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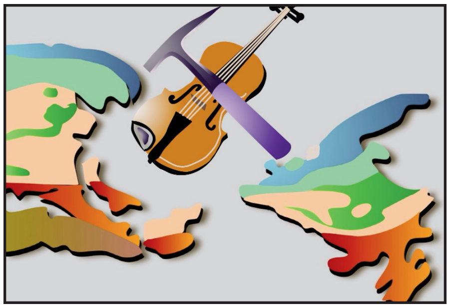

## Crustal Evolution of the Northeast Laurentian Margin and the Peri-Gondwanan Microcontinent Ganderia Prior to and During Closure of the Iapetus Ocean: Detrital Zircon U–Pb and Hf Isotope Evidence from Newfoundland

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### SUMMARY

Detrital zircon populations in sedimentary rocks from the Laurentian margin and the accreted microcontinent Ganderia on both sides of the main Iapetus suture (Red Indian Line) in central Newfoundland have been studied by combined U–Pb and Lu–Hf isotope analyses. Variation in $\varepsilon\mathrm{Hf}_{(t)}$ values with age of zircon populations of distal provenance (>900 Ma) reflect the crustal evolution within the source continents: in zircon derived from Laurentia, episodes of juvenile magma production in the source could be detected at 1.00 – 1.65 and 2.55 – 3.00 Ga, and mixing of juvenile and recycled crust in continental magmatic arcs occurred at 0.95 – 1.40, 1.45 – 1.60, 1.65 – 2.05 and 2.55 – 2.75 Ga. These ages are consistent with the crustal history of northeastern Laurentia. Similarly, zircon of distal provenance from Ganderia reveals times of juvenile magma production in the source at 0.70 – 0.90, 1.40 – 1.75, 1.85 – 2.40 and 2.7 – 3.5 Ga, and episodes of mixing juvenile and recycled crust at 0.95 – 1.35, 1.45 – 1.60, 1.70 – 2.15 and 2.6 – 2.8 Ga. These data reflect the crustal evolution in the present northern part of Amazonia, its likely source craton.

The evolution of magmatic arcs at the margins of both continents can be studied in a similar way using detrital zircon having a proximal provenance (<900 Ma). In contrast to the Laurentian margin, Ganderia is characterized by development of Neoproterozoic – Cambrian continental arcs (ca. 500 – 670 Ma) that were built on the margin of Gondwana. $\varepsilon\mathrm{Hf}_{(t)}$ values indicate recycling of Neo- and Mesoproterozoic crust. During and following accretion of the various elements of Ganderia to Laurentia, the syn-tectonic Late Ordovician to Silurian sedimentary rocks deposited on the upper plate (composite Laurentia) continued showing only detritus derived from Laurentia. These sedimentary rocks contain detrital zircon from Iapetan juvenile, continental and successor arcs that were active between ca. 440 and 550 Ma, and from continuing magmatic activity until 423 Ma. Arrival of the first Laurentian detritus at the outermost part of Ganderia indicates that the Iapetus ocean was closed at ca. 452 Ma. The magmatic arcs along the former Laurentian margin in Newfoundland evolved differently. In the northwestern part, $\varepsilon\mathrm{Hf}_{(t)}$ values point to recycling of Mesoproterozoic and Paleoproterozoic crust. In the southwest, $\varepsilon\mathrm{Hf}_{(t)}$ values indicate addition of juvenile crust, recycling of Mesoproterozoic crust and mixing with juvenile magma.

### SOMMAIRE

Les populations de zircons détritiques

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des roches sédimentaires issus de la marge laurentienne et du microcontinent d'accrétion de Ganderia, des deux côtés de la principale suture Iapetus (linéation de Red Indian) dans le centre de Terre-Neuve, ont été étudiés par analyses combinées U-Pb et Lu-Hf. Les variations des valeurs εHf$_{(t)}$ en fonction de l'âge des populations de zircons distaux (>900 Ma) reflètent l'évolution de la croûte des continents sources : les zircons de Laurentie ont permis de détecter des épisodes magmatiques juvéniles dans la source entre 1,00 - 1,5, et 2,55 - 3,00 Ga, ainsi que des épisodes de mélange de croûte juvénile avec des croûtes d'arcs magmatiques continentaux recyclés entre 0,95 – 1,40, 1,45 – 1,60, 1,65 – 2,05, et 2,55 – 2,75 Ga. Ces datations correspondent bien à l'histoire de la croûte de la portion nord-est de la Laurentie. De même, le zircon distal de Ganderia révèle des épisodes de production de magmas juvéniles dans la source entre 0,70 - 0,90, 1,40 - 1,75, 1,85 - 2,40, et 2,7 - 3,5 Ga, ainsi que des épisodes de mélanges de matériaux juvéniles et de croûtes recyclés entre 0,95 - 1,35, 1,45 - 1,60, 1,70 - 2,15, et 2,6 - 2,8 Ga. Ces données reflètent l'évolution de la croûte dans la portion nord actuelle de l'Amazonie, son craton source probable.

L'évolution des arcs magmatiques à la marge de ces deux continents peuvent être étudiées de la même manière en utilisant le zircon détritique proximal (<900 Ma). Contrairement à la marge laurentienne, celle de Ganderia est caractérisée par le développement d'arcs continentaux Néoprotéozoïque-Cambrien (env. 500 – 670 Ma) qui se sont constitués à la marge du Gondvana. Les valeurs de εHf$_{(t)}$ indiquent un recyclage de la croûte au Néoprotérozoïque et au Mésoprotérozoïque. Durant et après l'accrétion des divers éléments de Ganderia et de la Laurentie, les roches sédimentaires syntectoniques de la fin de l'Ordovicien et du Silurien qui se sont déposées sur la portion supérieure de la plaque (Laurentie composite) ne montrent toujours que des débris provenant de la Laurentie. Ces roches sédimentaires renferment des zircons détritiques juvéniles iapétiques, et d'arcs continentaux et d'arcs subséquents, qui ont été actifs entre (env. 440 et 550 Ma) et d'une activité magmatique continue jusqu'à 423 Ma. L'apport des premiers débris à la marge extrême de Ganderia indique que l'océan s'est fermée il y a env. 452 Ma. Les arcs magmatiques le long de l'ancienne marge laurentienne à Terre-Neuve ont évolué différemment. Dans la portion nord-ouest, les valeurs de εHf$_{(t)}$ indiquent un recyclage de la croûte au Mésoprotérozoïque et au Paléoprotérozoïque. Dans la portion sud-ouest, les valeurs de εHf$_{(t)}$ indiquent l'ajout d'une croûte juvénile, un recyclage de la croûte mésoprotérozoïque et un mélange avec un magma juvénile.

## INTRODUCTION

The Newfoundland Appalachians expose one of the most complete cross-sections through the Appalachian orogen, including several Lower Paleozoic magmatic arcs where juvenile mantle material was added, and microcontinents where continental crust was recycled (e.g. van Staal 2007; van Staal and Zagorevski 2012; Fig. 1). Hence this region provides an ideal natural laboratory for the study of processes related to crustal evolution, recycling and growth. Hf isotope composition of detrital zircon coupled with U–Pb dating represents a powerful tool to assess continental growth and recycling processes. Fractionation of Hf, which occurs between zircon and co-precipitating phases during magma generation (Vervoort et al. 1999; Kinny and Maas 2003; Scherer et al. 2007), preserves a record of the Hf isotope ratio of the magma and the crust where the magma originated, and allows Hf isotope composition to be utilized as a geochemical fingerprint of the magmatic source. In particular, times of juvenile material addition to the continental crust and crustal recycling can be recognized. Continental magmatic arcs represent a major site of juvenile additions and recycled continental crust, and the two may be mixed in varying proportions (Lucassen et al. 2004; Clift and Vannucchi 2004; Franz et al. 2006; Scholl and Huene 2007). The degree of juvenile addition and mixing with recycled crust is strongly related to the active continental margin setting (extensional vs. compressional). For instance, a high degree of continental recycling commonly occurs in overthickened compressional active continental margins (DeCelles et al. 2009). However, net crustal growth by juvenile magma addition may be balanced by continental destruction, e.g. by subduction erosion (Clift and Vannucchi 2004; Scholl and von Huene 2007).

Sm–Nd whole rock isotopy and detrital zircon U–Pb age spectra of both magmatic arcs and sedimentary basins in Newfoundland have been investigated to decipher the linkages to source terranes (e.g. Kerr et al. 1995, Whalen et al. 2006; Zagorevski et al. 2008, 2009; Waldron et al. 2012). However, Sm–Nd isotopes provide only a rough time-integrated average of the source terranes, limiting their utility for understanding crustal evolution. Detrital zircon U–Pb age spectra of sedimentary basins provide the age of the sources but do not generally constrain the evolution of source terranes. Hf isotope signatures of detrital zircon potentially provide a more reliable tool to study crustal evolution, but they have not yet been applied to study the evolution of accreted terranes in the Newfoundland Appalachians.

In this paper, we utilize a combination of U–Pb dating with Hf isotope compositions of detrital zircons to address crustal evolution, recycling and growth in the Newfoundland Appalachians, particularly in order to test and refine existing models. We provide Hf isotopic fingerprints for the Laurentian margin, the peri-Laurentian Dashwoods microcontinent, and the Gondwana-derived Ganderia microcontinent.

Detrital zircon from distal hinterland sources within a source continent mirrors the crustal evolution of that continent; i.e., crustal growth and recycling processes (Willner et al. 2008, 2013; Zeh et al. 2008; Zeh and Gerdes 2010; Collins et al. 2011; Abati et al. 2012). These relationships can be deciphered by comparing the Hf isotopic signature of detrital zircon with otherwise known processes of crustal evolution in the potential source continent. Patterns derived in this way can be used as a paleogeographic tool to identify the provenance of entire microcontinents or specific sedimentary rocks (overstep sequences) in continental collision zones.

Detrital zircon from proximal

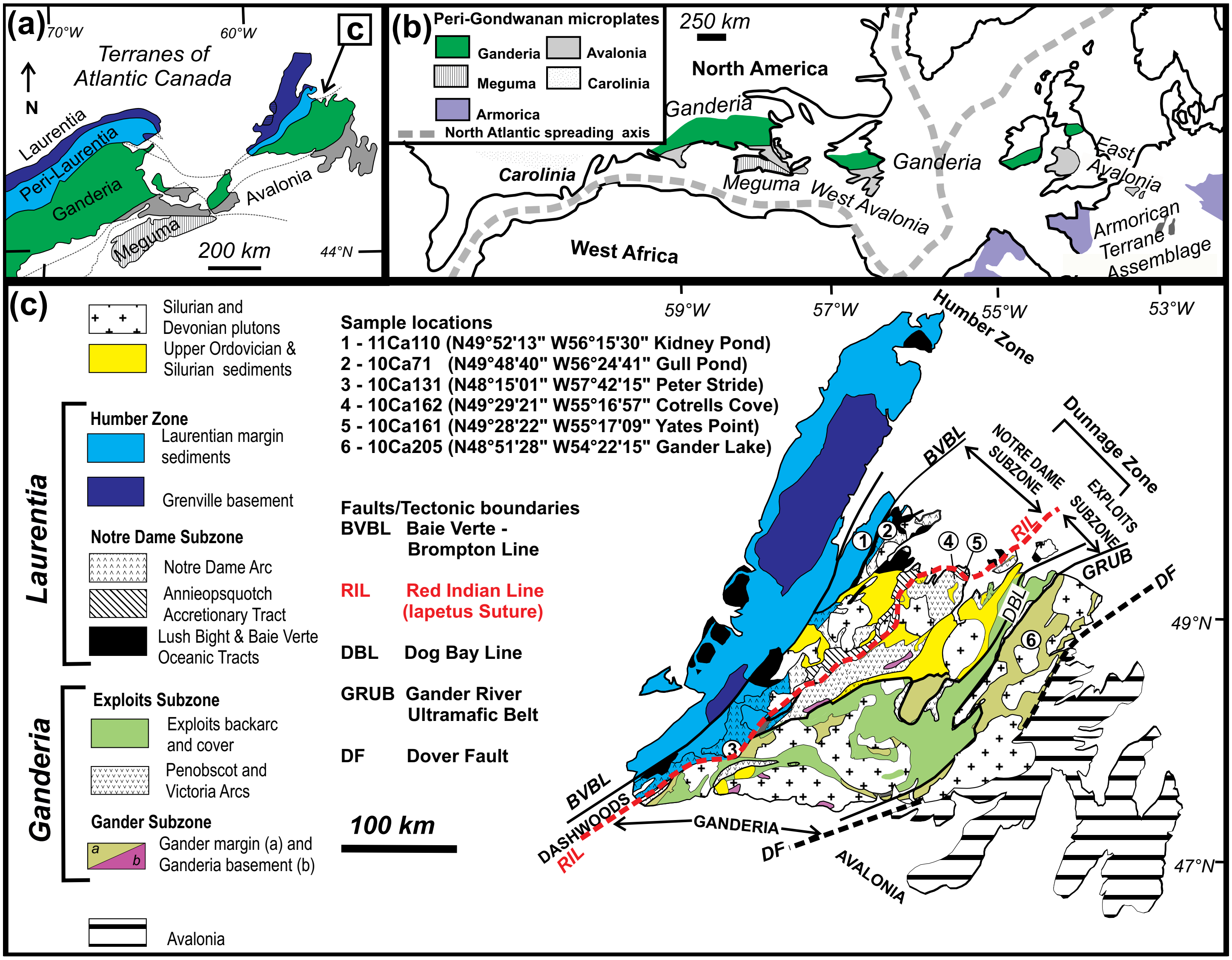

**Figure 1.** (a) Terranes in the northern Appalachians in eastern Canada. (b) Overview map showing the distribution of Ganderia within the assemblage of peri-Gondwanan microcontinents in the Appalachian – Caledonide orogen (redrawn after Pollock et al. 2009). (c) Geological map of Newfoundland showing the terrane boundaries (redrawn after van Staal and Zagorevski 2012 and Waldron et al. 2012). Further details of sample locations are given in Supplementary Table E1 in the electronic data base.

sources will be utilized to identify the provenance of pre-, syn- and postcollisional sedimentary rocks along the Iapetus suture. In these sedimentary rocks the youngest detrital zircon provides information about the timing and nature of the closure of the Iapetus Ocean, whereas the Hf isotopic composition of detrital zircon from proximal continental magmatic arcs is used to assess the amount of juvenile vs. recycled crust, and the nature of the crust underlying the arcs.

## GEOLOGICAL SETTING

The fundamental regional crustal blocks in Newfoundland were recognized by Williams (1979). Based on significant contrasts in lithology, stratigraphy, fauna, metamorphic evolution, geophysical signatures and magmatism of Lower Paleozoic and older rocks, four tectonostratigraphic zones were defined, namely, from west to east, the Humber, Dunnage, Gander and Avalon zones (Fig. 1). Williams et al. (1988) demonstrated that the 'oceanic' Dunnage zone is composite and subdivided it into a western (Notre Dame) subzone having lithological and faunal affinities with Laurentia, and an eastern (Exploits) subzone with a strong linkage to the Gander zone. These two subzones are separated by the Red Indian Line (Williams et al. 1988; Zagorevski et al. 2008), which in Newfoundland represents the main suture where the Iapetus Ocean closed. The substrate to most of the Notre Dame subzone later became known as the peri-Laurentian Dashwoods microcontinent (Waldron and van Staal 2001; Hibbard et al. 2007, van Staal and Barr 2012), which is separated from the Laurentian margin by the ophiolitic remnants of the Baie Verte oceanic tract exposed immediately east of the Baie Verte – Brompton Line. The Exploits subzone, originally presumed to be oceanic, is demonstrably built on Neoproterozoic arc basement (Rogers et al. 2006; Zagorevski et al. 2010) similar to the Gander Zone. Tectonic linkages (Colman-Sadd et al. 1992) suggest that the substrate to the Exploits subzone and the Gander zone constitute a fragmented microcontinent (Ganderia) having a west Gondwanan (Amazonian) provenance (e.g. van Staal et al. 1996, 2012; Rogers et al. 2006; Hibbard et al. 2007). East of the Ganderia microcontinent, another Gondwana-derived microcontinent – Avalonia – was added to the Appalachian terrane collage. The boundary between Ganderia and Avalonia is marked by the Dover – Hermitage Bay Fault in Newfoundland (van Staal 2005, 2007) and by other faults in Maritime Canada and New England (van Staal and Barr 2012).

The Humber zone consists of Grenvillian crystalline basement unconformably overlain by a cover sequence containing Late Neoproterozoic to Early Ordovician rift, continental margin and foreland basin units (Cawood et al. 2001). As such, it represents the Laurentian margin prior to the assembly of the northern Appalachians. This margin was subjected to a ~60 my period of Neoproterozoic extension and rifting (615 – 550 Ma), which opened the Iapetus Ocean and the Taconic seaway (Cawood et al. 2001; van Staal et al. 2013). The peri-Laurentian Dashwoods microcontinent formed as an extensional allochthon during hyperextension of the Laurentian margin and was separated from the margin by the Taconic seaway (van Staal et al. 2007, 2013). The Dashwoods microcontinent therefore constitutes a displaced fragment of Laurentia. During the Early Ordovician (480 – 455 Ma), the Taconic seaway was closed and oceanic rocks of the Notre Dame subzone (Baie Verte oceanic tract) were emplaced onto the Humber margin and progressively imbricated toward the foreland (Waldron and van Staal 2001; van Staal et al. 2007).

The Notre Dame and Exploits subzones comprise Iapetan realm arc and backarc complexes that were deformed and sequentially added to the composite Laurentian margin during closure of the Iapetus Ocean and its marginal basins (Zagorevski et al. 2010). The Notre Dame subzone is structurally largely underlain by Mesoproterozoic and older Laurentian crust of the Dashwoods microcontinent (van der Velden et al. 2004; van Staal et al. 2007), whereas most of the Exploit subzone is underlain by a Neoproterozoic – Early Cambrian arc basement of the Ganderia microcontinent (Zagorevski et al. 2010). In addition to basement characteristics, the Neoproterozoic – Early Paleozoic evolution differs significantly in the respective subzones. Most notably, Upper Ordovician – Lower Silurian terrestrial rocks unconformably overlie Ediacaran to Middle Ordovician rocks in the Notre Dame subzone (Williams et al. 1988; van Staal et al. 1998), whereas there is a conformable Ordovician – Lower Silurian marine sequence in the Exploits subzone. These differences arise from the contrasting tectonic histories of these subzones and their basements. The Middle Ordovician and older faunas have Laurentian affinities in the Notre Dame subzone, but are either Gondwanan or insular in the Exploits subzone (Harper et al. 2009), indicating derivation from the southern reaches of the Iapetus Ocean.

The Gander Zone is characterized by the Gander Group, a Cambrian to Tremadocian psammite and pelite sequence that conformably overlies Neoproterozoic to Early Cambrian magmatic arc rocks. The Gander Group and its correlatives in New England, New Brunswick and the British Isles (van Staal et al. 1996; Fig. 1b) are a distinctive cover sequence to the Ganderian microcontinental basement, which also underlies most of the Exploits Subzone (van der Velden et al. 2004; van Staal et al. 2012). The history of the Ganderia microcontinent, which is the first Gondwanan terrane to arrive at the Laurentian margin, is complex. Initially, Ganderia rifted off Gondwana prior to ca. 510 Ma, coeval with formation of the oldest phases of the extensional Penobscot magmatic arc-backarc system. The Late Cambrian Penobscot arc rifted, opening a narrow backarc basin between the active Penobscot arc and the trailing, passive margin of Ganderia. Closure of this backarc basin and obduction of the Penobscot backarc ophiolites onto the passive margin of Ganderia between 485 and 478 Ma (Penobscot orogeny; Colman-Sadd et al. 1992; Zagorevski et al. 2010) brought the fragments of Ganderia back together, but not for long.

Shortly following obduction, a new arc (the Popelogan – Victoria arc, ca. 476 – 474 Ma) was erected above the remnants of the Penobscot arc-back arc system and above an eastward-directed subduction zone. This arc was also extensional, and rifted approximately along the Penobscot suture, opening the Tetagouche – Exploits back-arc basin in its wake. It caused progressive separation of the Popelogan – Victoria arc and its Ganderian basement from the trailing edge of Ganderia and its new passive margin. The Popelogan – Victoria arc was active until ca. 455 Ma, when it first arrived as the outermost part of Ganderia at the Iapetus suture, resulting in arc-arc collision (Zagorevski et al. 2008). The Tetagouche – Exploits back-arc basin was closed during Early Silurian time, once again bringing the various fragments of Ganderia back together (440 – 430 Ma; van Staal 1994; van Staal et al. 2003; Pollock et al. 2007). The associated principal suture in Newfoundland is the Dog Bay Line (Williams et al. 1993; van Staal et al. 2009, 2014).

Outboard of the Gander Zone and to the east of the Dover Fault, the Avalon Zone is regarded as the type area of the peri-Gondwanan Avalonia microcontinent. This microcontinent, which also extended towards the British Isles and further south to Nova Scotia, New Brunswick and New England, mainly comprises a collage of arc-related Neoproterozoic – Early Cambrian volcano-sedimentary belts unconformably overlain by Middle Cambrian – Early Ordovician shale-rich platformal sedimentary rocks. Collision of Avalonia and subsequent deformation and metamorphism occurred during the Acadian orogeny between about 415 and 390 Ma (Murphy et al. 1999; van Staal et al. 2009, 2014).

## ANALYTICAL METHODS

### Cathodoluminescence Imaging

A cathodoluminescence (CL) detector mounted in a CAMECA SX100 microprobe at Universität Stuttgart was used to select homogeneous growth zones in individual zircon grains for analysis and to view internal zonation patterns.

Most analyzed zircon grains show characteristics of magmatic origin including well-developed concentric, narrowly spaced oscillatory growth zoning, sectoral zoning, resorption phenomena and very rarely age discordant (xenocrystic) cores. Irregular heterogeneous internal structures may be partly attributed to metamorphic zircon, which usually represents late or postmagmatic recrystallization phenomena in high temperature terranes. By the term 'irregular heterogenous fabric' we mean heterogeneous patchy zoning with sharp and curved sectoral boundaries, zones of dissolution and regrowth propagating through the zircon crystal, convolute zoning, and blurred former oscillatory zoning (Vavra et al. 1999; Corfu et al. 2003; Harley et al. 2007). Such features suggest the possibility of high temperature metamorphic processes in the source area, similar to unzoned crystals and thin overgrowth rims. However, metamorphic zircon cannot unambiguously be distinguished from magmatic zircon within a detrital zircon population. A low Th/U ratio of <0.1 is often used to identify metamorphic zircon (Rubatto 2002). But this criterion is also ambiguous: only 3 analyzed grains have such low ratios and only 14 grains have Th/U <0.2. Most of these grains show oscillatory growth zoning.

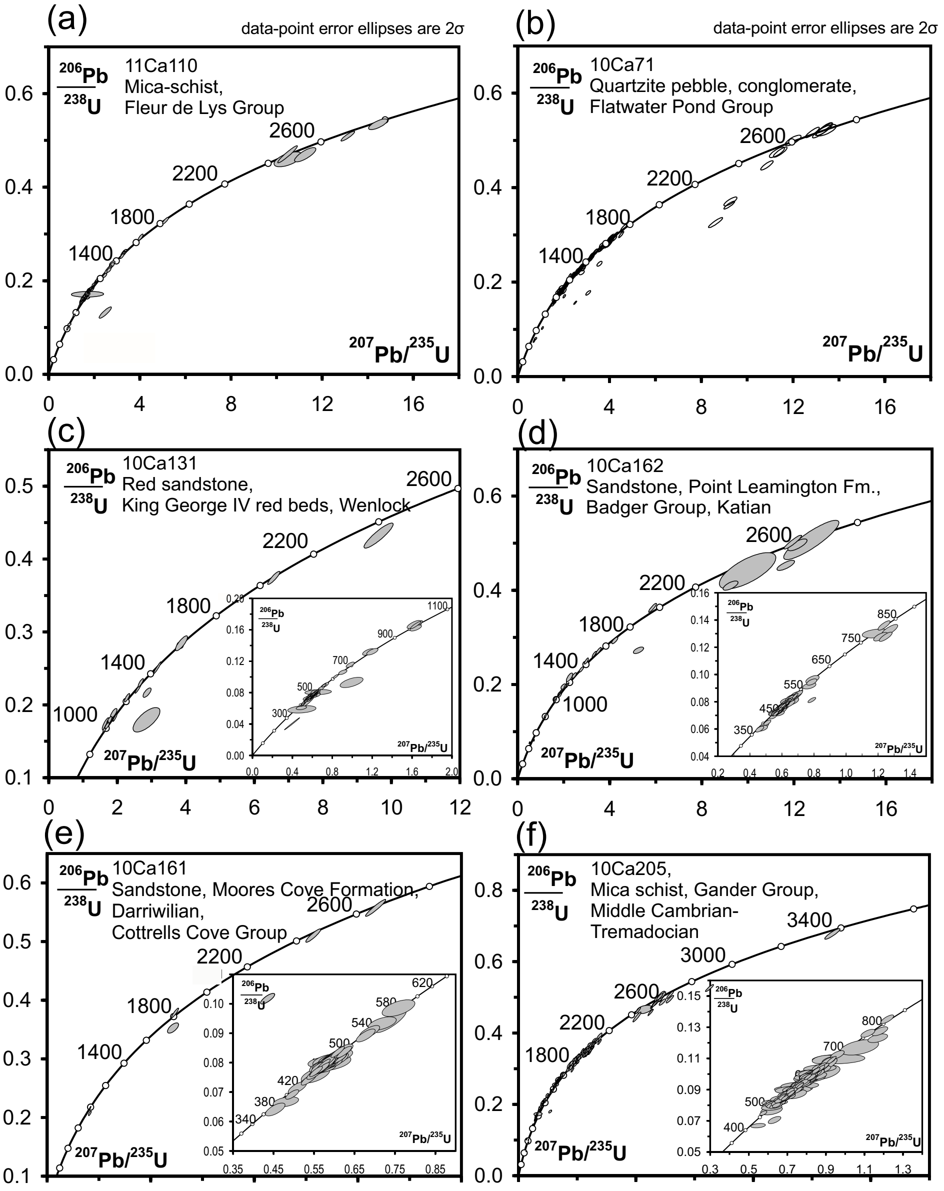

**Figure 2.** U–Pb concordia plots for samples shown in Figures 3 and 4. Data are available in Supplementary Table E1 in the electronic data base.

## U–Pb Dating

Between 83 and 301 detrital grains from each of six sandstone samples were dated. A total of 998 U–Pb ages were obtained by LA–ICP–MS techniques at Goethe University Frankfurt (GUF) using a Thermo-Scientific Element II sector field ICP–MS coupled to a RESOlution M50 193nm ArF Excimer (Resonetics) with a two-volume Laurin ablation cell following the method described in Gerdes and Zeh (2006, 2009). Laser spot selection (spot size = 23 μm; depth of crater ~15 μm for U–Th–Pb) was guided by internal structures as seen in CL images of the mounted and polished grains. Data were corrected for laser-induced elemental fractionation and mass bias by normalization to the reference zircon GJ-1 ($^{206}$Pb/$^{238}$U = 0.0986 ± 0.0004; $^{207}$Pb/$^{206}$Pb = 0.06016; JWG ID–TIMS). Prior to this correction, the drift in inter-elemental fractionation (Pb/U) during sample ablation was corrected by applying a linear regression through all measured ratios. Correction for common Pb was done whenever the corrected $^{207}$Pb/$^{206}$Pb was outside the internal errors of the uncorrected ratio and was based on the interference- and background-corrected $^{204}$Pb signal and the terrestrial Pb evolution model of Stacey and Kramers (1975). Reported uncertainties (2σ, standard deviation) were propagated by quadratic addition of the external reproducibility (2σ; standard deviation) obtained from the standard zircon GJ-1 (~0.6% and ~0.7% for the $^{207}$Pb/$^{206}$Pb and $^{206}$Pb/$^{238}$U, respectively; n = 12) during individual analytical sessions and the within-run precision of each analysis (2SE; standard error). The accuracy and reproducibility of the method were verified by 52 analyses of reference zircon 91500 (1062.7 ± 2.2 Ma, MSWD of concordance and equivalence = 0.69; weighted average $^{206}$Pb/$^{238}$U age = 1062.3 ± 2.2 Ma; ± 2SE; 2SD = 1.3%). Results are represented in Figures 2 and 3 and listed in an online electronic archive (Supplementary Table E1) at http://www.gac.ca/wp/?page_id=306. Final data presentation was made with

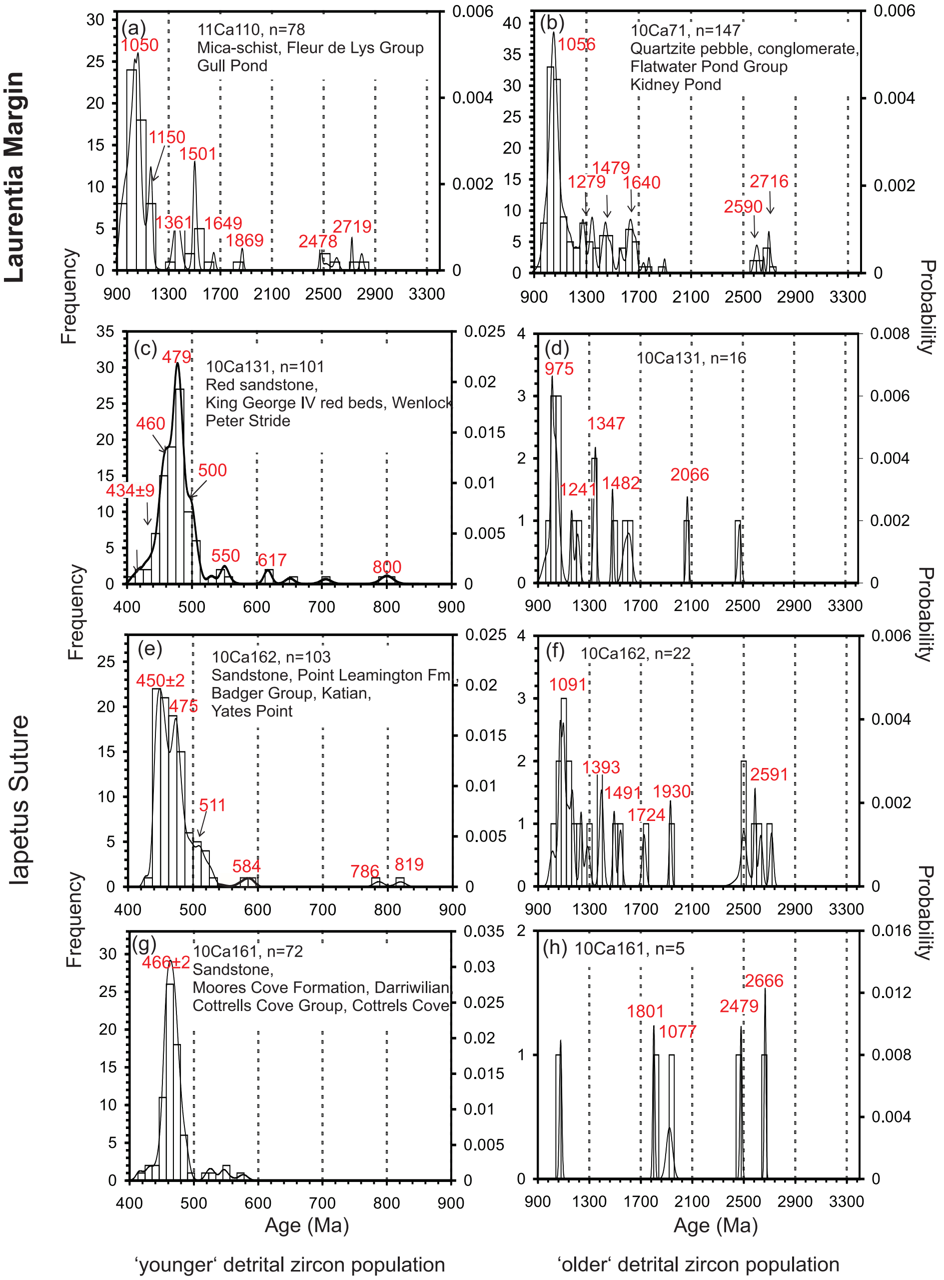

**Figure 3.** U–Pb age spectra (combined probability-histogram plots with single weighted means) for (on left) Neoproterozoic to early Cambrian and (on right) Mesoproterozoic, Paleoproterozoic and Archean detrital zircon ages for samples from the Laurentian margin (a and b) and the Iapetus suture (c to h). See Figure 1 for sample locations and Figure 2 for concordia diagrams. Data are from Supplementary Table E1 in the electronic data base.

Isoplot (Ludwig 2003).

For interpretation of the detrital zircon ages, only concordant or nearly concordant (>90% and <110% concordant) data were considered. For the probability and frequency plots the $^{238}U/^{206}Pb$ ages are used for grains <1 Ga and the $^{207}Pb/^{206}Pb$ ages for grains >1 Ga.

## Hf Isotope Analyses

Lu–Hf isotopic analyses were carried out on 411 zircon grains selected from five samples covering all measured age populations. Analyses were performed with a Thermo-Scientific Neptune multi-collector ICP–MS at Goethe-University Frankfurt (GUF) coupled to RESOlution M50 193 nm ArF Excimer (Resonetics) laser system following the method described in Gerdes and Zeh (2006, 2009). The Lu–Hf laser spot of 40 µm was ablated on top or directly next to the U–Pb laser spot. To correct for isobaric interferences of Lu and Yb on mass 176 the isotopes $^{172}Yb$, $^{173}Yb$ and $^{175}Lu$ were simultaneously monitored. The $^{176}Yb$ and $^{176}Lu$ were calculated using a $^{176}Yb/^{173}Yb$ of 0.796218 and $^{176}Lu/^{175}Lu$ of 0.02658 (GUF in-house values). The instrumental mass bias for Hf isotopes was corrected using an exponential law and a $^{179}Hf/^{177}Hf$ value of 0.7325. In case of Yb isotopes the mass bias was corrected using the Hf mass bias of the individual integration step multiplied by a daily βHf/βYb offset factor. All zircon LA–MC–ICP–MS analyses were adjusted relative to the JMC 475 $^{176}Hf/^{177}Hf$ ratio of 0.282160 and the reported uncertainties (2σ) were propagated by quadratic addition of the reproducibility of JMC 475 (2SD =0.0028%, n=8) and the within-run precision of each analysis (2SE). Accuracy and external reproducibility of the method were verified by repeated analyses of reference zircon GJ-1, Plesovice, and Temora2 (Supplementary Table E2), which yielded $^{176}Hf/^{177}Hf$ of 0.282008 ± 0.000025 (2SD, n=41), 0.0282476 ± 0.000020 (n=16), and 0.282675 ± 0.000022 (n=11), respectively. This is in agreement with previously published results (Woodhead et al. 2004; Slama et al. 2008; Gerdes and Zeh 2009) and with the LA–MC–ICPMS long-term average of GJ-1 (0.282010 ± 0.000025; n > 800), Plesovice (0.282478 ± 0.000025, n > 450), and Temora2 (0.282683 ± 0.000026; n > 200) reference zircon standards at GUF.

The initial $^{176}Hf/^{177}Hf$ values are expressed as $\varepsilon Hf_{(t)}$, which is calculated using a decay constant value of $1.865\times10^{-10}$ (Scherer et al. 2001), $^{176}Hf/^{177}Hf_{CHUR,today}$ = 0.282785 and $^{176}Lu/^{177}Hf_{CHUR,today}$ = 0.0336 (Bouvier et al. 2008). For the calculation of Hf two-stage model ages ($T_{DM}$) in billions of years, the measured $^{176}Lu/^{177}Hf$ ratio of each spot (first stage = age of zircon) and of 0.0113 for the average continental crust (Rudnick and Gao 2003), and a juvenile crust $^{176}Lu/^{177}Hf_{NC}$ = 0.0384 and $^{176}Hf/^{177}Hf_{NC}$ = 0.28316 (average depleted mantle, Chauvel et al. 2008) were used. The depleted mantle array (Figs. 4 to 6) was calculated using data for mid-ocean ridge basalts (MORB; Patchett et al. 1981), and the crustal evolution path was calculated assuming a crustal $^{176}$Lu/$^{177}$Hf ratio of 0.0113. Data are presented in Figures 4 to 6 and listed in Supplementary Table E2 in the electronic data deposit at http://www.gac.ca/wp/?page_id=306.

## RESULTS

### Characteristics of Samples and Detrital Zircon Populations

Samples for this study comprise three groups: (1) Two samples of sedimentary rocks that were originally deposited on the eastern Laurentian passive margin (11Ca110, 10Ca71; Figs. 1, 2a, b; 3a, b); (2) three samples of sedimentary rocks (10Ca161, 10Ca162, 10Ca131; Figs. 1, 2c–e, 3c–h) that were respectively deposited before, during and after closure of the main tract of the Iapetus Ocean (on both sides of the Red Indian Line); and (3) one metasedimentary rock sample (10Ca205; Figs.1, 2f, 4a, b) that was deposited on the Ganderia microcontinent.

#### *Samples from the Laurentian Margin*

***Sample 11Ca110*** is a Fleur de Lys Group mica-schist derived from Laurentian margin sedimentary rocks that were metamorphosed in the Early to Middle Ordovician at relatively high pressure and moderate temperature, in close vicinity to the Gull Pond eclogite (Jamieson 1990). The sample is characterized by a polygonal fabric typical of medium metamorphic grade, and consists of quartz (0.1–0.7 mm), K-feldspar, partially sericitized plagioclase, and oriented white mica and biotite. The sample yielded abundant 100 to 250 μm sized zircon grains. Most grains are slightly to moderately rounded (17% euhedral, 46% subhedral, 31% subrounded, 6% rounded) and typically show internal magmatic zoning (83% zoned; 15% irregular heterogeneous fabric; 2% unzoned; 6% very high CL; 2% very low CL; 3 grains with an overgrowth rim). U–Pb ages of detrital zircons in sample 11Ca110 are Meso- to Paleoproterozoic with maxima (weighted means) at 1.05 Ga, and minor peaks at 1.15, 1.36, 1.50, 1.65, 1.87, 2.48 and 2.72 Ga (Figs. 2a, 3a). Two grains contain inherited cores (rim/core: 0.91/1.02 Ga; 1.07/1.15 Ga).

***Sample 10Ca71*** is a boulder of coarse-grained, relatively mature quartz arenite from the polymictic Kidney Pond conglomerate near the base of the Flatwater Pond Group. The Kidney Pond conglomerate is likely Floian in age, because it contains ca. 479 Ma granitoid boulders and is overlain by ca. 476 Ma felsic volcanic rocks (Skulski et al. 2010). It was deposited on ophiolitic rocks of the Baie Verte oceanic tract after the latter had been amalgamated with the Dashwoods microcontinent and distal outboard rocks of the Humber margin (van Staal et al. 2013). Within the quartz arenite boulder, quartz clasts are subrounded to rounded (0.5–3.0 mm), and the quartzose matrix is slightly recrystallized. K-feldspar, plagioclase, white mica, zircon and rutile constitute minor mineral species. The sample yielded abundant 100 to 200 μm sized zircon grains, most of which are rounded or moderately rounded (5% euhedral, 13% subhedral, 41% subrounded, 40% rounded) and typically show internal magmatic zoning (72% zoned; 18% irregular heterogeneous fabric; 10% unzoned; 22% high CL; 18% low CL; 8 grains with an overgrowth rim). Detrital zircon in sample 10Ca71 yielded a Meso- to Paleoproterozoic age spectrum with a predominant peak at 1.06 Ga, and others at 1.28, 1.48, 1.64, 2.59 and 2.72 Ga (Figs. 2b, 3b). One grain contains an inherited core (rim 0.98 Ga; core 1.46 Ga).

Summarizing, the two samples display a very similar distribution of ages, consistent with other zircon age spectra from the eastern Laurentian margin of Newfoundland (Cawood et al. 2007; Allen 2009). An almost continuous age spectrum from 0.95 – 1.90 Ga is dominated by a ‘Grenvillian’ peak around 1.05 Ga. There is a marked lack of ages between ~1.9 and 2.5 Ga and a minor older peak between 2.5 and 2.7 Ga.

#### *Samples Near the Iapetus Suture*

***Sample 10Ca131*** is an immature, coarse-grained red sandstone from the Llandovery – Wenlock King George IV red beds (Chandler et al. 1987), situated immediately northwest of the Red Indian Line. Clasts are angular to subrounded (0.1–1.0 mm). Mineral clasts are quartz, K-feldspar, plagioclase, calcite, epidote, zircon and white mica. Lithoclasts comprise graphophyric aggregates, rhyolite, epidosite, siltstone, shale and basalt. Quartz and calcite form the matrix.

Sample 10Ca131 yielded two distinct detrital zircon populations. The U–Pb age spectrum is dominated by zircons in the 434 – 500 Ma range (101 grains; 85%), with age maxima (weighted means) at 434, 460, 479 and 500 Ma (Figs. 2c, 3c). Zircon of this ‘younger’ population mostly retains its original shape (43% euhedral, 45% subhedral, 5% subrounded, 7% rounded) and ranges in size from 100 to 250 μm. In CL images, concentric oscillatory zoning with strongly contrasting luminescence is conspicuous in 74% of the grains, whereas 20% are unzoned and 6% are characterized by an irregular heterogeneous fabric. Additionally, seven grains have an overgrowth rim. About 11% of the grains have very high CL, and 15% show very low CL. A notable gap containing only a few minor peaks at 550, 617 and 800 Ma occurs between the dominant ‘younger’ and an ‘older’ zircon population having an age range of 0.93 to 2.47 Ga, with probability peaks at 0.97, 1.24, 1.35, 1.48 and 2.08 Ga (Figs. 2b, 3d). Zircon of the ‘older’ population (18 grains; 15%) shows a higher degree of abrasion (50% subhedral, 20% subrounded, 30% rounded), but similar internal fabric (70% concentric magmatic zoning; 25% unzoned; 5% irregular heterogeneous fabric; 11% very low CL; 44% very high CL; 1 grain with an overgrowth rim; grain size 100 to 250 μm).

***Sample 10Ca162*** is an immature, coarse-grained sandstone from the Upper Ordovician Point Leamington Formation, located south of the Red Indian Line; the Point Leamington Formation is here characterized by broken formation that was grouped with the Boones Point Complex by Nelson (1981). At its occurrence near Yates Point it was reinterpreted as Moores Cove Formation by O´Brien (2003), but its structural position immediately south of the Boones Point Formation in Cottrells Cove and elsewhere suggests affiliation with the younger Point Leamington Formation (cf. Nelson 1981), which is supported by the ages of the youngest detrital zircon (see following). Angular clasts (0.5–5.0 mm) comprise quartz, feldspar, white mica and calcite, as well as lithic clasts of shale, siltstone, calcareous sandstone, basalt with ophitic texture, rhyolite and graphophyric aggregates. Quartz, calcite and white mica form a weakly recrystallized matrix. Sample 10Ca162 mainly contains detrital zircon within the 450 – 511 Ma range (107 grains; 80%); age maxima occur at 450, 475 and 511 Ma (Figs. 2d, 3e). Four concordant older grains are also observed in the 'younger' zircon population (590, 579, 786 and 819 Ma). Zircons of the 'younger' population mostly retain their original shape (55% euhedral, 41% subhedral, 7% subrounded) and range in size from 100 to 200 μm. In CL images, concentric magmatic zoning dominates in 90% of the grains, whereas 7% are unzoned and 3% are characterized by an irregular heterogeneous fabric; four grains have an overgrowth rim. About 14% of the grains have very high CL, and 23% show very low CL. A less pronounced 'older' zircon population (27 grains; 20%) is characterized by ages that range between 1.09 and 2.72 Ga and display probability peaks at 1.09, 1.39, 1.49, 1.72, 1.93 and 2.59 Ga (Figs. 2d, 3f). Zircons of the 'older' population show a higher degree of abrasion (11% euhedral, 26% subhedral, 33% subrounded, 30% rounded), and more variable internal fabric (59% concentric magmatic zoning; 15% unzoned; 26% irregular heterogeneous fabric; 4% very low CL; 15% very high CL; 3 grains with an overgrowth rim; grain size 100 to 200 μm) compared to the younger population. Three zircon grains in sample 10Ca162 contain inherited cores (rim/core: 0.49/2.49 Ga; 2.50/2.72 Ga; 0.90/1.28 Ga).

***Sample 10Ca161*** is an immature epiclastic sandstone from the Middle Ordovician Moores Cove Formation (Dec et al. 1997; O'Brien 2003). These rocks were deposited in a sedimentary basin to the south of the Middle Ordovician calcalkaline arc volcanic rocks of the Fortune Harbour Formation and immediately north of the Red Indian Line (Dec et al. 1997; O'Brien 2003). The sample contains angular clasts (0.5–1.0 mm) of quartz, K-feldspar, plagioclase, and white mica, as well as lithic clasts of quartzite, rhyolite and graphophyric aggregates. The quartzose matrix is recrystallized and contains abundant white mica neoblasts and titanite clusters. Sample 10Ca161 also yielded two distinct detrital zircon populations; predominant is a younger population having a single maximum at 466 ± 2 Ma (75 grains; 90%), suggesting a very proximal igneous source (Figs. 2e, 3g). Zircon grains of the younger population mostly retain their igneous shape (81% euhedral, 16% subhedral, 3% subrounded) and range in size from 100 to 250 μm. In CL images, concentric magmatic zoning is well developed in 98% of the grains, whereas 1% are unzoned and 1% are characterized by an irregular heterogeneous fabric. Two grains have an overgrowth rim. About 9% of the grains have very high CL, and 9% show very low CL. Five grains having concordant ages between 529 and 577 Ma occur within the younger zircon population. Zircon of the older population show maxima at 1.09, 1.80, 2.48 and 2.67 Ga (10%; 8 grains; Figs. 2e, 3h); they display a higher degree of abrasion (38% subhedral, 50% subrounded, 12% rounded), and more variable internal fabric (63% concentric magmatic zoning; 12% unzoned; 25% irregular heterogeneous fabric; 50% very low CL; 13% very high CL; 2 grains with an overgrowth rim; grain size 100 to 250 μm) compared to the younger population. Five zircon grains in sample 10Ca161 contain inherited cores (rim/core: 484/577 Ma; 471/546 Ma; 0.48/2.62 Ga; 1.08/1.18 Ga; 2.48/2.67 Ga).

Summarizing, in all three samples taken near the Iapetus suture a 'younger' zircon population in the approximate range of 435 – 511 Ma dilutes older zircon populations. Differences in the older zircon populations (> 900 Ma) of the three samples are related only to the small number of grains available. Nevertheless, detected ages fit into the age spectrum of zircon from Laurentian margin sedimentary rocks, discussed above. The large quantity of 'younger' zircons and their almost euhedral shape support a proximal source. The magmatic events at 435 – 511 Ma mainly correspond to the ages of the Lushs Bight oceanic tract, the Notre Dame arc and associated arcs (e.g. Red Indian Lake arc), which either developed on Laurentian crust or assimilated Laurentia-derived sedimentary rocks during episodic accretion at its outermost margin (O´Brien 2003; van Staal et al. 2007; Zagorevski et al. 2007).

### Samples from Ganderia

***Sample 10Ca205*** is a cordierite – andalusite mica-schist from the Gander Group. The sample is characterized by a dominant polygonal fabric of quartz (0.05–0.20 mm) and weakly oriented white mica and biotite (0.05–0.30 mm). This fabric is overgrown by dominant poikilitic 1–2 mm sized cordierite porphyroblasts, which show characteristic penetration twins and a variable degree of pinitization. Subordinate andalusite porphyroblasts of similar appearance are also present. A faint relict millimetre-scale banding defined by trails of opaques is locally visible. Zircon and tourmaline are common accessory minerals.

Detrital zircon in sample 10Ca205 is very abundant (301 grains) and also comprises two distinct age populations; 'younger' detrital zircons (51%; 155 grains; Figs. 2f, 4a) ranging from 507 to 770 Ma, and 'older' (>900 Ma) detrital zircons (49%; 146 grains; Figs. 2f, 4b) are present in almost equal amounts (grain size 70 – 200 μm). Zircons of the younger population show age maxima at 506, 525, 540, 580, 604, 640, 670 and 770 Ma, and have a moderately rounded shape (5% euhedral, 43% subhedral, 40% subrounded, 12% rounded). This population is characterized by concentric magmatic zoning in 88% of the grains, no zonation in 4% and an irregular heterogeneous fabric in 8% in CL. Five grains have overgrowth rims. About 7% of the grains have very high CL, and 12% show very low CL. Zircon grains in the older population show a higher degree of abrasion (10% subhedral, 39% subrounded, 51% rounded), and more variable internal fabric (72% concentric magmatic zoning; 5% unzoned; 23% irregular heterogeneous fabric; 10% very low CL; 12% very high CL; 9 grains with an overgrowth rim). These grains yielded age maxima at 0.95, 1.07, 1.23, 1.54, 1.64, 1.73, 1.88, 1.96, 2.08, 2.14 and 2.64 Ga and a notable age

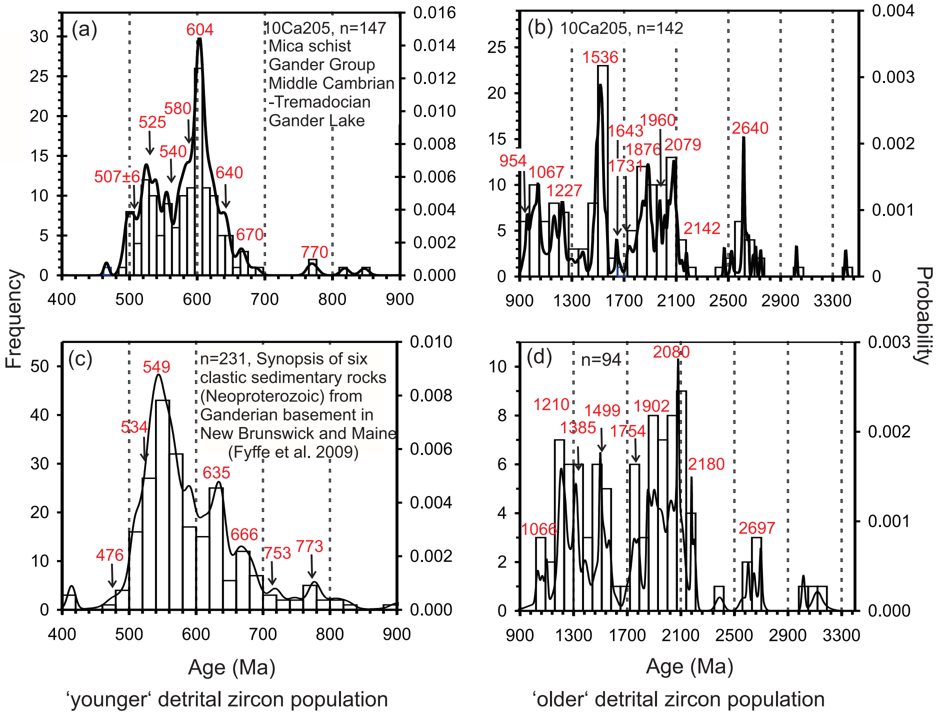

**Figure 4.** U–Pb age spectra (combined probability-histogram plots with single weighted means) for (on left) Neoproterozoic to early Cambrian and (on right) Mesoproterozoic, Paleoproterozoic and Archean detrital zircon ages for samples from a Ganderian sequence in Newfoundland (a, b), and for comparison, from Ganderian sequences in New Brunswick and Maine (c, d; Fyffe et al. 2009). See Figure 1 for sample locations and Figure 3 for concordia diagrams. Data are from Supplementary Table E1 in the electronic data base.

minimum at 2.2 – 2.5 Ga. Three zircon grains in sample 10Ca205 contain inherited cores (rim/core: 0.51/1.46 Ga; 1.49/1.56 Ga; 2.09/2.18 Ga).

Although detrital zircon from Ganderian sedimentary rocks is represented in only one sample in this study, we can regard the derived age spectrum as representative because of the unusually high number of analyzed grains. Comparison can be made with an age spectrum of zircon from six samples of suspected Ganderian sedimentary rocks in New Brunswick and Maine (Fyffe et al. 2009; Fig. 4c, d). The age ranges of the younger and older zircon populations are remarkably similar, although the position of the dominant peaks, particularly in the younger population (from a likely proximal source), differs. On the other hand, a comparison of the older zircon population in the Ganderia (Fig. 4b, d) and Laurentian margin (Fig. 3a, b) samples shows a very similar age range, although an age minimum at 2.2 – 2.5 Ga in the Ganderian spectrum is somewhat narrower than the 1.9 – 2.5 Ga age minimum in the Laurentian spectrum. However, the minor representation of the ~1060 Ma 'Grenvillian' age peak in the Ganderian age spectrum differs significantly from the Laurentian age spectrum, where Grenvillian ages are over-represented.

## Lu–Hf Isotope Compositions of the Detrital Zircon Populations

The 411 zircon grains selected for Lu – Hf analysis yielded measured $^{176}$Lu/$^{177}$Hf ratios of 0.00007 to 0.00500 and present-day $^{176}$Hf/$^{177}$Hf ratios of 0.2807 to 0.2828. The initial $^{176}$Hf/$^{177}$Hf, expressed εHf$_{(t)}$, reflects the Hf-isotopic signature of the magma at the time the zircon crystallized, and hence also the stage of evolution of the crust from which the magma was derived. Zircon grains in this study yield εHf$_{(t)}$ values of +12 to –21 and range in age from Archean to Paleozoic (Fig. 5a, b). The variation in εHf$_{(t)}$ and age allows us to investigate the long-term crustal evolution in both peri-Laurentian and peri-Gondwanan realms. In εHf$_{(t)}$ vs. age diagrams, data generally display two systematic trends: First, the 'crustal evolution' trend starts from juvenile zircon with strongly positive εHf$_{(t)}$ near the evolution line of the depleted mantle, and if no further younger juvenile crustal material is added, evolves with decreasing age towards more recycled zircon with negative εHf$_{(t)}$. This average crustal evolution path has a constant slope, which depends on the average $^{176}$Lu/$^{177}$Hf ratio of the continental crust (here 0.0113; Rudnick and Gao 2003), which is also used for calculation of the model crustal residence ages (T$_{DM}$). Second, the 'mixing' trend is commonly characterized by strong variation in εHf$_{(t)}$ in contemporaneous zircon, indicating a combination of processes including juvenile addition to the crust, crustal recycling, and mixing of juvenile and recycled crustal sources. In the case of mixing, calculated crustal residence ages have no meaning. This interpretation is supported by oxygen isotope signatures of zircon, in which the 'crustal evolution' trend is defined by zircon grains having identical oxygen isotope signatures, whereas 'mixing' trend zircon grains have strongly variable oxygen isotope compositions (Kemp et al. 2006). The combination of juvenile additions, crustal recycling and mixing of both sources commonly occurs in continental magmatic arcs (Lucassen et al. 2004; Franz et al. 2006).

### *Eastern Laurentia*

Sample 10Ca71 provides a εHf$_{(t)}$ vs. age pattern for detrital zircon that is consistent with derivation from a wide area of eastern Laurentia, identical to existing data from the literature (Allen 2009; Fig. 5a). The εHf$_{(t)}$ vs. age data for detrital zircon from the samples in the peri-Laurentian realm (10Ca131, 10Ca161 and 10Ca162) are also added to the data set (Fig. 5a). Distribution of crustal evolution trends, starting from highly positive εHf$_{(t)}$ values near or similar to those of MORB and depleted mantle, show two major times of addition of juvenile crust in the source continent, 1.00 – 1.65 Ga (Mesoproterozoic crust) and 2.55 – 3.00 Ga and likely older (Archean crust), both evolving toward more recycled crust with time and with similar crustal residence ages. A third, minor event of

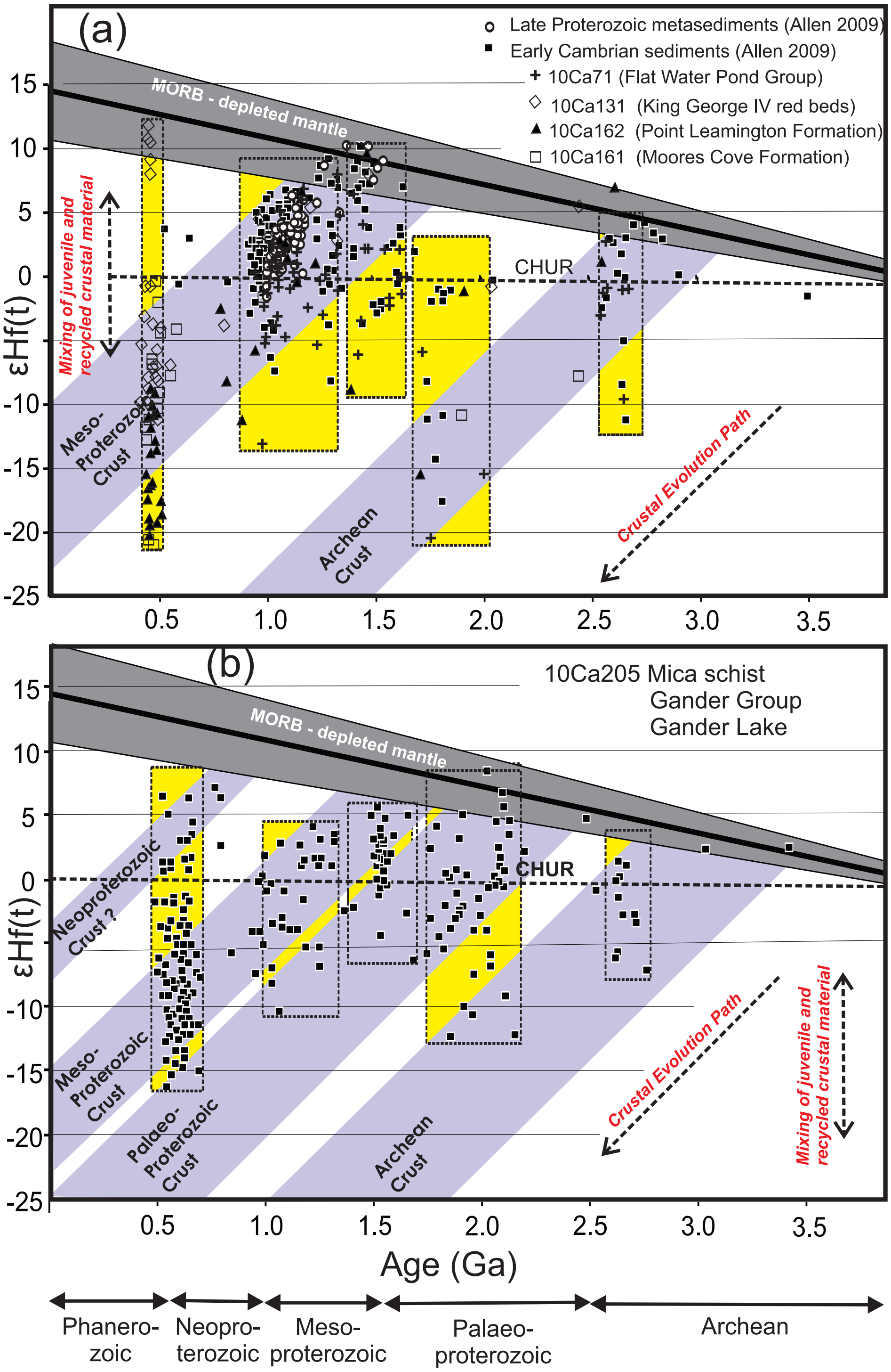

**Figure 5.** (a) Comparative $\varepsilon Hf_{(t)}$ evolution vs. age diagram for detrital zircon from Eastern Laurentia. (b) $\varepsilon Hf_{(t)}$ evolution vs. age diagram for detrital zircon from Ganderia. 2σ-errors are generally equal to or smaller than the size of symbols. Light blue shaded areas represent crustal evolution trends of zircon that might originate from common crustal domains. Yellow shaded vertical trends represent mixing between juvenile and recycled crustal material in magmatic arcs. Lu and Hf isotopic data are listed in Supplementary Table E2 in the electronic data base.

juvenile addition occurs at 0.45 Ga. Data indicate a lack of juvenile crustal addition during Paleoproterozoic (1.65 – 2.55 Ga) and Neoproterozoic (0.6 – 1.0 Ga) times. Hence, no Paleoproterozoic or Neoproterozoic crustal evolution trends appear in the $\varepsilon Hf_{(t)}$ vs. age pattern shown in Figure 5a.

Vertical 'mixing' trends indicate five time-restricted magmatic events in the source area of detrital zircon of distal provenance, including (1) an Archean event at 2.55 – 2.75 Ga ($\varepsilon Hf_{(t)}$ –12 to +4); (2) a Paleoproterozoic event at 1.65 – 2.05 Ga ($\varepsilon Hf_{(t)}$ –21 to +4) characterized by partial recycling of Archean crust with no juvenile input; (3) an Early Mesoproterozoic event at 1.45 – 1.60 Ga ($\varepsilon Hf_{(t)}$ –2 to +11); and (4) a Middle to Late Mesoproterozoic event at 0.95 – 1.40 Ga ($\varepsilon Hf_{(t)}$ –13 to +9). $\varepsilon Hf_{(t)}$ indicates recycling of Paleoproterozoic crust and addition of juvenile crust during both Mesoproterozoic events. A fifth event in the Early Paleozoic (0.43 – 0.51 Ga) is represented only by zircon from the sample taken near the Iapetus suture and corresponds to activity of the outermost margin of Laurentia. $\varepsilon Hf_{(t)}$ values vary strongly between –22 and +12 and mainly Mesoproterozoic crust is recycled.

### ***Ganderia***

In the distribution pattern of $\varepsilon Hf_{(t)}$ vs. age in detrital zircon from Ganderia (Fig. 5b), highly positive $\varepsilon Hf_{(t)}$ values near or similar to those of MORB and depleted mantle indicate four periods of production of juvenile crust from which four crustal evolution trends start: (1) at 2.7 – 3.4 Ga (formation of Archean crust); (2) at 1.85 – 2.40 Ga (formation of Paleoproterozoic crust); (3) at 1.40 – 1.75 Ga (formation of Mesoproterozoic crust); and (4) at 0.7 – 0.9 Ga (likely formation of Neoproterozoic crust). There is a notable 'gap' in the production of juvenile crust between 0.9 and 1.4 Ga that contrasts with the distribution pattern in northeastern Laurentia (Fig. 5a), whereas the 'gap' at 2.4 – 2.7 Ga is less pronounced than in the Laurentian pattern. Five distinct magmatic events involving juvenile additions, crustal recycling, and mixing of both sources in the source continent can be detected according to apparent vertical 'mixing trends' displaying strongly varying $\varepsilon Hf_{(t)}$ values (Fig. 5b): (1) an Archean event at 2.6 – 2.8 Ga ($\varepsilon Hf_{(t)}$ –7 to +2); (2) a Paleoproterozoic event at 1.70 – 2.15 Ga ($\varepsilon Hf_{(t)}$ –12 to +9) that has a high quantity of juvenile crustal material and recycled Archean crust; (3) an older Mesoproterozoic event at 1.45 –

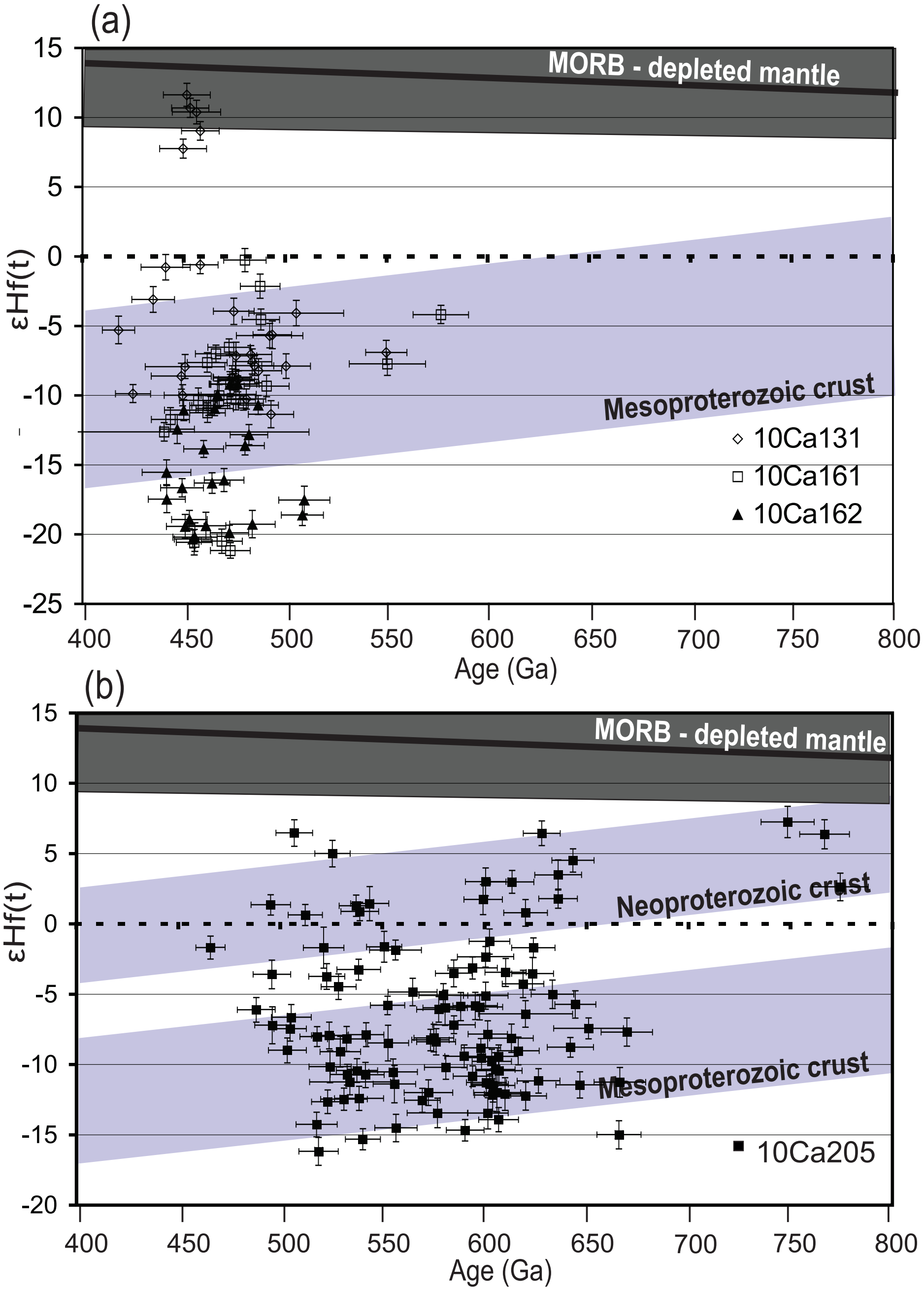

**Figure 6.** Detailed $\varepsilon Hf_{(t)}$ vs. age diagram for detrital zircon < 0.9 Ga in (a) samples from the Iapetus suture, and (b) Ganderia. 2σ errors for $\varepsilon Hf_{(t)}$ and age are given for each sample. Shaded fields are as in Figure 5. Lu and Hf isotopic data are listed in Supplementary Table E2 in the electronic data base.

1.60 Ga ($\varepsilon Hf_{(t)}$ –5 to +7) that recycled Meso- and Paleoproterozoic crust; and (4) a younger Mesoproterozoic ('Grenvillian') event at 0.95 – 1.35 Ga ($\varepsilon Hf_{(t)}$ –10 to +4 Ga) that also recycled Meso- and Paleoproterozoic crust, but without preserved juvenile additions. Most pronounced is (5) a Neoproterozoic – Cambrian event at 0.51 – 0.68 Ga that recycled Neo-, Meso- and likely Paleoproterozoic crust, reflected in the 'younger' detrital zircon population of proximal provenance.

Summarizing, the distribution pattern of $\varepsilon Hf_{(t)}$ vs. age in detrital zircon shows more differences between Ganderian (Fig. 5b) and Laurentian (Fig. 5a) detrital zircon data than just the age probability plots alone, which merely reflect a similar number of magmatic events at similar times. A lack of zircon from juvenile magmatism between ca. 1.65 and 2.55 Ga, and during most of the Neoproterozoic, but a pronounced appearance of juvenile zircon during the 'Grenvillian' magmatic event, mainly distinguishes Laurentia-derived zircon from Ganderia-derived zircon.

## DISCUSSION

### Detrital Zircon Signature and Crustal Evolution of Eastern Laurentia

The combination of $\varepsilon Hf_{(t)}$ values and age (Fig. 5a, b) shows characteristic patterns, especially for detrital zircon of distal provenance, if the number of analyzed grains is sufficiently high. Because the age of this zircon population ranges over most of the Precambrian, the zircon grains must have been collected over a large source area and should have been homogenized during multiple sedimentary recycling events. Hence the pattern of detrital zircon Hf isotopic signatures in sedimentary rocks from the original continental margin of Laurentia and in syn- to post-collisional sedimentary rocks along the Red Indian Line from peri-Laurentian terranes (Fig. 5a) should directly mirror crustal evolution in the northeastern part of Laurentia.

Most detrital zircon data follow the magmatic 'mixing' events in the age range of 0.95 – 1.60 Ga, which corresponds to continental magmatism in the adjacent Grenville orogen in eastern Laurentia (for summary see Gower and Krogh 2002). This includes the 1.60 – 1.71 Ga (Labradorian), 1.46 – 1.52 Ga (Pinwarian), and 1.23 – 1.18 Ga (Elzevirian) continental arc magmatism, and 1.46 – 1.23 Ga (Elsonian) bimodal magmatism. These magmatic episodes were overprinted by the Grenvillian continent-continent collision at 1.08 – 0.98 Ga and extensive post-tectonic magmatism that lasted until ca. 0.95 Ga. Magmatism in the Grenville orogen involved both addition of juvenile crust and recycling of existing crust. Recycling is supported by data such as ca. 0.9 – 1.1 Ga detrital grains having inherited discordant core ages of 1.02 – 1.46 Ga (see previous). Recycling and/or mixing with underlying Mesoproterozoic crust during the predominant Grenvillian event are notable in the Hf-isotopic distribution pattern in Figure 5a. However, much zircon in the 0.95 – 1.60 Ga range also plots in the field of a potential Paleoproterozoic crustal evolution trend (Fig. 5a), which is not apparent in the present data set because no detrital zircon was detected with positive $\varepsilon Hf_{(t)}$ values indicating a juvenile Paleoproterozoic source. Two explanations are possible: crust with Paleoproterozoic residence time may exist at depth within the present crust, or mixing of Mesoproterozoic and Archean crustal material may have occurred.

The mixing trend at 2.1 – 1.6 Ga shown by the detrital zircon pattern of Figure 5a could correspond to earlier continental arc magmatism at 1.91 – 1.70 Ga that reworked Archean crust to the west of the Grenville Province (Makkovikkian; Ketchum et al. 2002). The oldest mixing trend, at 2.55 – 2.75 Ga (Fig. 5a), correlates with the Archean Superior Province, which comprises rocks having ages mostly between 2.6 and 3.1 Ga (maximum 3.6 Ga; Card 1990), including a pronounced peak of calcalkaline magmatism at 2.60 – 2.75 Ga (Kenoran orogeny). Two detrital zircon grains, having ages of 2.48 and 2.50 Ga, contain discordant inherited cores with ages of 2.67 and 2.72 Ga, respectively, indicating crustal recycling during the Kenoran Orogeny.

Note that there is likely an increasing sampling bias in the detrital zircon available with respect to the possible oldest ages and potentially oldest crustal evolution trend in Laurentia. The oldest zircon crystallization ages detected in our samples are 2.72 – 2.80 Ga, and the oldest crustal residence ages ($T_{DM}$) are 2.91 – 3.47 Ga (Supplementary Table E2; see Allen 2009: 2.73 – 3.60 Ga and 2.91 – 3.79 Ga, respectively). Crustal residence ages of up to 3.4 – 3.8 Ga were detected by Hf isotopic studies of zircon in the western Lake Superior province (Davis et al. 2005). The oldest rock so far identified in the Canadian Shield is the Acasta Gneiss, which also represents the oldest known terrestrial rock (3.94 – 4.03 Ga; Iizuka et al. 2009). The crustal residence age of its protolith is 4.2 Ga (Iizuka et al. 2009).

In summary, the distribution pattern of Hf isotopic signatures and ages of detrital zircon from northeastern Laurentia mirrors the known zircon-forming events in the source continent and hence its crustal evolution. The overall characteristics of detrital zircon from eastern Laurentia are: (1) absence of any 0.7 – 1.0 Ga zircon that formed from juvenile magma sources; (2) abundant 1.0 – 1.2 Ga zircon formed by juvenile additions and crustal recycling during the Grenville orogeny and prior arc magmatism; (3) formation of most juvenile and recycled crust in the time range 0.95 – 1.60 Ga; (4) a gap in production of juvenile crust between 1.6 and 2.5 Ga; and (5) a gap of magmatic arc activity between 2.0 and 2.5 Ga.

## The Ganderia Detrital Zircon Signature and the Origin of Ganderia

In contrast to detrital zircons from Laurentia, the distal source of the 'older' Ganderian detrital zircon population is not apparent. However, at present there are many detailed arguments in the literature that the Ganderian microcontinent separated from the Amazonia craton within western Gondwana from the approximate present position of the Caribbean margin of Colombia (van Staal et al. 1996, 2012 and references therein). For example: (i) earlier-derived detrital zircon ages having a dominant Mesoproterozoic maximum could be matched with source areas overprinted by the Rondonian – San Ignacio orogeny and with Grenvillian basement inliers in the northern Andes; (ii) Cambrian cover sedimentary rocks in Colombia match coeval Ganderian strata containing trilobites of the same realm; (iii) truncation of Grenvillian inliers against the Caribbean coastline indicates removal of a crustal fragment; and (iv) a marked Cambrian transgression recorded in Colombian sedimentary rocks is interpreted to reflect separation of a large terrane. This proposed link between Ganderia and the northwestern margin of Amazonia can be tested by comparing the above-derived Ganderian detrital zircon signature (Fig. 5b) with the known crustal evolution in the northwestern part of South America, as follows:

(1) Three episodes of juvenile magma production in the Ganderian source continent, at 2.7 – 3.4 Ga, 1.85 – 2.40 Ga and 1.40 – 1.75 Ga (Fig. 5b), coincide well with periods of juvenile crust production detected in the Amazonian protocontinent, mainly based on Nd isotopic evidence (Tassinari and Macambira 1999; Cordani and Sato 1999; Cordani et al. 2009). For example, generation of Early Archean juvenile crust at ≥2.8 Ga was detected in the Central Amazonian craton, and Paleoproterozoic juvenile crust developed as a result of magmatic arc formation during the Transamazonian orogeny (1.80 – 2.25 Ga) as a result of formation of magmatic arcs. Furthermore, formation of Mesoproterozoic juvenile crust occurred during emplacement of magmatic arcs related to the Rondonian – San Ignácio orogeny (1.3 – 1.5 Ga), and during rift events in the Central Amazonian craton (1.60 – 1.95 Ga) and areas affected by the Rio Negro – Jurmena orogeny (1.2 – 1.4 Ga).

(2) Three periods of mixing of juvenile and recycled crustal material, at 2.6 – 2.8 Ga, 1.7 – 2.15 Ga, and 1.45 – 1.60 Ga (Fig. 5b), correspond well with calcalkaline arc magmatism in the Central Amazonian craton during the Transamazonian (1.90 – 2.25 Ga), Rio Negro – Jurmena (1.55 – 1.78 Ga), and Rondonian – San Ignácio (1.30 – 1.50 Ga) orogenies (Tassinari and Macambira 1999; Cordani and Sato 1999; Cordani et al. 2009). It should also be noted that Tassinari and Macambira (1999) reported an 'age minimum' at 2.0 – 2.4 Ga in the Central Amazonian craton. In the two younger arcs, Meso- and Paleoproterozoic crust is recycled, whereas the older arcs recycle Paleoproterozoic and Archean crust. This recycling is corroborated by observed xenocrystic cores of 1.56 Ga in a 1.49 Ga zircon and of 2.18 Ga in a 2.09 Ga zircon (see previous description).

(3) A marked over-representation of zircon ages of 1.45 – 1.55 Ga (Figs. 4, 5b), corresponding to magma production during the Rondonian – San Ignácio orogeny, can be an indicator of the proximity of Ganderia to its original position at the margin of Amazonia. In contrast, detrital zircon having Grenvillian ages (0.95 – 1.30 Ga) is pronounced in the Ganderian detritus, but does not show a marked over-representation as in the Laurentian detritus, where Grenvillian sources

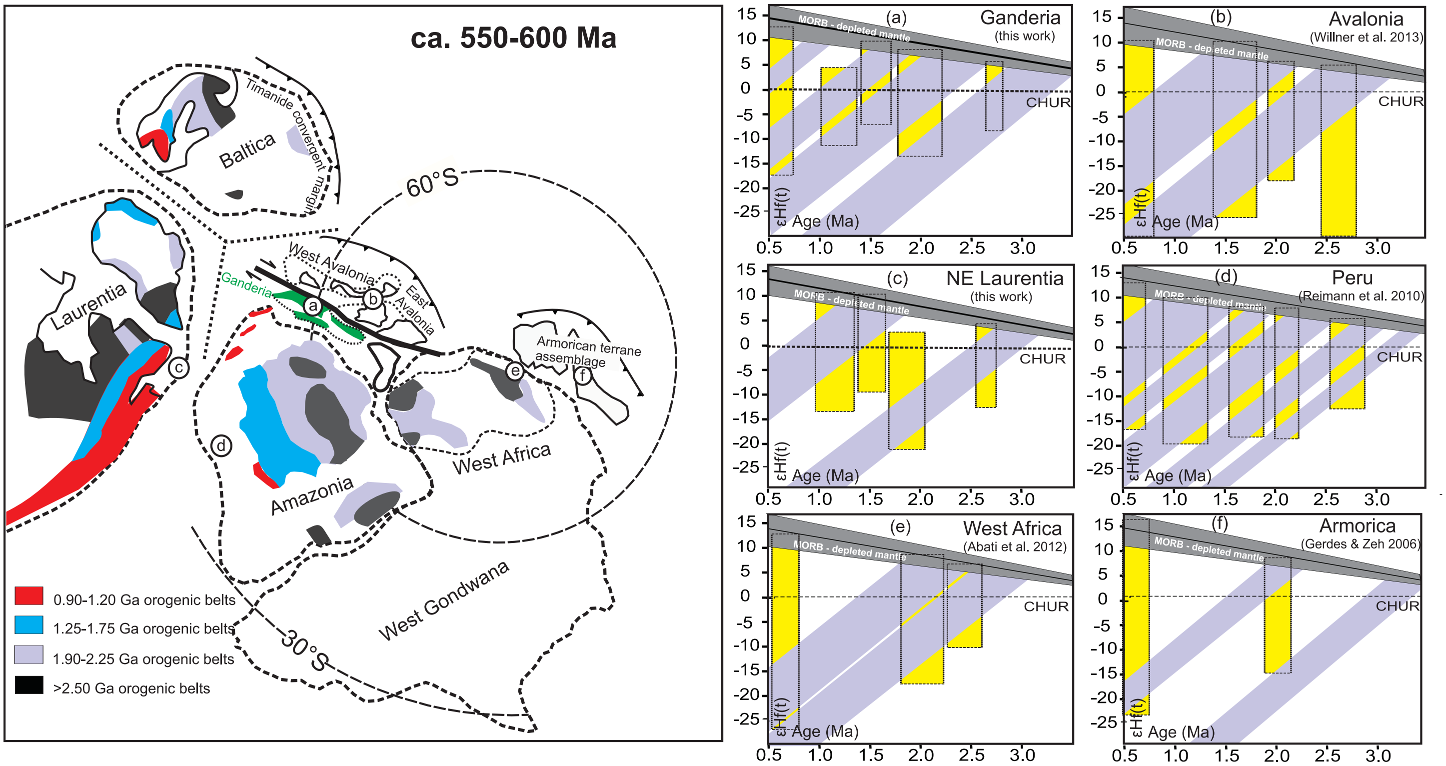

**Figure 7.** Paleogeographical map showing the postulated position of the Ganderia and Avalonia microcontinents with respect to West Gondwana at about 600 – 550 Ma, when Laurentia and Baltica just separated from West Gondwana. Restoration modified after Nance et al. (2012) and based on the paleocontinental reconstruction of Torsvik et al. (1996). (a) to (f) – Schematic $\varepsilon Hf_{(t)}$ vs. age for detrital zircon from data presented herein and from Gerdes and Zeh (2006), Reimann et al. (2010), Abati et al. (2012) and Willner et al. (2013).

are evidently proximal. The studied Grenvillian zircon population follows a vertical 'mixing' trend, but also a Mesoproterozoic 'crustal evolution' trend. However, there is a lack of juvenile zircon in that time range. The vertical trend likely reflects mere recycling of Meso- and Paleoproterozoic crust and mixing of both.

A detrital zircon signature very similar to that of Ganderia can also be detected in detrital zircon of distal provenance in sedimentary rocks of the Avalonia microcontinent (Willner et al. 2013; Fig. 7b), which also indicates derivation from the Amazonian margin. The main difference in the detrital zircon signatures of Ganderia and Avalonia is a significantly weaker representation of Mesoproterozoic zircon, particularly of Grenvillian age, within the Avalonian detrital zircon populations. Our data support the proposal of van Staal et al. (2012) with respect to the original position of Ganderia near the present Caribbean margin of Colombia. During the late Neoproterozoic, Avalonia was also sourcing Amazonian detritus, but was probably positioned more distally with respect to the source areas in Amazonia (Willner et al. 2013). The originally neighbouring West African craton can be eliminated as an alternative source craton for both microcontinents, because detrital zircon spectra with a West African source are characterized by a pronounced gap of Mesoproterozoic ages (Avigad et al. 2012; Abati et al. 2012; see Fig. 7e). It should be noted that, aside from Avalonia, three other terranes having remarkably similar zircon age spectra have closely been linked with Ganderia at its original location: Carolinia, in the present southern continuation of Ganderia along the margin of Laurentia (van Staal et al. 2012) and the Istanbul (Ustaömer et al. 2011) and Moesia (Balintoni et al. 2010) terranes, both at the present southeastern margin of Baltica in Turkey and Romania, respectively.

In summary, the following features characterize detrital zircon signatures of the Lower Paleozoic clastic rocks of Ganderia:

(i) A pronounced cluster of Grenvillian ages at 0.95 – 1.25 Ga point to a proximal source of this age, which, for example, occurs in Grenvillian basement inliers in the Caribbean Andes of Colombia.

(ii) A marked presence of zircon ages of 1.45 – 1.55 Ga corresponds to magma production during the Rondonian – San Ignácio orogenic cycle and supports proximity to this part of Amazonia.

(iii) A pronounced gap in the formation of juvenile crust exists between 0.9 – 1.4 Ma. These characteristics reflect derivation from the Caribbean part of the Amazonian source craton.

## Maximum Sedimentation Ages and the Time of Closure of the Iapetus Ocean

The 'younger' detrital zircon populations (Figs. 3c, e, g; 4a, c; 5a, b) have a rather proximal source (relative to the location of sedimentation), that can be relatively easily detected. In general, samples contain more zircon grains of this population than from the 'older', distal zircon population. This permits derivation of maximum sedimentation ages, which can be very close to the actual time of deposition if the igneous activity in the source region continues during sedimentation. Furthermore, 'overstep sequences' can be detected, i.e., sedimentary rocks sourced from one continent overstep a collision zone into a second (colliding) continent and hence provide a minimum age of the collision event.

Maximum depositional ages derived from the youngest U–Pb ages of detrital zircon in Late Ordovician to Silurian sedimentary rocks on the upper plate (composite Laurentia) close to the Red Indian Line are in agreement with the known sedimentation ages (Fig. 8; samples 10Ca162, 10Ca161 and 10Ca131). These sedimentary rocks contain detritus that appears to have been exclusively derived from the Laurentian margin, particularly from peri-Laurentian juvenile and continental arcs (e.g. Notre Dame Arc) in the Iapetus and from continuing non-arc magmatic activity until ca. 425 Ma after collision (Fig. 3c, e, g; Fig. 5a).

Epiclastic sandstone from the Middle Ordovician Moores Cove Formation (Nowlan 1996, 1997) yielded a youngest detrital zircon population with a weighted mean age (n=60) of 466 ± 2 Ma (10Ca161; point 5 in Figs. 1, 8). It is suggested that the Moores Cove sedimentary basin was sourcing a broadly coeval continental arc. The location of the Moores Cove basin along the Red Indian Line suggests that it may represent the forearc basin along the leading edge of composite Laurentia, which was magmatically active until 457 ± 4 Ma (Zagorevski et al. 2006). This phase of arc magmatism was terminated by arc-arc collision and closure of the main tract of the Iapetus along the Red Indian Line at ca. 455 Ma (Zagorevski et al. 2008).

Southeast of the Red Indian Line, the coeval Popelogan – Victoria arc system was active on the leading edge of Ganderia. The youngest ages from the Popelogan – Victoria arc system were obtained from tuffaceous sandstones along the Red Indian Line (453 ± 4 Ma: Zagorevski et al. 2007; 455 ± 3 Ma: Zagorevski et al. 2010). Following arc-arc collision and cessation of arc magmatism, the Popelogan – Victoria arc rocks were overlain by a blanket of black shale and turbiditic sedimentary rocks of the Upper Ordovician to Lower Silurian Badger Group. These sedimentary rocks represent the first Laurentia-derived detritus deposited on the accreted Ganderian rocks (see previous; Waldron et al. 2012), representing an accretionary forearc basin (Zagorevski et al. 2008; van Staal et al. 2014). In the upper section of the black shale horizon an early Katian *(Dicranograptus Clingani* zone) graptolite fauna was detected (Williams 1991); the absolute age of this faunal zone is 453 – 450 Ma (Cohen et al. 2013). Sandstone of the Point Leamington Formation was deposited on the Katian black shale and yielded a detrital zircon population of 472 Ma (Waldron et al. 2012), apparently not recording the immediately preceding Middle to Late Ordovician volcanism. Point Leamington Formation sample 10Ca162 (point 4 in Figs. 1, 8), collected from approximately the same stratigraphic horizon as the samples studied by Waldron et al. (2012), yielded a much younger detrital zircon population having a weighted mean age (n=43) of 450 ± 2 Ma. The collision-related black shale transgression on the Popelogan – Victoria arc therefore lasted from ca. 455 Ma (Zagorevski et al. 2007, 2010) until ca. 450 Ma (see previous).

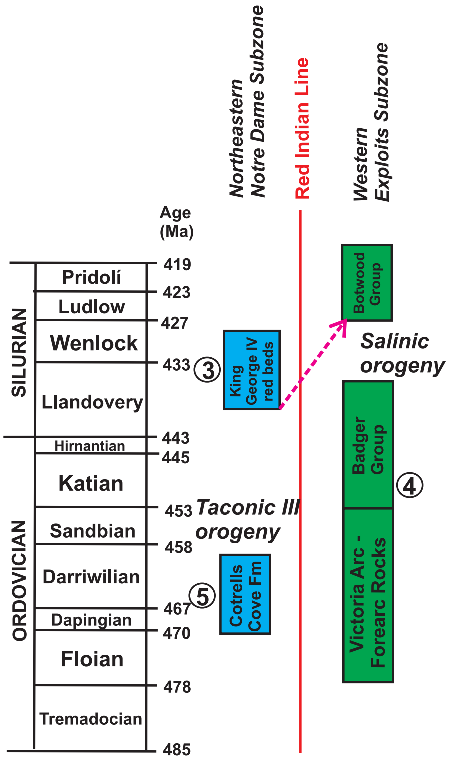

**Figure 8.** Schematic stratigraphic columns for Ordovician and Silurian rocks on both sides of the Iapetus suture. The time scale is that of the International Chronostratigraphical Chart (Cohen et al. 2013). Key to circled numbers (see Figure 1 for locations): 3 – 10Ca131; 4 – 10Ca161; 5 – 10Ca162. A trend in Silurian times shown by the arrow indicates increasing cratonization.

Silurian redbeds (Old Red Sandstone) diachronously overlie Laurentian and Ganderian rocks on both sides of the Red Indian Line. Red sandstone (King George IV red beds, a correlative of the Springdale Group; Chandler et al. 1987) deposited on the Laurentian side immediately northwest of the Red Indian Line (10Ca131; point 3 in Figs. 1, 8) yielded a youngest detrital zircon population having a poorly defined weighted mean age (n=11) of 434 ± 9 Ma. This age overlaps within error the age of interlayered volcanic rocks and consanguineous plutons (429 +6/–4 and 435 +6/–3 Ma, respectively; Dunning et al. 1990), and independently constrains the age of sedimentation. The red beds continued to source Laurentian detritus and show no conclusive evidence for derivation from adjacent Ganderian terranes. Southeast of the Red Indian Line, the Upper Silurian Botwood Group also continues to be essentially characterized by Laurentian detritus (Pollock et al. 2007) until the deposition of the Rogersons Lake conglomerate. The Rogersons Lake conglomerate unconformably overlies Ganderian Neoproterozoic basement and Cambro–Ordovician volcanic rocks, and displays zircon provenance of both Laurentian and Gondwanan provenance (Pollock et al. 2007). Hence the Ganderian sources were only exhumed to the surface following the Wenlock – Ludlow closure of the Tetagouche – Exploits back arc basin (van Staal 1994; van Staal et al. 2009, 2014; Fig. 1).

### Evolution of the Peri-Laurentian Arcs at the Composite Margin of Laurentia

The 'younger' detrital zircon population detected in our samples has the least degree of rounding and likely represents first-cycle detritus of local provenance. It mainly represents proximal detritus of magmatic arcs that developed on the leading edges of composite Laurentia and Ganderia (Zagorevski et al. 2009; van Staal and Barr 2012). The long-term evolution of these magmatic arcs can be studied using detrital zircon in a similar way as done for the crustal evolution of the source continents.

Prior to, during, and following accretion of various elements of Ganderia to Laurentia between ca. 455 and 425 Ma, the syntectonic Late Ordovician to Silurian sedimentary rocks deposited on the upper plate (composite Laurentia) continued showing only detritus derived from Laurentia (Figs. 4b, 6a). These sedimentary rocks (samples 10Ca162, 10Ca161 and 10Ca131) contain detrital zircon from Iapetan juvenile and continental arcs (e.g. Notre Dame Arc) that were active between ca. 510 and 435 Ma, and from continuing non-arc magmatic activity until ca. 425 Ma (Whalen et al. 2006).

Zircon derived from the peri-Laurentian magmatic arcs in Newfoundland displays a marked change in $\varepsilon Hf_{(t)}$ values from northeast to southwest (Figs. 1, 6a). In the northeast, strongly negative $\varepsilon Hf_{(t)}$ values (–5 to –20) point to recycling of both Mesoproterozoic and Paleoproterozoic crust (samples 10Ca161, 162). Furthermore, discordant inherited cores of detrital zircon having ages of 2.62 Ga and 2.59 Ga (see above) indicate that Early Archean magmatic rocks also underlie the composite Laurentian margin, or that underlying sedimentary rocks containing Archean detrital zircon were melted. In the southwestern part of the peri-Laurentian magmatic arcs, positive $\varepsilon Hf_{(t)}$ values (+7 to +12) indicate significant addition of juvenile crust, whereas time-equivalent negative $\varepsilon Hf_{(t)}$ values (0 to –10) suggest recycling of Mesoproterozoic crust and mixing with juvenile magma (Sample 10Ca131).

The oldest predominant age peak in the 'younger' zircon population of detritus from the composite Laurentian margin is 511 Ma, which could correspond with the age of the oldest suprasubduction zone rocks of the peri-Laurentian arcs in Newfoundland, the Twillingate trondhjemite (Elliott et al. 1991), and older volcanic rocks of the Sleepy Cove Group in the Lushs Bight oceanic tract (van Staal et al. 2007, 2009). However, fifteen concordant detrital zircon grains ranging in age from 529 to 617 Ma, and four grains ranging from 786 to 819 Ma are present in these three samples (Figs. 2, 3; Supplementary Table E1). The $\varepsilon Hf_{(t)}$ values of this zircon population follows the Mesoproterozoic crustal evolution trend (Figs. 4b, 6a; Supplementary Table E2), but are not related to a crustal mixing trend. Two inherited discordant cores of 546 and 577 Ma in zircon from the composite Laurentian margin indicate that equivalent Late Neoproterozoic rocks should underlie the peri-Laurentian arcs. These rocks most likely formed during the terminal rift events responsible for opening of the Iapetus Ocean and the Taconic seaway (Waldron and van Staal 2001).

### Evolution of the Peri-Gondwanan Arcs at the Margin of Ganderia

The pronounced 'younger' detrital zircon population in the detritus from Ganderia, having ages between 507 and 670 Ma and an earlier peak at 770 Ma (Figs. 2f, 4a), represents Neoproterozoic – Cambrian continental arcs that were built on the margin of Amazonia before the departure of Ganderia. The youngest probability peak at 507 ± 6 Ma is based on 12 single grains and represents the maximum sedimentation age of this part of the Gander Group. This age overlaps the oldest known ages of the Penobscot magmatic arc (515 – 485 Ma), which is interpreted to have developed on the Ganderian margin, before and during separation and drift of Ganderia from Gondwana as a microcontinent en route to Laurentia (Zagorevski et al. 2010; van Staal et al. 2012).

The $\varepsilon Hf_{(t)}$ values of detrital zircon derived from the Ganderian arcs consistently indicate that most zircon represents recycled Neoproterozoic and Mesoproterozoic crust (Figs. 5b, 6b). One discordant inherited core with an age of 1.46 Ga in a 0.51 Ga zircon (see previous description) supports this observation. The predominant peak at 1.54 Ga in the spectrum of the 'older' detrital zircon population most likely indicates close proximity to rocks of this age, which are evidently also underlying the Ganderian arcs. Three grains representing the earliest magmatic activity at the Ganderian margin (ca. 770 Ma) have positive $\varepsilon Hf_{(t)}$ values and follow the Neoproterozoic crustal evolution trend.

## CONCLUSIONS

The combination of U–Pb dating and Hf isotope analysis of detrital zircon provides very detailed information about the source cratons, including times of formation of juvenile crust and time ranges of magmatic arc activity, as well as the nature of recycled crust underlying these magmatic arcs. Provenance of Appalachian terranes can thus be identified and paleocontinental relationships can be reconstructed (Fig. 7). The $\varepsilon Hf_{(t)}$ vs. age patterns of detrital zircon for Ganderia, Avalonia and Peru (Fig. 7a, b, d) are comparable for ages >1.2 Ga and consistent with derivation from the Amazonian craton. In contrast to these patterns, $\varepsilon Hf_{(t)}$ vs. age plots for detrital zircon from northeast Laurentia (Fig. 7c) are characterized by a lack of juvenile crust formation at 1.8 – 2.5 Ga, and of mixing of juvenile and recycled material between 2.0 and 2.5 Ga. West Africa and associated Armorican terranes (Fig. 8e, f) lack crustal recycling and formation of juvenile crust between 1.8 and 0.7 Ga (e.g. Gerdes and Zeh 2006; Avigad et al. 2012; Abati et al. 2012), excluding West Africa as the source craton for Ganderia and Avalonia.

Zircon populations from northeast Laurentia (Allen 2009; this work) and Peru (Reimann et al. 2010) are dominated by 'Grenvillian' ages (0.9 – 1.2 Ga) and show a wide variation of $\varepsilon Hf_{(t)}$ values, indicating formation of juvenile crust, recycling of Meso- and Paleoproterozoic crust and mixing of juvenile and recycled crust. This marked Grenvillian signal further supports the contention that eastern Laurentia and the western Amazonian margin were once attached to one another (e.g. Cawood et al. 2001; Miškovic et al. 2009; Cardona et al.

2010). Although the Grenvillian orogen is a well-exposed chain on the eastern side of Laurentia, it is largely hidden along the Pacific margin of Amazonia. Nevertheless, detrital zircon from the Andean part of the orogen was abundantly present during multiple recycling events in most Phanerozoic sedimentary rocks at the Pacific Andean margin. In addition, Grenvillian crust underlying the Andean belt was recycled in Phanerozoic Andean arcs (Willner et al. 2008; Miškovic et al. 2009; Cardona et al. 2010; Reimann et al. 2010; Ramos 2010). The Grenvillian signal is much less pronounced in Middle Cambrian – Ordovician Ganderian zircon populations and it does not include zircon from juvenile crust. The Grenvillian signal is even weaker in Avalonian zircon populations, implying that Ganderia and Avalonia occupied a continuously distal position with respect to the Grenvillian belt (Willner et al. 2013). Ganderian and Avalonian zircon populations in the 1.4 – 1.7 Ga range are very pronounced, suggesting that the source crust involved in the Rondonian and Rio Negro orogenies should continue towards the present Caribbean coast of Colombia and Venezuela under the cover sedimentary rocks (see also Miškovic et al. 2009). Furthermore, it could be shown that crust forming at that time should underlie the Neoproterozoic – Cambrian arcs that developed on both Avalonian and Ganderian margins. Zircon formed in these arcs partly originated from melts that recycled Neo- and Mesoproterozoic crust. In contrast, the Ordovician and Silurian peri-Laurentian arcs in Newfoundland are characterized by recycling of underlying Meso- and Paleoproterozoic crust and variable additions of juvenile crust along strike.

When Ganderia left its position at the Amazonian margin along the present Caribbean coast of Colombia at ca. 500 Ma (van Staal et al. 2012), it traveled in ca. 45 my across the Iapetus Ocean to its present position in Canada and New England. The time of collision of the outermost part of Ganderia with composite Laurentia is constrained by arrival of the first Laurentian detritus on the active margin of Ganderia (i.e., the Popelogan – Victoria arc) at ca. 452 Ma, indicating that the main tract of the Iapetus Ocean had closed and subduction consequently stepped back into the Tetagouche – Exploits backarc basin (van Staal et al. 2009). During the Late Ordovician to Silurian, syn-tectonic sedimentation on composite Laurentia (i.e., Laurentia and leading elements of Ganderia) continued to feature detritus principally derived from Laurentia (see previous; Waldron et al. 2012), confirming that the trailing edge of Ganderia (Gander margin) remained isolated by a seaway from composite Laurentia until the Late Silurian.

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