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1	Further detrital zircon evidence for peri-Gondwanan blocks in the central Appalachian
2	Piedmont Province, USA
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13 0. ABSTRACT

14 Evidence for exotic terranes in the central Appalachian Piedmont Province is 15 fragmented between central Virginia, northern Maryland, and southeastern Pennsylvania. 16 Here we present LA-ICPMS data from detrital zircon that supports the presence of an 17 exotic terrane in this region. U/Pb dating of detrital zircon from new samples of the Storck 18 guartzite (central Virginia) and the Hoods Mill rocks (northern Maryland) confirms the 19 presence of a major age peak at ca. 630-610 Ma in these units. These ages are consistent 20 with derivation from Gondwana, but not Ediacaran Laurentia. Further, modern EHf values 21 of five of the ca. 670-580 Ma grains in these samples are inconsistent with derivation from 22 the few plutons of this age in Ediacaran Laurentia. The Loch Raven Schist and a 23 sedimentary xenolith in the Wilmington Complex contain a smaller proportion of ca. 670-24 580 Ma grains than the Storck quartzite and the Hoods Mill rocks, but more such grains 25 than in sediment derived from Ediacaran Laurentia, so we tentatively conclude that these 26 two units also received sediment from Gondwana. Detrital zircon ages from the type 27 localities of the Piney Run Formation, Pleasant Grove Schist, Prettyboy Schist, and 28 Wissahickon Formation indicate sediment provenance in Ediacaran Laurentia. We also 29 present new U-Pb and Lu-Hf isotopic data from western Newfoundland plutons for 30 comparison with such data from the detrital zircon. Intrusion ages of the Steel Mountain 31 Anorthosite, Disappointment Hill Tonalite, and Round Pond Granite are 608±12, 600±8, 32 and 590±9 Ma, respectively. None of these units was derived entirely from the depleted 33 mantle.

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34 **1. INTRODUCTION**

35 Successive accretion of magmatic arcs and ribbon continents to the eastern margin 36 of Laurentia during Early Ordovician to Early Mississippian time caused pulses of orogeny 37 in eastern Laurentia (van Staal et al., 2009; Pollock et al., 2012; Hibbard et al., 2012). In 38 the northern and southern Appalachian Orogen, many of these accreted terranes were exotic 39 to Laurentia, originally derived from the edges of Gondwana (Fig. 1; Pollock et al., 2012; 40 Macdonald et al., 2014). In the central Appalachian Piedmont Province, Early Ordovician 41 to Late Devonian deformation and metamorphism were widespread and locally intense 42 (Drake, 1989; Bosbyshell et al., 1999; 2016; Kunk et al., 2005; Aleinikoff et al., 2006; 43 Wintsch et al., 2010; Hughes et al., 2013), yet geologists have found only fragmentary 44 evidence for terranes derived from Gondwana. 45 The strongest published evidence for continental crust with Gondwanan affinity in 46 the central Appalachian Orogen consists of U/Pb isotopic ages of detrital zircon from 47 Cambrian-Ordovician meta-sandstone samples taken from three Piedmont Province 48 localities (Fig. 2; Hughes et al., 2014; Bosbyshell et al., 2015; Martin et al., 2015). The 49 three sample locations are in central Virginia (1 sample), northern Maryland (1 sample), 50 and southeastern Pennsylvania (4 samples). Some of the detrital zircon U/Pb ages from 51 these samples are not easily explained as originating from Ediacaran Laurentia but are 52 consistent with sediment derivation from a Gondwanan source. Most cogently, the 53 existence of abundant ages between 670 and 580 Ma in all these samples suggests 54 Gondwanan provenance. The 670-580 Ma ages are significant because felsic magmatism 55 of this age is known from only two locations in probable Ediacaran Laurentia. The 56 Goochland Terrane of central Virginia contains several small granitic plutons with 57 crystallization ages between ca. 660 and 580 Ma (Owens and Tucker, 2003), and several

58	granitic plutons in western Newfoundland crystallized between ca. 630 and 590 Ma
59	(Williams et al., 1985; van Berkel and Currie, 1988; Currie et al., 1992; Lin et al., 2013).
60	Further evidence for a Gondwanan sediment source to the sampled sandstone includes the
61	following two considerations. First, in some samples there is an absence of a spike in ages
62	at ca. 1200-950 Ma, the presence of which is a distinctive feature of Laurentia-derived
63	sediment deposited on the eastern margin of the continent (e.g., Macdonald et al., 2014,
64	Hughes et al., 2014; Martin et al., 2015). Second, some detrital zircon grains yielded U/Pb
65	ages between 950 and 780 Ma, which also was a period of scarce metamorphism or felsic
66	magmatism in Laurentia (Whitmeyer and Karlstrom, 2007). Bailey et al. (2008) reported
67	detrital zircon U/Pb ages between 950 and 780 Ma from a sandstone from central Virginia,
68	and likewise concluded that there was a Gondwanan sediment source to this sandstone
69	(Shores melange sample shown in Hughes et al., 2014). The existence of only a few meta-
70	sandstone samples with an apparent Gondwanan detrital zircon U/Pb age signature, and the
71	100 km that separates each of the central Virginia, northern Maryland, and southeastern
72	Pennsylvania sample localities, challenges secure recognition and reconstruction of a
73	putative peri-Gondwanan terrane in the Piedmont of the central Appalachian Orogen.
74	This paper investigates the possible presence of a Gondwana-derived terrane in the
75	central Appalachian Piedmont Province using a three-pronged approach. First, we confirm
76	the existence of sandstone that contains detrital zircon with a Gondwanan U/Pb age
77	signature by dating detrital zircon from new samples collected near the sites of the first
78	discoveries in central Virginia and northern Maryland (Hughes et al., 2014; Martin et al.,
79	2015). Second, we test whether the 670-580 Ma detrital zircon is incompatible with
80	derivation from Ediacaran Laurentia by comparing Hf isotope values in spots in the detrital
81	zircon grains to Hf isotope values in spots in zircon from the two possible Laurentian

82 sources, the granitoid in the Goochland Terrane and western Newfoundland. Third, we

83 search for more locations in the central Appalachian Piedmont Province that expose

84 sandstone with Gondwanan detrital zircon U/Pb age signatures.

85

86 **2. TECTONIC SETTING**

87 The Appalachian Orogen was part of a larger system of orogens that formed on the 88 eastern edge of Laurentia during the Paleozoic Era (Scotese and Langford, 1995; Pollock et 89 al., 2012). Geologists apply the term "Appalachian" to the portion of this larger orogen in 90 the eastern United States and Canada between Alabama and Newfoundland (Fig. 1; 91 Hibbard et al., 2006). In this article, we treat the southern Appalachians as that part of the 92 orogen southwest of the northeastern limit of Carolinia in central Virginia, the northern 93 Appalachians as the sector northeast of the southernmost exposure of Ganderia in southern 94 Connecticut, and the central Appalachians as the portion between the northeastern limit of 95 Carolinia and the southernmost exposures of Ganderia (Fig. 1). 96 Geologists use distinctions originally based on physiography to divide the central 97 and southern Appalachian Orogen into four tectonic provinces (Hatcher, 1989). From west 98 to east these are the Valley and Ridge, Blue Ridge, Piedmont, and Coastal Plain provinces. 99 The focus of this article is the Piedmont Province. Dominated by metasedimentary rocks, 100 the Piedmont Province is the formerly deepest and most outboard part of the orogen 101 currently exposed (Hibbard et al., 2006). Meta-volcanic, meta-plutonic, and 102 unmetamorphosed plutonic rocks also are present, particularly in eastern parts of the 103 Piedmont Province (Hibbard et al., 2006). Deposition and intrusion mostly occurred during 104 the Neoproterozoic and Paleozoic eras (Reinhardt, 1974; 1977; Fisher et al., 1979; 105 Aleinikoff et al., 2002; 2006; Owens and Tucker, 2003; Horton et al., 2010; Owens et al.,

106	2010; Pollock et al., 2010; Hughes et al., 2013; 2014; Bosbyshell et al., 2015; Martin et al.,
107	2015). Piedmont Province rocks constitute a collage of fault-bounded terranes of varying
108	origin (Fig. 2; Horton et al., 1989). The rocks in some terranes, such as the Westminster
109	Terrane and most of the Potomac Terrane, intruded or were deposited on the eastern edge
110	of Laurentia (Hughes et al., 2014; Martin et al., 2015). Others, such as the Carolinia
111	Domain, intruded and were deposited on the periphery of Gondwana (Pollock et al., 2012).
112	The depositional or intrusive origins of a few continental blocks are debated; examples
113	include the Goochland Terrane and the Wilmington Complex and adjacent rocks (Farrar,
114	1984; Horton et al., 1989; Glover et al., 1997; Aleinikoff et al., 2006; Bosbyshell et al.,
115	2015).
116	Geologists have assigned many names to and used shifting criteria to group the
117	metasedimentary rocks of the Piedmont Province in the central Appalachian Orogen over
118	the past 130 years (Williams, 1891; 1892; Bascom, 1902; 1905; Mathews, 1904; 1905;
119	Mathews and Grasty, 1909; Knopf and Jonas, 1923; Virginia Geological Survey, 1928;
120	Jonas and Stose, 1938; Hopson, 1964; Southwick and Fisher, 1967; Rodgers, 1970;
121	Higgins, 1972; Crowley, 1976; Horton et al., 1989; Hibbard et al., 2006; Southworth et al.,
122	2007; Bosbyshell et al., 2015; Martin et al., 2015). Neither detrital zircon U/Pb ages nor
123	other provenance indicators were used in the assignations prior to approximately 2006.
124	More recent U/Pb isotopic dating of detrital zircon made clear that some of the classical
125	lithology-based correlations and groupings are not supported by the new dating (Hughes et
126	al., 2014; Bosbyshell et al., 2015; Martin et al., 2015). It is beyond the scope of this article
127	to discuss the history of formation names or to provide a resolution to naming conflicts in
128	the central Appalachian Piedmont Province. Instead, we show sample locations in Figure 2
129	free of the biases produced by the old lithology-based formation names. In Table 1 we list
	6

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the formation names as currently applied but emphasize that in some cases these formation
names are not compatible with recent studies of detrital zircon U/Pb ages. One of the
samples listed in Table 1 came from a metasedimentary xenolith in the Brandywine Blue
Gneiss, which is a member of the Wilmington Complex. The Wilmington Complex is an
Early Ordovician island arc metamorphosed to granulite facies during the Silurian Period
(Aleinikoff et al., 2006).

136 From west to east, the island of Newfoundland consists of a Laurentian/peri-137 Laurentian domain and two peri-Gondwanan domains, Ganderia and Avalonia. Cawood et 138 al. (1995) divided part of the Laurentian domain into several fault-bounded tectonic blocks 139 with partially distinct geologic histories. Two of our Newfoundland samples come from a 140 central block, the Corner Brook Lake block (Fig. 3; Table 2). In the Corner Brook Lake 141 block, the oldest rocks are granulite facies gneiss and related rocks with protolith igneous 142 crystallization ages of ca. 1500 Ma (Currie et al., 1992; Cawood et al., 1996; Lin et al., 143 2013). These Mesoproterozoic units were intruded by a suite of granitic magmas at ca. 600 144 Ma (Williams et al., 1985; van Berkel and Currie, 1988; Currie et al., 1992; Lin et al., 145 2013) as well as by bimodal magmas at ca. 555 Ma (Cawood et al., 1995). Our third Newfoundland sample comes from the Steel Mountain Anorthosite, which intruded the 146 147 Mesoproterozoic Corner Brook Lake rocks in some locations and was thrust below the 148 southern edge of the Corner Brook Lake block in others (Fig. 3; Lin et al., 2013). The 149 intrusion age of the Steel Mountain Anorthosite heretofore was assumed to be 150 Mesoproterozoic (Lin et al., 2013) but in this contribution is shown to be ca. 608 Ma. The 151 Proterozoic rocks are unconformably overlain by Ediacaran to Ordovician clastic and 152 carbonate deposits (Cawood and Nemchin, 2001). Lin et al. (2013) concluded that the Corner Brook Lake block, traditionally held to be the para-autochtonous former eastern 153

margin of Laurentia, is in fact allochthonous, placed in its present position by motion on
Paleozoic transcurrent faults.

Laurentia consisted of an Archean nucleus surrounded by Proterozoic and Paleozoic 156 157 terranes, some of which originated far from Laurentia (Whitmever and Karlstrom, 2007). 158 Regardless of their origin, after accretion to the continent these terranes became part of 159 Laurentia. We therefore consider a Laurentian sediment source to be a source in Laurentia 160 as the continent existed at the time of deposition. The sedimentary rocks considered in this 161 article were deposited during or after the Ediacaran Period, so we refer to the continent as 162 Ediacaran Laurentia when discussing sediment provenance. We use modern orientations to 163 indicate compass directions, though it is important to consider that Laurentia rotated in map 164 view during the Neoproterozoic and Paleozoic eras (Li et al., 2008; Pollock et al., 2012). 165 Previous authors have referred to named high strain zones using the term "fault" or "shear 166 zone" without careful attention to deformation mechanisms in some cases. We retain the 167 historical uses of these words to ease comparison with previous publications and because 168 deformation mechanisms are irrelevant for the conclusions in this article.

169

170 **3. METHODS**

171 **3.1 Sample collection and zircon processing**

Martin collected all samples. Each meta-sandstone sample came from the least
micaceous, coarsest-grained part of the outcrop. Photomicrographs of thin sections from
every sample are shown in Figure S1.

Martin et al (2015) reported Gondwanan zircon in a meta-sandstone in the Mather
Gorge Formation of northern Maryland. In this paper we assign the informal name "Hoods
Mill rocks" to these exotic rocks because their provenance is distinct from that of the

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178 Mather Gorge Formation near its type section, which was derived from Ediacaran 179 Laurentia. For the current paper, Martin collected a new sample of the Hoods Mill rocks to 180 verify the presence of Gondwanan detrital zircon (Table 1). The new sample was collected 181 south of the south branch of the Patapsco River and east of Maryland State Route 97 182 whereas the sample reported in Martin et al. (2015) was located along the railroad tracks 183 north of the river and directly west of the road. For the same purpose, Martin collected a 184 new sample of the informally-named Storck quartzite of central Virginia near the location 185 of the sample with Gondwanan zircon described by Hughes et al. (2014).

186 Zircon was separated and mounted in the laboratories in the Department of Geology 187 at the University of Maryland. We isolated zircon from each sample by first crushing the 188 rock using a mortar and pestle, then removing silt- and clay-sized particles by hand-panning 189 in water, removing magnetic grains with a Frantz magnetic barrier separator, and removing 190 less-dense grains in methylene iodide. We poured the now nearly-pure zircon onto double-191 sided tape and cast the grains in an epoxy disk together with shards or loose grains of 192 reference zircon crystals. We then sanded and polished the disks by hand to expose the 193 interiors of the grains. Prior to isotopic analysis, we imaged the grains using backscattered 194 electrons and cathodoluminescence in the JEOL JXA-8900R electron probe microanalyzer 195 at the University of Maryland. The images were used to avoid multiple 196 cathodoluminescence zones, cracks, and inclusions during selection of spots for isotopic 197 analysis.

198

199 **3.2 Zircon mass spectrometry**

Zircon from the western Newfoundland igneous rocks and all central Appalachian
Piedmont Province meta-sandstone was analyzed in the Arizona LaserChron Center at the

202 University of Arizona (Tables 1 and 2). Gehrels and Pecha (2014), Pullen et al. (2014), and 203 Ibanez-Meija et al. (2015) described the analytical procedures in detail: in this sub-section 204 we briefly summarize these methods. We first analyzed spots in zircon grains for uranium 205 and lead isotopes. We then selected zircon from samples 314002 (Storck quartzite) and 206 1213002 (Hoods Mill rocks) for in situ analysis of lutetium and hafnium isotopes in a 207 different analysis session. We focused on grains from these samples with ages between ca. 208 670 and 580 Ma; we also analyzed some younger and older grains. The laser spot for the 209 hafnium isotopic analyses was centered on the pit excavated by laser ablation for the 210 uranium and lead isotope measurements. All uncertainties are reported at the 2-sigma 211 level.

212

213 **3.2.1** Uranium-lead isotope measurements

214 A 20 µm-diameter spot was ablated in each zircon grain using a Photon Machines 215 Analyte G2 excimer laser equipped with a HelEx ablation cell. The ablated zircon was 216 carried in helium into the plasma source of a Thermo Element2 single-collector high 217 resolution inductively coupled plasma mass spectrometer, which sequences rapidly through 218 measurement of U, Th, and Pb isotopes. Each analysis consisted of 5 s of background 219 measurement on peaks with the laser off, 10 s with the laser firing, and a 20 s delay to 220 purge the previous sample and save files. The resulting ablation pits were approximately 221 12 µm deep. Analyses of reference zircon FC52 and SL bracketed every 5 analyses of 222 sample zircon; these analyses were used to correct fractionation of the ²⁰⁶Pb/²⁰⁷Pb and 223 206 Pb/ 238 U ratios, respectively. We also bracketed every 15 analyses of sample zircon with 224 an analysis of reference zircon R33, treated as an unknown in this situation, to check the

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225	age determinations. Accepted ages for these standards are 1098.47±0.16, 563.5±3.2, and
226	420.53±0.16 Ma for FC, SL, and R33, respectively (Gehrels et al., 2008; Mattinson, 2010).
227	Data reduction was performed offline using an in-house Python decoding routine
228	and an Excel program. Data reduction included the following steps. (1) Using the natural
229	isotopic ratio ${}^{202}\text{Hg}/{}^{204}\text{Hg} = 4.3$, subtraction of the contribution from ${}^{204}\text{Hg}$ to the mass 204
230	signal to yield ²⁰⁴ Pb intensity. This mercury correction was not significant for most
231	analyses because the mercury backgrounds were low. (2) Correction for common lead
232	based on the measured ratio 206 Pb/ 204 Pb and the assumed composition of common lead from
233	Stacey and Kramers (1975). (3) Correction for mass fractionation during analysis using the
234	bracketing analyses of the reference zircon. (4) Calculation of U and Th concentrations
235	using the measured intensity and known concentrations of reference zircon FC52. For
236	detrital grains, we removed from consideration analyses with greater than 20% normal
237	discordance, greater than 5% reverse discordance, or greater than 10% measurement
238	(internal) uncertainty. We used ²⁰⁶ Pb/ ²³⁸ U dates for grains with ²⁰⁶ Pb/ ²⁰⁷ Pb dates younger
239	than 900 Ma and ²⁰⁶ Pb/ ²⁰⁷ Pb dates for older grains.

240

241 **3.2.2 Lutetium-hafnium isotope measurements**

All Lu-Hf isotope data reported in this paper were acquired during a single analysis session. We used a Nu multi-collector high resolution inductively coupled plasma mass spectrometer to analyze hafnium isotopes. Instrument settings were established first by measurements of 10 ppb solutions of JMC475 and a Spex Hf solution followed by measurements of 10 ppb solutions containing Spex Hf, Yb, and Lu. These mixtures contained a range of concentrations of Yb and Lu, with ¹⁷⁶(Yb+Lu) up to 70% of the ¹⁷⁶Hf. After instrument tuning using the solutions, the settings on the mass spectrometer were

249	optimized for laser ablation by a Photon Machines Analyte G2 excimer laser with a beam
250	diameter of 40 μ m. We then analyzed shards or loose grains of seven different zircon
251	standards (R33, Temora, Mud Tank, Plesovice, 91500, FC52, and Sri Lanka) that were
252	included in the epoxy disks with the sample zircon. After precision and accuracy were
253	acceptable, we began analysis of zircon grains from the Appalachian samples using the
254	same acquisition parameters. Each analysis consisted of background measurement via a
255	single 40-second integration on peaks with the laser off followed by sixty 1-second
256	integrations with the laser firing. The ablation rate was about 0.8 μ m per second. Analysis
257	of each standard bracketed about 20 analyses of sample zircon.
258	Data reduction was performed offline using an Excel program developed at the
259	Arizona LaserChron Center. We corrected for mass fractionation during analysis following
260	the method of Woodhead et al. (2004). All corrections were performed line-by-line. For
261	each 60-second analysis, the mean and standard error of the ratio ¹⁷⁶ Hf/ ¹⁷⁷ Hf were
262	calculated from the 1-second integrations after removing values more than two standard
263	deviations from the mean. We calculated the ratio ¹⁷⁶ Hf/ ¹⁷⁷ Hf at the time of zircon
264	crystallization using the measured ¹⁷⁶ Hf/ ¹⁷⁷ Hf and ¹⁷⁶ Lu/ ¹⁷⁷ Hf values and the ¹⁷⁶ Lu decay
265	constant of 1.867 x 10 ⁻¹¹ year ⁻¹ (Scherer et al., 2001; Soderlund et al., 2004).
266	
267	3.3 Uranium-lead data analysis and presentation
268	We prepared concordia, weighted mean, and relative age-probability diagrams using
269	Isoplot version 4.15 (Ludwig, 2008). The weighted means were calculated with weighting
270	according to the square of the internal uncertainties. The total uncertainty on the intrusion

- age determination for each western Newfoundland igneous sample was calculated by
- 272 quadratic addition of the measurement (internal) and systematic (external) uncertainties.

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Age-probability diagrams for individual meta-sandstone samples show the age of each dated zircon and its measurement error as a normal distribution, summed into a single curve. Stacked age-probability plots from multiple meta-sandstone samples were prepared using an Excel program written at the Arizona LaserChron Center. This program normalizes the age-probability curves according to the number of constituent analyses such that each curve contains the same area, allowing direct comparison of multiple datasets, each composed of different numbers of analyses.

281 **3.4 Determination of maximum depositional ages**

282 Calculation of the maximum possible depositional age of a metasedimentary rock 283 using its youngest detrital zircon grains commonly is complicated by lead loss, growth of 284 metamorphic zircon, multiple age zones in a single grain, analytical error, and dating a very 285 small fraction of the total zircon present in the sedimentary rock. Using the youngest single 286 age from a set of detrital zircon U/Pb isotopic ages can result in a determination of 287 maximum depositional age that is younger than the true depositional age (Dickinson and 288 Gehrels, 2009). Instead, determining maximum depositional age using multiple analyses 289 that overlap within uncertainty yields a more robust constraint on the maximum possible 290 depositional age (Dickinson and Gehrels, 2009). We therefore calculate the maximum 291 depositional age as the weighted mean of the two or more youngest analyses that overlap at 292 the 1-sigma level plus the 2-sigma error on this weighted mean, rounded up to the nearest 293 10 M.y. interval.

294

4. WESTERN NEWFOUNDLAND IGNEOUS ZIRCON SPOT U-PB AND LU-HF ISOTOPE DATA

297 4.1 Western Newfoundland zircon U-Pb isotope data

We placed one 20 µm-diameter analysis spot in each zircon crystal because most
crystals were small and many contained only one cathodoluminescence domain (Fig. 4).
Table S1 and figure 5 present the results of these analyses. The dates of the standard zircon
analyzed along with the unknowns mostly overlap the accepted ages within uncertainty
(Table S2).

Analyses of spots in 50 zircon crystals from sample 1013002 produced ²⁰⁶Pb/²³⁸U ages between 627 and 581 Ma (Table S1, Figs. 4, 5). Analyses of spots in 16 zircon grains from sample 1013005 mostly yielded ²⁰⁶Pb/²³⁸U ages between 634 and 590 Ma plus one concordant analysis with a ²⁰⁶Pb/²⁰⁷Pb age of ca. 1140 Ma (Table S1, Figs. 4, 5). Spot analysis of 46 crystals from sample 1013006 yielded ²⁰⁶Pb/²³⁸U ages from 657 to 440 Ma (Table S1, Figs. 4, 5). Several of these grains are discordant with ²⁰⁶Pb/²⁰⁷Pb ages between ca. 1429 and 713 Ma.

310

311 4.2 Western Newfoundland zircon Lu-Hf isotope data

312 Repeated analysis of the seven zircon standards throughout the data acquisition

313 session yielded the following weighted mean values of the measured ¹⁷⁶Hf/¹⁷⁷Hf ratios.

314 R33: 0.282751±6, Temora: 0.282668±7, Mud Tank: 0.282540±5, Plesovice: 0.282500±6,

315 91500: 0.282316±7, FC52: 0.282179±6, Sri Lanka: 0.281684±5 (Fig. S2). These values

316 mostly overlap the accepted values within uncertainty; note, however, that different articles

317 report different values for some of the standards (Woodhead and Hergt, 2005; Kemp et al.,

318 2006; Slama et al., 2008; Fisher et al., 2014).

319 In order to fit the 40 µm-diameter laser spot into a single cathodoluminescence

320 domain while avoiding large cracks and inclusions, we were forced to analyze some

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321	slightly discordant zircon (Tables S1, S3). Analysis of fifteen spots in zircon from sample
322	1013002 of the Disappointment Hill Tonalite yielded a range of current EHf values between
323	-13.0 and -16.5 (Fig. 6; Table S3). This range was +0.2 to -3.6 at the time of intrusion, ca.
324	600 Ma. Analysis of five spots in non-xenocrystic zircon from sample 1013005 of the Steel
325	Mountain Anorthosite gave present-day EHf values from -10.4 to -13.7 (Fig. 6; Table S3).
326	At 600 Ma, these values were +2.8 to -0.6. The modern ϵ Hf value of the ca. 1140 Ma
327	crystal from sample 1013005 is -15.7; this value was +9.3 at 1140 Ma. The current eHf
328	values of eight spots in non-xenocrystic zircon from sample 1013006 of the Round Pond
329	Granite ranges from -7.8 to -12.2; these values were +5.1 to -0.1 at 600 Ma (Fig. 6; Table
330	S3). These eight spots do not include the analysis from grain 31, which may be a xenocryst
331	(see section 6.1). Including the analysis of this grain would not change our interpretation
332	because its modern EHf value of -7.9 falls within the range of EHf values from the non-
333	xenocrystic zircon extracted from sample 1013006.
334	
335	5. CENTRAL APPALACHIAN PIEDMONT DETRITAL ZIRCON U-PB AND LU-
336	HF ISOTOPE DATA

337 5.1 Central Appalachian Piedmont detrital zircon U-Pb isotope data

We placed one 20 µm-diameter analysis spot in each zircon grain. Table S4
contains the U-Pb isotope data from these spots and Figure S3 shows the ages in histograms
and relative probability plots. For analysis of Lu-Hf isotopes in zircon from samples
314002 and 1213002, we were forced to use six 5-16% reversely discordant analyses in
order to fit the 40 µm-diameter laser spot into the zircon grains without intersecting
multiple cathodoluminescence domains or large cracks or inclusions. These six U-Pb
isotope analyses are listed in red in Table S4. We used these spots only for the Lu-Hf

345	isotope analyses, we did not use the U-Pb isotope analyses for determination of sediment
346	provenance or maximum depositional ages. The mean ages of the zircon standards
347	analyzed with each sample overlap the accepted ages within uncertainty (Table S2).
348	Zircon from sample 314002 of the Storck quartzite (formal name: Mine Run
349	Complex I) yielded 269 analyses with ages between ca. 3064 and 499 Ma. Similarly,
350	zircon from sample 1213002 of the Hoods Mill rocks (formal name: Mather Gorge
351	Formation) produced 252 analyses with ages between ca. 3034 and 521 Ma.
352	194 analyses of zircon in sample 1213003 of the Loch Raven Schist yielded ages
353	from ca. 1801 to 397 Ma and one age of ca. 2601 Ma. Many grains younger than
354	approximately 460 Ma had high U/Th ratios, up to 380 (Table S4). Zircon from sample
355	114002 of the Piney Run Formation yielded 166 analyses with ages between ca. 2775 and
356	1033 Ma plus one analysis at ca. 514 Ma (167 analyses total). Sample 114003 of the
357	Pleasant Grove Schist yielded 271 ages between ca. 3005 and 955 Ma plus one analysis at
358	ca. 629 Ma and one at ca. 518 Ma (273 analyses total). Analyses of zircon from sample
359	314001 of the Prettyboy Schist yielded 284 ages from ca. 1915 to 910 Ma plus additional
360	ages at ca. 2478, 561, and 536 Ma (287 analyses total). 245 analyses in zircon from sample
361	314003 of the sedimentary xenolith in the Brandywine Blue Gneiss yielded ages between
362	ca. 1993 and 423 Ma. Finally, zircon from sample 114005 of the Wissahickon Formation
363	yielded 207 analyses with ages from ca. 2848 to 939 Ma plus two analyses at ca. 540 Ma
364	(209 analyses total).
365	

366 5.2 Central Appalachian Piedmont detrital zircon Lu-Hf isotope data

We analyzed spots in 40 zircon grains from sample 314002 of the Storck quartzite
with crystallization ages between ca. 927 and 513 Ma (Tables 1; S3). The range of present-

369 day εHf values in these crystals is -4 to -27 (Fig. 7, Table S3). We analyzed spots in 22

370 crystals from sample 1213002 of the Hoods Mill rocks with crystallization ages between ca.

371 911 and 541 Ma (Tables 1; S3). The modern εHf values in these grains range from -1 to -

372 40 (Fig. 7, Table S3). There is no correlation between zircon crystallization age and

373 modern εHf value (Fig. 7).

374

375 6. DISCUSSION

376 6.1 Intrusion ages of western Newfoundland plutons

For zircon from sample 1013002 of the Disappointment Hill Tonalite, removing from further consideration analyses more than 10% normally discordant or 5% reverse discordant leaves 30 analyses. The weighted mean ${}^{206}Pb/{}^{238}U$ age of these 30 analyses is 600 ± 8 Ma (MSWD = 0.7) including systematic errors (Fig. 8). We take this as the intrusion age of the tonalite. This age overlaps within uncertainty the 606±2 Ma intrusion age published by Lin et al. (2013).

383 Zircon grain 13 in sample 1013005 of the Steel Mountain Anorthosite yielded a 384 ²⁰⁶Pb/²⁰⁷Pb age of 1141±64 Ma, which is more than 500 M.y. older than the other ages from 385 this sample. We interpret this grain to be a xenocryst inherited from the Mesoproterozoic 386 rocks of the Corner Brook Lake block. Excluding the analysis of this xenocryst as well as 387 analyses more than 10% normally discordant or 5% reverse discordant leaves 11 analyses. 388 The weighted mean of these 11 analyses is 608 ± 12 Ma (MSWD=0.6) including systematic 389 errors (Fig. 8). We take this as the intrusion age of the anorthosite. The Ediacaran 390 intrusion age of the Steel Mountain Anorthosite is considerably younger than the 391 Mesoproterozoic intrusion age assumed in the absence of radiometric dating (Lin et al., 392 2013). Based on our new dating, we now recognize that intrusion of the anorthosite was

393	broadly coeval with ca. 600 Ma intrusion of a granitic suite including the Disappointment
394	Hill Tonalite and the Round Pond Granite. Because of the similarity in intrusion ages, the
395	Steel Mountain Anorthosite perhaps should be considered part of the Corner Brook Lake
396	block, as is this granitic suite.
397	Analysis of spot 42 in sample 1013006 of the Round Pond Granite was 4.5%
398	reverse discordant and its ²⁰⁶ Pb/ ²³⁸ U age of 657 Ma is about 50 M.y. older than the
399	206 Pb/ 238 U age of the next oldest concordant analysis. We interpret this grain to be a
400	xenocryst and do not use it for calculating the intrusion age of the granite. The 206 Pb/ 238 U
401	age from spot 31 of 479 Ma is about 90 M.y. younger than the next youngest concordant
402	analysis. Crystals 12, 30, and 11 have ²⁰⁶ Pb/ ²³⁸ U ages of ca. 440, 555, and 557 Ma and
403	corresponding ²⁰⁶ Pb/ ²⁰⁷ Pb ages of 801, 713, and 739 Ma, respectively, and thus we
404	recognize these grains as xenocrysts. Based on the results from grains 12, 30, and 11, we
405	suggest grain 31 also is a xenocryst and do not use it in the calculation of the intrusion age.
406	Excluding spots 42 and 31 as well as analyses more than 10% normally discordant or 5%
407	reverse discordant leaves 21 analyses. The weighted mean of these 21 analyses is 590±9
408	Ma (MSWD=0.8) including systematic errors (Fig. 8). We take this as the intrusion age of
409	the granite. Although this age overlaps with the multi-crystal thermal ionization mass
410	spectrometer (TIMS) age of 602±10 Ma published by Williams et al. (1985) because of the
411	large uncertainties on both ages, the central value of the TIMS age is 12 M.y. older than the
412	central value of our laser ablation spot age. We now identify inherited zircon in this
413	sample, leading us to suggest that the TIMS age may be slightly too old because Williams
414	et al. (1985) did not recognize that some parts of the grains they analyzed were xenocrysts.
415	

416 **6.2 Sources of magma to western Newfoundland plutons**

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417 The main goal of this paper is not to elucidate the sources of melt for the Ediacaran 418 western Newfoundland plutons. Nevertheless, the zircon spot U-Pb and Lu-Hf isotope data 419 allow us to recognize that none of the three studied plutons was derived solely from the 420 depleted mantle. This conclusion is based on two types of data. First, the presence of 421 xenocrystic zircon in samples 1013005 and 1013006 requires assimilation of preexisting 422 crust into the Steel Mountain Anorthosite and Round Pond Granite magmas. Second, the 423 εHf values of zircon in all three plutons were -7.8 to -16.5 at 600 Ma whereas the depleted 424 mantle ε Hf value at this time was approximately +13 (Martin et al., 2018). The much more 425 negative values of all the analyzed zircon in the western Newfoundland plutons rule out 426 magma derivation purely from the depleted mantle. 427

428 6.3 Maximum possible depositional ages of central Appalachian Piedmont sandstone 429 In sample 314002 of the Storck quartzite, the two youngest analyses came from 430 grain 238 (499±44 Ma) and grain 76 (513±28 Ma) (Table S4). The weighted mean of these 431 two analyses is 509 ± 23 Ma, which gives a maximum possible depositional age of 540 Ma. 432 If instead we reject grain 238 as too young and use the ages from grain 76 and grain 299 433 $(524\pm22 \text{ Ma})$, the weighted mean is 520 ± 17 , which again yields a maximum depositional 434 age of 540 Ma. Accordingly, we take 540 Ma as the maximum possible depositional age of 435 the Storck quartzite. Hughes et al. (2014) similarly found that the maximum depositional 436 age of their sample of the Storck quartzite (their sample KSH-11-40) was Cambrian. 437 In sample 1213002 of the Hoods Mill rocks, grains 151 and 307 yielded the two 438 voungest ${}^{206}Pb/{}^{238}U$ ages, 521±40 and 529±22 Ma, respectively (Table S4). The weighted 439 mean of these ages is 528±19 Ma, which gives a maximum depositional age of 550 Ma. 440 Adding grains 92 (531±30 Ma) and 144 (531±30 Ma), the weighted mean of the four

441	youngest analyses is 529±14 Ma, which likewise gives a maximum depositional age of 550
442	Ma. We thus take 550 Ma as the maximum depositional age of the Hoods Mill rocks based
443	on this sample. The youngest detrital zircon in the Hoods Mill rocks sample studied by
444	Martin et al. (2015) (their sample 1010002) had similar ages and the maximum depositional
445	age determined from their sample was comparable at 540 Ma.
446	Interpretation of the zircon ages in sample 1213003 of the Loch Raven Schist is
447	complicated by post-depositional growth of metamorphic zircon as well as lead loss from
448	detrital grains. Grains 253 through 165 yielded the youngest ²⁰⁶ Pb/ ²³⁸ U ages from this
449	sample and most of these crystals had U/Th ratios between 380 and 102 (Table S4).
450	Because of their youth and high U/Th ratios (see Hoskin and Schaltegger, 2003), we
451	interpret these 17 grains to have crystallized during metamorphism after deposition;
452	therefore we do not use these grains for determining maximum depositional age or
453	provenance. Grains 232 through 288 are older and had lower U/Th ratios than these 17
454	metamorphic crystals (Table S4). However, most of these grains were only 86-91%
455	concordant, and grain 324 was 4% reverse discordant with an age uncertainty of 100 M.y.
456	It is possible that these 8 grains are detrital grains that actually crystallized between ca. 488
457	and 459 Ma. However, the discordance of the analyses suggests that these could be detrital
458	grains with older crystallization ages. We conservatively choose not to use these 8 grains
459	for determination of the maximum depositional age or provenance in order to avoid
460	erroneously young ages. The next two youngest analyses came from grain 203 (507±22
461	Ma; 97% concordant) and grain 213 (510±44 Ma; 96% concordant). The weighted mean of
462	these two ages is 507±20 Ma, which gives a maximum depositional age of 530 Ma.
463	Adding spot 310 (526±24 Ma), the weighted mean of these three analyses is 515±15 Ma,

464	which likewise gives a maximum depositional age of 530 Ma. We therefore take 530 Ma
465	as the maximum depositional age of the Loch Raven Schist.
466	With a ²⁰⁶ Pb/ ²³⁸ U age of 514±28 Ma, grain 180 produced the youngest analysis
467	from sample 114002 of the Piney Run Formation (Table S4). However, no other ages from
468	this sample overlap the age of grain 180 within uncertainty, so we do not use the age of
469	grain 180 to determine the maximum depositional age of this formation. Grain 185
470	(1033±46 Ma), grain 228 (1036±38 Ma), and grain 148 (1037±52 Ma) are the next three
471	youngest grains in sample 114002. The weighted mean of their ages is 1035±25 Ma, which
472	gives a maximum depositional age of 1060 Ma for the Piney Run Formation.
473	Sample 114003 of the Pleasant Grove Schist produced two young grains with
474	²⁰⁶ Pb/ ²³⁸ U ages that do not overlap with any other analyses within uncertainty: grain 10 at
475	519±32 Ma and grain 309 at 629±24 Ma (Table S4). The four youngest grains with ages
476	that do overlap within uncertainty are grain 165 (955±132 Ma), grain 187 (969±100 Ma),
477	grain 206 (988±62 Ma) and grain 186 (989±146 Ma). The weighted mean of these four
478	ages is 980±46 Ma, which results in a maximum depositional age of 1030 Ma.
479	The ages of the two youngest grains in sample 314001 of the Prettyboy Schist, grain
480	124 (536±22 Ma) and grain 240 (561±28 Ma) overlap within uncertainty. The weighted
481	mean of these two ages is 546 ± 17 Ma. We therefore use 570 Ma as the maximum
482	depositional age of the Prettyboy Schist.
483	Interpretation of the ages of the zircon extracted from sample 314003 of the
484	sedimentary xenolith in the Brandywine Blue Gneiss is complicated by the presence of
485	metamorphic zircon (see also Aleinikoff et al., 2006). The youngest 37 zircon crystals,
486	from grain 45 to grain 183, have ²⁰⁶ Pb/ ²³⁸ U ages that overlap the intrusion age of the
487	Brandywine Blue Gneiss protolith (476±6 Ma; Aleinikoff et al., 2006), considering
	21

488	uncertainties (Table S4). We conservatively choose not to use these 37 ages for
489	interpretation of maximum depositional age or provenance in order to avoid inclusion of
490	grains with an age that is incorrectly too young. The two youngest grains with $^{206}Pb/^{238}U$
491	ages that do not overlap the gneiss protolith intrusion age within uncertainty are grain 242
492	(515 \pm 24 Ma) and grain 162 (515 \pm 34 Ma). The weighted mean of these two ages is 515 \pm 19
493	Ma, which produces a maximum depositional age of 540 Ma. Adding the next youngest
494	analysis, 517±24 Ma from grain 50, the weighted mean of the three ages becomes 516±15
495	Ma, for a maximum depositional age of 540 Ma. We thus take 540 Ma as the maximum
496	depositional age of the sedimentary xenolith in the Brandywine Blue Gneiss.
497	The two youngest analyses from sample 114005 of the Wissahickon Formation
498	produced ${}^{206}Pb/{}^{238}U$ ages of 537±48 Ma (grain 114) and 542±34 (spot 23) (Table S4). The
499	weighted mean of these ages is 540±28 Ma. The maximum depositional age of the
500	Wissahickon Formation therefore is 570 Ma.
501	
502	6.4 Central Appalachian Piedmont sandstone sediment provenance
503	The tallest age peak in samples 114002, 114003, and 114005 of the Piney Run
504	Formation, Pleasant Grove Schist, and Wissahickon Formation lies at ca. 1460-1380 Ma
505	(Fig. S3). Each sample also has a major peak at ca. 1200-1170 Ma, a smaller Neoarchean
506	peak, a continuum of ages between ca. 1800 and 950 Ma, and one or two grains between
50 7	

507 550 and 500 Ma. The results from sample 314001 of the Prettyboy Schist are similar, with

- a main age peak at ca. 1050 Ma, one analysis at ca. 2500 Ma, a continuum of ages between
- 509 ca. 1500 and 950 Ma, and two analyses between ca. 560 and 530 Ma (Fig. S3). The
- 510 Mesoproterozoic main age peak in all four samples also is present in late Ediacaran-
- 511 Cambrian quartzite from the Blue Ridge Province directly to the west (Fig. 9; Satkoski,

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512	2013). The source of the Blue Ridge sediment was Ediacaran Laurentia (Satkoski, 2013;
513	Smoot and Southworth, 2014). The spectrum of detrital zircon ages from the four new
514	samples also is similar to the age spectrum from many other Ediacaran-Cambrian quartzite
515	samples from the Piedmont Province of the central part of the Appalachian Orogen (Fig. 9).
516	Hughes et al. (2014) and Martin et al. (2015) showed that the sources of the zircon to these
517	Piedmont meta-sandstone samples could have been rocks in Ediacaran Laurentia. The
518	abundant ca. 1550-950 Ma grains could have come from the Granite-Rhyolite and
519	Grenville provinces directly west of the Appalachian Piedmont Province, whereas most of
520	the grains with other ages may have originated farther west (Whitmeyer and Karlstrom,
521	2007; Martin et al., 2015). This analysis holds for our four new samples as well. Because
522	of the viable sources in Ediacaran Laurentia and the similarity of the new ages to those
523	from the Blue Ridge Province, we interpret Ediacaran Laurentia as the source of sediment
524	to the type localities of the Piney Run Formation, Pleasant Grove Schist, Prettyboy Schist,
525	and Wissahickon Formation.

526 One motive for analyzing a new sample of the Storck quartzite and the Hoods Mill 527 rocks is to verify the detrital zircon age spectra reported by Hughes et al. (2014) and Martin 528 et al. (2015). Figure 10 confirms that the detrital zircon age spectrum from each of our new 529 samples is nearly identical to the previously reported age spectrum from each unit. Figure 530 10 also highlights the similarities in ages of detrital zircon from the Storck quartzite and the 531 Hoods Mill rocks. In both units, the main age peak is at ca. 630-600 Ma. Each unit 532 additionally contains zircon with ages both younger and older than this spike, with no large 533 gaps between ca. 2200 and 530 Ma.

Hughes et al. (2014) and Martin et al. (2015) concluded that Ediacaran Laurentia
was an unlikely source for the detrital zircon in the Storck quartzite and Hoods Mill rocks,

536 respectively, for two reasons. First, the main detrital zircon age peak in these samples is at 537 ca. 630-610 Ma, but Ediacaran Laurentia contained few felsic rocks with crystallization 538 ages between ca. 670 and 580 Ma (Whitmever and Karlstrom, 2007). Second, the ages of 539 detrital zircon from the Storck quartzite and Hoods Mill rocks do not form a prominent 540 Mesoproterozoic peak, but a major Mesoproterozoic age peak is a key signature of detrital 541 zircon derived from Ediacaran Laurentia. Hughes et al. (2014) and Martin et al. (2015) 542 instead proposed that Gondwana was the most likely source of the sediment to these units. 543 Figure 9 emphasizes that the ages of detrital zircon from the Storck quartzite and Hoods 544 Mill rocks are similar to detrital zircon ages from known peri-Gondwanan terranes but are 545 dissimilar to detrital zircon ages from sediment derived from Ediacaran Laurentia, such as 546 the late Ediacaran-Cambrian sandstone of the central Blue Ridge Province. Based on the 547 detrital zircon ages, we concur with the previous interpretations: Gondwana is a plausible 548 source of the sediment to the Storck quartzite and Hoods Mill rocks but Ediacaran Laurentia is not. 549

550 To test whether the Neoproterozoic plutons in the Goochland Terrane or western 551 Newfoundland could have provided detrital zircon to the Storck quartzite or Hoods Mill 552 rocks, we compare the modern *e*Hf values of spots in zircon in Neoproterozoic plutons 553 from each area to the modern EHf values of spots in the detrital zircon in samples 314002 554 and 1213002 that overlap the age range 670-580 Ma within uncertainty (Fig. 11). Most of 555 the ε Hf values of the detrital zircon overlap the ε Hf values of the zircon from the plutons. 556 However, one detrital grain from the Hoods Mill rocks has an EHf value less negative than 557 the pluton zircon and three grains from the Hoods Mill rocks and one grain from the Storck 558 quartzite have an EHf value more negative than the pluton zircon, outside uncertainty. It is 559 impossible to rule out the Goochland Terrane or western Newfoundland as sources of the

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560 detrital zircon based on EHf values because it is possible that a magmatic rock bearing 561 zircon with the appropriate Lu-Hf isotopic composition existed at the time of deposition of 562 the Storck quartzite or Hoods Mill rocks but that magmatic rock has since been eroded 563 away, leaving it unavailable for sampling. Nevertheless, the presence of detrital zircon 564 with modern *e*Hf values both less and more negative than any of the values measured in 565 zircon from the Neoproterozoic plutons is consistent with the derivation of Storck and 566 Hoods Mill sediment from a source other than the Goochland Terrane or western 567 Newfoundland. Moreover, the present-day EHf values of the detrital zircon in the Storck 568 quartzite and Hoods Mill rocks mostly overlap the modern EHf values of ca. 670-580 Ma 569 detrital zircon from a Ganderian sandstone, which range from -7 to -30 (Willner et al., 570 2014).

571 Neither the U/Pb ages nor the present-day *E*Hf values of detrital zircon in the Storck quartzite and Hoods Mill rocks are consistent with sediment derivation from Ediacaran 572 573 Laurentia. Further, the U/Pb ages of detrital zircon from these units are similar to the ages 574 of detrital zircon from late Ediacaran-Cambrian sandstone in known peri-Gondwanan 575 terranes. We therefore conclude that Gondwana was the most likely source of sediment to 576 the Storck quartzite and the Hoods Mill rocks. The Storck quartzite lies directly west of the 577 Chopawamsic Fault in northern Virginia and the Hoods Mill rocks crop out directly west of 578 the Plummers Island Fault in northern Maryland (Fig. 2). Based on the occurrence of these 579 exotic rocks directly west of these two faults, we speculate that the faults might be 580 correlative.

581 In contrast to the other samples, interpretation of the provenance of the Loch Raven 582 Schist (sample 1213003) and the sedimentary xenolith in the Brandywine Blue Gneiss 583 (sample 314003) is not clear-cut. These units have late Mesoproterozoic to earliest

584	Neoproterozoic major age peaks, broadly similar to sediment derived from Ediacaran
585	Laurentia (Fig. 9). However, both samples also contain multiple detrital grains with U/Pb
586	ages between 670 and 580 Ma (Fig. S3; Table S4). The quantities of grains of this age are
587	intermediate between the numbers of such grains in the Storck quartzite and Hoods Mill
588	rocks and the quantities in samples with sediment derived from Ediacaran Laurentia. 24%
589	and 37% of the detrital zircon in the Storck quartzite and the Hoods Mill rocks,
590	respectively, have ages that overlap the range 670 to 580 Ma including uncertainties. These
591	proportions are 6% and 5% in the Loch Raven Schist and the sedimentary xenolith,
592	respectively, but fall to 1% and 0.4% in most central Piedmont quartzite of the same age
593	and central Blue Ridge late Ediacaran-Cambrian quartzite, respectively. Many central
594	Piedmont samples contain zero 670-580 Ma grains. Excluding the Storck quartzite, Hoods
595	Mill rocks, Loch Raven Schist, and the sedimentary xenolith, the central Piedmont sample
596	with the most grains in this age range is sample 909001 of the northern Sykesville
597	Formation, with 3% (Martin et al., 2015).
598	There are two plausible interpretations of the provenance of the detrital zircon in the
599	Loch Raven Schist and the sedimentary xenolith. One possibility is sediment derivation
600	from Ediacaran Laurentia. In this scenario, the ca. 670-580 Ma grains came from the
601	Goochland Terrane, western Newfoundland, or an unrecognized source in Ediacaran
602	Laurentia. The second option is sediment derivation from Gondwana. In this scenario, the
603	Loch Raven Schist and the sedimentary xenolith received more ca. 1200-900 Ma zircon
604	than is present in most other Gondwana-derived late Ediacaran-Cambrian sandstone due to
605	normal variations in the proportions of sediment transported by separate networks that
606	crossed large areas of a continent.

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607 A challenge to option 1 is that no other units unequivocally derived from Ediacaran 608 Laurentia have so many ca. 670-580 Ma detrital grains. Sykesville Formation sample 609 909001 contains 3% detrital zircon of this age (Martin et al., 2015), but this remains a 610 lower percentage than in the Loch Raven Schist or the sedimentary xenolith. Further, the 611 Laurentian provenance of the Sykesville Formation is not unimpeachable. One point in 612 favor of option 2 is that there are examples of Gondwanan late Ediacaran-Cambrian 613 sandstone with age spectra similar to those in the Loch Raven Schist and the sedimentary 614 xenolith. For example, the Puncoviscana Formation in northwestern Argentina contains 615 many detrital grains with ages between ca. 1200-900 Ma (Fig. 9), including sample PMXX-616 2 in Adams et al. (2011), which has a prominent ca. 1200 Ma age spike and no zircon 617 younger than 900 Ma. Schwartz and Gromet (2004) likewise found a metamorphosed 618 equivalent of the Puncoviscana Formation with a major age peak at ca. 1000 Ma; this peak 619 is larger than the ca. 600 Ma peak in this unit. Option 2 also has the advantage that other 620 units geographically near and geologically related to the Wilmington Complex contain 621 Gondwana-derived zircon. These units include the Chester Park Gneiss and rocks labeled 622 as the Wissahickon Formation (Bosbyshell et al., 2015). The latter unit is intercalated with 623 rocks of the Wilmington Complex. Based on the problem with option 1 and the merits of 624 option 2, we tentatively conclude that option 2 is the best interpretation. That is, the Loch 625 Raven Schist and the sedimentary xenolith might contain Gondwana-derived zircon, but 626 this conclusion is less certain for these units than for the Storck quartzite and the Hoods 627 Mill rocks. If the sedimentary xenolith in the Brandywine Blue Gneiss does contain 628 Gondwanan zircon, the Wilmington Complex island arc intruded sedimentary rocks that 629 were derived from Gondwana.

631 **7. CONCLUSIONS**

- 632 Our new U-Pb and Lu-Hf isotope data from spots in zircon support the following633 conclusions.
- Intrusion of the Steel Mountain Anorthosite, Disappointment Hill Tonalite, and
 Round Pond Granite in western Newfoundland occurred at 608±12, 600±8, and
- 636 590±9 Ma, respectively. Intrusion of the Steel Mountain Anorthosite is now
- 637 recognized to have been broadly coeval with intrusion of a granitic suite in western
- 638 Newfoundland at ca. 630-590.
- 639 2. The source of magma to the Steel Mountain Anorthosite, Disappointment Hill
 640 Tonalite, and Round Pond Granite was not solely depleted mantle.
- 641 3. The maximum depositional age for most of our central Appalachian Piedmont
 642 Province quartzite samples is late Ediacaran or Early Cambrian.
- 6434. Sediment in our samples from the type localities of the Piney Run Formation,
- 644 Pleasant Grove Schist, Prettyboy Schist, and Wissahickon Formation was derived
 645 from Ediacaran Laurentia.
- 5. Sediment in the Storck quartzite and the Hoods Mill rocks was derived from
 Gondwana. We confirm the presence of a ca. 630-610 Ma peak in detrital zircon
- ages in these units. The large proportions of detrital grains with ages between 670
- and 580 Ma in these units is not consistent with derivation from Ediacaran
- 650 Laurentia but is similar to the age spectra of detrital zircon in much late Ediacaran-
- 651 Cambrian sandstone with a Gondwanan provenance. Modern εHf values of some of
- the 670-580 Ma detrital zircon likewise are inconsistent with derivation from the
- 653 scarce Neoproterozoic plutons in Ediacaran Laurentia.

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654	6.	The provenance of the sediment in the Loch Raven Schist and a sedimentary
655		xenolith in the Brandywine Blue Gneiss is less certain. However, these units
656		contain more ca. 670-580 Ma detrital zircon than any late Ediacaran-Cambrian
657		sandstone unequivocally derived from Ediacaran Laurentia, so we tentatively
658		conclude that Gondwana was the source of zircon to these units. If correct, this
659		conclusion means that the Brandywine Blue Gneiss, which is part of the
660		Wilmington Complex island arc, intruded sedimentary rocks derived from
661		Gondwana.

662

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965	Fl	GURE CAPTIONS
966	1.	Generalized geologic map of the Appalachian Orogen. Modified from Hibbard et al.
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968	2.	Geologic map of part of the central Appalachian Piedmont Province. Modified from
969		Hibbard et al. (2006), Southworth et al. (2007), Hughes et al. (2014), and Martin et al.
970		(2015).
971	3.	Geologic map of part of western Newfoundland. Modified from Brem et al. (2006) and
972		Lin et al. (2013).
973	4.	Backscattered electron (left column) and cathodoluminescence (right column) images
974		of zircon from the three samples of western Newfoundland intrusive rocks. The circles
975		show the locations of 20 μ m-diameter laser spots that yielded the indicated $^{206}Pb/^{238}U$
976		ages.
977	5.	Concordia diagrams for U/Pb isotope analyses of spots in zircon from the three samples
978		of western Newfoundland intrusive rocks.
979	6.	Modern ϵ Hf values of spots in zircon from the three samples of western Newfoundland
980		intrusive rocks.
981	7.	Modern EHf values versus crystallization age for detrital zircon from the Storck
982		quartzite and the Hoods Mill rocks. There is no correlation between zircon
983		crystallization age and modern EHf value.
984	8.	Plots of concordant, non-xenocrystic U-Pb isotope analyses (vertical red bars) from
985		each sample of western Newfoundland intrusive rocks showing the weighted mean
986		²⁰⁶ Pb/ ²³⁸ U age (horizontal green bar). The reported uncertainty includes both
987		measurement and systematic errors.

988	9. Normalized relative probability plots of detrital zircon ages from our samples co	mpared
989	to detrital zircon ages from other late Ediacaran-Cambrian Laurentian and peri-	
990	Gondwanan sandstone. Data were normalized so that the area under each curve	is the
991	same. Data sources and processing details are given in Table S5 and the ages an	ıd
992	corresponding errors are in Table S6. The sample numbers for the Piney Run	
993	Formation, Pleasant Grove Schist, Prettyboy Schist, and Wissahickon Formation	n are
994	114002, 114003, 314001, and 114005, respectively.	
995	10. Normalized relative probability plots of detrital zircon ages from the Storck quar	rtzite
996	and the Hoods Mill rocks. The detrital zircon age spectra from our new samples	are
997	nearly identical to the previously-published spectra from these units. Data were	
998	normalized so that the area under each curve is the same. Data processing is des	scribed
999	in Table S5.	
1000	11. Comparison of present-day εHf values of 670-580 Ma detrital zircon from the St	torck
1000	11. Comparison of present-day EHf values of 670-580 Ma detrital zircon from the St	erozoic
1000 1001	11. Comparison of present-day εHf values of 670-580 Ma detrital zircon from the St quartzite and the Hoods Mill rocks with modern εHf values of zircon in Neoprot	erozoic εHf
1000 1001 1002	11. Comparison of present-day εHf values of 670-580 Ma detrital zircon from the St quartzite and the Hoods Mill rocks with modern εHf values of zircon in Neoprot plutons in the Goochland Terrane and western Newfoundland. The present-day	erozoic εHf e zircon
1000 1001 1002 1003	11. Comparison of present-day EHf values of 670-580 Ma detrital zircon from the St quartzite and the Hoods Mill rocks with modern EHf values of zircon in Neoprot plutons in the Goochland Terrane and western Newfoundland. The present-day values of the detrital zircon extend to values both less and more negative than the	erozoic εHf e zircon s that
1000 1001 1002 1003 1004	11. Comparison of present-day εHf values of 670-580 Ma detrital zircon from the St quartzite and the Hoods Mill rocks with modern εHf values of zircon in Neoprot plutons in the Goochland Terrane and western Newfoundland. The present-day values of the detrital zircon extend to values both less and more negative than th in the plutons, outside uncertainty. The plot shows only detrital grains with ages	erozoic εHf e zircon s that 18 in
1000 1001 1002 1003 1004 1005	11. Comparison of present-day EHf values of 670-580 Ma detrital zircon from the St quartzite and the Hoods Mill rocks with modern EHf values of zircon in Neoprot plutons in the Goochland Terrane and western Newfoundland. The present-day values of the detrital zircon extend to values both less and more negative than th in the plutons, outside uncertainty. The plot shows only detrital grains with ages overlap the age range 670-580 Ma within uncertainty. These are grains 131 to 2	erozoic εHf e zircon s that 18 in values
1000 1001 1002 1003 1004 1005 1006	11. Comparison of present-day ɛHf values of 670-580 Ma detrital zircon from the St quartzite and the Hoods Mill rocks with modern ɛHf values of zircon in Neoprot plutons in the Goochland Terrane and western Newfoundland. The present-day values of the detrital zircon extend to values both less and more negative than th in the plutons, outside uncertainty. The plot shows only detrital grains with ages overlap the age range 670-580 Ma within uncertainty. These are grains 131 to 2 sample 314002 and 69 to 266 in sample 1213002 (Table S3). The range of ɛHf v	erozoic εHf e zircon s that 18 in values plutons
1000 1001 1002 1003 1004 1005 1006 1007	11. Comparison of present-day ɛHf values of 670-580 Ma detrital zircon from the St quartzite and the Hoods Mill rocks with modern ɛHf values of zircon in Neoprot plutons in the Goochland Terrane and western Newfoundland. The present-day values of the detrital zircon extend to values both less and more negative than th in the plutons, outside uncertainty. The plot shows only detrital grains with ages overlap the age range 670-580 Ma within uncertainty. These are grains 131 to 2 sample 314002 and 69 to 266 in sample 1213002 (Table S3). The range of ɛHf v for the zircon in the plutons includes uncertainties. The bands of values for the plutons of values for the plutons of values of values of values of values of the plutons of values of values of values of values of the plutons of values of values of values of values of values of the plutons of values o	erozoic εHf e zircon s that 18 in values plutons
1000 1001 1002 1003 1004 1005 1006 1007 1008	11. Comparison of present-day EHf values of 670-580 Ma detrital zircon from the St quartzite and the Hoods Mill rocks with modern EHf values of zircon in Neoprot plutons in the Goochland Terrane and western Newfoundland. The present-day values of the detrital zircon extend to values both less and more negative than th in the plutons, outside uncertainty. The plot shows only detrital grains with ages overlap the age range 670-580 Ma within uncertainty. These are grains 131 to 2 sample 314002 and 69 to 266 in sample 1213002 (Table S3). The range of EHf for the zircon in the plutons includes uncertainties. The bands of values for the plutons of the plutons are shown stretching horizontally across the entire diagram to ease viewing, but	erozoic εHf e zircon s that 18 in values plutons the

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- 1013 TABLES
- 1014 1. Central Appalachian Piedmont meta-sandstone sample summary.
- 1015 2. Western Newfoundland igneous rocks sample summary.
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1017 ELECTRONIC SUPPLEMENT

- 1018 FIGURES
- 1019 Figure S1: Photomicrographs of thin sections of each sample. The scale is the same for
- 1020 each image. In (B), all minerals in the field of view are plagioclase feldspar.
- 1021 Abbreviations: amp-amphibole, bt-biotite, chl-chlorite, fsp-feldspar, grt-garnet, ms-
- 1022 muscovite, q-quartz.
- 1023
- 1024 Figure S2: Measured ¹⁷⁶Hf/¹⁷⁷Hf ratios of spots in the seven zircon standards acquired
- 1025 throughout the Lu-Hf isotope analysis session. The horizontal green bar indicates the
- 1026 weighted mean.
- 1027
- 1028 Figure S3: Histograms and relative probability plots of the U/Pb isotopic ages of detrital
- 1029 zircon from each central Appalachian Piedmont meta-sandstone sample. Best age is the
- 1030 ²⁰⁶Pb/²³⁸U date for grains with a ²⁰⁶Pb/²⁰⁷Pb date younger than 900 Ma and ²⁰⁶Pb/²⁰⁷Pb date
- 1031 for older grains.
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- 1033 **TABLES**
- 1034 Table S1: Western Newfoundland igneous zircon spot U-Pb isotope data.
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- 1036 Table S2: Summary of U/Pb ages of standard zircon analyzed along with zircon from the
- 1037 samples.
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- 1039 Table S3: Zircon spot Lu-Hf isotope data.
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- 1041 Table S4: Central Appalachian Piedmont detrital zircon spot U-Pb isotope data.
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- 1043 Table S5: Data sources and processing details for Figure 9.
- 1044
- 1045 Table S6: Data used to make Figure 9.



Sample number	Formation								
number			Latitude	Longitude	U/Pb analyses	age (Ma),	analyses used to determine	Number of	
number	name	Informal name	°N	°W	kept	this study	max depo age (Ma)	Hf analyses	Reference
Central Virgini	a								
314002 N	1ine Run Complex I	Storck quartzite	38.41843	77.63513	269	540	499, 513	40	Hughes et al., 2014
Northern Mary	/land								
1213002	Mather Gorge	Hoods Mill rocks ^b	39.35212	77.01553	252	550	521, 529	22	Martin et al., 2015
1213003ª	Loch Raven Schist	-	39.50111	76.62347	169	530	507, 510	not analyzed	Crowley, 1976
114002 ^a	Piney Run	-	39.53998	76.77674	167	1060	1033, 1036, 1037	not analyzed	Crowley, 1976
114003 ^a Pl	easant Grove Schist	-	39.54285	76.80614	273	1030	955, 969, 988, 989	not analyzed	Crowley, 1976
314001 ^a	Prettyboy Schist	-	39.61900	76.70773	287	570	536, 561	not analyzed	Crowley, 1976
Northern Dela	ware								
314003 Bra	ndywine Blue Gneiss	sedimentary xenolith ^b	39.77823	75.50748	208	540	515, 515	not analyzed	Plank et al., 2000
Southeastern I	Pennsylvania								
114005ª	Wissahickon	-	40.05671	75.21819	209	570	537, 542	not analyzed	Bascom, 1905
				Totals:	1834	analyses fro	m 8 samples	62 a	nalyses from 2 sampl

TABLE 1. SUMMARY OF META-SANDSTONE SAMPLES FROM THE CENTRAL APPALACHIAN PIEDMONT

^bInformal name assigned in this study.

TABLE 2. SUMMARY OF WESTERN NEWFOUNDLAND IGNEOUS SAMPLES

Sample number	Unit name	Latitude °N	Longitude °W	Number of zircon Hf analyses	Igneous crystallization age (Ma)	Igneous crystallization age reference
1013002	Disappointment Hill Tonalite	48.58884	58.14222	15	606 ± 2	Lin et al., 2013
1013005	Steel Mountain Anorthosite	48.55948	58.15767	6	608 ± 12	this study
1013006	Round Pond Granite	49.06184	57.70439	9	590 ± 9	this study
			Total:	30	analyses from 3 samples	

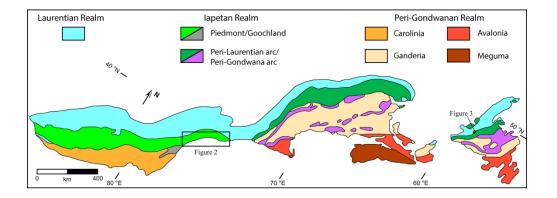


Figure 1 (Martin and Bosbyshell)

Generalized geologic map of the Appalachian Orogen. Modified from Hibbard et al. (2006).

231x119mm (300 x 300 DPI)

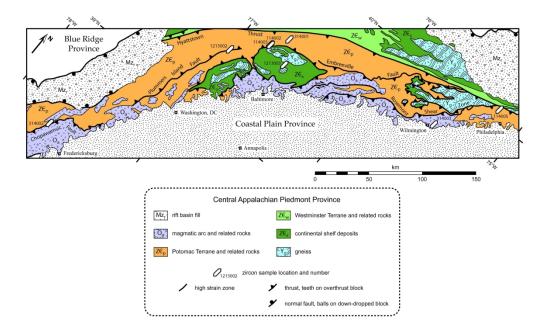
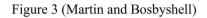


Figure 2 (Martin and Bosbyshell)

Geologic map of part of the central Appalachian Piedmont Province. Modified from Hibbard et al. (2006), Southworth et al. (2007), Hughes et al. (2014), and Martin et al. (2015).

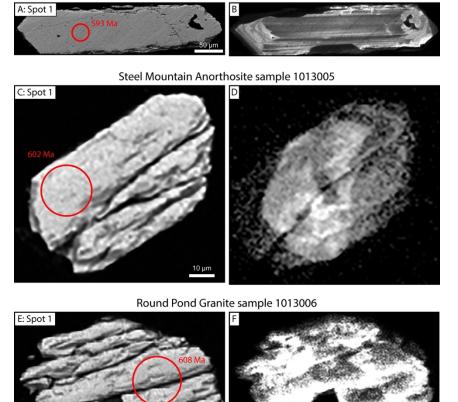
219x163mm (300 x 300 DPI)





Geologic map of part of western Newfoundland. Modified from Brem et al. (2006) and Lin et al. (2013).

94x183mm (300 x 300 DPI)

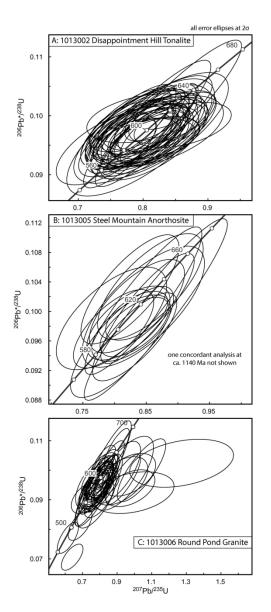


Disappointment Hill Tonalite sample 1013002

Figure 4 (Martin and Bosbyshell)

Backscattered electron (left column) and cathodoluminescence (right column) images of zircon from the three samples of western Newfoundland intrusive rocks. The circles show the locations of 20 μ m-diameter laser spots that yielded the indicated 206Pb/238U ages.

154x217mm (300 x 300 DPI)





Concordia diagrams for U/Pb isotope analyses of spots in zircon from the three samples of western Newfoundland intrusive rocks.

101x253mm (300 x 300 DPI)

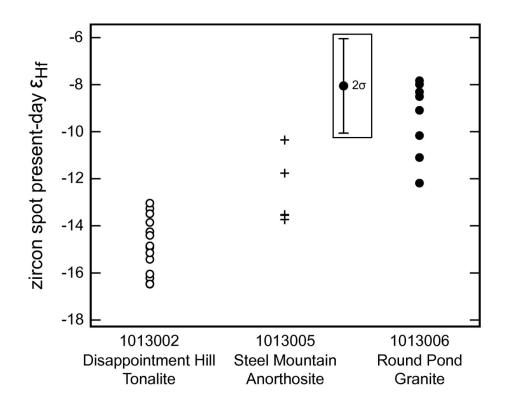
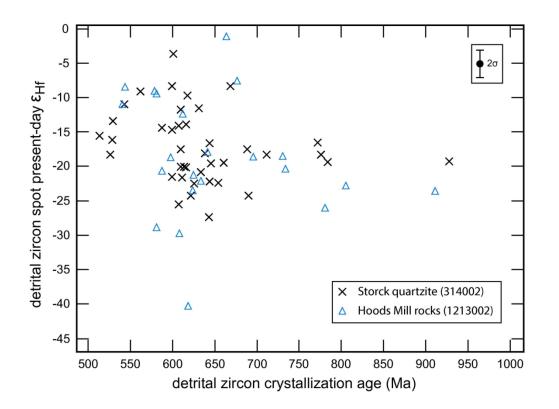
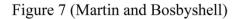


Figure 6 (Martin and Bosbyshell)

Modern ɛHf values of spots in zircon from the three samples of western Newfoundland intrusive rocks.

94x123mm (300 x 300 DPI)





Modern ɛHf values versus crystallization age for detrital zircon from the Storck quartzite and the Hoods Mill rocks. There is no correlation between zircon crystallization age and modern ɛHf value.

131x157mm (300 x 300 DPI)

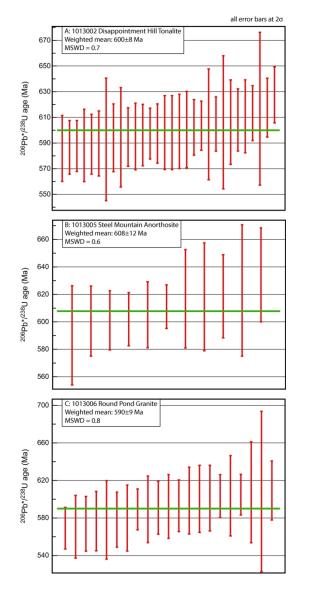


Figure 8 (Martin and Bosbyshell)

Plots of concordant, non-xenocrystic U-Pb isotope analyses (vertical red bars) from each sample of western Newfoundland intrusive rocks showing the weighted mean 206Pb/238U age (horizontal green bar). The reported uncertainty includes both measurement and systematic errors.

108x241mm (300 x 300 DPI)

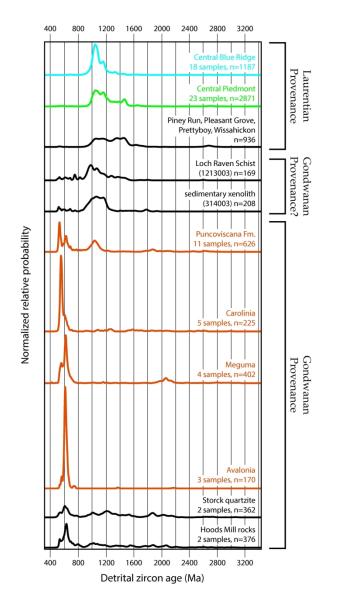


Figure 9 (Martin and Bosbyshell)

Normalized relative probability plots of detrital zircon ages from our samples compared to detrital zircon ages from other late Ediacaran-Cambrian Laurentian and peri-Gondwanan sandstone. Data were normalized so that the area under each curve is the same. Data sources and processing details are given in Table S5 and the ages and corresponding errors are in Table S6. The sample numbers for the Piney Run Formation, Pleasant Grove Schist, Prettyboy Schist, and Wissahickon Formation are 114002, 114003, 314001, and 114005, respectively.

115x236mm (300 x 300 DPI)

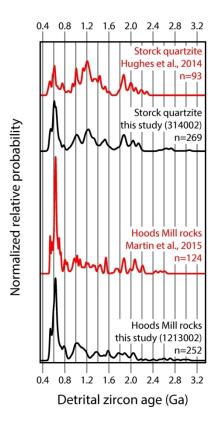


Figure 10 (Martin and Bosbyshell)

Normalized relative probability plots of detrital zircon ages from the Storck quartzite and the Hoods Mill rocks. The detrital zircon age spectra from our new samples are nearly identical to the previously-published spectra from these units. Data were normalized so that the area under each curve is the same. Data processing is described in Table S5.

67x184mm (300 x 300 DPI)

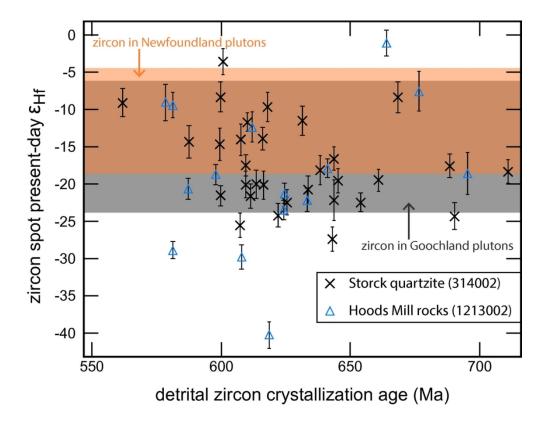


Figure 11 (Martin and Bosbyshell)

Comparison of present-day εHf values of 670-580 Ma detrital zircon from the Storck quartzite and the Hoods Mill rocks with modern εHf values of zircon in Neoproterozoic plutons in the Goochland Terrane and western Newfoundland. The present-day εHf values of the detrital zircon extend to values both less and more negative than the zircon in the plutons, outside uncertainty. The plot shows only detrital grains with ages that overlap the age range 670-580 Ma within uncertainty. These are grains 131 to 218 in sample 314002 and 69 to 266 in sample 1213002 (Table S3). The range of εHf values for the zircon in the plutons includes uncertainties. The bands of values for the plutons are shown stretching horizontally across the entire diagram to ease viewing, but the actual range of intrusion ages is 630-590 and 660-580 Ma for the western Newfoundland and Goochland plutons, respectively. εHf values for the Goochland Terrane zircon from Martin et al. (2018).

108x118mm (300 x 300 DPI)