominated by zircon between 2.0 and 2.2 Ga in age; recording 330 similar peak ages of 2.06 Ga and 2.04 Ga, respectively (Fig. A2). Only 5 grains of earliest 331 Paleoproterozoic or Archean in age (i.e., > 2.3 Ga) were identified (~2% of the total), and these 332 grains are generally late Archean (2.6-2.8 Ga) (Fig. 8). All zircon U-Pb analyses are older than 333 the depositional age range of the  $1.78 \pm 0.03$  Ga  $- 1.98 \pm 0.08$  Ga Roraima Supergroup (Santos 334 335 et al. 2003).

336

#### Discussion

Guyana's alluvial diamond deposits represent an economic resource that also preserve valuable insights into the evolution of surface and subsurface processes of the Guiana Shield. Hypotheses regarding the mantle paragenesis and surface provenance of Guyana's diamonds exist, but each are based largely on speculation. Our morphology, composition, and inclusion

datasets provide empirical based reasoning to characterize the mantle conditions of diamond
 formation and identify the possible provenance of placer alluvial deposits.

The Guiana Shield has existed as a thick craton with lithospheric roots extending >130 343 km depth since at least 1.9 Ga (Gibbs and Barron 1993; Schulze et al. 2006). Such depths are 344 enough to intersect the diamond stability field beneath the craton (Stachel and Harris 2009), 345 depending on the thermal regime during that time. However, kimberlite deposits in Guyana have 346 not been discovered, unlike elsewhere in the Guiana Shield (Kaminsky et al. 2000; Channer et al. 347 2001; Kaminsky et al. 2004; Svisero et al. 2017, Cabral et al. 2017). Instead, diamonds are 348 recovered from Quaternary to modern alluvial deposits. Tectonic and sedimentologic processes 349 may have mixed diamonds from different temporal and geographic sources into the same alluvial 350 basins. Thus, Guyana's diamonds reflect complex surface and mantle processes across the 351 Guiana Shield, since the Paleoproterozoic. 352

#### 353 **Paragenesis**

Diamonds preserve both compositional and physical evidence of their parental mantle 354 source. During crystal growth, diamonds may entrap particular suites of minerals as inclusions. 355 Guyanese diamonds primarily contain inclusions of forsteritic olivine, enstatitic orthopyroxene, 356 and chromite. This assemblage indicates most of the diamonds crystallized in, and were 357 extracted from, the harzburgite peridotite dominated upper mantle. Less common diamonds 358 preserve an eclogitic assemblage containing rutile, almandine garnet, clinopyroxene, and coesite 359 360 inclusions. Cratonic lithospheric mantle assemblages are typically stable between 1 and 7 GPa and less than 1300°C (Mather et al. 2011). Both peridotite and eclogite assemblages spatially 361 coexist beneath stable cratons, consistent with model geothermal gradients of 38-42 mWm<sup>-2</sup> 362 (Pollack and Chapman 1977; Stachel and Harris 2009). 363

The carbon isotopic composition of diamonds can be used to further resolve mantle 364 paragenesis. Guyana's diamonds yield an average  $\delta^{13}$ C value of -5.82 ± 2.45‰ (Fig. 6), falling 365 within the range of -8 to -2‰ found in most P-type diamonds sampled from the peridotitic upper 366 mantle (Cartigny 2005). Few diamonds have values less than -10‰. These light  $\delta^{13}$ C values are 367 E-type, having crystallized from eclogite and contain a possible organic carbon source derived 368 from metasedimentary rocks (Cartigny et al. 2014). Very light  $\delta^{13}$ C values (< -16‰) might be 369 indicative of websteritic paragenesis (Deines and Harris 2004) but are rare in Guyana's 370 diamonds. Together, Guyana's diamond inclusion suites and carbon isotopic compositions 371 confirm that the diamonds are largely sourced from the lithospheric mantle and derived from 372 predominantly peridotite and eclogite rocks. 373

Accessory mineral compositions can also be used to infer the paragenesis of Guyana's 374 diamond deposits. Accessory garnet is spessartine (Alm<sub>27±19</sub>Py<sub>2±5</sub>Sp<sub>70±23</sub>) in composition and 375 most are likely derived from granite and granodiorite rocks, which are common throughout the 376 Guiana Shield. A subpopulation of the spessartine preserves Mg/Ca ratios indicative of a mantle 377 derived affinity (Fig. 7; Mg#/Ca# ~ 3.80). These mantle spessartines (Alm<sub>3±3</sub>Py<sub>12±2</sub>Sp<sub>84±3</sub>) have 378 low Cr and low TiO<sub>2</sub> concentrations and are likely derived from eclogitic source rocks (Schulze 379 et al. 2003). Although sourced from the mantle, these garnets are not likely of kimberlitic origin. 380 Instead, these low-Cr and low-TiO<sub>2</sub> spessartine were more likely derived from subducted oceanic 381 crust rich in Mn. In Guyana there are early Proterozoic manganiferous metasedimentary rocks 382 383 (Gibbs and Barron 1993); the largest of which are at Mathews Ridge which outcrops north of the Roraima Supergroup (Westerman 1969). These could be remnants of subducted oceanic crust 384 from which the mantle affiliated garnets are derived. Accessory ulvospinels ( $Wu_{72\pm 16}Ru_{28\pm 16}$ ) are 385 386 Mg-poor and are similarly derived from non-kimberlite mafic intrusions. Most chromites have

moderate to low  $TiO_2$  and  $Al_2O_3$  concentrations indicative of ARC and MORB tectonic

environments which is typical of the Guiana shield; consisting of volcanic arc segments accreted

over an Archean core (Tassinari 1997; Cordani & Teixeira 2007; Daoust et al. 2011; Reis et al.

- 2000; Fraga et al. 2009; Kroonenberg et al. 2016).
- **391 Paleo-thermal conditions of the mantle**

The concentration and aggregation state of lattice bound nitrogen in diamond provides 392 additional constraints on the crystallization temperature and mantle residence conditions of 393 Guyanese diamonds. Nitrogen is the most abundant impurity in diamond, with concentrations 394 sometimes reaching >0.3 wt% (Woods et al. 1990; Cartigny et al. 2001; Stachel and Harris 2009; 395 Smart et al. 2011). Quantitative and FTIR derived nitrogen concentrations range from trace to 396 1971 ppm, with population means from different localities ranging between 120 and 620 ppm 397 (Fig. 5a). Guyanese diamonds are predominantly Type I, including Type IaAB, Type IaA, and 398 Type IaB. When compared to other South American diamonds (Kamarang, Kurupung, 399 Konawaruk and Maikwak), nitrogen concentrations are within similar ranges to diamonds from 400 Canastra and Arenápolis, Brazil (Table 6). The aggregation state of nitrogen within diamond is 401 largely a function of residence temperature (Chrenko et al. 1977; Evans and Qi 1982). Kinetic 402 modelling of the change in nitrogen aggregation state can be used to quantify the thermal 403 evolution of the continental lithosphere and paleotectonic processes (Taylor et al. 1990). 404 Individual total N ppm vs. %N<sub>B</sub> (degree of nitrogen aggregation) ratios were used to calculate 405 mean residence temperatures of  $1124 \pm 100^{\circ}$ C (Fig. 5) using the kinetics of the nitrogen A-B 406 center aggregation reaction (e.g., Taylor et al. 1990; Taylor et al. 1996). Residence temperature 407 distribution is slightly negatively skewed and slightly leptokurtic (Table 7). We considered a 408 409 minimum age of eruption of 2.0 Ga and an assumed residence time of 1.3 Ga, if Guyana's

422	Magma composition
421	evidence for crystallization in the lithospheric mantle (Stachel and Harris 2009).
420	38 - 42 mW/m <sup>2</sup> gives minimum crystallization depths between 165 and 185 km, which is
419	emplacement ages. Projecting the average residence temperature along geothermal gradients of
418	similar thermal conditions beneath the Guiana Shield, despite a wide range in possible
417	3%. These formation temperatures indicate that both diamond populations can be derived from
416	Paleoproterozoic of 1.3 Ga and Neoproterozoic of 2.6 Ga vary residence temperature by less than
415	Guaniamo kimberlites ~ 0.7 Ga). Assumptions regarding residence times ranging from the
414	residence time of 2.6 Ga (maximum age of peridotitic diamonds 3.3 Ga – maximum age of
413	of the Guiana Shield have an eruption age of $\sim 0.7$ Ga (Channer et al. 2001) and an assumed
412	that Guyana's diamonds might be derived from more recent kimberlites, of which the youngest
411	3.3 Ga – maximum age of Roraima Supergroup ~ 2.0 Ga). We also considered the possibility
410	diamonds are derived from the Roraima Supergroup (maximum age of peridotitic diamonds $\sim$

Diamond habit, size, and form reflect crystal development during growth in the mantle, 423 whereas dissolution and abrasion textures reveal resorption during ascent and transport, 424 respectively. Guyana's diamonds are predominantly octahedral (Fig. 3a), indicating slow growth 425 in near-equilibrium conditions found beneath a thick and stable subcratonic lithosphere in the 426 diamond stability field (Harrison and Tolansky 1964; Seal 1965; Sunagawa 1984; Stachel and 427 Harris 2009). Dodecahedrons (Fig. 3b) represent the second most common crystal habit for 428 429 diamonds across Guyana; this form is particularly observed at Ekereku. Dodecahedrons may form in the mantle or during magmatic ascent via dissolution of octahedron apices and edges in 430 response to disequilibria produced by high temperature oxidation in the presence of CO<sub>2</sub> and 431 432 H<sub>2</sub>O fluid phases (Fedortchouk et al. 2007). Diamond populations from other regions in Guyana

do not preserve a distribution so enriched in dodecahedral form, providing evidence for different
 conditions of mantle departure or host magma volatile contents.

Additional aspects of the diamond-entraining magmatic fluid can be inferred from 435 dissolution features on crystal faces. Chemical oxidation caused by the interaction of fluid 436 volatile phases in a kimberlite or related rock, exploits surface defects on a diamond, causing it 437 to dissolve. Dissolution produces pits and hillocks, whose geometries are controlled by internal 438 dislocations, crystal defects, and intensive variables including pressure, temperature, and  $fO_2$ 439 (Fedortchouk 2015). Dissolution features (Fig. 3f) occur on 95% of the diamonds. Teardrop 440 hillocks are the most common resorption texture and indicate moderate resorption (Tappert and 441 Tappert 2011). Many different pit dissolution textures are recognized but are indistinguishable 442 amongst different geographic locations. Flat and point bottomed trigons, tetragons, and hexagon 443 pits are observed. Coarse flat bottom trigons are evidence for pre-eruptive mantle/melt 444 dissolution at temperatures ranging from 1250 to 1300°C (Fedortchouk 2015). Average diamond 445 residence temperatures of ~1120°C and mantle/melt dissolution temperatures of 1250 to 1300°C 446 suggest diamonds encounter variable mantle thermal conditions during residence, such as higher 447 temperatures of kimberlitic melt during ascent. The greater abundance of flat bottom trigons 448 relative to point bottom trigons in abraded diamond populations indicates that the erupting host 449 magma was enriched in H<sub>2</sub>O relative to CO<sub>2</sub> (Fedortchouk 2015). Experimental data confirm that 450 dissolution pit textures begin as point bottom features. In a fluid enriched in H<sub>2</sub>O relative to 451 452 CO<sub>2</sub>, dissolution of the diamond proceeds more rapidly in the lateral direction rather than the vertical or {111} direction, creating coarse and flat-bottomed pits (Fedortchouk 2015). Late 453 stage etching microdisk textures (Fig. 3g) found on ~5% of the diamonds also provide evidence 454 for an H<sub>2</sub>O-rich magma. These features form when the surface of the diamond is etched by a 455

H<sub>2</sub>O fluid phase in the ascending host magma (Fedortchouk et al. 2007). Kimberlite magmas are 456 inferred to be often CO<sub>2</sub>-rich and have a high CO<sub>2</sub>/H<sub>2</sub>O ratio (Sparks 2013). The dissolution 457 textures observed in Guyana's diamonds instead suggest that both abraded and non-abraded 458 diamond populations were hosted by a magma with more  $H_2O_2$  and therefore a lower  $CO_2/H_2O_2$ 459 ratio. This characteristic is more commonly inferred for lamproitic melts (Bergman 1987; 460 Mitchell and Bergman 1991) although H<sub>2</sub>O-rich kimberlite melts are possible (e.g., Ekati mine 461 kimberlites, see Fedortchouk et al. 2010). Because the non-abraded diamonds present a greater 462 proportion of point bottom dissolution textures than abraded diamonds, these diamonds may 463 have been entrained in a different host melt with a different ratio of  $H_2O/CO_2$ . 464

#### 465 Abraded versus non-abraded diamonds

Diamond is strongly resistant in the natural environment to abrasion. It is brittle, which 466 permits only microcleavage abrasion in the faceting process, and it is extremely hard (Wilks and 467 Wilks 1972; Oganov et al. 2013). Accordingly, percussive marks, scratches, and edge abrasion 468 textures require repeated cycles of erosion, transport and deposition and/or a lengthy residence 469 time within surface environments. Using abrasion textures, we recognized two populations of 470 alluvial diamonds. Pristine, non-abraded diamonds account for 15% of all diamonds. Ekereku 471 has the highest relative proportion of non-abraded diamonds. Jawalla, Konawaruk, Maikwak and 472 Monkey Mt. have no abraded diamonds, but we acknowledge that the sample sizes from those 473 regions are small. 474

The non-abraded stones are found in the same deposits as the abraded diamonds. Nitrogen concentrations indicate that non-abraded and abraded groups may have statistically distinct subpopulations, six of which are common to both groups. These shared subpopulations suggest that some diamonds are sourced from similar mantle residence and thermal conditions.

The distinct subpopulations identified in only the abraded or non-abraded groups may reflect 479 isolated mantle and emplacement conditions. Statistically distinct spectroscopic responses 480 between abraded and non-abraded populations also present evidence for different provenance 481 and source (Table 4). Ultraviolet luminescence responses in abraded diamonds are 482 overwhelmingly green and non-responsive, whereas non-abraded diamonds are split between 483 non-responsive or green (Fig. 4). Luminescence is caused by the presence of impurities, such as 484 nitrogen, and related defects which create vacancy centers in diamonds. Combined substitutional 485 nitrogen and vacancy centers (H3 and H4 defects) within the diamond produced by irradiation 486 followed by high temperature annealing are commonly ascribed to green luminescence (Shigley 487 and Breeding 2013). This suggests that Guyana's abraded diamond population, which have a 488 greater proportion of luminescent green stones were subjected to some form of metamorphic 489 overprint. Abraded diamonds cathodoluminesce predominantly green, with moderate to minor 490 blue and other colors. The majority of non-abraded diamonds cathodoluminesce blue. 491 Worldwide, most diamonds from unmetamorphosed deposits cathodoluminesce blue with zero 492 phonon line emissions at 415-440 nm and 480-490nm (Bulanova et al. 1995; Lindblom et al. 493 2005). This is attributed to the N3 defect where three nitrogen atoms surround a vacancy. With 494 increasing grades of metamorphism and mild annealing at higher temperatures, nitrogen related 495 defect centers become increasingly more complex and blue CL shifts to other colors, with 496 emissions between 490-670 nm (Bruce et al. 2011; Kopylova et al. 2011). In diamond, vacancies 497 and interstitials become more mobile at 500-800°C and ~300°C respectively (Clark et al. 1992; 498 Collins et al. 2005; Collins and Kiflawi 2009). Enhanced diffusion and entrapment of vacancies 499 and interstitials at higher temperatures at the site of various nitrogen forms to produce new 500 501 optical centers is likely how blue CL shifts to other colors (Iakoubovskii and Adrianssens 1999;

Collins and Ly 2002). In the crust, these temperatures are typically inferred for zeolite to 502 greenschist facies metamorphic environments. The abraded population likely experienced some 503 metamorphic overprint. Comparatively, non-abraded diamonds may have been less 504 metamorphosed. This can be also influenced by pre-existing nitrogen and vacancy related defects 505 (N3, NV<sup>o</sup>, and NVN) in Type Ia diamonds and manifest as photoluminescence emissions more 506 commonly at 575 nm and less so at 430-450 nm but correlates rarely with cathodoluminescence 507 (Bruce et al. 2011). Alternatively, irradiation damage during long placer residence would create 508 charge vacancies and interstitials in the diamond lattice, which would also shift CL emittance 509 from blue to other colors, but this damage is usually more common within the first 25 microns of 510 the diamond surface (Vance et al. 1973; Breeding et al. 2018). Abraded diamonds are also larger 511 in mass  $(9.3 \pm 2.2 \text{ mg})$  than non-abraded diamonds  $(5.5 \pm 4.2 \text{ mg})$ , present fewer dodecahedral 512 forms, and a greater ratio of flat-bottom to point-bottom dissolution textures (5:1 compared to 513 1:1). This is evidence that abraded and non-abraded diamond populations may have been 514 entrained in different host melts. At present, both abraded and non-abraded populations are 515 mixed into the same alluvial deposits, reflecting a complex interplay of mantle storage, eruptive 516 history, and fluvial transport throughout the > 2.5 Ga evolution of the Guiana Shield. 517

### 518 **Provenance**

Abrasion, nitrogen concentration, CL, and UV observations indicate that alluvial deposits from each productive region across Guyana host both abraded, metamorphosed, nitrogen-poor diamonds as well as non-abraded, less metamorphosed, nitrogen-rich diamonds. Guyana's abraded diamonds are most likely derived from recycled paleo-placers with a metamorphic overprint. The probable source is the greenschist facies pebbly conglomerates or volcaniclastics of the 1.98 to 1.78 Ga Roraima Supergroup. Diamondiferous alluvial deposits occur in close

proximity to the margin of the Roraima Supergroup. In addition, diamond-bearing alluvial 525 deposits contain abundant guartzite and jasper clasts derived from the nearby Roraima 526 Supergroup. The Roraima Supergroup has been exposed at the surface since at least the early 527 Mesozoic (McConnell 1968). Increased weathering and erosion associated with rifting and 528 opening of the Atlantic since the Jurassic produced initial alluvial deposits derived from the 529 Roraima Supergroup, which have been continually exhumed and redeposited since that time by 530 repeated cycles of river capture, rejuvenation, degradation and aggradation (McConnell 1968, 531 Gibbs and Barron 1993). The majority of diamond populations in Guyana were likely abraded 532 within this complex polycyclic alluvial environment. Non-abraded diamonds were also likely 533 sourced from the Roraima Supergroup basal conglomerates and volcaniclastic horizons but were 534 trapped in sediment packages that experienced less energetic deposition, reworking and 535 experienced less metamorphic overprint and longer primary storage. 536

The Roraima Supergroup source is supported by comparison to alluvial diamond deposits 537 in Mutum, Brazil. Mutum diamonds are mined from alluvial terraces shed from conglomerates of 538 the Tepequém Formation in Brazil (Santos et al. 2003; Reis et al. 2017). The diamonds are 539 similar to Guyana's diamonds in size, skin color,  $\delta^{13}$ C, and nitrogen concentrations. These 540 diamonds, however, also present more flattened forms (60%), grey diamonds (40%), and fewer 541 proportions of diamonds with trigons (Araújo et al. 2011). This suggests that these diamonds 542 may be derived from a different magma source than Guyana's diamonds. The Arai Formation of 543 the Roraima Supergroup is the local age and lithologic equivalent of the Tepequém Formation. 544 Accordingly, conglomerates and volcaniclastics of the Arai Formation may be the detrital source 545 of Guyana's diamonds, representing a regional Paleoproterozoic high energy, siliciclastic 546

depositional basin that was a sink for diamonds emplaced by several episodes of kimberlite 547 volcanism within the Guiana Shield, during the middle Paleoproterozoic or older (>2.0 Ga). 548 Other hypotheses regarding the source of the diamonds have been put forward over the 549 past decades, including sources in West Africa, Boa Vista (Brazil), Dachine (French Guiana), 550 and Guaniamo (Venezuela). West Africa is home to kimberlite and alluvial diamond deposits. 551 Most diamonds from West Africa are alluvial in origin, but diamonds have also been mined from 552 kimberlites (Deines and Harris 1995; Skinner et al 2004). The alluvial diamonds are largely 553 weathered from Mesozoic kimberlite pipes and dykes. Colorless to brown body color and green 554 skins are common; similar to those in Guyana (Sutherland 1982; Janse 1996; Stachel et al. 2002). 555 Size and quality are inversely related to distance from the kimberlite source (Knopf 1970; 556 Norman et al. 1996; Grantham and Allen 1960; Janse 1996). The smallest carat sizes and best 557 quality gems are found in beach placers along the West African coastline. The oldest Mesozoic 558 sources may have produced microdiamonds that could have reached Guyana's eastern coast prior 559 to rifting associated with the Atlantic Ocean, but diamonds are not found on the coast. The more 560 recent Mesozoic kimberlites are too young and shed detrital diamonds into the juvenile Atlantic 561 Ocean instead. We conclude that Guyana's diamonds are unlikely to be related to sources in 562 West Africa. 563

The closest primary diamond-bearing igneous sources in the modern tectonic setting are the Guaniamo kimberlite dykes in Venezuela and metamorphosed ultramafic and pyroclastic shoshonites or lamprophyres in Dachine, French Guiana (Capdevila et al. 1999; Channer et al. 2001; Kaminsky et al. 2000; Kaminsky et al. 2004; Smith et al. 2016). Although some similarities exist, including upper mantle temperatures, sizes, and colors between Guyana's and Guaniamo's diamonds, we contend that those sources are unlikely to be genetically associated

with Guyanese diamonds based on geographic distance, distinct <sup>13</sup>C signatures, nitrogen
aggregation, predominantly eclogitic inclusions, and CL responses (Channer et al. 2001;
Kaminsky et al. 2000; Kaminsky et al. 2004). Furthermore, Dachine diamonds are smaller (< 1</li>
mm), have a lower total nitrogen range from trace to 110 ppm, are strictly Ib to IaA, and have
mostly sulfide inclusions (Smith et al. 2016).

Detrital zircon geochronology provides additional compelling evidence supporting 575 provenance. Detrital zircon U-Pb ages from both colluvial and alluvial sediment demonstrate that 576 these diamondiferous deposits are likely derived from rocks of late Trans-Amazonian age, with 577 lesser contributions of Archean grains (Fig. 8). These deposits are inferred to be Pleistocene to 578 recent in age (McConnell 1968; Gibbs and Barron 1993). The similar ages between the diamond-579 bearing colluvium and alluvium is consistent with the interpretation that both types of placer 580 diamond deposit are derived from similarly aged bedrock sources of the Barama-Mazaruni group 581 such as Surumu group volcaniclastics (Basei 1978; Schobbenhaus et al. 1994; Reis et al. 2017), 582 or alternatively, as inherited minerals from the once extensive Roraima Supergroup in a foreland 583 basin fill (Santos et al. 2003). The dominantly unimodal characteristic of these detrital zircon age 584 spectra is consistent with derivation from a relatively small geographic region, as a more multi-585 modal age distribution would be expected if the diamond-bearing sediment was sourced from a 586 large fluvial catchment. Furthermore, the narrow range of ages suggesting a local geographic 587 origin, preponderance of coarse quartz and jasper lithic fragments, and presence of non-588 589 kimberlitic or related rock type indicator minerals, lends evidence to the Roraima Supergroup being the provenance area for the Guyana detrital zircons and diamonds. 590

591 Implications

In an environment prone to extreme tropical weathering, Guyana's diamonds are likely 592 the only remaining mantle xenocrysts which can offer insight into volcanic and tectonic controls 593 that existed during the early evolution of the Guiana Shield. Detrital zircon U-Pb ages suggest 594 that the kimberlite or related rock sources that erupted Guyana's diamonds represent episodes of 595 kimberlite volcanism during at least the middle Paleoproterozoic or during the Archean. Also, 596 the surfaces of diamonds demonstrate they were emplaced by various mantle-derived melts 597 enriched or balanced in H<sub>2</sub>O relative to CO<sub>2</sub>. Guyana's diamonds are likely xenocrysts from 598 some of the Earth's oldest kimberlites or lamproites. These source rocks have long since eroded 599 or are still undiscovered. 600

Guyana's diamonds are divided into two populations of recycled and abraded diamonds 601 and primary and non-abraded diamonds. Both abraded and non-abraded diamonds, can be 602 distinguished according to unique spectroscopic, luminescence and morphological 603 characteristics. This classification can aid in characterizing the prospectivity of underexplored 604 diamond fields, especially when other mantle xenocrysts are absent. The abraded diamonds are 605 likely recycled detrital grains eroding from rocks of the Roraima Supergroup, which could 606 therefore represent a regional placer diamond source terrane with a lateral extent of at least 607 450,000 km<sup>2</sup>. The provenance of non-abraded diamonds still remains enigmatic. It may be that 608 there are undiscovered primary kimberlites but it is more likely that these non-abraded diamonds 609 may also be sourced from the Roraima Supergroup basal conglomerates and volcaniclastic 610 horizons which have experienced less metamorphic overprint. Both diamond populations, 611 inclusion suites,  $\delta^{13}$ C, and nitrogen concentrations suggest diamond formation at residence 612 temperatures of approximately 1120°C in the peridotitic and eclogitic lithospheric mantle of the 613 Guiana Shield. 614

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912	Figure captions
913	Figure 1. Geological Map of the Guiana Shield and inferred adjacent West African craton during
914	the Jurassic, showing primary and placer diamond deposits. (AM – Amatuk, AR –
915	Arenápolis, BA – Baoule, BV – Boa Vista, CA – Canastra, CO – Los Coquitos, CR –
916	Coromandel, DC – Dachine, EK – Ekereku, GU – Guaniamo, JW – Jawalla, KM –
917	Kamarang, KU – Kurupung, KW – Konawaruk, MG – Mano Gadua, MK –
918	Maikwak, MM – Monkey Mt., NA – Nassau, RO – Rosebel, SE – Seguela, TE –
919	Tepequém, TO – Tongo, TY – Tortiya) (Modified from Santos et al. 2003, Nadeau
920	and Heesterman 2010, Daoust et al. 2011, Kroonenberg et al. 2016, and Bassoo and
921	Murphy 2018).
922	Figure 2. Body and skin color. a) colorless, b) white, c) yellow, d) brown, e) brown skins f)
923	green skins
924	Figure 3. Morphological features. a) octahedral, b) dodecahedral, c) flattened/macle, d) trigons,
925	e) hexagons, f) hillocks, g) microdiscs, h) fully abraded, i) edge abrasion, j) percussive
926	marks

927 Figure 4. Cathodoluminescence and UV luminescence optical responses.

928	Figure 5. Nitrogen aggregation of Guyana's diamonds. a) Total N (ppm) vs $N_B$ (%) plot of
929	abraded and non-abraded diamonds (Total N $\pm$ 24 ppm, %N <sub>B</sub> $\pm$ 1.7%), b) nitrogen
930	aggregation type classification, c) representative FTIR spectrum showing carbon and
931	nitrogen in diamond regions.
932	Figure 6. $\delta^{13}$ C values (in % <sub>VPDB-LSVEC</sub> ) of Guyana's diamonds compared to other South
933	American diamonds. ( <sup>1</sup> Tappert et al. 2006; <sup>2</sup> Kaminsky et al 2000; <sup>3</sup> Smith et al. 2016)
934	Figure 7. Ca# vs Mg# plot of mantle and crustal derived accessory garnets (Schulze et al. 2003).
935	Figure 8. Comparative detrital zircon U-Pb age distributions from alluvial and colluvial samples
936	at Kurupung.
937	Supplementary figure captions
938	Figure A1. Kernel density plot of N (ppm) between a) abraded and b) non-abraded diamonds,
939	and c) Bayesian Information Criteria vs component plot showing a 21 component mixture
940	as the most appropriate number of nitrogen populations of the FMM.
941	Figure A2. Detrital zircon U-Pb age distributions plotted as a) cumulative and b) relative
942	probability density plots (Sharman et al. 2018). Vertical grey bar indicates the
943	depositional age range of the Roraima Supergroup. Peak ages are labeled.
944	Table captions
945	Table 1. Summary morphology.
946	Table 2. Pit dissolution textures of Guyana's diamonds.
947	Table 3. Optical spectroscopy.
948	Table 4. Spectroscopic comparisons between abraded and non-abraded diamonds.
949	Table 5. Guyana diamond inclusion types.

- Table 6. Nitrogen concentrations of Guiana Shield diamonds.
- Table 7. Formation temperatures of Guiana Shield diamonds.

952	Supplementary table captions
953	Table A1. Abraded vs non-abraded diamond comparisons.
954	Table A2. Comparative FTIR and $\delta 13C$ of Guyana's diamonds.
955	Table A3. Comparative FTIR derived N concentrations, residence temperatures, CL, and UV
956	responses for Guyana's diamonds.
957	Table A4. Accessory mineral compositions (wt%).
958	Table A5. U-Th-Pb ages of detrital zircons from diamondiferous alluvium and colluvium
959	analyzed at the University of Arkansas.

960

End



60°W

50°W

#### Figure 1 - Legend





Figure 3





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#### Figure 5



# Figure 6



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#### Figure 8



Table 1.	Summary	morphol	ogy
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		Colo	or						Habit						Spott	ing			Late Stag	e Etching	Sedimentilo	gical abrasion
Location	п	В	Br	Y	Gr	Р	W	Х	0	D	Co	С	Fl	Unk	Gr	Br	Со	Х	Etching	No Etching	Abraded	Non Abraded
Ekereku	226	0	9	6	0	0	0	211	69	103	29	9	11	5	97	9	18	205	90	208	159	53
Jawalla	19	0	1	2	0	0	0	16	6	7	4	1	1	0	8	0	0	11	6	13	19	0
Kamarang	154	0	6	2	0	0	1	145	44	16	35	0	40	19	95	4	9	46	53	101	140	13
Konawaruk	6	0	0	0	0	0	0	6	3	1	0	0	2	0	3	0	1	2	0	5	6	0
Kurupung	81	0	1	4	0	0	1	75	25	17	11	3	18	8	30	2	6	90	28	55	78	5
Maikwak	3	0	0	0	0	0	1	2	1	2	0	0	0	0	3	0	0	0	0	3	3	0
Monkey Mt.	3	0	0	0	0	0	0	3	2	1	0	0	0	0	2	0	0	1	0	3	3	0
%		0.0	3.5	2.8	0.0	0.0	0.6	92.7	30.4	29.8	16.0	2.6	14.6	6.5	37.1	2.3	5.3	55.3	31.3	68.7	85.2	14.8

B - blue, Br - brown, Y - yellow, P - pink, W - White , X - no colour/spotting O - octahedral, D - dodecahedral, Co - combination, C - cubic, Fl - flattened/elongate, Unk - unknown

Tuble 2. The dissolution textures of Guyunu's diamonds										
Туре	Trigons	Tetragons	Hexagons	Trapezoids						
Point Bottom (%)										
very fine (< 25 μm)	7.7	1.3	0.0	0.0						
fine (25 - 50 µm)	2.6	3.8	0.0	0.0						
medium (50 - 75 μm)	1.3	5.1	1.3	0.0						
coarse (75 - 100 µm)	0.0	6.4	0.0	0.0						
very coarse (> 100 µm)	0.0	1.3	0.0	0.0						
Total %	11.5	17.9	1.3	0.0						
Flat Bottom (%)										
very fine (< 25 μm)	5.1	0.0	5.1	0.0						
fine (25 - 50 µm)	14.1	0.0	9.0	5.1						
medium (50 - 75 μm)	5.1	0.0	1.3	1.3						
coarse (75 - 100 µm)	11.5	3.8	3.8	0.0						
very coarse (> 100 µm)	1.3	0.0	1.3	1.3						
Total %	37.2	3.8	20.5	7.7						

Table 2. Pit dissolution textures of Guyana's diamonds

error  $\pm 5~\mu m,$  # of diamonds - 370, # of diamonds with dissolution pits - 78

Table 3.	Optical	spectroscopy

	Cathe	Cathodoluminescence				UV flourescence				п					
Location	В	Gr	Т	0	R	Y	Х	Ζ	Gr	0	R	Y	Х	Ζ	
Ekereku	80	53	32	9	3	1	28	2	154	1	3	1	50	0	209
Jawalla	3	4	7	4	0	0	0	1	18	1	0	0	0	0	19
Kamarang	20	66	26	12	5	1	19	3	127	0	7	5	13	1	152
Konawaruk	1	3	1	0	0	0	1	0	6	0	0	0	0	0	6
Kurupung	10	36	16	4	6	1	3	4	65	2	3	3	7	0	80
Maikwak	1	2	0	0	0	0	0	0	3	0	0	0	0	0	3
Monkey Mt.	2	0	1	0	0	0	0	0	3	0	0	0	0	0	3
%	24.8	34.8	17.6	6.2	3.0	0.6	10.8	2.1	79.5	0.8	2.7	1.9	14.8	0.2	
	•								-						

Cathodoluminescence, n - 471, UV flourescence, n - 473

B - blue, Gr - green, T - turquoise, Y - yellow, O - orange, R - red , X - no response, Z - combination

Table 4.	Spectroscopic	comparisons	between	abraded a	and non-	-abraded	diamonds
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Spectroscopy	Abraded		Non-abraded	
FTIR <sup>1</sup>	N (ppm)	n	N (ppm)	n
mean	$271 \pm 21$	349	$621\pm47$	44
median	109		590	
σ	353		473	
skewness	1.82		0.26	
kurtosis	6.62		1.91	
FMM populations				
Pop. 1	$1911 \pm 150$	2	—	—
Pop. 2	$1234\pm99$	16	—	—
Pop. 3	—	_	$968\pm79$	18
Pop. 4	$610\pm52$	54	$604\pm48$	7
Pop. 5	$351\pm29$	60	$390\pm26$	4
Pop. 6	$193\pm16$	30	$180 \pm 16$	6
Pop. 7	$84\pm8$	34	_	_
Pop. 8	$39 \pm 3$	46	$47 \pm 3$	1
Pop. 9	$21 \pm 2$	26	$23 \pm 1$	4
Pop. 10	$13 \pm 1$	16	_	_
Pop. 11	$6\pm0.4$	5	$4\pm0.3$	2
Cathodoluminescence <sup>2</sup>	%	n	%	n
Blue	19.0	76	58.6	41
Green	38.7	155	12.9	9
Turquoise	19.0	76	10.0	7
Orange	7.0	28	1.4	1
Red	2.7	11	4.3	3
Yellow	0.7	3	—	—
No Response	10.5	42	12.9	9
Combination	2.5	10	—	—
UV luminescence	%	п	%	n
Blue	_	_	_	_
Green	84.3	339	52.1	37
Turquoise	_	_	_	_
Orange	1.0	4	—	—
Red	3.0	12	1.4	1
Yellow	2.0	8	1.4	1
No Response	9.5	38	45.1	32
Combination	0.2	1	_	_

 $^{1}$  Abraded vs Non-abraded log N (ppm), |t|-test statistic: 12.7 > |t|-threshold value: 1.3, with 95% compatibility interval and p < .00001

 $^{2}$  Abraded vs Non-abraded CL (%),  $\chi$ 2- test statistic: 287 >  $\chi$ 2-threshold value: 14.1, with 7 degrees of freedom, a 90% compatibility interval, and p < .00001

Table 5. Guyana diamond inclusion types	5
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2	21	
Inclusion	# of diamonds	Paragenesis
Chromite	11	Peridotitic
Enstatite <sup>1</sup>	15	Peridotitic
Forsterite <sup>2</sup>	42	Peridotitic
Cr-pyrope garnet	3	Peridotitic
Almandine garnet	1	Eclogitic
Coesite	1	Eclogitic
Clinopyroxene	1	Eclogitic
Rutile	15	Eclogitic

<sup>1</sup>Raman spectra doublet peaks indicate a more Mg rich and enstatite composition. <sup>2</sup>Raman spectra doublet peak and lower intensity of v3 mode peak suggest a more forsteritic composition (Mg#  $\sim$  90)

Location	Total N $\pm \sigma$ (ppm)	$N_{\rm B}$	n	
Guyana				
Ekereku	$407\pm472$	$38\pm34$	134	
Jawalla	$123\pm201$	$27\pm34$	19	
Kamarang	$275\pm310$	$38\pm 34$	150	
Kurupung	$242\pm352$	$36\pm41$	79	
Konawaruk	$249\pm265$	$18\pm19$	6	
Maikwak	$250\pm78$	$34\pm5$	3	
Monkey Mt.	$610\pm138$	$65 \pm 16$	3	
Venezuela				
Los Coquitos <sup>1</sup>	$641\pm324$	$62\pm19$	77	
Guaniamo <sup>2</sup>	620	69	192	
French Guiana				
Dachine <sup>3</sup>	$20 \pm 29$	NA	18	
Brazil				
Boa Vista <sup>4</sup>	$345\pm412$	$39\pm 30$	34	

Table 6. Nitrogen concentrations of Guiana Shield diamonds

<sup>1</sup> Kaminsky et al 2006, <sup>2</sup> Kaminsky et al 2000, <sup>3</sup> Smith et al 2016, <sup>4</sup>Kaminsky et al 2001, <sup>4</sup>Tappert et al 2006

Table 7. Formation temperatures of Gulana Shield diamonds							
Location T (°C)		n	Derivation				
Guyana							
Abraded	$1128\pm104$	208	N aggregation (1.3Ga residence)				
Non-abraded	$1101\pm72$	34	N aggregation (2.6Ga residence)				
mean	$1124\pm100$	242					
median	1148	242					
skewness	-1.49	242					
kurtosis	1.72	242					
Brazil <sup>1</sup>							
Boa Vista	$1208\pm74$	27	Chromite-Olivine inclusion equilibration				
Venezuela <sup>2</sup>							
Guaniamo	$1116 \pm 99$	6	Garnet-Diopside inclusion equilibration				

Table 7. Formation temperatures of Guiana Shield diamonds

<sup>1</sup> Tappert et al 2006, <sup>2</sup> Kaminsky et al 2000