# Modelling U–Pb discordance in the Acasta Gneiss: implications for fluid–rock interaction in Earth's oldest dated crust

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Abstract

The U–Pb isotopic system in zircon is the tool of choice to interrogate high-temperature geological processes, yet this system has potential to investigate lower temperature fluid–rock interaction as well. In some cases, removal of radiogenic Pb is incomplete, potentially allowing regression of discordant U–Pb data on a concordia diagram to determine both the age of crystallization and the timing of fluid-driven isotopic disturbance. However, in rocks preserving more complex histories, simple regression is not effective at resolving multiple Pb loss events. Here, we use a 'concordant–discordant comparison' (CDC) test to establish the times of U–Pb disturbance in the Acasta Gneiss Complex (AGC), Canada. AGC c. 4.03 to c. 3.40 Ga orthogneisses experienced a long and complex post-crystallization history, for which U–Pb zircon data reflects not only the heterogeneous nature of the rock, but also the varying degrees and duration of crustal reworking that inevitably involved open system processes. The CDC test calculates the similarity between the concordant age structure and a modelled age structure, the latter inferred from discordant analyses, over a wide range of potential disturbance times. Our analysis reveals concordant zircon components implying new growth and/or recrystallization at $3992 \pm 5$, $3501 \pm 6$, $3442 \pm 5$ and $3126 \pm 6$ Ma. In addition, we establish episodes of radiogenic-Pb loss driven by fluid–rock interaction at $3150 \pm 50$ Ma, and probably at $2875 \pm 50$ Ma and c. 2590 Ma. These Pb-loss episodes correlate with previously recognised events recording growth of zircon rims during metamorphism, granite emplacement, and unroofing. Pb-loss within the AGC shows an antithetic relationship in different samples that are in close geographic proximity. We suggest that zircon alteration and associated new growth effectively rendered those grains that underwent Pb-loss at a particular time less susceptible to alteration during the next episode of fluid interaction.

KEYWORDS: Zircon, Acasta Gneiss Complex, Pb-loss, U–Pb geochronology; Discordance

## Introduction

U–Pb isotopes are the system of choice to address many temporal aspects of our planet's formative history, founded in part on the physical robustness of the mineral zircon and its tendency to incorporate U but exclude Pb during initial crystallization (see U-Pb zircon publication metrics in Spencer et al.,

36  2016). However, the very process that permits radiogenic dates to be calculated, the slow decay of

37  isotopes of U to Pb, also causes radiation damage to the crystal structure of zircon over time (Holland

38  and Gottfried, 1955). This radiation damage leads to radiogenic Pb loss from zircon and hence

39  discordance between the U–Pb isotopic systems, such that the extent of discordance in different parts of

40  zircon grains typically correlates with the abundance of U and accompanying non-formula elements

41  (including common Pb) (Fig. 1) (Köppel and Sommerauer, 1974; Ludwig and Cooper, 1984).

42  Discordance, which can be defined as the degree of dissimilarity between any pair of isotopic dates, is the

43  principal measure by which open system alteration processes in zircon may be tracked (Geisler et al.,

44  2002; Pidgeon et al., 2013; Stern et al., 1966).

45  Radiogenic-Pb loss is generally regarded as detrimental to U–Pb geochronology, in particular where the

46  goal is in determining a primary crystallization age. However, radiogenic-Pb loss and the resulting trends

47  of discordant data on concordia diagrams offer the possibility to understand the timing of events that

48  stripped Pb from zircon (Kirkland et al., 2017). In many cases this Pb mobility corresponds to important

49  metamorphic, anatectic and/or fluid-mediated events (Morris et al., 2015; Zamyatin et al., 2017).

50  Figure 2 plots two different measures of discordance versus age for a global dataset of zircon U–Pb

51  analyses. In Figure 2a, the centroid ([$^{207}$Pb/$^{206}$Pb age – $^{238}$U/$^{206}$Pb age]/ $^{207}$Pb/$^{206}$Pb age x 100) of the

52  analytical error ellipse is compared to age; in Figure 2b the proximity of the edge of the 2σ error ellipse to

53  the concordia curve is used, whereby a zero value indicates an error ellipse that intersects this curve.

54  These plots illustrate several salient points with regard to discordance: (i) For Neoproterozoic and

55  younger zircons the discordance of the centroid increases consistent with the precision of the $^{207}$Pb/$^{206}$Pb

56  ratio decreasing, as $^{238}$U/$^{206}$Pb remains relatively more constant as it is dictated by standard defined

57  calibration uncertainties (Fig. 2a). In contrast, the amount of discordance of the two-sigma ellipse is not

58  significantly different between Archean versus younger zircons (Fig. 2b). (ii) Although older grains

59  generally will have accumulated more radiation damage and younger analyses will be associated with

60  greater relative imprecision, on average greater discordance of the error ellipse is not clear for older

61  analyses. (iii) Normal discordance ($^{207}$Pb/$^{206}$Pb age > $^{238}$U/$^{206}$Pb age) is vastly more prevalent than reverse

62  discordance, consistent with a dominance of Pb loss over Pb gain.

63  Linear regression of discordant U–Pb data resulting from episodic Pb loss has been used to calculate

64  concordia intercept ages ever since the Wetherill (1956) and Tera and Wasserburg (1972) concordia

65  diagrams were introduced, and since Pb loss by continuous diffusion was ruled out as the dominant

66  process (c.f. Tilton, 1960). Although this approach is powerful, such discordia regressions become

67  challenging or impossible to fit with any degree of confidence in more complex samples that have

68  undergone multiple phases of radiogenic-Pb loss and/or that contain multiple age populations. In such

69  situations, an approach that considers multiple regressions of discordant data against the concordant age

70  structure may have greater success at resolving the absolute timing of Pb mobility.

71 An important suite of rocks for which zircon U–Pb geochronology has been an essential tool in
72 understanding early terrestrial evolution, but one complicated by U–Pb discordance, is the Acasta Gneiss
73 Complex (AGC), Northwest Territories, Canada. The AGC contains the oldest known evolved rocks on
74 Earth, with zircon U–Pb crystallization ages up to c. 4.03 Ga (Stern and Bleeker, 1998; Bowring and
75 Williams, 1999; Reimink et al. 2014, 2016b). Consequently, these rocks provide important constraints on
76 Hadean to Archean terrestrial evolution, including the timing of formation and early geochemical
77 evolution of the continental crust and depleted mantle (Moorbath et al., 1997; Reimink et al., 2018;
78 Reimink et al., 2014; Reimink et al., 2016a).

79 Several different age components have been identified among the orthogneisses of the AGC: at c. 4.20
80 Ga, 4.03–3.92 Ga, c. 3.75 Ga, 3.65–3.55 Ga, c. 3.4 Ga, and c. 2.95 Ga. The oldest identified components
81 of the AGC, the Idiwhaa Tonalitic Gneisses (ITG), have crystallization ages of 4.03–4.02 Ga (Bowring
82 and Williams, 1999; Mojzsis et al., 2014; Reimink et al., 2016a; Stern and Bleeker, 1998), whereas the
83 oldest analyzed zircon, a xenocrystic zircon core in a younger granitoid sample, is c. 4.2 Gyr old (Iizuka et
84 al., 2007b). These ages have been interpreted to represent times when crustal magmas were added to the
85 AGC (Reimink et al., 2019). Nonetheless, there are significant challenges in making robust age
86 determinations for rocks of the AGC, with issues related to the inherent complexity of its zircon
87 populations. This complexity, that occurs on the scale of individual zircon grains and sub-grain domains,
88 reflects a combination of factors, including different crystallisation ages, the presence of inherited cores,
89 the development of overgrowths and/or resorption during high-grade metamorphism and/or
90 hydrothermal alteration, and discordance of the U–Pb isotopic systems due to multistage Pb loss (in
91 particular early Pb loss; Reimink et al., 2019). These factors, along with physical mixing between rocks of
92 different ages during deformation, and mixing/mingling of coexisting magmas, likely have contributed to
93 the difficulty in assigning accurate and precise ages to rocks of the AGC, in particular its oldest
94 components (Iizuka et al., 2007a). In an effort to resolve these difficulties and understand the timing of
95 radiogenic-Pb mobility, we apply Pb-loss modelling to a formerly unstudied sample of the ITG and
96 compare our results with existing data.

97 Method

98 Two different samples were investigated for U-Pb geochronology and for both samples zircon grains
99 were separated from a crushed sample by picking from a heavy liquid non-magnetic concentrate. Sample
100 CWX was analysed by SIMS as follows: U–Pb analyses were undertaken using the SHRIMP II ion
101 microprobe facility at the John de Laeter Centre, Curtin University. Operating procedures for U, Th, and
102 Pb isotopic measurements using the SHRIMP II ion microprobe are detailed in Wingate and Kirkland
103 (2010). Uranium concentrations and $^{238}$U/$^{206}$Pb calibration were determined using the 91500 reference
104 zircon (Wiedenbeck et al., 1995). Nineteen analyses of the 91500 were obtained during the session and
105 indicated an external spot-to-spot uncertainty (reproducibility) of 0.5% (1σ), and a $^{238}$U/$^{206}$Pb* calibration
106 uncertainty of 0.3% (1σ). Calibration uncertainties are included in both the errors of $^{238}$U/$^{206}$Pb* ratios

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107 and dates listed in electronic appendix Table 1. Common Pb corrections were applied to all analyses using
108 the measured $^{204}$Pb and the contemporaneous isotopic composition from Stacey and Kramers (1975) Pb
109 evolution model. Five analyses of the OG-1 reference zircon were undertaken during the session and
110 yielded a weighted mean $^{207}$Pb*/$^{206}$Pb* date of 3467 ± 5 Ma (MSWD = 1.2), within uncertainty of the
111 accepted age (Stern et al., 2009).

112 Sample AC-10 was analysed by Laser Ablation Quadrupole Inductively Coupled Plasma Mass
113 Spectrometry at the John de Laeter Centre, Curtin University. Zircon grains were ablated using a
114 Resonetics RESOlution M-50A-LR sampling system (incorporating a Compex 102 excimer laser) coupled
115 to an Agilent 8900 ICPMS. After two cleaning pulses, and following 20 seconds of background analysis,
116 samples were ablated for 30 seconds at a 5 Hz repetition rate using a 22 μm spot with laser energy of 1.8
117 J/cm². The sample cell was flushed by ultrahigh purity He (323 mL min$^{-1}$) and N$_2$ (1.2 mL min$^{-1}$). During
118 time-resolved processing, contamination resulting from inclusions and compositional zoning was
119 monitored, and only the relevant part of the signal was integrated. The primary reference material used
120 for U–Pb dating in this study was zircon standard OG-1 (3465.4 ± 0.6 Ma Stern et al. (2009)), which has
121 an ablation response similar to the Archaean grains of interest. Secondary standard zircon 91500 (1062.4
122 ± 0.4 Ma; Wiedenbeck et al. (1995)), GJ-1 (601.7 ± 1.4 Ma; Jackson et al. (2004)), and Plešovice (337.13 ±
123 0.37 Ma; Sláma et al. (2008)) yielded $^{206}$Pb/$^{238}$U ages within 2% of the accepted value when reduced using
124 appropriate matrix matched standards (i.e. using a primary reference material for reduction with similar U
125 content to the secondary reference material). The time-resolved mass spectra were reduced using the U–
126 Pb Geochronology data reduction schemes in IoliteTM (Paton et al., 2011), and in-house Microsoft Excel
127 macros. No common lead corrections were deemed necessary for sample AC-10 due to low $^{204}$Pb counts.

128 Sample details

129 Sample CWX
130 This material was supplied from a specimen in the collection of Crystal World Australia and was
131 collected from 65°10'25.9"N 115°33'12.1"W (Fig. 3) and is a leucosome-rich portion of a plagioclase–
132 quartz–hornblende–biotite–garnet-bearing mafic tonalite. The sample is from the same general
133 location as the ITG (Reimink et al., 2014), although given the complex gneissic layering that
134 juxtaposes several generations of gneisses at a decimetre scale (Reimink et al., 2016), may not directly
135 correspond to the specific component investigated by Reimink et al. (2016). While previous whole-
136 rock major-element compositions for the ITG unit indicate moderate SiO$_2$ (57.9–66.9 wt.%), low
137 Al$_2$O$_3$ (13.8–14.1 wt.%), high total iron (8.6–15.3 wt.% FeO$_t$) and low Mg# (100 × molar MgO/(MgO
138 + FeO = 3–18) (Reimink et al., 2014), the investigated sample is more felsic, reflecting the abundance
139 of leucosome. The sample yielded subhedral to euhedral, brown, turbid, equant to acicular, zircon
140 crystals, which are up to 300 μm long, and have aspect ratios up to 6:1. Most zircon grains are rich in
141 inclusions (mainly of apatite) and show a predominantly low CL-response, with rare blurred indications of
142 what probably represents original concentric growth zoning (Fig. 4). Small patches of homogeneous high

143 CL-response zircon are preserved in cores between low CL-response, inclusion-rich domains. Inclusion-
144 free low CL-response rims overgrow core domains (Fig. 4). All grains show evidence for alteration and
145 metamictization manifest as spongy inhomogeneous portions with residual patches of resorbed and
146 relatively unaltered zircon in the cores of some grains. The altered portion of grains are probably
147 composed of hydrolysed amorphous zircon and zircon crystallites (Rayner et al., 2005).

## Sample AC-10

149 To further understand the veracity of the Pb loss modelling results, we also investigate a sample of Acasta
150 gneiss from the collections of Oxford University. This sample, AC-10, was collected by Prof. Stephen
151 Moorbath on 22nd July 1995 from a locality 150–200 m northwest of the Acasta camp (Moorbath et al.,
152 1997). Sample AC-10 is a quartzofeldspathic gneiss with patches of pink feldspar. In thin section, sample
153 AC-10 is weakly foliated and volumetrically dominated by plagioclase and quartz, with clots and highly
154 discontinuous and irregular thin mafic layers comprising mostly chlorite and epidote. Accessory minerals
155 include apatite and zircon.

## Results

### SIMS (Sample CWX)

158 The analyses are predominantly normally discordant, with only 12 out of 92 analyses (13%) within $2\sigma$
159 analytical uncertainty of concordia (Fig. 5; electronic appendix Table 1). The analyses define a broadly
160 triangular shaped array when plotted on a Tera–Wasserburg diagram, spreading away from both para-
161 Hadean and Paleoarchean concordant components. Importantly, no single time of Pb loss (i.e. single
162 'event') can describe the distribution of all data points. Those analyses that are within analytical
163 uncertainty of concordia range in age from 3994 to 3126 Ma, with probability peaks centred around c.
164 3950 Ma (Group A), c. 3500 Ma (Group B) and c. 3100 Ma (Group C). Nonetheless, the spread within
165 any of these concordant age components can be significant, implying either mobility of Pb or multiple
166 punctuated growth events. The lack of any correlation between U content and age, along with the
167 dissimilarity in U contents between these concordant groups, supports an interpretation of punctuated
168 growth for at least some of the concordant analyses. Additionally, measured versus calculated Th/U
169 (where calculated Th/U is derived from the $^{207}Pb^*/^{206}Pb^*$ age and the measured $^{208}Pb/^{206}Pb$ ratio
170 assuming an undisturbed system) is similar in the concordant analyses, except for a single high f204%
171 analysis (spot ID 41.2) (Fig. 5). This general correspondence between measured and calculated Th/U for
172 Groups A, B and C implies no significant ancient disturbance to the Pb isotope composition of these
173 analysed volumes, and so supports the interpretation that the U–Pb dates record growth rather than
174 alteration (e.g. Kirkland et al., 2017; Pidgeon et al., 2017). A group of five analyses (Group A-1 and
175 Group D-1) located on homogeneous, low CL-response, inclusion-free zircon defines a linear regression
176 with an upper intercept at 3992 ± 5 Ma (MSWD = 0.39) and an imprecise Mesoproterozoic (c. 1440 Ma)
177 lower intercept. These analyses have low common Pb (f204% < 0.04%) and comparable Th/U ratios
178 (~0.69). Analyses of zircon defining the Group B component were located on high CL response core

179 regions that appear texturally similar to the older Group A core regions. Two analyses (spot IDs 41.2 and

180 38.1) from this age group have high common Pb, and a third is located on a region preserving blurred

181 zonation (spot ID 25.1). The remaining four analyses define two weighted mean $^{207}Pb*/^{206}Pb*$ age groups

182 at 3501 ± 6 Ma (MSWD = 0.003; Group B-2) and 3442 ± 5 Ma (MSWD = 0.001; Group B-3). The

183 youngest concordant analysis is situated on a rim and yields a $^{207}Pb*/^{206}Pb*$ age of 3126 ± 6 Ma (Group

184 C-4).

185 The dates of 3992 ± 5 Ma, 3501 ± 6 Ma, 3442 ± 5 Ma, and 3126 ± 6 Ma for concordant groups A/D-1,

186 B-2, B-3, and C-4 respectively, yield the best estimates for the crystallization age(s) of the protoliths to the

187 gneiss and/or the age(s) of metamorphism or recrystallization. Due to the complex initial age structure

188 (Fig. 5), a substantial portion of the U–Pb age data (87 %) is not interpretable within a conventional

189 discordia regression framework (e.g. Kirkland et al., 2017; Morris et al., 2015; Reimink et al., 2016b). Such

190 a framework typically evaluates regression fit parameters (e.g. MSWD) to substantiate the timing of

191 episodic Pb loss (Spencer et al., 2016). This sample illustrates the need for a tool to predict the most likely

192 time of radiogenic-Pb mobility in more complex discordant U–Pb populations.

LA-ICPMS (Sample AC-10)

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194 Seventy-six analyses were undertaken on ten different grains from sample AC-10. The data are mainly

195 concordant with only 19 analyses outside of 2σ analytical uncertainty of concordia, these discordant

196 analyses predominantly appear to have lost radiogenic-Pb, given their normal discordance (Fig. 6). The

197 remaining data scatters along concordia, but there are several discrete age groups associated with specific

198 grain internal textures. The oldest concordant component yields $^{207}Pb/^{206}Pb$ ages between 3630 and c.

199 3500 Ma. These analyses have a general negative correlation between age and U content (younger ages

200 higher U content), consistent with some of the spread on concordia for this age component being a

201 function of ancient radiogenic-Pb loss near parallel to concordia rather than prolonged growth. In any

202 case, the oldest statistically coherent component yields a weighted mean $^{207}Pb/^{206}Pb$ age of 3586 ± 4 (n =

203 24; MSWD = 0.88; Group I), interpreted as the age of magmatic crystallization. These analyses were

204 located on core regions with indications of oscillatory zonation (Fig. 7). Another concordant component

205 clusters around concordia with a weighted mean $^{207}Pb/^{206}Pb$ age of 3396 ± 10 (n = 5; MSWD = 0.58;

206 Group M1), this group is associated with a single euhedral, high length to width oscillatory zoned grain as

207 well as overgrowths. This group is interpreted to reflect partial melting, new zircon growth of discrete

208 grains, as well as modification to pre-existing zircon. Other discrete concordant components have

209 $^{207}Pb/^{206}Pb$ ages of 3225 ± 36 Ma (n = 1; Group M2), 3097 ± 29 Ma (n = 1; Group M3), and 2928 ± 32

210 Ma (n = 2; MSWD = 0.22; Group M4). Group M2 is associated with the core of a grain which has radial

211 fractures, that are interpreted to have facilitated radiogenic-Pb loss, and hence this date likely reflects Pb

212 loss (Fig. 7). Groups M3 and M4 are both associated with edges of grains and are interpreted to reflect

213 periods of metamorphic zircon growth / recrystallization. Those analyses that spread between these

214 groups are interpreted to have undergone variable degrees of ancient radiogenic-Pb loss (Group P).

## Discussion

In order to extract additional information on the post-crystallization history of the AGC from the high proportion of discordant zircon analyses that may not be otherwise apparent from the concordant subset, we explore in detail the U–Pb systematics of sample CWX. We utilize a concordant–discordant comparison test (CDC) (Kirkland et al., 2017) to evaluate the most likely time(s) of radiogenic Pb loss and compare the results of the CDC test for sample CWX to the results of a series of modelled zircon populations of increasing complexity to demonstrate the sensitivity and efficacy of this test.

The CDC test applies a Bayesian approach, by first utilizing the measured age of any concordant population as *a priori* knowledge, and then comparing this age to the upper intercepts of regressions through the discordant fraction of analyses for every possible time of Pb loss. The test makes the implicit assumption that the discordant population was derived from zircon with a similar age structure to the concordant population (Kirkland et al., 2017). For each discordant U–Pb analysis in a population, an upper concordia intercept may be calculated for a given time of disturbance. For each grain and for each successive time of Pb loss, the resultant modelled population can be compared to the concordant population. A number of different non-parametric statistical tests could be employed to compare the concordant age structure with the modelled age populations. Here we use the "likeness" metric of Satkoski et al., (2013), which quantifies the degree of overlap between pairs of probability density plots (PDPs). The "likeness" metric represents the similarity between two unitized PDPs, and is calculated as one minus the sum of the absolute differences between all PDP values divided by two. A similarity value of 1 represents matching concordant and modelled age probability density values, whereas a value of zero indicates dissimilarity. This procedure is then repeated using different assumed times of disturbance, and the similarity function computed across the entire range of possible times of Pb loss. Analysis of this data then defines the most likely time of radiogenic Pb loss, which corresponds to the closest similarity between the concordant and modelled population.

## Concordant–discordant comparison models

In order to investigate both the sensitivity and systematics of the CDC test we ran a range of synthetic datasets subjected to variable degrees and timings of radiogenic Pb loss (Fig. 8).

## Simple models

Models refer to those plots shown in figure 8. **Model A**: To begin, we chose a simple situation of a normally-distributed population of 40 concordant analyses with an age of 4.0 Ga and a standard deviation of five million years. For this population we modelled a single Pb-loss event at 2.9 Ga that caused 30% radiogenic Pb loss in all of the synthetic data. Running the modelled discordant population through the CDC test resolves a single well-defined similarity peak centred at 2.9 Ga. Unsurprisingly, the CDC test performs well in this scenario and is able to accurately determine the timing of Pb loss. The similarity structure of the CDC test function shows a slightly negative skew, which is a function of the shape of the

7

250 concordia curve, whereby a regression through a younger modelled Pb loss time will lead to a shallower

251 slope and greater variance in this slope to the Concordia curve for the next modelled Pb loss time.

252 **Model B**: We next modelled a situation in which half of the population of discordant zircon was subject

253 to a second overprinting radiogenic Pb loss event. The model was structured such that 20 analyses of the

254 original concordant 4.0 Ga population lost 30% Pb at 2.9 Ga, and a second set of 20 analyses underwent

255 two Pb loss events, losing 30% Pb at 2.9 Ga and another 30% Pb at 1.9 Ga. The resultant CDC similarity

256 function resolves the single stage of Pb loss at 2.9 Ga, along with a spurious peak at c. 2.2 Ga, offset

257 some c. 300 Ma from the true timing of the second Pb loss event. Examination of the concordia diagram

258 that corresponds to this model shows clearly why such a spurious secondary similarity peak is present in

259 the CDC test result. The spurious age represents the lower intercept of a secondary regression line joining

260 the primary concordant population and the two stage synthetic data, as though the latter had experienced

261 only single stage Pb loss. Unfortunately, the position of the secondary similarity peak will vary with the

262 amount of, and timing of the overprinting Pb loss such that the true overprinting 1.9 Ga Pb loss event

263 cannot be resolved.

264 **Models C and D:** represent variations on Model B in which higher proportions of analyses have

265 undergone two episodes of Pb loss (80% of analyses in model C and 100% in model D are two stage Pb

266 loss). Model C produced a similar test result to Model B, correctly resolving the first stage of Pb loss at

267 2.9 Ga, but with a larger spurious second-stage Pb loss peak, again at c. 2.2 Ga. Only in Model D where

268 no analyses recording single stage evolution does the CDC approach fail to capture the 2.9 Ga event, nor

269 any of the true Pb loss times.

270 Two population models

271 To further explore the performance of the CDC test in complex scenarios involving overprinting Pb loss,

272 we ran a similar sequence of models involving one or more Pb loss events affecting a mixture of two

273 concordant populations (at 4.0 Ga and 3.5 Ga).

274 **Model E** replicates the simple situation in model A except two concordant components (20 grains with

275 an age of 4.0 Ga and 20 with an age of 3.5 Ga) are each losing 30% Pb at 2.9 Ga. The CDC test returns a

276 well-defined similarity peak centred at the correct time of Pb loss. This model illustrates a situation where

277 the CDC test is much better able to extract useful timing information, as no geologically meaningful

278 single regression fit to all the data is possible using a simple discordia regression.

279 **Models F, G and H** are similar to models B and C, but again with the additional 3.5 Ga age component

280 that has undergone Pb loss. Model F represents a situation where 50% of the 4.0 Ga and 3.5 Ga

281 populations underwent 20% Pb loss at 2.9 Ga and the remaining 50% underwent 30% Pb loss at 2.9 Ga,

282 and then again at 1.9 Ga. Model G represents a situation where even more grains underwent two stages

283 of Pb loss, with 20% of the grains undergoing a single Pb loss event at 2.9 Ga, and 80% of the grains

284 experiencing Pb loss at both 2.9 Ga and again at 1.9 Ga. Model H considers an extreme situation where

285 all grains underwent both stages of Pb loss (i.e. at 2.9 Ga and 1.9 Ga).

286 The CDC test similarity spectra from models F, G, and H are comparable in that they all feature spurious

287 secondary similarity peaks generated from fitting regressions through the two stage Pb loss data points.

288 The secondary similarity peaks increase in height in relation to the proportion of two stage grains to single

289 stage grains. The apparent ages of the similarity peaks correspond to the lower intercepts of regressions

290 through the second stage Pb loss node and the concordant points. Importantly, as with the simple

291 models, the first stage Pb loss event at 2.9 Ga is accurately determined for models F and G, and the CDC

292 test is strongly weighted to resolve single stage events due to the geometry of the Tera–Wasserburg plot.

293 The combined results for models A, B, C, E, F and G illustrate that the CDC test is effective in

294 determining accurately the time of the first stage of Pb loss even when only a very small number of

295 analyses in the data set represent domains that experienced the single Pb loss event without additional Pb

296 loss at a later time. However, the CDC test cannot deconvolve multiple overprinting Pb loss events if

297 there are no such domains retained.

298 Complex models

299 Finally, we present five models to illustrate other features in the similarity spectra generated from the

300 CDC test (Fig. 9). In **Model I** we take the same concordant age structure used in models E, F, G and H

301 and introduce single stage Pb loss at both 2.9 Ga and 1.9 Ga. This results in four batches of 10 grains

302 each that have undergone respectively (i) Pb loss at 2.9 Ga from an initial concordant age of 4.0 Ga, (ii)

303 Pb loss at 2.9 Ga from 3.5 Ga grains, (iii) Pb loss at 1.9 Ga from 4.0 Ga grains and (iv) Pb loss at 1.9 Ga

304 from 4.0 Ga grains. Plus an additional 20 grains from the first stage (2.9 Ga) event are also subjected to a

305 30% Pb loss event at 1.9 Ga in a second stage of Pb loss. The CDC test similarity plot defines a peak at

306 2.9 Ga, successfully identifying this as a Pb loss event, consistent with at least some of the population

307 having undergone a single stage of Pb loss at this time. A broad peak at around c. 1.9 Ga is also defined,

308 consistent with the younger Pb loss event. The breadth of the c. 1.9 Ga peak is partly a function of the

309 length and shallowness of the slope of the projection back to concordia. The implications are that short

310 projections result in greater confidence in the CDC test, and that older Pb loss events will be better

311 resolved by the CDC test provided they are not fully overprinted. The peaks at 2.2–2.3 Ga and 1.1–1.2

312 Ga are spurious and the result of secondary Pb loss lines. It is important to note that the oldest significant

313 peak on the CDC test similarity plot will always reflect a real event if any grains preserve only a single

314 stage of Pb loss.

315 **Model J** presents a more complex situation whereby the 4.0 Ga and 3.5 Ga concordant populations lost

316 Pb during events at 3360 Ma (5% loss), 2900 Ma (10% loss), and 1900 Ma (10% loss), and 10 of the

317 grains that lost Pb at 3360 Ma underwent a second stage of Pb loss at 2900 Ma (10% loss), while another

318 10 underwent a third stage of Pb loss at 1900 Ma (10% loss). The resultant CDC test similarity spectrum

319  is complex, although a few grains preserve a peak at 2.9 Ga, consistent with Pb loss at this time. Many

320  other younger peaks are spurious, as is the long similarity tail towards younger apparent Pb loss ages,

321  which is a consequence of the multiple phases of overprinting Pb loss bringing points from the older (4.0

322  Ga) concordant age group into close proximity (in terms of $^{207}Pb/^{206}Pb$ ratio) with the younger (3.5 Ga)

323  concordant age component.

324  **Model K** applies a geologically realistic scenario and models two episodes of radiogenic-Pb loss of

325  variable intensity from an original concordant 4000 Ma population. The first event at 2900 Ma results in a

326  random amount of Pb loss between 0 and 20%, whereas the second event at 1900 Ma involves a random

327  amount of Pb loss between 0 and 10%. The CDC test similarity spectrum rises to greatest similarity at

328  2900 Ma, successfully resolving the age of the first Pb loss event. However, the second Pb loss event is

329  not determined by the CDC test, as only grains that have seen both events record this overprint. **Model**

330  **L** takes the same population as in Model K with the added complexity of a variable amount of recent

331  radiogenic-Pb loss. Importantly, the CDC similarity spectra is essentially identical to model K, resolving

332  the same first stage of Pb loss age, albeit at a lower similarity.

333  **Model M** depicts variable Pb loss at 2900 Ma and 1900 Ma on 4.0 Ga and 3.5 Ga concordant

334  populations. The degree of Pb loss during stage one is randomly set between 1 and 10 %, whereas during

335  stage two it is randomly set between 0 and 10%. The CDC test yields a similarity spectrum that rises to

336  peaks at about 2.9 Ga and 2.4 Ga with minor >3.0 Ga peaks. The salient point in this model is that the

337  oldest significant similarity peak (as in all other models) approximates the initial time of Pb loss.

338  Pb loss models for the Acasta Gniesses

339  We now apply a series of CDC tests to U–Pb geochronology datasets from the Acasta Gneiss.

340  Uncertainties on the Pb loss model are estimated using bootstrapping (random sampling with

341  replacement). In **comparison one** we apply the CDC test to all analyses from sample CWX that lie

342  beyond 2σ analytical uncertainty from concordia and that have f204 values less than 1%. This includes 76

343  analyses but excludes three analyses with up to 6.5% f204, as the latter may be compromised due to the

344  combined effects of Pb loss and common Pb incorporation. Specifically, their calculated age is overly

345  reliant on the correct common Pb composition having been applied. The test compares this discordant

346  population, modelled through various Pb loss events between zero and 4200 Ma, with the concordant

347  components (i.e. those analyses within 2σ analytical uncertainty of concordia). We exclude two analyses

348  from the concordant group with >1% f204 for the reason stated previously (Fig. 10).

349  The CDC test similarity plot (red line in Fig. 10) rises towards greatest similarity for recent (zero age) Pb

350  loss but also shows significant peaks at 3150 ± 50 Ma and 2870 ± 50 Ma. As per our synthetic CDC

351  results, and assuming that at least some of the analyses are from domains that have only undergone a

352  single Pb loss event, the oldest peak in the similarity structure (c. 3150 Ma) likely reflects a real radiogenic

353  Pb loss event. The long tail on the recent Pb loss peak is comparable with models in which prolonged

354 open system behaviour produced a broad similarity peak. More specifically, the recent broad similarity
355 peak in the CWX data is a consequence of the presence of discordant analyses lying immediately to the
356 right of the Paleoarchean (Group B) concordant component (Fig. 5). Analyses plotting within this region
357 of Tera–Wasserburg space could reflect either recent Pb loss from Group B, ancient Pb loss from the
358 oldest component (Group A), or some combination of these processes.

359 One mechanism to further evaluate this data is to split the CDC test into additional discrete components
360 for comparison (Fig. 10). **Comparison two** evaluates the Group A concordant analyses and those
361 discordant analyses (Group D) with a $^{207}Pb/^{206}Pb$ age older than Group B (i.e. >3450 Ma). **Comparison**
362 **three** compares Group A with those Group D analyses with $^{207}Pb/^{206}Pb$ ages younger than, or the same
363 age as, the Group B component (i.e. with $^{207}Pb/^{206}Pb$ age ≤3450 Ma). **Comparison four** compares
364 concordant Groups B and C with those Group D analyses with a $^{207}Pb/^{206}Pb$ age ≤3450 Ma. The
365 purpose of these additional comparisons is to reduce the effects of multi-stage Pb loss that are manifest in
366 the similarity spectra as apparent recent Pb loss. In comparisons two and three, similarity peaks at 3150
367 Ma and 2870 Ma are again resolved, but with the broad recent peak that was prominent in comparison
368 one much reduced. Comparison four has an entirely different shape, rising to a single similarity peak at c.
369 980 Ma. These additional comparisons support our interpretation that the recent broad similarity peak in
370 comparison A is principally a function of ancient Pb loss from the c. 4.0 Ga component, which has
371 generated $^{207}Pb/^{206}Pb$ ratios close to those of the Paleoarchean concordant component, and is unlikely to
372 mainly reflect recent Pb loss. This interpretation is further supported by the lack of analyses plotting in
373 the region outside (i.e. with higher $^{238}U/^{206}Pb$ than) a discordia stretching from c. 4.0 Ga to 1.4 Ga.

374 It is important to state that the CDC test takes a fundamentally different approach to Pb loss modelling
375 than the method advocated by Reimink et al., (2016b). The Reimink et al., (2016b) method is coded to
376 distinguish discordia *lines* within a dataset, but uses no *a priori* information, rather than *comparing*
377 discordant to concordant components. The Reimink approach has been demonstrated to be well suited to
378 cases where long discordant tails are apparent in complex populations. However, in situations where the
379 degree of Pb loss is not sufficient to generate a distinct tail or there is clusters rather than cords the
380 method rapidly loses resolving power. Applying the Reimink et al., (2016b) algorithm to the CWX dataset
381 results in a broad poorly resolved likelihood spectrum that reaches a maximum around 1800 Ma,
382 essentially between the similarity peaks at zero and the early Archean resolved by the CDC test. A
383 conspicuous shift in low likelihood levels around the c. 3150 Ma CDC test similarity peak is also indicated
384 by this method. This difference in apparent Pb loss timings between these two approaches is likely a
385 result of the clustered nature of the Acasta data, which appears better suited to interrogation using the
386 CDC test.

## Likelihood that the grains were affected by a single Pb loss event
388 It has been shown from the modelling above that a pre-requisite to acceptance that the similarity peaks
389 record real Pb loss events is that some grains, or analytical domains, must have been affected by a single

390 stage of Pb loss. We propose two ways to evaluate this requirement, firstly an analysis of the alpha dose

391 received by the zircon populations and secondly by means of regional CDC comparisons. With regard to

392 alpha dose, if grains were at various stages of metamictization, their susceptibility to Pb mobility would

393 also vary (Nasdala, 1998; Krogh, 1982). Variable susceptibility to Pb loss is conducive to analysis using

394 the CDC test, as the probability of all grains undergoing the same degree of overprinting Pb loss is very

395 low, and hence the influence of geologically meaningless similarity peaks is greatly reduced in real data

396 sets (relative to peaks representing real Pb loss events). A plot of calculated apparent alpha dose for the

397 CWX zircons demonstrates this point (Fig. 11). Sample CWX shows a wide range in the number of alpha

398 events experienced by different domains within the zircon crystals at any given point in time, with most

399 areas having values corresponding to a highly metamict state were they to have remained unannealed

400 since crystallization (Murakami et al., 1991; Weber, 1990).

401 In order to make further comparisons of the timing of Pb loss events affecting the Acasta gneisses, the

402 CDC test has been applied to several additional samples of the ITG. Sample TC-3, a tonalitic gneiss

403 studied by Reimink et al., (2014), contains both unzoned and oscillatory-zoned zircon dated at c. 4.0 Ga,

404 that is interpreted as the age of magmatic crystallization. This sample also contains metamorphic

405 overgrowths and/or recrystallized domains dated at c. 3.2 Ga. Sample TC-3 shows less discordance than

406 other samples of the ITG (95 of 160 analyses are within analytical uncertainty of concordia), prompting

407 the suggestion that this sample largely avoided much of the fluid alteration that affected most other

408 samples of the ITG (Reimink et al., 2016a). The similarity structure for the CDC test of this sample (Fig.

409 10) shows a prominent similarity peak at 3080 Ma, a minor peak at 2590 Ma, and a broad rise towards

410 geologically recent times. The broad rise is comparable to that shown in sample CWX, which has been

411 shown to be, at least in part, a function of ancient Pb loss affecting the older age component, bringing

412 $^{207}Pb/^{206}Pb$ ratios into juxtaposition with those of a younger concordant component. Another previously

413 reported sample of the ITG (sample JR12-141) shows greater discordance (7 of 25 analyses are within

414 analytical uncertainty of concordia) (Reimink et al., 2016a). This sample was analysed by LA–ICP–MS

415 rather than SIMS, and is from a more felsic sample than TC-3 (Reimink et al., 2019). The CDC test

416 similarity spectrum (Fig. 10) has peaks at c. 2870 Ma and c. 2590 Ma along with a broad rise indicating

417 recent apparent Pb loss.

418 On the basis of the repeated appearance of a c. 3150 Ma similarity peak in the comparison test results

419 (tests one to three) for sample CWX, we interpret the similarity peak at c. 3150 Ma to date a real

420 radiogenic Pb loss event. This peak is absent, however, from the CDC test spectrum from sample TC-3

421 which instead features a similarity peak at c. 3080 Ma. The latter is probably also geologically significant

422 given that it is at the older end of the CDC test array for that sample and is thus much less prone to the

423 effects of multiple phases of overprinting Pb loss and geologically spurious similarity peaks. At c. 2870

424 Ma, the oldest similarity peak in sample JR12-141 is also present in the spectra from sample CWX,

425 supporting the interpretation that this peak too represents the timing of a real episode of radiogenic Pb

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426 loss. Comparable CDC test peaks at c. 2590 Ma, in samples JR12-141 and TC3 are also suggestive of a

427 real Pb loss event. However, as discussed previously, the broad rise to geologically recent times in the

428 spectra from all samples cannot confidently be ascribed to single-stage recent Pb loss alone. Further

429 verification of the CDC test Pb loss ages of 3150 ± 50 Ma and 2870 ± 50 Ma from sample CWX is

430 provided by rim ages in sample AC-10, which indicate rims of broadly similar age.

## Implications for regional geology

432 Although some much older (c. 4200 Ma) zircon xenocrysts have been identified within samples of felsic

433 gneiss from the AGC (Iizuka et al., 2006; Iizuka et al., 2007a; Iizuka et al., 2007b), various authors have

434 typical identified a c. 4030 Ma population as the oldest zircon components in layered gneisses of the AGC

435 (Iizuka et al., 2009; Mojzsis et al., 2014; Reimink et al., 2019; Reimink et al., 2014; Reimink et al., 2016a;

436 Stern and Bleeker, 1998). Mojzsis et al., (2014) considered that only c. 3960 Ma magmatic zircon crystals

437 were in chemical equilibrium with their host rock, and hence viewed this age as the most likely time of

438 crystallization for much of the AGC magmatic protoliths. Younger, c. 3600 Ma concordant ages from

439 different AGC samples are attributed to zircon growth during high-grade metamorphic events (Bowring

440 and Housh, 1995; Bowring et al., 1990; Guitreau et al., 2018; Iizuka et al., 2007a; Iizuka et al., 2007b) and

441 also magmatism (Bauer et al., 2017; Reiminik et al., 2019). Subsequent to the emplacement of tonalitic

442 protoliths at c. 4200–3920 Ma (Iizuka et al., 2006), a protracted history of tectono-metamorphic events

443 overprinted the AGC (Bowring et al., 1989; Stern and Bleeker, 1998). Two early Archean thermal events

444 dated at c. 3850–3720 Ma and 3660–3590 Ma involved igneous intrusion, melting and recrystallisation of

445 older units (Bowring et al., 1989; Iizuka et al., 2006; Iizuka et al., 2007b). Later modifications include

446 granitic intrusion and the formation of migmatites, with renewed growth of zircon at c. 3371 Ma (Bleeker

447 and Stern, 1997; Moorbath et al., 1997) and again later at c. 2900 Ma (Stern and Bleeker, 1998).

448 Deposition of supracrustal rocks (the Yellowknife Supergroup) over ancient basement rocks was

449 accompanied by a renewed period of granitic magmatism at c. 2700 Ma, and followed by the intrusion of

450 multiple Paleoproterozoic dyke swarms and c. 1800 Ma bodies of syenite (Firth et al., 1977; Sano et al.,

451 1999; Reimink et al., 2019). This protracted history culminated in regional metamorphism associated with

452 Wopmay tonalite intrusion between about 1800 and 1900 Ma (Bleeker and Stern, 1997; Iizuka et al.,

453 2007b).

454 Some of the interpreted ages of Pb loss identified using the CDC test correspond with geological events

455 known to have affected the western margin of the Slave Province (Isachsen and Bowring, 1994).

456 Specifically, the c. 3150 Ma similarity peak is comparable to the age of a concordant 3126 ± 12 Ma zircon

457 overgrowth (38.2) in ITG sample CWX, and a rim age of 3097 ± 29 Ma, in sample AC-10. The CWX

458 overgrowth mantles a metamict core with an apparent $^{207}Pb^*/^{206}Pb^*$ age of c. 3480 Ma. Around 120 km

459 south east of Acasta River, the Augustus granite, an altered rock consisting of plagioclase, quartz,

460 microcline and chloritized mafic minerals, yielded zircon with an upper intercept age of 3156 ± 3 Ma,

461 interpreted as the magmatic crystallization age (Henderson, 1998). Furthermore, a c. 3150 Ma age peak is

462  evident from an analysis of detrital zircon from the Central Slave Cover Group (Sircombe et al., 2001).

463  The widespread areal distribution of c. 3150 Ma dates suggests that crystalline basement of this age was

464  extensive (Ketchum et al., 2004).

465  Unroofing of the basement components of the Slave Craton by the late Mesoarchean is inferred from the

466  widespread development of a c. 2900–2800 Ma cover sequence (Bleeker et al., 1999a; Bleeker et al.,

467  1999b). This emergence and erosional unroofing has been considered to have occurred in an aggressive

468  $CO_2$-rich atmosphere (Bleeker et al., 1999a; Bleeker et al., 1999b; Sircombe et al., 2001). A causal link

469  between this unroofing and the c. 2880 Ma Pb loss peak is plausible because the zircon in the AGC will

470  have moved into a location accessible to near surface reactive fluids. Furthermore, the similarity peak at

471  $2870 \pm 50$ Ma is comparable to ages of moderately deformed granites at c. 2875 Ma in the vicinity of the

472  AGC (Mojzsis et al., 2014; Stern and Bleeker, 1998), an age also comparible to rim growth in AGC

473  sample AC-10.

474  Beginning at around 2600 Ma, the entire Slave Craton was affected by significant shortening,

475  characterized by broadly north–south structural trends, which arguably developed in response to far-

476  field stresses associated with collision at a distal active margin (Bleeker et al., 1999a; Bleeker et al.,

477  1999b). Moderate overthickening of the crust led to high temperature, low pressure metamorphism

478  and associated widespread anatexis, culminating in extension at c. 2590 Ma and the production of a

479  regional-scale granite bloom. A Pb mobility event at c. 2590 Ma is comparable to the age of a tonalite

480  near Grant Lake in the Wopmay Orogen ($2583 \pm 6$ Ma), some 30 km west of the Acasta River, which

481  correlates with cessation of orogenesis throughout the Slave Province (Jackson et al., 2013).

Process of radiogenic Pb loss in the Acasta Gneisses

483  Comparing the data from different samples within the general area of the ITG, equating to a scale of tens

484  of metres, the dominant Pb loss age as determined by CDC analysis is not consistent (Fig. 10). This

485  inconsistency, if interpreted in a geological framework, suggests that radiogenic Pb loss in the interval

486  3.4–2.2 Ga was highly localised. The Pb loss process appears to render certain rocks and their constituent

487  zircon grains resistant to the next phase of overprinting Pb loss, while other samples remained

488  susceptible. One explanation for this is episodic fluid flow that could have preferentially affected zircons

489  that had reached a critical threshold of radiation damage. The scale of lithological heterogeneity and

490  hence zircon average U content would strongly control the domain size of Pb loss susceptible versus

491  resistant horizons. In some cases, we suggest that such fluid-mediated alteration can be associated with

492  new zircon growth driven by the release of essential elements from the damaged grains (e.g. at c. 3150

493  Ma). Surprisingly, the oldest component within sample CWX (Group A) has the highest calculated alpha

494  dose trajectory, yet is concordant, which indicates either that annealing has occurred, that this component

495  has been impermeable to reactive fluids, or that cryptic Pb loss and gain occurred (e.g. Kusiak et al.,

496  2013). Experimental studies have shown that, depending on the level of initial damage, lattice recovery

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497 can occur at temperatures as low as 427°C (Ming et al., 2000). Additionally, discrete grains or grain

498 domains may be isolated from further fluid interaction by being shielded by zircon overgrowths, thus

499 limiting Pb mobility. In this way a rock, or specific mineral components within it, can be rendered less

500 susceptible to overprinting radiogenic Pb loss than others in close proximity, until a specific interval of

501 time has passed with attendant alpha damage or until dissolution of protective overgrowths has occurred.

502 Fundamentally, such highly-localised cryptic overprinting Pb loss is probably related to initial

503 inhomogeneities in the zircon chemistry, which presumably reflect intrinsic differences in bulk rock

504 composition. Such differences in zircon chemistry would lead to different susceptibilities to radiogenic-Pb

505 loss driven by fluids. Furthermore, it is well known that water by itself is not effective at removing Pb

506 from metamict zircon, but Na or HCl solutions can rapidly strip radiogenic Pb from metamict zircon

507 (Geisler et al., 2002; Nasdala, 1998; Nasdala et al., 2001). Hence, weathering solutions have been

508 proposed as an effective agent of Pb loss (Pidgeon et al., 2017).

509 Zircon grains are multiphase systems with respect to the resistivity of domains with different Th and U

510 content to Pb loss (Nasdala et al., 1996). U–Pb discordance in zircon corresponds with complex internal

511 zircon morphologies, depletion in radiogenic-Pb, U, Si, Zr, Th and HREE, and gain of common Pb, Ca,

512 Fe, Al, LREE and water. Such observations have led to a percolation model of radiation damage whereby

513 initial lattice damage is limited to random spots of $\alpha$ recoil-induced crystal defects. In such a situation,

514 element loss is by volume diffusion requiring high temperatures. After this point, recoil-damaged areas

515 increase in number until they join to form a continuous amorphous network that provides a pathway for

516 rapid fluid and element migration. This point, defined as the first percolation point, corresponds to an $\alpha$-

517 dose of around $3 \times 10^{18}$ $\alpha$-decay events $g^{-1}$ (Salje et al., 1999). As alpha damage progresses, the lattice

518 framework is further degraded resulting in the formation of lattice fragments that are surrounded by an

519 amorphous matrix. This is defined as the second percolation point, and corresponds to an $\alpha$-dose of

520 around $8 \times 10^{18}$ a-decay events $g^{-1}$ (Crocombette, 1999; McLaren et al., 1994; Ríos et al., 2000; Salje et al.,

521 1999). Comparison of time versus calculated alpha dose plots for different zircon components within the

522 AGC, including those which have been measured for their REE contents (Mojzsis et al., 2014), indicates

523 that some of the highest apparent alpha doses occur in the most discordant analyses (Group D; sample

524 CWX), consistent with a percolation model of radiation damage (Fig. 12). However, in sample CWX,

525 Group A zircons show the highest calculated alpha doses, implying complexity in this model potentially

526 through shielding of susceptible core domains by zircon overgrowths or annealing.

527 Many U–Pb analyses from the AGC have been targeted on zircon overgrowths. Many of these analyses

528 have been interpreted as zircon growth and/or recrystallization during younger metamorphic events, the

529 majority within the 3.4 – 3.2 Ga age range (Reimink et al., 2016b). A previously identified large-scale

530 metamorphic/fluid event within the AGC at c. 3.37 Ga (Moorbath et al., 1997; Reimink et al., 2014; Stern

531 and Bleeker, 1998), corresponding to intrusion of large granodioritic-granitic bodies a few km west of

532 ITG, may explain some of these AGC zircon overgrowths. However, it is noteworthy that the CDC tests

15

533 do not indicate that any significant Pb loss occurred during that time interval or during that particular

534 metamorphic/fluid event. Rather, the test results show that significant radiogenic-Pb mobility occurred

535 later in the history of the AGC commencing at c. 3150 Ma and more likely related to episodic highly

536 localised fluid alteration in a strongly heterogeneous rock mass.

## Implications for the evolution of the Acasta Gneiss Complex and early Earth processes

537

538 The AGC represents the oldest continental crust yet identified and contains concordant zircon

539 components with ages between 4.2 Ga and 3.7 Ga. The age of the protoliths that comprise portions of

540 the AGC has been interpreted to be c. 3920 Ma, given the correspondence between zircon trace elements

541 and host rock chemistry (Mojzsis et al., 2014). However, an older component with an age of 4030 Ma is

542 recognised, namely the ITG. Hadean zircons with ages up to 4.2 Ga have been considered to be

543 inherited, generated during earlier episodes of crust formation (Reimink et al., 2014; Reimink et al.,

544 2016a). Such Hadean input into the AGC is consistent with evolved Hf isotopic signatures and negative

545 [142]Nd anomalies (Reimink et al., 2018).

546 On the basis of time constrained zircon oxygen and whole-rock chemistry, Reimink et al., (2016a)

547 proposed that rocks of the AGC formed during 3.94 to 3.75 Ga magmatic events through shallow partial

548 melting of hydrated mafic rocks (i.e., not in the garnet stability field). In addition, the 4.02 Ga ITG, had

549 acquired its negative εHf signature from partial melting of enriched mantle or through interaction with

550 older Hadean mafic crust (Reimink et al., 2019; Reimink et al., 2018). Whole rock geochemistry and

551 petrogenetic modelling of the ITG are consistent with formation by shallow (low *P*) fractional

552 crystallization of low-$H_2O$ basaltic magmas (Reimink et al., 2014). By contrast, Johnson et al., (2018)

553 proposed that moderate degrees of partial melting of hydrated Fe-rich rocks in the uppermost few

554 kilometres of a fractionated, dominantly mafic, Hadean crust was a plausible environment in which the

555 rocks of the IGC formed. Such shallow melting implies geothermal gradients in excess of

556 ~4,000 °C GPa−1, which greatly exceed regional metamorphic rocks of any age worldwide, requiring a

557 significant additional source of heat which was inferred to require bolide impact. Later, at c. 3.6 Ga, the

558 AGC experienced the introduction of isotopically juvenile material that has been proposed as being

559 derived from melting of hydrated mafic crust at significant depths (>40 km) (Reimink et al., 2019;

560 Reimink et al., 2016a).

561 Discordance modelling implies that by 3.1 Ga zircon crystals within the ITG had substantially different

562 propensities to radiogenic-Pb loss. This grain-scale feature is consistent with a heterogeneous

563 composition of the ITG by this stage in its development. Furthermore, to facilitate radiogenic-Pb loss at

564 3.1 Ga implies that the ITG was in an environment where near surface reactive fluids could interact with

565 zircon to strip radiogenic Pb from unannealed crystals. Episodic Pb loss over the course of the next 200

566 Ma is consistent with a modified percolation model of radiation damage where shielding of zircon cores

567 by new growth, and a fluid rich environment can account for the observed zircon U–Pb systematics.

568 Granitoid magmas were emplaced into the AGC at c. 3.4 and c. 2.95 Ga. These felsic rocks have evolved

569 compositions and are interpreted to have been derived through infracrustal partial melting mixed with

570 juvenile additions (Reimink et al., 2019). Regional unroofing around the end of this magmatic period is

571 consistent with further fluid–rock interaction and Pb loss at this time.

## Conclusions

572

573 • SIMS U-Pb geochronology from a hitherto unstudied, legacy, sample of tonalitic gneiss, Acasta

574 Gneiss Complex, yields concordant zircon components implying new growth and/or

575 recrystallization at 3992 ± 5, 3501 ± 6, 3442 ± 5 and 3126 ± 6 Ma. However, significantly, a large

576 component of the zircon population indicates highly disturbed U–Pb isotope systematics

577 consistent with multiple phases of ancient radiogenic Pb loss.

578 • Applying the Concordance–discordance comparison test to the same sample, we establish

579 radiogenic-Pb loss at 3150 ± 50 Ma and likely also at 2870 ± 50 Ma, while other published

580 samples of the Acasta Gneiss also indicate Pb loss at c. 2590 Ma. These Pb loss events

581 respectively correlate with other regional and local geological processes including growth of

582 zircon rims and granite emplacement, unroofing, and late orogenic processes including additional

583 pulses of granite generation.

584 • LA-ICPMS U-Pb geochronology grain mapping on zircon from a quartzofeldspathic gneiss

585 yields magmatic zircon with crystallization age of 3586 ± 4 Ma, and also records either new

586 zircon growth / overgrowth or recrystallization / Pb loss at 3396 ± 10 Ma, 3225 ± 36 Ma, 3097

587 ± 29 Ma, and 2928 ± 32 Ma. Importantly, the latter two of these episodes are within uncertainty

588 of the radiogenic-Pb loss events established by the concordance–discordance comparison test on

589 sample CWX.

590 • Modelling of the sensitivity of the Concordance–discordance comparison test indicates it is most

591 powerful in resolving older periods of radiogenic-Pb loss, but necessitates that overprinting Pb

592 loss does not affect all zircon crystal equally. That is, some grains must remain unaffected by any

593 multistage overprinting processes. Variable recent Pb loss principally serves to reduce

594 concordance–discordance comparison similarity peaks rather than shifting them. Evaluation of

595 calculated alpha doses for zircon grains of the Acasta Gneiss Complex indicate that it is highly

596 unlikely that all grains would become susceptible to radiogenic Pb mobility at the same time and

597 hence the concordance–discordance comparison test is effective in resolving at least the oldest

598 Pb loss event.

599 • Different components of a gneiss will respond differently to fluid alteration events and associated

600 radiogenic-Pb loss, influenced by the dissimilar zircon chemistry. This observation prompts a

601 modified percolation model for zircon radiation damage. On obtaining a metamict crystal

602 structure Pb is able to be effectively stripped from zircon by hydrothermal solutions. However,

603 Pb mobility in some cases appears synchronous with new zircon growth, due to the release of

17

604         necessary elements for zircon precipitation. In such cases shielding of susceptible core domains

605         by zircon overgrowths is feasible and appears to limit Pb loss. Such model is able to account for

606         the highly localised antithetic relationship between zircon Pb loss, where some domains may be

607         losing Pb whereas other domains may be growing zircon and resistant to that episode of Pb loss.

608         A natural consequence of this process is that through time, and alpha dose accumulation, that

609         domains will oscillate between being robust to susceptible and back again to Pb mobility. The

610         scale of the domain is a function of the compositional heterogeneity of the rock mass and hence

611         its zircon cargo.

612     •   On the basis of Pb loss modelling and the necessity of fluids to strip Pb from an unannealed

613         zircon crystal structure, components of the Acasta Gneiss, resided at depths accessible to reactive

614         fluids at least between c. 3.2 Ga to 2.6 Ga.

## Acknowledgements

## Figure captions

619 Figure 1a: Discordance (expressed as the percentage difference between the $^{238}$U/$^{206}$Pb and $^{207}$Pb/$^{206}$Pb

620 ages) versus U (ppm) for a dataset of 34,733 SIMS analyses of zircon. The red curve is a LOWESS

621 (LOcally WEighted Scatterplot Smoothing; Cleveland, 1981) curve fit to the data and highlights a

622 stepwise increase in discordance at higher U ppm. Figure 1b: Discordance (%) versus f204(%) from the

623 same SIMS dataset. f204% is the percentage of common (non-radiogenic) $^{206}$Pb as calculated from the

624 measured $^{204}$Pb/$^{206}$Pb and the estimated contemporaneous composition of terrestrial Pb from the Stacey

625 and Kramers Pb evolution model. The red curve denotes the LOWESS fit which shows a function that

626 diverges to higher common Pb away from zero discordance. Data set available from

627 http://www.dmp.wa.gov.au/geochron.

628 Figure 2a: Zircon age versus percentage discordance of centroid ([$^{207}$Pb/$^{206}$Pb age / $^{238}$U/$^{206}$Pb

629 age]/$^{207}$Pb/$^{206}$Pb age x 100), contoured for data density. Figure 2b: Zircon age versus percentage

630 discordance of error ellipse. Age is based on $^{207}$Pb/$^{206}$Pb ratio for analyses >1600 Ma and otherwise uses

631 the $^{238}$U/$^{206}$Pb ratio. Analyses are TIMS, SIMS and LA-ICPMS, n = 319,147, extracted from Puetz et al.,

632 (2018). Both plots are contoured for data density.

633 Figure 3a: Sketch map of the Acasta Gneiss Complex (AGC) on the basis of age and composition after

634 (Reimink et al., 2014). The yellow star marks the location of the Crystal Word specimen CWX. Figure 3b:

635 Regional map of the Northwestern territories, Canada, with the AGC shown as a white star. The city of

636 Yellowknife is shown as a circle that corresponds with the circle shown in c. Figure 3c: map of Canada

637 showing the location of the Slave Province shown. The box indicates the location of map b.

638 Figure 4: Representative zircon images from CWX, a leucosome from the Acasta Gneiss. The lower

639 image is a false colour BSE image where green represents higher BSE response and higher U content, and

640 blue represents lower BSE response and lower U content. The upper shows an enlargement of grain 38 in

641 both BSE (left) and CL (right).

642 Figure 5: Tera–Wasserburg concordia diagram for zircons from sample CWX. Error crosses are at the

643 two sigma level. Green filled squares denote concordant Group A. Blue filled squares denote concordant

Group B. The red filled square denotes concordant Group C. Grey filled squares (opaque and transparent) denote discordant analyses (Group D). The histogram on the Y-axis is for $^{207}$Pb*/$^{206}$Pb* age. Those analyses defining a c. 4.0 Ga regression discussed in the text are Group A and opaque grey squares in Group D. Inset shows calculated Th/U (based on $^{208}$Pb*/$^{206}$Pb* and age) versus measured Th/U with points scaled for common Pb content.

Figure 6: Tera–Wasserburg concordia diagram for zircons from sample AC-10. Error crosses are at the two sigma level. The histogram on the Y-axis is for $^{207}$Pb/$^{206}$Pb age. Weighted mean $^{207}$Pb/$^{206}$Pb ages are shown in black filled boxes. Grey horizontal bars denote Pb-loss ages as estimated from the Concordant–discordant comparison test on sample CWX.

Figure 7: Gridded $^{207}$Pb/$^{206}$Pb ages from sample AC-10 as a transparent overlay on BSE images.

Figure 8: Concordant–discordant comparison test similarity spectra and corresponding Tera–Wasserburg plots for models A to H. Black filled sections of the similarity spectra are times of Pb loss, grey filled curves are spurious peaks (secondary similarity peaks) caused by multiple phases of Pb loss affecting the same isotopic ratio. First stage Pb loss lines are shown as solid black lines, second stage lines are shown as dashed lines. Growth of secondary similarity peaks due to increase in pervasiveness of overprinting is highlighted by a rising dashed line through models B–D and F–H. Concordant starting material is indicated by the red filled squares. The first stage of Pb loss is shown by the blue filled squares, and the second stage of Pb loss by the green filled squares. Black filled squares are the time of radiogenic-Pb loss.

Figure 9: Concordant-discordant-comparison test similarity spectra and corresponding Tera-Wasserburg plots for models I, J, K, L and M. First stage Pb loss lines are shown as solid black lines, second stage shown as dashed line. Symbols as per figure 8. Brown squares denote third stage of Pb loss. Grey fill in Concordia plot for models K, L and M is cone of discordance due to variable amounts of second stage Pb loss. Red similarity curve denotes model L with recent radiogenic-Pb loss.

Figure 10: Concordant–discordant–comparison (CDC) similarity values for samples of the Acasta Gneiss. Subset test of CWX permits evaluation of the effect of geologically spurious similarity peaks caused by juxtaposition of $^{207}$Pb/$^{206}$Pb ratios by overprinting ancient Pb loss. Also shown is results of the Reimink et al., (2016b) Pb loss modelling method applied to CWX. Note the results of this method are expressed on the secondary Y axis as likelihood values. A probability density plot of zircon crystallization ages in the Slave Craton as recorded in the Canadian Geochronology Knowledgebase (https://www.nrcan.gc.ca) is provided on the upper X-axis of the figure. The shaded confidence bar is a representation of probability of similarity peak for sample CWX being geologically meaningful.

Figure 11: Plot of calculated alpha dose (Murakami et al., 1991) versus accumulation duration for zircon crystals in sample CWX. The first and second percolation thresholds are shown with changes in grey scale (Salje et al., 1999). The yellow data points represent the values for analysed zircon grains/domains in sample CWX assuming alpha damage accumulation from their apparent $^{207}$Pb*/$^{206}$Pb* age without annealing. Inset shows measured Th versus U for concordant zircon.

Figure 12: Plot of calculated alpha dose (Murakami et al., 1991) versus accumulation duration for the average composition of zircon crystals in the Acasta Gneiss. Zircons with REE compositional data are from Mojzsis et al., (2014). The first and second percolation points are shown with a change in grey scale (Salje et al., 1999). Note Group A has significantly higher alpha dose for all durations. Group D and discordant analyses of Mojzsis et al., (2014) are essentially identical.

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## Electronic Appendix

Appendix Table 1: SHRIMP U-Pb zircon geochronology from sample CWX, Idiwhaa Tonalitic Gneiss, Acasta Gneiss, Canada.

Appendix Table 2: LA-ICPMS U-Pb zircon geochronology from sample AC-10, Acasta Gneiss, Canada.
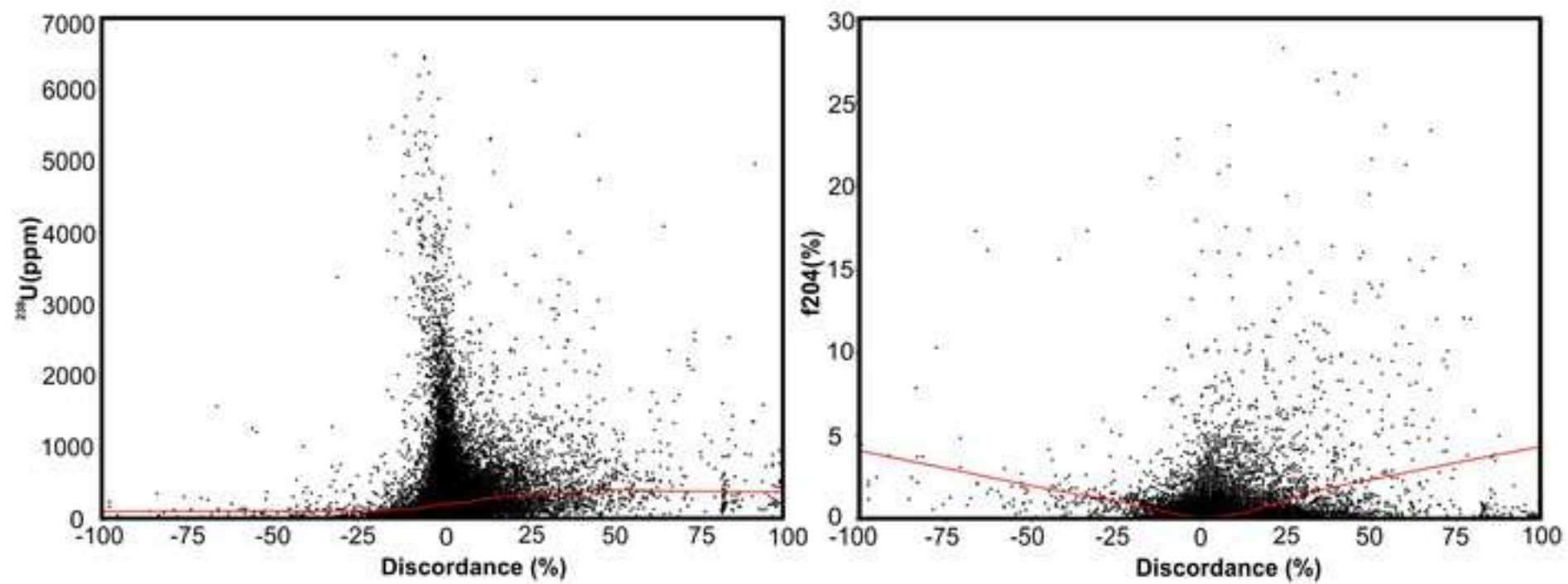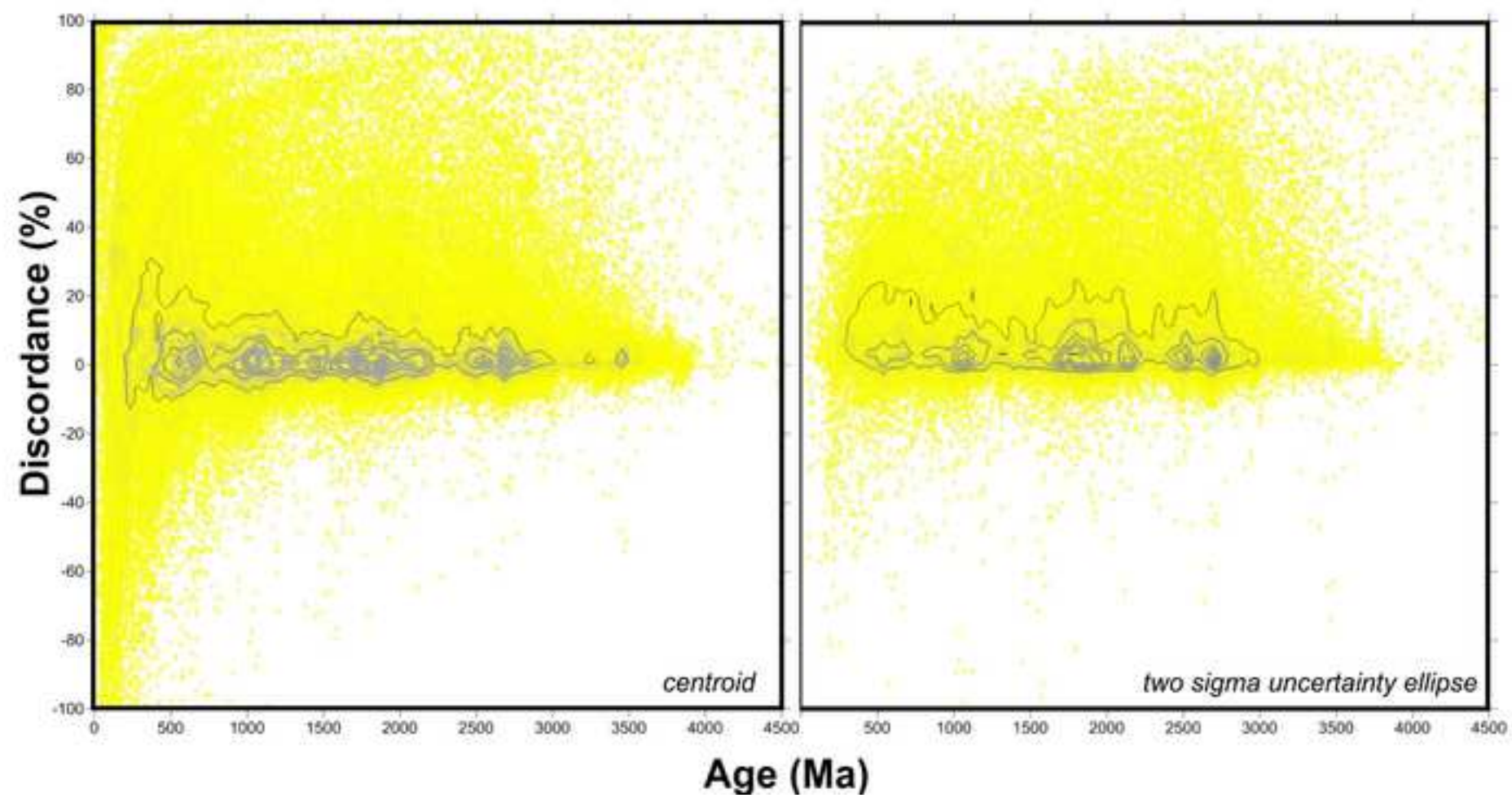
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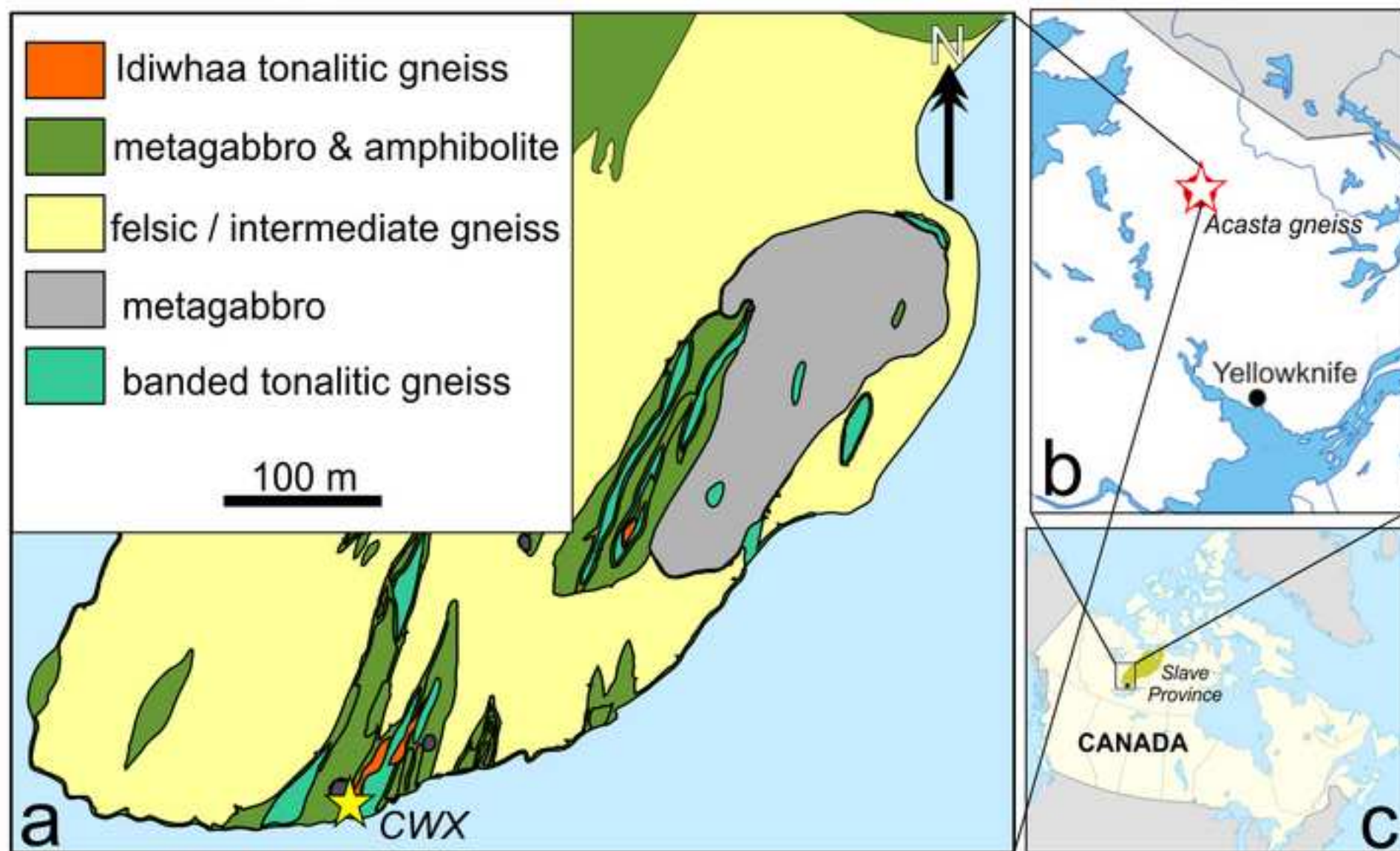847     from the Aduiskii massif, middle Urals, Russia. Mineralogy and Petrology 111, 475-497.