Radiation-enhanced fission track annealing revisited and consequences for apatite thermochronometry

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Abstract

Apatite fission track (AFT) analyses for granitoid and metamorphic bedrock samples from the Western Superior Province (Ontario), the Churchill-Rae Province (Melville Peninsula and Southampton Island, Nunavut), and the Slave Province (Northwest Territories) show a broad range of single grain effective uranium concentrations (eU) (less than 1 to ~300 ppm) and some of the oldest reported AFT ages in North America. Although most of our samples have a typical fluorapatite composition (effective Cl less than 0.1 apfu) with implied low track retentivity, single grain AFT ages are overdispersed and decrease with increasing eU content. This eU-age relationship is resonant of the Hendriks and Redfield (Earth and Planetary Science Letters, 236, 443-458, 2005) argument for α-radiation enhanced fission track annealing (REA) and is analogous to the negative age-eU correlation observed in published zircon and titanite (U-Th)/He data from slowly-cooled cratonic rocks. The high intra-sample age variability for low-Cl bedrock apatites with protracted histories (greater than 200-500 m.y.) at less than 100°C since the Precambrian suggests strong REA control on AFT ages. Conversely, some low Cl AFT samples with a narrower eU range show less age dispersion and a weak apparent age-eU correlation. A complex trade-off between radiation damage and chemical composition (e.g. low Cl and REE enrichment) is implied when eU and rmr0 (and equivalent effective Cl) are correlated. In all cases, the samples fail the canonical χ^2 test to evaluate if grains are from a single age population (χ^2 less than 5%) and have characteristic "open jaw" radial plots, generally considered to indicate multiple age populations. Previous assessments of the influence of REA on AFT age were based on evaluating central age and mean track length, which potentially mask high single-grain age scatter and REA effects. Therefore, it is crucial that bedrock samples exhibiting high age scatter are evaluated in terms of intra-sample compositional heterogeneity. AFT samples with relatively low Cl concentrations are especially prone to greater REA control of cooling ages and this underscores the need for routine acquisition of compositional data for AFT datasets. Our broad range in single-grain AFT ages (with no other clear, strong compositional controls) supports the notion that radiation damage affects both the AFT and (U-Th)/He thermochronometers in slowly-cooled settings and must be accounted for during thermal modeling and interpretation.

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ABSTRACT 8

9 Apatite fission track (AFT) analyses for granitoid and metamorphic bedrock samples from the 10 Western Superior Province (Ontario), the Churchill-Rae Province (Melville Peninsula and Southampton Island, Nunavut), and the Slave Province (Northwest Territories) show a broad 11 range of single grain effective uranium concentrations (eU) (<1 to ~300 ppm) and some of the 12 oldest reported AFT ages in North America. Although most of our samples are characterized by 13 near-endmember fluorapatite composition with implied low track retentivity (<0.1 apfu Cl, r_{mr0}) 14 $\sim 0.85-0.82$), single-grain AFT ages are statistically overdispersed and ages decrease with 15 increasing eU content. This eU-age relationship is resonant of the Hendriks and Redfield (2005) 16 *Earth Planet*. Sci. Lett. **236** (443-458) argument for α-radiation enhanced fission track annealing 17 (REA) and is analogous to the negative age-eU correlations observed in published zircon and 18 titanite (U-Th)/He data from slowly-cooled cratonic rocks. In all cases, the samples fail the 19 canonical χ^2 test (<5%), generally considered to indicate that the ages are unlikely to be drawn 20 from a single Poissonian distribution with a discrete mean value and may represent multiple 21 populations. The high intra-sample age variability for low-Cl bedrock apatites with protracted 22 23 histories (>200-500 m.v.) at <100°C since the Precambrian suggests strong REA control on AFT ages. Conversely, some low Cl AFT samples with a narrower eU range show less age dispersion 24 and a weak apparent age-eU correlation. A complex trade-off between radiation damage, 25 chemical composition (e.g. low Cl and REE enrichment), and thermal history is implied when 26 27 eU and r_{mr0} are positively correlated. Previous assessments of the influence of REA on AFT age were based on evaluating central age and mean track length, which potentially mask high single-28 29 grain age scatter and REA effects due to the modal nature of central age determination. REA is 30 also supported by and compatible with materials science and nuclear waste studies of radiation damage in different apatite groups, therefore it is crucial that bedrock samples exhibiting high 31 32 age scatter are evaluated in terms of intra-sample compositional heterogeneity. AFT samples 33 with relatively low Cl concentrations are especially prone to greater REA control of cooling ages and this underscores the need for routine acquisition of compositional data for AFT datasets. Our 34 35 broad range in single-grain AFT ages (with no other clear, strong compositional controls) 36 supports the notion that radiation damage affects both the AFT and (U-Th)/He 37 thermochronometers in slowly-cooled settings and must be accounted for during thermal history modeling and interpretation. 38

39 Keywords: apatite; fission track; alpha damage; radiation-enhanced annealing; uranium

41 **1. Introduction**

42 Uranium and thorium-rich minerals accumulate radiation damage over geological time, with the primary spontaneous decay process in apatite being ⁴He α -particle emission during ²³⁸U decay 43 (Donelick et al., 2005 for review). The majority of radiation damage results from α -recoil in the 44 apatite lattice, whereas fission events are a minor contribution (for every ~ 2 million ²³⁸U nuclei 45 undergoing α -decay only one fission event occurs, e.g. Donelick et al., 2005). The ion-spike 46 47 explosion model of Fleischer et al. (1965) states that positive ions created by the transmission of highly charged fission fragments repel one another and form a cylindrical region of crystal 48 49 damage. The fission track dating method is based on the accumulation of these crystallographic 50 damage trails due to spontaneous nuclear fission in U-bearing minerals such as apatite Ca₅(PO₄)₃(OH,F,Cl) (Price and Walker, 1963; Wagner, 1968; Naeser and Faul, 1969). Apatite 51 fission track (AFT) thermochronology is established on the principle that crystal damage from 52 53 fission fragments is annealed with increasing temperature, which results in a quantifiable reduction to track lengths and track density in the apatite volume (e.g. Gleadow and Duddy, 54 1981; Green, 1988). However, difficulties remain in fully characterizing the thermally-activated 55 56 annealing behaviour of fission tracks at geologic timescales and the temperature of complete 57 track annealing, as these factors are influenced by duration of heating (Duddy et al., 1988; Green et al., 1986; Green, 1988; Green et al., 1989), variable apatite composition (i.e. common 58 59 fluorapatite vs. chlorapatite; Carlson, 1990; Barbarand et al., 2003; Carlson et al., 1999), and crystal anisotropy (e.g. Donelick et al., 1999; Ketcham et al., 2007; Nadzri et al., 2017). Chlorine 60 61 content is considered the dominant compositional control (Green et al., 1986) on fission-track retentivity when >0.1 atom per formula unit, apfu or >0.35 wt.% (Barbarand et al., 2003), along 62 63 with secondary Ca-site (substituting) cations, such as Fe, Na, Mn, Sr, and Mg, and some rareearth elements (REE) such as La and Ce (Barbarand et al., 2003; Carlson et al., 1999; Ravenhurst 64 65 et al., 2003). However, the trade-offs between kinetic variability, elemental substitutions, and 66 track annealing behaviour are multifaceted and not fully understood (Barbarand et al., 2003; Ketcham et al., 1999; Ketcham et al., 2007). 67

The internal structure of fission tracks in apatite control the annealing process, which is very different from zircon annealing where amorphous tracks "heal" by recombination of vacancies and interstitials in the track interior (Li et al., 2010; Li et al., 2011; Li et al., 2012). Track 71 shortening in apatite occurs due to thermo-emission of vacancies from the porous track core to 72 the adjacent crystalline matrix (Li et al., 2010; Li et al., 2011) and tracks become discontinuous 73 or segmented due to Rayleigh instability, Brownian motion, or motion of track segments due to 74 the high surface energy and diffusivity of voids (Li et al., 2011). To complicate annealing 75 behaviour further, it has been well known for decades that there are small length differences 76 between spontaneous and induced tracks and that apatite FT annealing of up to 10-15% or ~1.5-77 2.5 µm (fading) can occur at ambient temperatures over millions to billions of years (e.g. Crowley et al. 1991; Donelick et al., 1990). 78

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80 In contrast to fission events, α -decay damage accumulates in minerals through energetic α -81 particle exchange by ionization and electronic excitation and by α -recoil "ballistic" collisions 82 with atomic nuclei in the mineral lattice (e.g. Ewing and Weber, 2010; Weber et al., 2015). The α -recoil collisions produce more damage due to the greater destructive interaction with the 83 crystal structure (Holland and Gottfried, 1955). The accumulation of energetic particles in the 84 crystal and a high-energy absorption rate can result in self-heating from α -decay and permanent 85 structural defects can form as a result of energetic electron interactions, which produce electron-86 hole pairs - resulting in charged defects, bond rupture, and increased defect diffusion in 87 88 fluorapatite (Ewing and Weber, 2010; Wang et al., 1994). Damage results in the formation of 89 point defects and defect clustering within the mineral lattice and causes a crystalline-to-90 amorphous transition at high levels of damage (e.g. Ewing et al., 2000; Ewing and Weber, 2010; 91 Weber et al., 2012). The primary α -radiation damage mechanisms include the "direct impact" 92 and "defect accumulation" models (Ewing et al., 2000 and refs. therein). The direct impact 93 model (e.g. Gibbons 1972; Morehead and Crowder, 1972) is predicated on the assumption that 94 an amorphous (non-crystalline) region is formed within the core of a displacement cascade, and complete amorphization is reached when the amorphous domains increase with ion dose, 95 96 eventually occupying the entire sample. The point defect accumulation model (e.g. Gibbons 1972; Jackson 1988) assumes that emitted particles create discrete point defects and that 97 98 amorphization is achieved when the local defect concentration reaches a critical level. For 99 example, the α -decay of U and Th in natural apatites containing REE and Si leads to a 100 crystalline-to-amorphous "metamict" change due to radiation damage accumulation - contingent

101 upon composition, crystalline structure, irradiation level, and temperature (Weber et al., 1997). 102 The general view is that at low temperatures, typical phosphate apatite compositions require less 103 radiation to become amorphized than silicate apatites (e.g. Wang et al., 1994; Weber et al., 104 1997). Durango fluorapatite ($Ca_{10}(PO_4)_6F_2$) becomes amorphized at lower temperatures and at 105 lower radiation dose than silicate-apatite (Ca₂La₈(SiO₄)₆O₂), however at temperatures \geq 77°C, 106 fluorapatite required 5x higher dose to become amorphous than silicate apatite (Wang et al., 107 1994). The critical temperature for amorphization varies depending on the irradiation dose 108 (Ewing et al., 2000), however apatite composition and temperature directly play a role in the 109 persistence of α -radiation damage and the example of increased silica content shows a reduction 110 in the recrystallization tendency of apatite (Ewing et al., 2000). This raises the question of 111 whether and how (other) unusual elemental substitutions known to affect apatite retentivity (e.g. 112 Carlson et al., 1999; Gautheron et al., 2013; Djimbi et al., 2015; Ravenhurst et al., 2003) coupled with slow cooling ($\leq 1-3^{\circ}$ C) and minor, episodic thermal annealing events over billion year 113 timescales play a role in modulating bulk radiation damage accumulation, annealing, and 114 115 recrystallization in apatite.

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117 Radiation damage has implications for multiple diffusion-based thermochronometers, and even 118 though fission track dating is established on constraining crystalline damage, the suspected 119 effects of α-damage on track annealing have been controversial (e.g. Hendriks and Redfield, 120 2005). A drawback of fission-track dating with respect to visualizing damage is that the process 121 of increasing the optical visibility of tracks requires chemical etching, which erases the initial 122 damage structure and essential information about the physical scale of radiation damage (Afra et al., 2011). Ion tracks are similar in character to fission tracks (Rabone et al., 2008) and ion track 123 124 annealing experiments from swift heavy ion bombardment in apatite verify unit-cell increases during irradiation, crystal strain and cracking at high ion fluence, high-pressure reduction of 125 amorphization, radiation-enhanced diffusion, and anisotropic lattice damage response (e.g. Liu et 126 127 al., 2008; Miro et al., 2005; Miro et al., 2012; Weikusat et al., 2011). Latent ion particle tracks in 128 Durango apatite were found to anneal via a two-stage process of structural relaxation followed 129 by recrystallization (Afra et al., 2011), which has also been observed in silicate apatite (Bae et 130 al., 2007). Ionization-annealing of ballistic damage in SiC revealed a surprisingly low threshold

for electronic energy loss (a few MeVs) to initiate annealing at room temperature and repair
defect clusters, crystalline structure, and displacements at the atomic level (Zhang et al., 2015).
Carpéna (1998) proposed that annealing in low-temperature conditions is also possible for
silicate apatite (fluorbritholite) with lanthanide enrichment (e.g. La, Ce) or cation substitutions
such as Na, Mg, Sr, Mn, Fe, U, and Th.

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Radiation damage is often discussed in terms of "alpha dose," which is a quantitative estimate of 137 the self-irradiation (driven by the α -fluence) in a mineral from time-integrated damage 138 139 accumulation (e.g. Nasdala et al., 2001). The first-order control on α -damage is U content, or 140 "effective U" concentration (eU=U+0.235×Th+0.0046×Sm; Gastil et al., 1967) of the host grain 141 weighted for parent α -productivity. Extreme α -damage levels cause crystallographic 142 amorphization in zircon due to relatively high U content (Chakoumakos et al., 1987; Murakami 143 et al., 1991; Nasdala et al., 2001), whereas α -damage is more easily recoverable in apatite 144 (Weber et al., 1997; Li et al., 2017). (U-Th)/He dating of zircon is an excellent example of a 145 mineral demonstrating a relationship between diffusivity and α -dose/eU. The expectation is that 146 a positive age-eU correlation exists at low dose and a negative relationship is evident after the 147 damage percolation threshold (or point at which damage interconnects) is reached, causing increased diffusivity (Guenthner et al., 2013). There is an expected decrease in ⁴He diffusivity at 148 the α -recoil percolation dose of ~2.5-3.1×10¹⁶ α/g and then an increase in diffusivity at fission 149 track percolation achieved at a higher damage level of ~ 1.9×10^{18} α/g (Ketcham et al., 2013). The 150 zircon radiation damage and accumulation model (ZRDAAM; Guenthner et al., 2013) predicts a 151 152 complex diffusive relationship between damage accumulation/annealing (eU) and derived He 153 age that is strongly influenced by thermal history. Self-irradiation over geological timescales 154 results in high α -doses above the percolation point and a negative age-eU relationship should 155 become apparent in grains that have accumulated high radiation damage and spent significant time below the He partial retention zone. This age-eU pattern is achieved either through He loss 156 157 at low temperatures or alternatively, rocks spending considerable time at low temperatures and 158 experiencing a thermal pulse during their history (Guenthner et al., 2013). The model for zircon 159 radiation damage has been recently improved upon to show that bulk radiation damage annealing 160 requires longer durations and higher temperatures than fission track annealing, and when fission

161 tracks are fully annealed, the total radiation damage has only been annealed \sim 30-50% (Ginster et 162 al., 2019). Fission track annealing characteristics between zircon and apatite are different (Li et 163 al., 2011), but α -recoil damage is known to diminish fission track thermal stability in U-bearing phases such as zircon (Garver et al., 2004; Kasuya and Naeser, 1988), titanite (Lumpkin et al., 164 165 1991), and apatite (Carpéna and Lacout, 2010; Ritter and Märk, 1986). These findings imply that 166 the presence of α -damage lowers the stability of fission tracks in minerals commonly used in 167 thermochronology and that α -damage can persist above temperatures typically believed to 168 completely anneal fission damage.

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The apatite (U-Th)/He (AHe) thermochronometer is affected by radiation damage (e.g. Shuster et 170 al., 2006; Shuster and Farley, 2009). There are consequences for He diffusion in a damaged 171 172 mineral lattice, with the main assertion being that radiation-induced damage impedes helium 173 diffusivity and that damage evolves as a function of temperature and eU concentration (e.g. Farley, 2000; Flowers et al., 2009; Gautheron et al., 2009; Ketcham et al., 2017; Recanati et al., 174 175 2017; Shuster et al., 2006; Shuster and Farley, 2009). Radiation damage effects on He retentivity 176 have been explored and described through models of diffusive loss (Shuster et al., 2006; Flowers 177 et al., 2009; Gautheron et al., 2009; Gerin et al., 2017; Willett et al., 2017). There is now a better 178 understanding of the numerous factors affecting He diffusivity in apatite including compositional 179 and crystallographic controls on α -damage annealing (Djimbi et al., 2015; Gautheron et al., 180 2013; Gautheron et al., 2009), crystallographic microvoids (Zeitler et al., 2017), vacancy damage 181 (Gerin et al., 2017), and strain-induced dislocation traps (McDannell et al., 2018b). The α -182 damage recoil track percolation threshold has been recently described for He diffusivity in 183 apatite (Ketcham et al., 2017; Recanati et al., 2017), and seems genetically similar to damage 184 modes in zircon. A similar age- α -dose relationship has been observed for titanite (Baughman et 185 al., 2017; Guenthner et al., 2017), however a titanite radiation damage model has yet to be 186 formally described. Due to the difficulty in direct damage observation in past studies and in 187 determining α -recoil kinetics, α -damage annealing in apatite has long been considered analogous 188 to fission track annealing behaviour, and the most commonly used empirical diffusion models of 189 damage accumulation and annealing are established on this principle (Flowers et al., 2009; 190 Gautheron et al., 2009). However, it has been previously proposed that the rate of α -damage 191 annealing is slower than fission-track annealing (e.g. Ritter and Märk, 1986), which would

require revision of existing models. The dilemma is that we really do not know what the kinetics are for α -damage annealing, and most importantly in this context, we should not assume that it is the same for both damage types. We have done so for the purposes of trying to model damage effects on He diffusion in apatite, but with arguably limited success especially in geologic settings where the kinetics for each damage mode diverge.

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198 The developments in understanding how α -damage acts to modify He diffusion are integral for 199 using apatite as a viable thermochronometer and undoubtedly have implications for AFT analysis 200 and fission-track retention in rocks that have experienced slowly-cooled histories. The relatively 201 subtle kinetic and compositional controls on apatite annealing and retentivity in rapidly-cooled 202 settings become more complex and magnified in cratonic interiors due to their protracted thermal 203 histories at low temperatures (e.g. Green and Duddy, 2006; Flowers, 2009). There have been 204 many problems associated with the interpretation of both AFT and AHe data in slowly-cooled 205 settings, including significant intra- and inter-sample age dispersion (e.g. Fitzgerald et al., 2006; 206 McKeon, 2012; McDannell et al., 2018b) and AFT<AHe cooling age inversion (e.g. Hendriks 207 and Redfield, 2005; Green et al., 2006; Flowers and Kelley, 2011). Hendriks and Redfield (2005) 208 suggested that elevated U concentration in apatite has a strong influence on fission-track 209 annealing, invalidating prior thermochronologic interpretations for cooling signals across the 210 Fennoscandian Shield. In a series of papers, this matter and the robustness of AFT versus AHe 211 dating was debated (Green and Duddy, 2006; Hendriks and Redfield, 2006; Larson et al., 2006). 212 Hendriks and Redfield's argument for AHe dates being more reliable than AFT ages was 213 contested and attributed to variability in He retentivity, rather than variability in AFT annealing 214 behaviour (Green et al., 2006). The apparent U control on AFT ages was instead attributed to 215 lithologic differences and elevated Cl content for the analysed apatites (Kohn et al., 2009).

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The effect of U on AFT ages was perhaps dismissed prematurely, and analytical advances in the past 15 years have motivated changes in AFT methodology to better address this problem. The potential relationship between U and AFT age has long been recognized and an early example of a negative correlation between zircon FT age and U concentration was given in Carter (1990) using the external detector method (EDM). Discussion around this relationship focused on

222 single-grain U variability, track density, and potential counting biases, as well as the fact that 223 both AFT age and U estimates are derived from the same induced track count, and therefore related. The recommendation in Carter (1990) for determining if a real association exists 224 225 between age and U is to measure U independently. The introduction of laser ablation inductively 226 coupled plasma mass spectrometry (LA-ICP-MS) for AFT dating (Hasebe et al., 2004) made the 227 direct measurement of U procedurally advantageous over the EDM, which uses an irradiated, low-U muscovite detector as a proxy for apatite ²³⁸U content through reactor-induced fission. 228 The main drawback for ICP-MS 238 U measurement is that very low U content (<< 0.5 ppm) is 229 230 difficult to measure and leads to modestly older ages compared to the EDM (Seiler et al., 2013), nonetheless U can still be reliably measured at <1 ppm. It is also well established that apatites 231 232 with high fission-track density (i.e. old grains or high U) are more difficult to measure using the EDM, consequently biasing AFT measurements towards younger or low U grains (e.g. Carter, 233 1990; Seiler et al., 2013). In a series of analyses on laboratory age standard AFT grains, Hasebe 234 235 et al. (2004) showed that the EDM underestimated the U content of high U, whole-grain 236 laboratory age standard apatites with respect to LA-ICP-MS measurements.

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In light of advances in precise, in situ U concentration measurements for AFT analysis, re-238 evaluation of α -radiation enhanced annealing (REA) is warranted. Assessing the viability of 239 240 REA in slowly-cooled terranes is important for the understanding of the annealing kinetics of 241 apatite fission tracks and the implications for other thermochronometers that are affected by the same α -damage and annealing processes, such as the case with the apatite (U-Th)/He system 242 243 (Gautheron et al., 2013; Gautheron et al., 2009; Recanati et al., 2017; Shuster et al., 2006). We present new LA-ICP-MS AFT data from Archean-Paleoproterozoic plutonic granitoids and 244 245 gneissic bedrock samples from localities across the Canadian Shield and assess the REA potential related to U content. In general, grains that are characterized by highly variable eU and 246 247 low Cl concentrations (<0.1 apfu) typically show a strong, negative age-eU correlation, whereas 248 contrasting metasedimentary samples from southern Baffin Island with "typical apatite" eU 249 levels (less variability, ~20-30 ppm) and low Cl (<0.1 apfu) show seemingly weak U control on 250 age, less single-grain age scatter, or display complex relationships between retentivity proxies 251 (i.e. eU and r_{mr0}).

Our intent is not aimed at generating a calibrated model of REA behaviour, nor wholly reconciling observations between AFT and AHe single-grain ages in slowly-cooled settings for geologic interpretation – but rather to bring REA back into the communal foreground and assert that this phenomenon exists, although it is poorly understood due to complexities associated with fission-track annealing.

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259 2. Regional Geologic Setting

260 The AFT samples presented here all come from exposed Precambrian granitic and gneissic 261 bedrock across Canada. An overview of the Precambrian geologic history of the Canadian Shield 262 is provided by Hoffman et al. (1989) and here we focus mainly on areas of Archean (>2.5 Ga) 263 crust that are currently exposed across the Canadian interior that were sutured during Trans-264 Hudson orogenesis and Laurentia assembly at ca. 1.9-1.8 Ga. The major terranes that were 265 sampled are the Western Superior Province (Percival et al., 2012), rocks of the northeastern 266 Churchill Province-Rae domain at Southampton Island (Berman et al., 2013) and Melville 267 Peninsula (Berman et al., 2015), and the Slave Province (Isachsen and Bowring, 1994) all shown 268 on figure 1. Results from southern Baffin Island are also shown for comparison to the 269 aforementioned data (see discussion).

270 The Archean-Proterozoic tectonomorphic history and setting for each terrane is thoroughly 271 described in their respective reference. All of these regions have presumably been exposed at near surface conditions or in the upper crust since the late Proterozoic and experienced minor, 272 273 episodic burial throughout the Phanerozoic (Burgess, 2008; Miall and Blakey, 2008). The 274 Archean-Paleoproterozoic age of these terranes and the limited Phanerozoic sedimentary strata 275 provide few constraints on the burial and exhumation history of these rocks. The Phanerozoic 276 sedimentary succession in central Canada is preserved in Hudson Bay (mainly Paleozoic, ~2500 277 m thick; fig. 1) and unconformably overlies Canadian Shield bedrock (Pinet et al., 2013). The 278 Paleozoic platform sequence is exposed in the Western Superior Province at Hudson and James 279 Bay, and Ordovician-Silurian sediments onlap much of the southern Arctic, northern Melville 280 Peninsula (Corrigan et al., 2013), and Baffin Island crystalline bedrock (fig. 1).

282 In the absence of formal models for the time-temperature history of many of these areas, 283 geologic evidence and the sparse Phanerozoic sedimentary record yield clues about the thermal 284 history experienced by Shield rocks. Kimberlites can include sedimentary xenolith evidence of the pre-existing Paleozoic cover that was removed by erosion. The Jericho kimberlites in the 285 286 central Slave craton have limestone xenoliths of Middle Devonian age (L. Eifelian-E. Givetian) 287 (Cookenboo et al., 1998) and the ca. 75-45 Ma Lac de Gras kimberlite field suggests up to ~1.4 288 km of Cretaceous strata existed at that time (Nassichuk and McIntyre, 1995; Stasiuk et al., 2006). 289 The Mesozoic Kirkland Lake (Ontario) kimberlites contain Ordovician-Devonian xenoliths in 290 the Superior Province (McCracken et al., 2000) and the Jurassic Chidliak kimberlites of southern 291 Baffin Island also contain Late Ordovician-Early Silurian xenoliths (Zhang and Pell, 2014). 292 These areas all indicate that most of interior Canada was inundated by a shallow sea in the middle Paleozoic and experienced some level of regionally cohesive burial and exhumation 293 294 through Paleozoic-Mesozoic time, which is reinforced by regional studies using low-temperature 295 thermochronology (Ault et al., 2009; Ault et al., 2013; Feinstein et al., 2009; Kohn et al., 2005). AFT studies suggest that most of the Trans-Hudson region remained at <80-90°C since the 296 Ordovician (Pinet et al. 2016) and that rocks of the Superior Province in Ontario were never 297 298 hotter than ~90°C during episodic burial and exhumation since the Cambrian (Kohn et al., 2005). 299 Generally speaking, the majority of the Canadian Shield has been at temperatures <100-120°C at 300 least since the latest Precambrian-Cambrian (~600-500 Ma).

301 **3. Analytical Methods**

302 3.1 LA-ICP-MS apatite fission track analysis

303 AFT ages were determined using the LA-ICP-MS method (Hasebe et al. 2004; Donelick et al. 304 2005; Chew and Donelick 2012), which gives similar results to the traditional external detector 305 method but avoids the requirement for sample irradiation in a nuclear reactor (e.g. Seiler et al., 2013). A single grain mount was used to acquire AFT age, length, and D_{par} data for each sample 306 307 according to the procedures described in Donelick et al. (2005). Following mineral separation 308 procedures, apatite separates were mounted in epoxy, polished, and etched in 5.5M HNO₃ for 309 20s at 21°C to reveal all natural fission tracks intersecting the polished grain surface. For each 310 age grain, locations were recorded and spontaneous AFT densities were counted using a microscope at 2000x magnification under unpolarized light.²³⁸U concentrations were determined 311

312 for the track count areas on each age grain using the Washington State University (WSU) Finnigan Element II Magnetic Sector ICP-MS by measuring the weighted mean ratio of ²³⁸U to 313 ⁴³Ca from multiple spot analyses at a fixed laser point (²³²Th and ¹⁴⁷Sm were also measured). 314 The volume of ablated material is estimated using ⁴³Ca, assuming that Ca occurs in 315 stoichiometric amounts. AFT ages were calculated using the LA-ICP-MS ζ -calibration approach 316 (Donelick et al., 2005; Vermeesch, 2017) based on the Durango apatite age standard. Apatite U-317 318 Pb age data were acquired for AFT age grains as described in Chew and Donelick (2012). Apatite U-Pb age density plots were created using DensityPlotter v. 8.2 software (Vermeesch, 319 320 2012) using a Kernel Density Estimator with an adaptive bandwidth and logarithmic 321 transformation of the single-grain common Pb-corrected isotopic sum ages and their 2σ errors. 322 The quoted "mixture model" ages (table 1) incorporate all measured isotopic sum ages and are 323 for a single age population.

324 3.2 Apatite elemental analyses and the "effective" Cl kinetic parameter

325 Sample apatite grains were analysed for 13 elements (F, Na, Mg, P, S, Cl, Ca, Mn, Fe, Sr, Y, La, and Ce) by electron probe microanalysis (EPMA) at the Washington State University Peter 326 327 Hooper Geoanalytical Laboratory using a JEOL JXA8500F Field Emission Electron Microprobe 328 operated at 15 kV (20 nA current) with a beam size of 5 µm. Laboratory weight % oxide values 329 were converted to atom per formula unit (apfu) values, including estimation of OH content from Cl and F values, using the apatite stoichiometric model of Ketcham (2015). The r_{mr0} kinetic 330 331 parameter was calculated for each analysed apatite grain by substituting the apfu values into the 332 multivariate equation of Carlson et al. (1999). The nonlinear rmr0 values were converted into "effective" Cl (eCl) values (in apfu) using the empirical r_{mr0} -Cl relation of Ketcham et al. (1999) 333 334 for better visualization of data on a linear scale, for calculating arithmetic means of single-grain 335 kinetic parameters, and for comparison with the commonly used measured Cl kinetic parameter. D_{par} (mean etch pit diameter perpendicular to c-axis; e.g. Donelick et al., 2005) data are not 336 discussed in detail here. However, D_{par} has been found to be a lower precision kinetic proxy 337 when compared to r_{mr0} or measured Cl (Issler et al., 2018), and in other localities across Canada 338 339 it has demonstrated limited utility in successfully separating apatite grain age populations (e.g. 340 Issler and Grist, 2008; McDannell et al., 2019; Powell et al., 2017; Schneider and Issler, 2019). In the majority of our samples, measured Cl and D_{par} show no relation with AFT age and in 341

342 general, eCl is preferred as a kinetic parameter over measured Cl because it takes into account the contribution of multiple elements that influence track retentivity (e.g. Barbarand et al., 2003; 343 344 Carlson et al., 1999). Negative eCl values ($r_{mr0} > 0.84$) indicate that AFT track retentivity is 345 lower than that of the apatite used for the laboratory annealing experiments (Ketcham et al., 346 1999). We use the Carlson et al. (1999) r_{mr0} equation rather than the Ketcham et al. (2007) 347 equation because in our experience with multi-kinetic AFT populations the former generally 348 produces a broader range of r_{mr0} values than the latter equation for apatites displaying a range of kinetic behavior. The greater spread in values can result in better discrimination between kinetic 349 350 age populations. Ketcham et al. (2007) demonstrates that both experimental datasets can be reconciled but this does not necessarily mean that the latter equation is superior. Refitting of the 351 352 model parameters in the newer equation may be influenced by: (1) the narrower compositional range of the Barbarand et al. (2003) experimental apatite compared with those of Carlson et al. 353 354 (1999), which may be biasing the fit due to the overriding influence of Cl and (2) there may be 355 residual analyst bias between experiments even after corrections were applied.

356

357 **4. Apatite fission track results**

358 We present new LA-ICP-MS AFT results for 12 samples taken from cratonic crystalline bedrock 359 across the Canadian interior (table 1; refer to the Supporting Information [SI] for all raw 360 analytical data). Two other new samples from Baffin Island are not from crystalline bedrock, but 361 are from Paleoproterozoic metasediments overlying Archean basement that have internally 362 consistent apatite U-Pb ages and are treated as homogeneous samples. The Baffin Island AFT, apatite U-Pb, and EPMA datasets (McDannell et al., 2018a) are available from 363 http://dx.doi.org/10.1594/IEDA/111241. There is also an additional AFT sample with 364 365 accompanying AHe data previously published for the Hudson Bay region in Pinet et al. (2016) 366 that is examined in the discussion (see table 1). Published AFT data from Canada and other areas 367 worldwide are included for comparison only to illustrate other types of complex relationships 368 between AFT single-grain age, eU/α -dose, and other kinetic parameters (see discussion). We 369 want to emphasize that this dataset is complex, and in specific cases this complexity may 370 preclude thermal history interpretation (at this time) without a proper understanding of the 371 kinetics governing REA or at the very least, differences between fission track and α -damage

annealing rates. However, we discuss some possible ways forward when dealing with AFT and
 accompanying elemental data from slowly-cooled terranes, which may serve as a framework to
 assist in future interpretation.

375

376 A plot of central AFT age/MTL versus eU is shown in figure 2A, B. This visualization of AFT data (generally utilizing ²³⁸U only) has been the convention for previous studies addressing REA 377 (e.g. Hendriks and Redfield, 2005; Kohn et al., 2009). However, central age plots are non-ideal 378 379 for displaying age-eU relationships because of the modal tendency of this metric, which masks 380 high age scatter between individual grains. In our case there is large geographic distance between 381 sample locations and discrete thermal histories should be expected for each sample, yet there is a 382 consistent decrease in age with increasing eU. Our apatites are relatively mono-compositional. 383 Apatites characterized by >0.35 wt. % Cl (>0.1 apfu) are considered structurally controlled by Cl 384 and imply higher fission track retentivity (Barbarand et al., 2003), whereas our data are almost 385 entirely low retentivity F-apatite (fig. 2C) or from apatite varieties with greater hydroxyl and Ca-386 site cation substituents (e.g. REE-rich F-apatite) There is also the possibility that a very minor apatite component (<10 grains total) is in the belovite or britholite groups based on wt. % P₂O₅ 387 (Pan and Fleet, 2002). Radial plots are useful for assessing mixtures of single-grain ages and 388 AFT age precision (Galbraith, 1990). All of our samples fail the χ^2 test when plotted on radial 389 plots, indicative of high age dispersion (fig. 3). Detrital AFT data often fail the χ^2 test due to 390 391 variable provenance (and composition), however for slowly-cooled bedrock samples failure may 392 be expected, $d \square \square \square \square \square$ the range of single grain kinetic responses to annealing under slow-393 cooling conditions (Galbraith and Laslett, 1993). An interesting feature of these data overall are that samples with fewer grains typically have much lower dispersion (fig. 3), which is probably a 394 result of "under-sampling" and is an issue that may be unwittingly pervasive in older vintage 395 AFT datasets from slowly-cooled terrains. 396

397

Fission track length distributions for each sample are shown in figure 4, and in the absence of thermal modeling, can be qualitatively assessed to understand thermal history complexity to first order. Length distributions are presented as frequency (%) in 1 μ m bins with equivalent normal distributions shown for the mean $\pm 1\sigma$ (solid, blue curve) and the mode (dashed, red curve). The 402 track length distributions are mostly unimodal and shortened (centered at $\sim 12-13$ µm), signifying 403 monotonic slow cooling or slow cooling and reheating (partial annealing) to ~65-80°C (Gleadow 404 et al., 1986). Bimodal distributions, skewed distributions, or those with 'shoulders' can signify 405 single-phase reheating to higher temperatures of ~80-100°C or episodic reheating in the partial 406 annealing zone (~60-110°C; Gleadow et al., 1986). Baffin Island sample 09SRB-M100 and Melville Peninsula sample SNB-01-M2055 are examples that exhibit "out-of-phase" mean and 407 408 mode distributions (fig. 4), which is indicative of bimodality and implies multi-stage thermal 409 histories (ignoring differences in *n* tracks). These areas are both characterized by geologic and 410 thermochronologic evidence of Paleozoic and/or Mesozoic burial (McDannell et al., 2019; Pinet et al., 2016). 411

412

We discuss the geologic context and other relevant high-temperature dating performed on each sample (when applicable) for each Canadian AFT sample below. All samples have summarized AFT information in the text and table 1. Likewise, all published samples are presented and discussed with known information relevant to their histories.

417 4.1 Western Superior Province

Sample PBA-98-743 is from a biotite tonalite-gneiss in the Harmon Lake Gneiss Complex of the 418 Wabigoon Subprovince. The gneiss has a mean ²⁰⁷Pb/²⁰⁶Pb zircon core metamorphic protolith 419 age of 2890 ± 8 Ma (MSWD=1.1) and a U-Pb titanite age of 2678 ± 2 Ma (Percival et al., 2004). 420 421 The corresponding sample apatite U-Pb mixture model single age peak is 2971 ± 174 Ma (2σ ; n=25). The AFT sample has a pooled age of 572.3 ± 15.6 Ma (1 σ ; n=40) and a central age of 597 422 423 \pm 37 (1 σ) with 36% age dispersion. The mean track length (MTL) is 12.07 \pm 1.94 μ m (1 σ ; 424 n=130). The mean kinetic parameters for PBA-98-743 are: measured D_{nar} of 1.89 μ m, eU of 7 ppm (range 1-19 ppm), calculated r_{mr0} of 0.84, and measured Cl of 0.008 apfu. 425

426 Sample 03-GRS-013 is from a tonalite-quartz diorite in the Mesoarchean Sachigo Subprovince. 427 The apatite U-Pb mixture model single age peak is 3009 ± 94 Ma (2σ ; n=29). The AFT sample 428 has a pooled age of 414.0 ± 14.1 Ma (1σ ; n=40) and a central age of 447 ± 34 (1σ) with 46% age

dispersion. The MTL is $12.51 \pm 1.63 \mu m$ (1 σ ; n=131). The mean kinetic parameters for 03-GRS-

430 013 are: measured D_{par} of 1.91 μ m, eU of 33 ppm (range 1-241 ppm), calculated r_{mr0} of 0.84, and 431 measured Cl of 0.003 apfu.

432 4.2 Slave Province

Sample 12-DRA03-001 is from a plagioclase-bearing porphyritic intrusion with a turbidite 433 434 deposit near Wheeler Lake in the Northwest Territories. This sample is at the approximate location of the Isachsen and Bowring (1994) lithic tuff sample (within turbidites) that has a 435 436 reported U-Pb zircon age of 2612 ± 1 Ma. The sample apatite U-Pb mixture model single age 437 peak is 2628 ± 96 Ma (2σ ; n=36). The AFT sample has a pooled age of 335.7 ± 11.3 Ma (1σ ; 438 n=40) and a central age of 344 ± 14 (1 σ) with 22% age dispersion. The MTL is 12.04 ± 2.39 µm (1σ; n=131). The mean kinetic parameters for 12-DRA03-001 are: measured D_{par} of 1.87 μm, eU 439 440 of 16 ppm (range 2-55 ppm), calculated r_{mr0} of 0.83, and measured Cl of 0.019 apfu.

441 Sample BNB97-035 is plutonic med-coarse grained biotite granitoid with strong lineations taken from the west flank of the Sleepy Dragon Complex in the Yellowknife Domain of the Slave 442 443 craton with nearby basement crystallization ages of ca. 2900 Ma (Bleeker et al., 1999), similar to 444 the sample apatite U-Pb mixture model single age peak of 2858 ± 124 Ma (2σ ; n=22). The AFT 445 sample has a pooled age of 229.3 ± 22.1 Ma (1 σ ; n=40) and a central age of 272 ± 16 (1 σ) with 34% age dispersion. The MTL is $12.75 \pm 1.80 \ \mu m$ (1 σ ; n=116). The mean kinetic parameters for 446 BNB97-035 are: measured Dpar of 1.72 µm, eU of 21 ppm (range 0.2-98), calculated r_{mr0} of 447 0.85, and measured Cl of 0.006 apfu. 448

Sample 12NK-L18A3 is from granitic gneiss in Nunavut with a zircon U-Pb age of 2487 \pm 5.4 Ma and a metamorphic overprint U-Pb age of 2377 \pm 3.5 Ma (Davis et al., 2014). The apatite U-Pb mixture model single age peak is significantly younger at 1788 \pm 40 Ma (2 σ ; n=24). The AFT sample has a pooled age of 392.4 \pm 9.0 Ma (1 σ ; n=26) and a central age of 397 \pm 13 (1 σ) with 14% age dispersion. The MTL is 11.91 \pm 2.06 µm (1 σ ; n=78). The mean kinetic parameters for 12NK-L18A3 are: measured Dpar of 1.85 µm, very high eU of 339 ppm (range 139-655), calculated r_{mr0} of 0.79, and measured Cl of 0.012 apfu.

456 4.3 Churchill-Rae Province, Melville Peninsula region

457 Sample SNB-01-M2055 is from Melville Peninsula, ~150 km southwest of Committee Bay and is
458 a foliated biotite tonalite that cuts a komatiite at the base of the Howling Wolf section with an U-

- 459 Pb zircon igneous crystallization age of 2606 ± 4 Ma (Cairns et al., 2005). The apatite U-Pb 460 mixture model single age peak is 1940 ± 68 Ma (2σ ; n=26). The AFT sample has a pooled age of 461 348.5 ± 9.8 Ma (1σ ; n=30) and a central age of 356 ± 18 (1σ) with 24% age dispersion. The 462 MTL is $12.19 \pm 2.30 \mu m$ (1σ ; n=130). The mean kinetic parameters for SNB-01-M2055 are: 463 measured D_{par} of 2.00 μm , eU of 14 ppm (range 0.2-46 ppm), calculated r_{mr0} of 0.84, and 464 measured Cl of 0.003 apfu.
- 465 Sample 10CXAD-086A is from eastern Melville Peninsula on the Foxe Basin margin and was collected from a plutonic gabbro-anorthosite with nearby detrital U-Pb ages of 1899 ± 7 Ma and 466 467 1897 ± 15 Ma in the overlying Paleoproterozoic Penrhyn Group (Partin et al., 2014). The sample 468 apatite U-Pb mixture model single age peak is 1803 ± 73 Ma (2σ ; n=24). The AFT sample has a 469 pooled age of 363.1 ± 28.5 Ma (1 σ ; n=40) and a central age of 460 ± 44 (1 σ) with 57% age dispersion. The MTL is $12.60 \pm 1.79 \ \mu m$ (1 σ ; n=132). The mean kinetic parameters for 470 471 10CXAD-086A are: measured D_{par} of 2.12 μm, eU of 13 ppm (range 0.2-73 ppm), calculated r_{mr0} 472 of 0.82, and measured Cl of 0.029 apfu.
- 473 *Sample 11CXAN017* is from the Prince Albert block on Melville Peninsula and was collected 474 from a Neoarchean Bt-Kfs porphyritic monzogranite. The sample apatite U-Pb mixture model 475 single age peak is 1676 ± 59 Ma (2σ ; n=18). The AFT sample has a pooled age of 418 ± 11 Ma
- 476 (1 σ ; n=18) and a central age of 452 ± 31 (1 σ) with 28% age dispersion. The MTL is 12.67 ± 1.92
- 477 μ m (1 σ ; n=120). The mean kinetic parameters for 11CXAN017 are: measured D_{par} of 1.95 μ m,
- eU of 21 ppm (range 3-43 ppm), calculated r_{mr0} of 0.85, and measured Cl of 0.002 apfu.
- 479 *Sample 13LVA04* is from a gabbro at the Discovery outcrop that cuts a banded iron formation

and the Sam Fm. at the Thelon Basin in S. Nunavut. The apatite U-Pb mixture model single age

- 481 peak is 2224 ± 125 Ma (2σ ; n=39). The AFT sample has a pooled age of 546.0 ± 33.8 Ma (1σ ;
- 482 n=40) and a central age of 557 ± 31 (1 σ) with 21% age dispersion. The MTL is $12.44 \pm 2.06 \mu m$
- 483 (1 σ ; n=121). The mean kinetic parameters for 13LVA04 are: measured D_{par} of 1.92 μ m, eU of 7
- 484 ppm (range 3-42 ppm), calculated r_{mr0} of 0.80, and measured Cl of 0.012 apfu.

- 485 Sample 09SZ-23-01 is from Precambrian granitic gneiss at the northern margin of Melville 486 Peninsula at the Fury and Hecla Strait, <200 m from the Paleozoic unconformity presented in 487 Pinet et al. (2016). The AFT sample has a pooled age of 462.7 ± 29.9 Ma and a central age of
- 488 486 \pm 27 Ma with 21% age dispersion. The MTL is 12.18 \pm 1.58 μ m (1 σ ; n=100). The mean

kinetic parameters for 09SZ-23-01 are: measured D_{par} of 1.67 µm, ²³⁸U of 6 ppm (range 3-12 489 490 ppm), calculated r_{mr0} of 0.85, and measured Cl of 0.016 apfu. There are no apatite U-Pb data for this sample but there are coexisting apatite (U-Th)/He data with a weighted mean age of $347 \pm$ 491 492 65 Ma (1σ ; n=5) and an additional nearby (U-Th)/He sample 09SZ-24-01 with a weighted mean 493 age of 211 ± 80 Ma (1 σ ; n=7). Sample 09SZ-23-01 is not entirely comparable to our ICP-MS AFT data because only ²³⁸U was measured and is not necessarily representative of the eU, as Th 494 495 and Sm contributions could make the eU much greater and add greater variability between 496 grains. To add more approximate values for eU we took the average Th and Sm concentrations 497 of the two apatite (U-Th)/He samples and used those to calculate eU for the AFT data. This yields a mean eU of 14 ppm (range 11-20 ppm). Further information regarding AFT analytical 498 499 methods, Cl compositional data, and accompanying AHe data (sample 09SZ-24-01 at about the 500 same location) are given in Lavoie et al. (2013) and Pinet et al. (2016).

501 4.4 Churchill-Rae Province, Southampton Island

502 Sample 07CYA-M38B is from gabbroic anorthosite bedrock on Southampton Island with a U-Pb 503 zircon inherited primary crystallization age of 3005 ± 22 Ma and a recrystallization age of 1870 504 \pm 10 Ma (MSWD=1.2) due to Trans-Hudson overprinting (Rayner et al., 2011). The apatite U-Pb 505 mixture model single age peak is 1834 ± 60 Ma (2σ ; n=35). The AFT sample has a pooled age of 506 341.6 ± 10.5 Ma (1 σ ; n=40) and a central age of 384 ± 21 (1 σ) with 30% age dispersion. The 507 MTL is $12.22 \pm 2.0 \ \mu m$ (1 σ ; n=132). The mean kinetic parameters for 07CYA-M38B are: measured D_{par} of 1.90 µm, eU of 22 ppm (range 1-71 ppm), calculated r_{mr0} of 0.84, and measured 508 509 Cl of 0.024 apfu.

- 510 *Sample 07CYA-M133A* is a plutonic diorite from Southampton Island with a U-Pb igneous 511 crystallization age of 1842 ± 5 Ma (Rayner et al., 2011). The apatite U-Pb mixture model single
- 512 age peak is 1812 ± 79 Ma (2σ ; n=13). The AFT sample has a pooled age of 364.2 ± 10.6 Ma (1σ ;
- 513 n=15) and a central age of 375 ± 17 (1 σ) with 16% age dispersion. The MTL is $11.57 \pm 2.16 \,\mu\text{m}$
- 514 (1 σ ; n=101). The mean kinetic parameters for 07CYA-M133A are: measured D_{par} of 2.26 μ m,
- eU of 74 ppm (range 7-258 ppm), calculated r_{mr0} of 0.80, and measured Cl of 0.086 apfu.

516 4.5 Rae craton, southern Baffin Island

517 Sample 09SRB-M100 is a metasedimentary semipelite within the Paleoproterozoic Hoare Bay 518 Group at Kumlien Fiord on the Cumberland Peninsula, Baffin Island. The apatite U-Pb mixture 519 model single age peak is 1674 ± 35 Ma (2σ ; n=28). The AFT sample has a pooled age of 97.2 \pm 520 4.3 Ma (1σ ; n=35) and a central age of 102 ± 8 (1σ) with 37% age dispersion. The MTL is 12.94 521 \pm 2.23 µm (1σ ; n=41). The mean kinetic parameters for 09SRB-M100 are: measured D_{par} of 2.14 522 µm, eU of 22 ppm (range 1-69 ppm), calculated r_{mr0} of 0.83, and measured Cl of 0.023 apfu.

523 *Sample 14SUB-H43A* is a metamorphic garnet-biotite psammite from a Paleoproterozoic 524 metasediment/layered mafic intrusion sequence on northern Hall Peninsula, Baffin Island. The 525 apatite U-Pb mixture model single age peak is 1782 ± 38 Ma (2σ ; n=25). The AFT sample has a 526 pooled age of 440.2 ± 11.4 Ma (1σ ; n=25) and a central age of 442 ± 12 (1σ) with 10% age 527 dispersion. The MTL is 12.46 ± 1.72 µm (1σ ; n=130). The mean kinetic parameters for 14SUB-528 H43A are: measured D_{par} of 2.29 µm, eU of 26 ppm (range 16-35 ppm), calculated r_{mr0} of 0.82, 529 and measured Cl of 0.022 apfu.

530 4.6 Grenville Orogen, southeast Quebec

531 Sample 02NKL-871 is from a granitic orthogneiss in southeast Quebec. It is shown here for 532 comparative purposes, as it is much younger (Grenvillian) than the majority of our other samples 533 taken from Archean bedrock. The apatite U-Pb mixture model single age peak is 951 ± 38 Ma $(2\sigma; n=25)$. The AFT sample has a pooled age of 160.8 ± 4.5 Ma $(1\sigma; n=35)$ and a central age of 534 177 ± 13 (1 σ) with 38% age dispersion. The MTL is $12.27 \pm 1.80 \mu$ m (1 σ ; n=135). The mean 535 kinetic parameters for 02NKL-871 are: measured D_{par} of 1.83 µm, eU of 31 ppm (range 2-160 536 537 ppm), calculated r_{mr0} of 0.85, and measured Cl of 0.008 apfu. This sample will also be discussed with AFT and AHe data from the Anticosti Island (Quebec) Grenville basement sample of 538 539 Powell et al., (2018a), see table 1.

540

541 **5. Discussion**

542 5.1 AFT age relationships between eU and kinetic parameters r_{mr0} and effective Cl

543 Using plots of central age versus U, Hendriks and Redfield (2005) proposed that REA could 544 explain the observed decrease in AFT age with increasing U content for samples from the

545 Fennoscandian Shield. Kohn et al. (2009) compared central ages with eU values for a larger set 546 of Precambrian shield rocks and reported weaker or inconsistent trends, suggesting that variable 547 Cl content rather than REA was controlling AFT annealing and age dispersion. These contrasting results are not surprising given the methods that were used. Kinetic variability or potential REA 548 549 effects should be evaluated for individual grains, particularly for the samples of this study that 550 exhibit "open jaw" radial plots (O'Sullivan and Parrish, 1995; fig. 3) that are a common feature 551 of multi-kinetic AFT samples (e.g. Issler et al., 2005; Powell et al., 2018b; Schneider and Issler, 552 2019). Furthermore, it can be difficult to infer compositional controls on AFT annealing using Cl 553 content alone, when OH and various cations can also have a significant effect on AFT annealing 554 (Barbarand et al., 2003; Carlson et al., 1999). Combining samples of different apatite 555 composition and/or with different thermal histories may obscure any potential relationship 556 between AFT age and eU.

557 Unlike the previous studies that used the external detector method where typically up to 20 age 558 grains are measured, we use larger sets of AFT single grain ages (up to 40 grains per sample; 559 table 1) obtained using the LA-ICP-MS method to investigate intra-sample age dispersion with 560 respect to eU and annealing kinetic parameters derived from detailed elemental data. Samples 561 with the largest number of measured grains (30-40) have the largest age dispersion (21-57%) 562 whereas those with <30 grains have much lower dispersion (10-21%; fig. 3). Clearly it is 563 necessary to measure a sufficient number of grains in order to sample as broad a range of eU 564 values and AFT ages as possible. Although we also observe a weak negative correlation between 565 central age and eU (fig. 2), single grain ages spanning hundreds of millions of years, or over a 566 billion years in some cases, show a well-defined relationship between increasing eU and younger 567 AFT age (fig. 5). Our bedrock AFT samples generally have near end-member fluorapatite 568 compositions based on measured Cl concentration. Some samples show a broader range in track 569 retentivity when other kinetic parameters such as r_{mr0} (or equivalent eCl) and D_{par}, are examined 570 but there is no clear correlation between age and apatite composition (fig. 5). Instead, the 571 extreme variations in eU seem to offer the best explanation for the observed AFT age 572 heterogeneity (clear correlation with age, fig. 5). The effect of eU on fission-track retentivity may have a discernible link to long residence at low temperatures in or below the partial 573 574 annealing zone.

575

576 The interplay between eU and the other dominant kinetic parameter (i.e. Cl) becomes complex in 577 some instances where Cl concentrations are comparatively variable or elevated, or there is relative enrichment of cations such as Fe or REE (fig. 5 and fig. 6). This is clearly demonstrated 578 by a counterintuitive association between eU and eCl (or inverse correlation with r_{mr0} , fig. 6). 579 The Baffin Island semipelite sample 09SRB-M100C1 demonstrates a spurious correlation 580 between eU and eCl/r_{mr0} , which implies there is a confounding variable (e.g. thermal history) or 581 other kinetic parameter controlling retentivity. Higher eCl (and low r_{mr0}) values signify higher 582 retentivity, and high eU should hypothetically lower retentivity if greater damage is a proxy for 583 enhanced annealing. Chemically-retentive grains with elevated eU that have experienced a 584 585 thermal pulse may differentially anneal α -damaged FT regions with respect to grains that are 586 lower eCl, and suggest cumulative thermal annealing in this sample was sufficient to obscure 587 expected associations between retentivity proxies. A first-order filter for samples is examining 588 the AFT age-eU relationship. If a sample shows that eCl is heterogeneous in 'age-eU space' with 589 a clear negative (curvi-) linear age-eU correlation, then it can be assumed that eU is primarily 590 driving retentivity, see Southampton Island and Superior Province samples. If clear eCl 591 'domains' or divisions are apparent when viewing age-eU relationships (sample 09SRB-592 M100C1), this suggests Cl or another cation-site element is playing a greater role in controlling 593 retentivity. Samples with more homogenous chemical compositions and low Cl show no such relationship between eU and eCl/rmr0 and denote eU as a primary driver of intra-sample age 594 595 dispersion, which is reflected in plots of eU versus age that show age scatter across the entire 596 range of Cl. There is likely some threshold where Cl begins to counteract the effects of REA or 597 dominates fission-track retention, however this seems variable between samples and conceivably 598 has an association with thermal history.

599 5.2 Elemental substitutions and compositional variation

The formula for apatite when considering substitutions (Barbarand et al., 2003) can be written as $X_{10}YO_4Z_2$ where, X = mainly Ca, Y = P, and Z = F, Cl, or OH. The most common substitutions on the X site are Fe²⁺, Mn²⁺, Na⁺, REE³⁺ Sr²⁺ and U⁴⁺, whereas Si and S occur on the Y site. The trivalent (REE³⁺) cations substitute in pairs to maintain charge balance: REE³⁺ + Si⁴⁺ proceeds to Ca²⁺ + P⁵⁺ and REE³⁺ + Na⁺ to 2Ca²⁺ (Barbarand et al., 2003 and refs. therein). Cation

substitutions directly govern the structural integrity of the apatite lattice and there is an inverse 605 606 correlation between the number of substitutions on the cation (X) and anion (Z) sites (Carpéna, 607 1998). It has also been shown that preferential U and Th substitution in fluorapatite or chlorapatite causes a change in volume (decrease and increase, respectively) of the apatite Ca (I) 608 609 and Ca (II) polyhedron (e.g. Carpéna et al., 1988; Carpéna, 1998; Luo et al., 2009). This change 610 results in an increase in size and distortion of the Ca (II) site, allowing for preferential 611 substitution of U and Th at both Ca sites in chlorapatite and is an interesting observation because 612 the Carlson et al., (1999) dataset documents that Ca substitutions tend to reduce annealing rates 613 in apatite but may also depend on a complex relationship with mixing at the halogen site (i.e. F/Cl/OH). The results of Luo et al., (2009) demonstrate that U and Th are structurally preferred 614 615 in chlorapatite, so it is expected that previous work attributed differences in AFT ages to Cl 616 rather than eU, as Cl is a dominant control on track retentivity.

617

We show two examples where we try to further investigate the compositional controls on AFT grain ages from samples with high and low age dispersion. Sample PBA-98-743 from the Superior Province and 12NK-L18A3 from the northeast Slave craton are characterized by 36% and 14% dispersion, respectively (fig. 3). PBA-98-743 exhibits strong eU control on age (figs. 4 and 6), however 12NK-L18A3 does not (fig. 4). Moreover, the Slave sample is characterized by old ages despite high eU (high α -damage; see fig. 9), suggesting another kinetic control on retentivity.

625 Linear correlation matrices were performed on the elemental data (apfu, including OH estimates) for our samples to assess the dominant interactions between the known controls on retentivity 626 627 (fig. 7; all samples shown in SI). The data were standardized first to reduce any extreme variance 628 between elements. Correlation coefficients associated with positive correlations are shown in 629 black and variables that are anti-correlated are in white with the size of the circle corresponding 630 to larger or smaller correlation coefficients (fig. 7A). For the Superior Province sample all linear 631 correlations are shown in fig. 7A, however to assess the robustness of the relationship, only those that correspond to p-values of ≤ 0.05 or pass at 95% confidence level were retained (fig. 7B). F 632 633 and Cl are anti-correlated as expected and in this case eU is most highly correlated with Ce (r =634 0.68), La (r = 0.56), Cl (r = 0.50), Y (r = 0.46), Mn (r = 0.45) and is anti-correlated with F (r = -

635 0.41). La and Ce are the most-highly correlated elements (r = 0.90). However, caution is advised 636 in interpreting these relationships because in some cases many of the grains contain low or 637 negligible elements but only a few grains are enriched and produce a statistically robust 638 correlation (fig. 7C), as is the case with eU and La/Ce. However, an overall assessment of 639 elemental abundances and apatite lattice site "preference" are contained in these data.

640 Figure 7D is a case where no clear age-eU relationship is observed and this is revealed in the 641 correlation matrix (p-values of ≤ 0.05) as eU is not robustly correlated with any other element. 642 The Slave craton sample is incorporating REE, Mn, Fe, Na, and Cl. The most highly correlated 643 elements are La-Ce (r = 0.97), Na-Cl (r = 0.79), Na-Mg (r = 0.76), Mn-Fe (r = 0.76), Mn-Y (r = 0.74), Fe-Y (r = 0.66), and Fe-La (r = 0.63). A relationship that is clear from our samples is that 644 645 the preferential incorporation of cation elements such as REE and U-Th (table 2) seems to 646 dominate in cases where AFT ages exhibit a negative age-eU relationship, suggesting a dominant 647 retentive control on age due to elevated eU, which agree with the results of Carpéna (1998). Our 648 elemental data suggest that in addition to common apatite varieties, we may have more "exotic" 649 apatites that heavily incorporate REE, Na, U, and Si. We have not probed for Si but that may be 650 important to monitor in future AFT studies. These associations yield some insight into retentivity 651 (AFT age) and they indicate the elements that are being incorporated into apatite grains from a 652 particular bedrock sample, which may ultimately and indirectly aid in assessment of age 653 dispersion. Our correlation matrices (see SI) also demonstrate that fluorapatite is characterized by lower Cl and U-Th affinity, whereas chlorapatite shows a positive correlation between Cl and 654 655 eU. These relationships also suggest that if actinides are incorporated into chlorapatite, then there 656 may be a delicate balance between elevated eU (retentivity decrease) and elements that are 657 known to increase track retentivity, which may be ultimately governed by damage accumulation 658 time and specific elemental abundance. It is extremely important to point out that while many of 659 these elements are highly correlated with one another, eU is the only element that exhibits a very 660 consistent and strong (curvi)-linear relationship with AFT single-grain ages in virtually all of our 661 samples. This observation advocates for α -radiation as a governing factor for AFT annealing in 662 slowly-cooled settings.

663

664 5.3 Enhanced dissolution and fission track etching rates

665 Previous work has demonstrated α -radiation-enhanced dissolution is a concern for U-rich or 666 actinide-bearing accessory mineral phases (e.g. Dran et al., 1984; Petit et al., 1985). Petit et al. 667 showed that implantation of low energy (~1 keV/amu) Pb ions, which simulate α -recoil, greatly increases dissolution rate above a certain critical damage threshold of ~2.5×10¹⁸ α/g , dependent 668 upon mineral U (Th and Sm) concentrations and accrued time of damage accumulation. Weber 669 670 and Matzke (1986) examined actinide-host phases, one of which was the apatite structure Ca₂Nd₈(SiO₄)₆O₂ that was doped with ²⁴⁴Cm to investigate the effects on microstructure from 671 self-irradiation by α -decay and fission. They found that α -damage and fission tracks overlap at 672 high doses and leads to amorphization at ~ 1.1×10^{19} α/g , ~8% swelling of the crystalline volume, 673 and a factor of 10 increase in leachability. Ewing et al. (1981) document similar enhanced 674 675 dissolution (by a factor of ~ 100) in increasingly older zircons with greater accumulated damage. 676 Intuitively or hypothetically, if higher U caused enhanced track annealing, the FT analyst could 677 measure the track lengths on grain mounts where U has also been directly obtained and observe a 678 positive correlation between shorter lengths and higher U. We have examined some unpublished 679 samples where we have U from both age and length grains and there is no clear relationship between length and U, accounting for both measured and c-axis projected lengths. This may be 680 681 due to any number of factors affecting track length collection, such as differences in etching conditions (e.g. etch time, temperature, or acid strength), etching and/or annealing anisotropy, 682 683 variability in measured length with respect to the c-axis, U heterogeneity, or simply and most 684 importantly, the sub-random sampling of tracks for measurement. However, high U apatite (highly damaged) may reveal apparently longer tracks because short tracks are difficult to 685 686 observe on a high track density surface – invalidating the assumption of high eU being correlated with shorter tracks and suggesting that the observation of longer tracks may be more likely for 687 688 high eU grains. Ultimately, for AFT dating there may be no discernible differences in etching for 689 typical apatites because the induced α -damage levels in laboratory experiments are rarely 690 achieved in the geological environment.

691

692 5.4 Considering α-recoil and fission track damage accumulation and annealing

693 The α-damage present in apatite will increase with time as a function of the parent nuclide 694 content, but will also decrease with heating (Shuster and Farley, 2009). The negative age-eU trends in our dataset (not commonly observed for apatite data) imply long residence at low temperatures below those required for track annealing (or significant He retention). Our data empirically support the persistence of α -recoil damage over extremely long timescales in the AFT system (figs. 8 and 9) and that α -damage is characterized by slower annealing rates than fission-track damage, as previously suggested (e.g. Fox and Shuster, 2014; Ritter and Märk, 1986; Willett et al., 2017).

701

We further interrogate our AFT samples by estimating the amount of α -damage they have accumulated. Alpha dose is calculated following equation 5 of Nasdala et al. (2005) and is expressed as α/g .

$$705 \qquad D_{\alpha} = 8 \cdot \frac{C_U \cdot N_A \cdot 0.9928}{M_{238} \cdot 10^6} \cdot \left(e^{\lambda 238t} - 1\right) + 7 \cdot \frac{C_U \cdot N_A \cdot 0.0072}{M_{235} \cdot 10^6} \cdot \left(e^{\lambda 235t} - 1\right) + 6 \cdot \frac{C_T h \cdot N_A}{M_{232} \cdot 10^6} \cdot \left(e^{\lambda 232t} - 1\right)$$
(1)

Where, C_U and C_{Th} are the actinide concentrations in ppm, N_A is Avogadro's number, and M_{238} (etc.) are the molecular weights of the parent isotopes, and λ_{238} , λ_{235} , λ_{232} are the decay constants for each. The *t* in the equation refers to the integration or accumulation time for α -particle selfirradiation. Traditionally, for zircon this time is taken as the time since crystallization and is presumably a reasonable assumption, however this becomes more difficult to assess for other minerals such as apatite that are more prone to greater damage annealing at moderate temperatures of <200°C.

713 The most difficult assumption regarding α -dose estimation is the integration time for selfirradiation, and here we refer to the integration time as the "effective dose accumulation time" 714 715 (t_{EDA}). Typically it is believed that α -damage is fully annealed at temperatures of ~150°C (Weber et al., 1997) and if fission tracks are fully annealed at ~125°C (Green et al., 1986) then the oldest 716 AFT age would yield a conservative estimate for t_{EDA}. A limitation in adhering to strict 717 718 temperature limits is that apatite composition can introduce differences in the thermal sensitivity 719 of an individual grain, and that the time spent at a given temperature is just as important, if not 720 more so for cratonic rocks. The trade-off between time and temperature, with respect to 721 annealing, is analogous to laboratory annealing experiments where the same degree of annealing 722 can be achieved at higher temperatures in a shorter time, or at lower temperatures over a longer 723 duration. Therefore, we optimistically estimate the t_{EDA} for apatite as the mean of the 2σ

724 minimum apatite U-Pb isotopic sum age for the sample. The apatite U-Pb age estimate for AFT 725 t_{EDA} seems to be a suitable approximation, however this should not be viewed as a rigorous 726 temperature-based appraisal because the large U-Pb apatite errors are in essence providing a 727 crude intermediary between the oldest representative AFT age (~track retention age) and isotopic 728 closure for the U-Pb system. We are currently more interested in observing data trends rather than establishing absolute relationships, and as more data are collected on the ²³⁸U percolation 729 730 dose threshold and the temperatures at which α -damage accumulation in different apatite 731 compositions is appreciable, there can be improvements to this approach as the t_{EDA} uncertainties 732 are high.

733

734 An example of the relationship between α -damage and cooling age is shown in figure 8 where 735 AFT and AHe data from the same location (Pinet et al., 2016) are plotted against eU and 736 transposed into α -dose. The apatite grains show a modest range in eU but interestingly the AFT 737 data only capture the low eU and the AHe data have a greater spread in eU (fig. 8A,B). This 738 observation certainly suggests preferential selection of grains for each method, perhaps outside 739 of the established routine of choosing pristine whole grains for (U-Th)/He analysis. The black dashed line is the ²³⁸U α -recoil track percolation threshold of 1.9x10¹⁶ α/g from Ketcham et al., 740 741 (2017) and is the point where diffusivity is expected to increase due to damage connectivity 742 creating fast-path diffusion for the (U-Th)/He system. These data clearly demonstrate the increase in AHe age going from low eU/low α -dose and increasing in age until the percolation 743 744 threshold is reached whereby a decrease in age follows (this is very similar to the observed 745 behaviour for zircon U-Th/He data for fission-track percolation Guenthner et al., 2013). The AFT data are mostly above the U percolation threshold even though those grains have low eU, 746 747 whereas the AHe grains have higher eU concentrations/damage and are even younger. The chronometer ages plot above the damage threshold due to long t_{EDA}, which dominates any effects 748 of high or variable eU between single grains. When examining samples, the difference in t_{EDA} 749 750 can explain why very different U and Th amounts between samples can result in similar overall 751 age-eU trends. Visually the data display remarkable adherence to the α -recoil track percolation 752 threshold, despite the fact that the data are from different thermochronometers, suggesting α -753 damage affects apatite diffusive processes and has repercussions for both damage annealing and

diffusivity. Sample 13JP10 from the Grenville basement of Anticosti Island (Powell et al., 2018a) potentially spent ~275 Ma at temperatures >120-150°C, or experienced an episodic pulsed history to these high temperatures, which suggests that thermal resetting completely decoupled α -damage and fission track damage accumulation from one another (fig. 8C,D). It is also worth mentioning that 13JP10 underwent significant thermal annealing and is also one of the only samples shown in figure 2 with 40 dated age grains and low age dispersion (16%).

760

Our α -dose plots imply that at least some fission tracks are impacted by α -recoil tracks. At this time it is not entirely clear why AFT ages behave similarly to AHe ages with respect to the recoil damage percolation threshold, however it may be due to their shared dependence on diffusive mechanisms that affect track annealing and ⁴He retentivity. We envision difficulty in trying to develop a model to jointly explain these damage types, as the dimensions, frequencies, and timescales (t_{EDA}) are very different. Now we examine our Canadian Shield AFT data with respect to the ²³⁸U α -recoil percolation threshold and discuss these relationships.

768

769 Figure 9A shows AFT data from the Canadian Shield selected from table 1. All AFT samples 770 with long (and similar) $t_{EDA} > 1$ Ga plot to the right of the damage threshold, whereas Grenvillian basement samples 02NKL-871 and 13JP10 from Powell et al. (2018a) with shorter $t_{EDA} < 1$ Ga 771 772 show an increase and subsequent decrease in AFT ages before and after the damage threshold, 773 respectively. To better understand the damage relation with AFT age, it is notable that a very 774 high eU sample 12NK-L18A3 displays high damage and high eU variance (σ^2) between single grains (mean eU: 339 ± 158 ppm, 1σ) resulting in a relatively low spread in single-grain ages 775 776 $(403 \pm 71 \text{ Ma} (2\sigma, n=26))$ but moderate spread in damage level. There is no clear age-eU relationship for this sample (fig. 5). Conversely, sample 13LVA04 has very low eU (and low σ^2 ; 777 778 mean eU: 6 ± 1 ppm) and plots on the percolation threshold resulting in extreme age variability 779 $(560 \pm 329 \text{ Ma} (2\sigma, n=40))$. High age scatter is expected for samples exhibiting high eU 780 variability and especially for those near the percolation threshold – as this would be the region of 781 greatest sensitivity to subtle changes in crystal damage and therefore changes in track annealing 782 and effects on AFT age.

783

784 We also chose younger samples from the recent literature that have been dated by AFT and U-Pb 785 (when possible) to compare to our Canada dataset. AFT samples from younger bedrock terranes in active tectonic settings including those from Sakhalin Island (Okhostsk), Russia (Glorie et al., 786 787 2017), the Yukon-Tanana area of central Alaska (Dusel-Bacon et al., 2016), the Wrangellia Terrane near Mt. Logan, Alaska (Enkelmann et al., 2017), and the northern Chilean Andes 788 789 (Rodríguez et al., 2018). We also compared these to single-grain analyses of the Durango (eU =790 81 ± 7 ppm, 1σ , n=300) and Fish Canyon Tuff (eU = 42 ± 15 ppm, n=300) AFT standards dated 791 by P. O'Sullivan. This comparison was done to assess if there is similar behaviour with respect 792 to damage accumulation and adherence to the α -recoil percolation threshold that is demonstrated 793 by our older cratonic samples. The major limitation for finding suitable published data is that the 794 majority of studies do not publish single-grain AFT analyses (only AFT sample summaries) and 795 that apatites from younger rocks are not typically dated via LA-ICP-MS U-Pb because it is 796 impractical due to the low amounts of U, Th, and Pb and the high initial common Pb 797 incorporated in apatite (Chew et al., 2011). Published samples where Th was not reported or the 798 EDM was used, we simply used the U amount to estimate the α -dose (Wrangellia and Russia), and examples where U-Pb data was unavailable we used the oldest individual AFT grain age or 799 another high-T thermochronometer, i.e. 40 Ar/ 39 Ar biotite for Wrangellia to estimate t_{EDA}. 800

We give a brief description of each sample here, although for more complete information the 801 802 reader should consult the cited reference. The Russian Sakhalin Island sample SK14-03 comes 803 from a granodiorite with a zircon U-Pb age of 43.3 ± 0.6 Ma and an apatite U-Pb age of $38.9 \pm$ 5.5 Ma. Twenty-six single-grain LA-ICP-MS AFT analyses yield a pooled age of 19.3 ± 2.1 Ma 804 and central age of 18.4 ± 1.6 Ma (avg. ²³⁸U=32 ppm; Glorie et al., 2017). The Yukon Alaska 805 sample 09AD-240 is from a leucogranite with a SHRIMP zircon U-Pb igneous crystallization 806 807 age of 191 ± 5 Ma and a pooled fission track age of 48.9 ± 3.9 Ma (avg. eU=12 ppm, n=40). The t_{EDA} for this sample was estimated by evidence that Mesozoic granitoids intruded the country 808 809 rock and cooled to <300°C by ~185 Ma (Dusel-Bacon et al., 2016). Sample LL07 was collected 810 from the Chilean Andes Main Cordillera north of 31.5°S from the Upper Cretaceous-Paleocene 811 intrusive belt (Rodríguez et al., 2018). This sample has an AFT age of 31 ± 1.8 Ma (avg. eU=73)

812 ppm, n=40) with a biotite K/Ar age of c. 67 Ma, taken as an approximation for t_{EDA} (all ages are 813 quoted at the 1σ level).

Figure 9B shows that all of the younger samples from the literature plot to the left of the α -recoil percolation threshold and do not display the same age-dose patterns as the old cratonic samples. This further demonstrates that apatites characterized by high or variable eU still require

significant time to accumulate radiation damage to produce an effect on AFT ages.

818 5.5 Multi-kinetic interpretation and thermal history modeling

819 The goal of most AFT studies is to model the age and track length data to make inferences about 820 past thermal history and geologic processes. This can only be achieved if we have a robust 821 understanding of the kinetics that govern the thermochronometers that we use. There are multiple 822 fission track annealing proxies such as measured Cl, r_{mr0}, and D_{par} that assist us in deconvolving 823 the annealing complexity contained within samples, however these tools are not perfect and in 824 some cases offer little help in understanding multi-kinetic samples. Our Canadian Shield samples 825 are typically compositionally homogeneous, which makes multi-kinetic interpretation 826 exceedingly problematic. Figure 10 shows two examples of an exploratory attempt at utilizing 827 the AFT age- α -dose (eU) relationship that is common to all of our cratonic samples. Samples 828 02NKL-871 and 11CXAN017 are very different in terms of calculated t_{EDA} and are used as 829 examples for multi-kinetic interpretation. We display the grain ages for each sample on a radial 830 plot (fig. 10 A, E) and perform age mixture modeling as before to identify statistically significant 831 age populations. These age populations are then plotted in terms of their eCl (fig. 10B, F) and 832 there is complete overlap between all populations, which invalidates clear compositional 833 separation due to chemical homogeneity between grains spanning the typical eCl error range. If 834 traditional compositional proxies suggest our sample is probably common F-apatite (fig. 10B, F), 835 but there is high age dispersion, we can be certain that another factor such as eU may be 836 controlling retentivity (fig. 10C, G). When these samples are examined in terms of α -dose it 837 becomes obvious that there are grains of similar age – for different reasons. The low eU grains 838 for sample 02NKL-871 have accumulated less damage or are generally low retentivity that have 839 probably experienced some thermal annealing, whereas the high eU/highly damaged grains have 840 experienced REA. There is undoubtedly a competition between fission track and α -damage 841 ingrowth and annealing that occurs and is further complicated by minor thermal annealing

842 events, as fission tracks and α -damage vary in both temperature sensitivity and natural 843 abundance. The same level of thermal annealing would preferentially remove more fission 844 damage, but the remaining α -damage would still play a role in overall retentive behaviour. The 845 separation of kinetic populations is more apparent for grains that have migrated across the α -846 recoil percolation threshold due to longer t_{EDA} (fig. 10H). The age peaks picked by the radial plot 847 mixture modeling are based on age (and U-related age precision) and match the age populations 848 identified by the differences in α -dose between the two groups for sample 11CXAN017. In these 849 cases we have identified a potential path forward in terms of separating kinetic populations by 850 eU or α -dose for apatites in slowly-cooled rocks.

851 5.6 Implications for the apatite fission track and (U-Th)/He thermochronometers

852 Carpéna et al. (1988) discuss one of the earliest known examples of the effects of U-Th on AFT 853 ages from the In Ouzzal carbonatites in Africa where they suggested that higher radiation 854 damage causes apatite lattice expansion and increases the thermal sensitivity of fission tracks to 855 thermal annealing events. Carpéna and Lacout (2010) carried out thermal annealing experiments on synthetic apatites doped with ²³⁵U and irradiated with thermal neutrons to induce fission. 856 857 They compared the synthetic apatite (no α -damage) to natural apatite to demonstrate that fission 858 tracks in the synthetic apatite needed higher temperatures and longer time to reach the same 859 degree of annealing when compared to natural apatite. Experimental evidence has also shown 860 that α -particle irradiation-induced annealing occurs in buried amorphous layers in natural Durango fluorapatite (Ouchani et al., 1997) and similar behaviour has been observed in silicate 861 862 apatite structures, which are more thermally stable than natural apatite (Weber et al., 1986; 863 Weber et al., 1997). The latter studies were performed on thick amorphous layers, which should be more difficult to anneal than amorphous tracks, supporting the idea that fission track 864 865 annealing could be enhanced by α -radiation. Non-thermal annealing of α -recoil damage and recovery of crystalline structure from (pre-damaged) fully-amorphous Durango apatite 866 867 specimens was observed by *in situ* transmission electron microscopy (Li et al., 2017). Damage 868 recovery through α -healing has also been recently demonstrated for monazite, suggesting that 869 natural monazite, like apatite, never becomes amorphous due to the effects of α-recoil (Seydoux-870 Guillaume et al., 2018). These experiments and observations support REA and imply that cratons 871 would be the ideal natural laboratory because rocks reside at low temperatures (<100°C) over

hundreds of millions to billions of years favouring high α -damage accumulation in natural apatite compositions. In the case of lower eU concentrations, a long t_{EDA} could also create the same conditions for lowering annealing activation energy.

875

876 We are unsure if α -particles directly anneal fission tracks, as posited by Hendriks and Redfield (2005), or that perhaps α -recoil acts to destabilize fission tracks and therefore affects annealing 877 rates. More work is required to properly evaluate the thermal and temporal sensitivity of 878 879 different apatite compositions and the kinetics of α -recoil and fission-track damage. The process 880 or mechanism by which α -recoil damage destabilizes fission tracks is not fully understood. 881 However, some hypotheses are that (1) lattice distortion from accrued α -damage may increase 882 the thermal annealing susceptibility of apatite, (2) α -decay ionization and electrical excitation 883 cause fission track defect annealing via self-heating over 100s-1000 My timescales, or (3) α damage interferes with or "connects" with fission-tracks directly, disrupting the crystal lattice, 884 885 which in turn yields a fission track configuration that is easier to anneal. Elemental substitutions 886 may act in a similar manner as hypothesis (3) by elements of different size or lattice site 887 affecting annealing behaviour.

888

We know that α -damage zones (Farley, 2000; Shuster et al., 2006), vacancies (Gerin et al., 889 890 2017), porous voids (Zeitler et al. 2017), and dislocations (McDannell et al., 2018b) increase ⁴He 891 retentivity and act as diffusive traps for (U-Th)/He dating. In the case of AFT analysis, REA may 892 be a purely structural effect from lattice distortions or convolved with the fact that in some cases 893 those substitutions happen to be high α -emitters, thus introducing the likelihood for the 894 degradation of fission-track integrity that is most evident when thermal annealing is a secondary 895 concern. We can envision the structural relaxation phase of α -damage annealing being very 896 important in apatite that has accumulated greater damage, which may lead to differences in 897 annealing due to enhanced lattice recovery during any thermal perturbation with respect to an 898 undamaged apatite. It is not clear how other Ca-site substituting elements such as Fe that 899 increase fission track retentivity, would play a role in counteracting REA and would obviously 900 only do so if present in sufficient quantity within the lattice. A corollary exists in zircon where 901 greater U and Th content increases microstructural deformation as further development and creep

902 of lattice dislocations cause enhanced diffusivity of these elements (Timms et al., 2006). Crystal 903 lattice defects related to α -recoil might be a primary contributor to fission track instability. This 904 could either be directly, or by association with substituting α -emitting parent nuclides such as U 905 and Th. Interference between lattice defects due to a-recoil and fission-track defects could 906 invariably result in α -recoil causing sufficient damage to facilitate annealing, especially given the 907 rarity of fission tracks with respect to α-recoil tracks. These effects could presumably overwhelm 908 apatite fission tracks from rocks with variable U and Th concentrations that have been residing at 909 near-surface conditions for hundreds of millions of years. In other cases involving more complex 910 shallow burial and exhumation events, some amount of α -damage may be preserved for up to a 911 billion years or more and never fully annealed. In these scenarios, thermal annealing is mostly insignificant at <60°C, but α -induced damage could cumulatively modify the apatite lattice in 912 913 proportion to the U and Th, making fission tracks unstable and driving differential annealing at 914 low temperatures – thereby yielding large AFT single-grain age dispersion. This is likely 915 amplified by longer damage accumulation times and is more noticeable in samples with a range 916 of eU. Fission tracks and α -damage both undergo annealing via diffusive processes (Ritter and Märk, 1986), which are regulated by crystallographic damage level; therefore they should be 917 918 viewed and treated as a damage continuum with different timescale and kinetic dependencies.

919 6. Recommendations and questions for future work

920 The effects of REA on AFT ages in slowly-cooled settings is a complex problem that requires 921 sufficient data to be able to evaluate relationships between chemical composition and age. Some 922 factors to consider in future work to assist in addressing REA include:

- Acquire enough age grains to capture and to characterise intra-sample AFT age
 dispersion. We recommend counting a minimum of 35-40 grains (dependent on sample
 quality), especially given the ~10-20% dispersion for <25 measured grains versus ~30-
 40% dispersion for >30 grains quoted in figure 3.
- 927 2. Collect detailed elemental data for every sample including length and age grains; e.g.
 928 measured Cl is insufficient and could give misleading results. The acquisition of F, Na,
 929 Mg, P, S, Cl, Ca, Mn, Fe, Sr, Y, La, Ce, and Si at the very least should be carried out,
 930 since many of these elements are known to affect apatite damage retentivity. The

measurement of U, Th, and Sm on length grains (we did not do this for our samples) mayalso be beneficial for interrogating potential AFT length-eU relationships.

- Acquire apatite U-Pb age data to estimate t_{EDA}. In cases where U-Pb data are lacking and
 a broad range of U has been sampled within an AFT dataset, the oldest reliably measured
 AFT age seems to be a satisfactory minimum time estimate of dose accumulation.
- 9364. If cratonic bedrock samples are to be treated as multi-kinetic for thermal history937modeling, evaluating grains in terms of eU may be a suitable kinetic proxy alternative for938separating kinetic populations. The influence of apatite composition on annealing needs939refinement and currently the choice of r_{mr0} equation is important and further work is940needed to constrain the relationship at high and low retentivity where existing models do941not sufficiently capture variability in retentivity.

942 **7. Conclusions**

943 Apatites from bedrock samples across many areas of the Canadian Shield show coincident high 944 single-grain age dispersion and variable eU. There are no other clear intra-sample, compositional 945 controls such as Cl, Fe, or REE substitution that demonstrate a strong relation with AFT age. In 946 some isolated cases there are some grains enriched in elements known to enhance fission-track 947 retentivity, however these are typically only a few grains from within a sample. The complexity 948 of our AFT dataset further establishes the necessity of acquiring compositional data for 949 interpreting apatite fission-track analyses and our data strongly support the notion that REA from 950 α -damage is related to the eU content within apatite. We stress that this does not invalidate AFT 951 analysis, although care should be taken to properly evaluate slowly-cooled rocks. Complications 952 arise between co-existing, specific elemental substitutions that independently act to either 953 enhance or reduce fission track annealing. The exact mechanism for enhanced annealing is likely 954 multifaceted but may be due to α -damage affecting tracks directly, α -damage and elemental 955 substitutions causing lattice defects that promote track instability and lower thermal annealing 956 resistance, α -damage self-healing reducing lattice damage and affecting fission tracks, or a 957 combination of these effects that are exacerbated in settings where thermal annealing is minimal 958 and timescales for α -damage accumulation are long.

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1397 FIGURE CAPTIONS

1398 Figure 1. Map of the Canadian Shield with simplified geologic domains modified from Whitmeyer and 1399 Karlstrom (2007). Bedrock AFT samples are shown as circles with their corresponding central age, red 1400 points are LA-ICP-MS AFT samples and the orange point is the published Pinet et. al. (2016) sample. Areas in purple show Paleozoic succession of Hudson Bay, sub-basins, and surrounding Arctic platform, 1401 1402 which imply bedrock has been at or near the present surface since (at least) the Ordovician. Refer to table 1403 1 and text in section 4 for more information on AFT samples. Missing section in central Hudson Bay is 1404 younger overlying Mesozoic-Cenozoic sediment. Quebec Grenvillian and Cumberland Peninsula, Baffin 1405 Island samples are shown and discussed for comparison only, as they have a more complex thermal history compared to the Canadian Shield samples. Sample ID shown in box near central age value for 1406 1407 each sample, refer to table 1. THO = Trans-Hudson Orogen; GSL = Great Slave Lake shear zone; STZ = Snowbird Tectonic Zone; CP = Cumberland Peninsula; HP = Hall Peninsula; AB = Athabasca Basin; TB 1408 1409 = Thelon Basin.

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1411 Figure 2. (A) Conventional plot of eU content versus AFT age as well as mean track length for all 1412 bedrock samples. Other than the two high eU examples there is a decrease in age with eU across all 1413 samples. A simple least-squares linear regression (removing the two eU outliers) produces an R^2 value of 0.36. (B) Highest eU grains clearly have the shortest MTL, however there are not enough intermediate-to-1414 1415 high eU data to confidently extrapolate the relationship. (C) All single grains shown in F and Cl weight percent. Barbarand et al., (2003) demonstrate Cl does not dominate the apatite structure (retentivity) until 1416 >~0.35 wt. % or >0.1 apfu. Our dataset is dominated by F-apatite and/or hydroxyl-bearing F-apatite 1417 1418 varieties.

1419 Figure 3. Radial plots (log transformed) from DensityPlotter v. 8.2 for LA-ICP-MS AFT data. "Open jaw" radial plots are typical of our dataset and are generally interpreted as meaning that there is a complex 1420 mixture of AFT ages that are non-Poissonian in character. Refer to table 1 for sample information. As an 1421 1422 example of single-grain age dispersion effects, we have shown the two-component mixture model ages 1423 associated with age peaks of ca. 160 and ca. 75 Ma for sample 09SRB-M100. This illustrates how the 1424 central age for a sample can be misleading in cases of high dispersion. For sample 14SUB-H43A the 1425 opposite is true as this sample only has 10% age dispersion and lower n. Stars denote published samples from Pinet et al. (2016) and Powell et al., (2018a). 1426

1427Figure 4. Fission track length distributions for each sample shown in figure 3. Length distributions are1428presented as frequency (%) in 1 μ m bins with equivalent normal distributions shown for the mean $\pm 1\sigma$ 1429(solid, blue curve) and the mode (dashed, red curve). A mismatch between the equivalent normal1430distributions is taken to suggest bimodality and more complex, episodic thermal histories.

1431Figure 5. Bedrock sample AFT age versus eU and eCl for each sample in Table 1. Notice that each1432sample has a negative age-eU trend for single grains, only sample 12NK-L18A3 does not and this is1433discussed in section 5. There is no clear age relationship with eCl (r_{mr0}). Note: samples are coloured the1434same in figure 5 and figure 9. In the few cases where a grain was not probed, we used the average sample1435eCl value as a placeholder.

1436

1437Figure 6. Examples of the relationship between eU and AFT age, eCl, and r_{mr0} . (A) The Baffin Island1438sample exhibits behaviour indicative of Cl or another element controlling retentivity (> than any eU1439effect), whereas the other samples suggest eU is controlling retentivity (B-C). The panels in C are age-1440normalized to show both samples PBA-98-743 and 03-GRS-013 together. Unfilled circles are values

1441 outside the color ramp maximum. Trend lines (reduced major axis regression) are for visual purposes

between eU and eCl (or r_{mr0}). The color ramp change from red to blue is arbitrarily chosen to be the 1443 1444 approximate mean eCl value of all the grains. Panels D-F: Scatter plots of other Canadian Shield AFT examples. (D) An example from Pinet et al. (2016) showing the relationship between ²³⁸U and age. The 1445 relation is similar to our data except not as well defined given that there are a low number of grains dated 1446 and eU was not calculated, and eCl values are all similar and negative suggesting end-member apatite 1447 1448 composition. (E) Sample 09SRB-M100C1 from Baffin Island shows a spurious relationship between AFT 1449 age and kinetic parameters eU and eCl/r_{mr0} (discussed in text, shown in fig. 6A) that is not readily apparent when eCl is plotted against AFT age shown here. This sample also underwent episodic thermal 1450 1451 annealing. (F) Sample 14SUB-H43A also from Baffin Island displays lower eU variability than most 1452 samples and all single grains are in a range of "typical apatite" eU, it also has characteristic highly 1453 clustered positive eCl values suggesting low kinetic variability between grains. This relationship results in 1454 the appearance of less age variability, which may be naturally inherent to the sample or a result of 1455 unintended grain selection preference.

1456

1457 Figure 7. Correlation matrices for standardized EPMA compositional analyses. Analyses are for Cl. eU. 1458 F, Fe, Mg, Mn, Na, S, Sr, and REE (Ce, La, Y) in wt. % shown in hierarchical clustering of correlation 1459 coefficients and (B and D) only show correlations with p-values <0.05 or at the 95% confidence level. 1460 Positive (+1, black) or negative (anti-) correlation (-1, white) is shown with the size of the corresponding circle equal to the linear correlation coefficient (i.e. small circle is low correlation coefficient and vice 1461 1462 versa). Notice the anti-correlation between F and Cl as expected. (A, B) Examples are shown for the Superior Province sample PBA-98-743 that displays eU variability in relation with single-grain AFT age 1463 1464 (fig. 3 and fig. 5) versus sample 12NK-L18A3 with very high eU (>>100 ppm) and no obvious age-eU 1465 relationship (D). Note that care is required during interpretation of these relationships, as some grains 1466 may seem highly correlated but are represented by only a few EPMA analyses/grains, where the majority 1467 of grains are zero/below detection limit, which is typically the case for REE, seen in panel C plots of 1468 normalized eU, Ce, and La for sample PBA-98-743. Best-fit linear trend (black line) and correlation 1469 coefficient (r) are shown for each plot in C. The SI document shows correlation coefficients for each AFT 1470 sample (table S1) and correlation matrices for each sample (fig. S1).

Figure 8. Example of both AFT and AHe data from Melville Peninsula with respect to α -dose from Pinet 1471 1472 et al., (2016). (A) AFT (sample 09SZ-23-01; circles) and AHe ages (samples 09SZ23-01 and 09SZ24-01; hexagons) with respect to eU. The measured ²³⁸U value is used as a proxy for eU here. This is a good 1473 approximation because most accompanying AHe single grains have low Th, <10 ppm. (B) AFT and AHe 1474 ages plotted against the estimated α -dose. The AFT data are older vintage without U-Pb ages; therefore 1475 we used the t_{EDA} from nearby AFT sample 10CXAD-086. (C-D) The same plot as A and B, but for 1476 sample 13JP10 on Anticosti Island (Powell et al., 2018a). There is no clear relationship between age and 1477 1478 eU for this sample.

Figure 9. Plot of estimated α-dose vs. age for selected Canadian Shield AFT samples summarized in table 1479 1480 1 and shown on fig. 1. Samples with long t_{EDA} plot at high damage levels above the threshold and show negative age- α -dose correlations, whereas samples with shorter t_{EDA} show the expected positive 1481 relationship at low damage below the percolation threshold. Collectively these samples capture the full 1482 1483 range in α -dose or damage level for our dataset. 13LVA04 and 12NK-L18A3 demonstrate the effect of low/high eU variance (σ^2) on α -dose. Black dashed line is the percolation threshold from Ketcham et al., 1484 (2017). Lower panel: estimated α -dose from young samples taken from published literature. All grains 1485 fall below the ²³⁸U damage percolation threshold, implying no significant damage effects on AFT age. 1486 Sample 13JP10 shown on both plots for reference. Single-grain age errors not shown on plots for clarity 1487 1488 but AFT age percent error range (1 σ) for each sample is shown beside the sample ID. Note: Wrangellia 1489 sample plots off of scale at low α -dose.

Figure 10: An example of visualizing multi-kinetic samples in terms of composition and radiation damage for thermal history modeling. Panels are read vertically for each sample. (**A**, **E**) Samples 02NKL-

1492 871 from Ouebec and 11CXAN017 from Melville Peninsula are characterized by high dispersion and fail 1493 the X^2 test. Radial plots show age populations based solely on mixture modeling. Note that age peak 1 for 11CXAN017 contains noticeably higher precision grains (i.e. high eU). (B, F) AFT age plotted against 1494 1495 eCl for each sample. Points are colors by their respective age population shown in the radial plot panels. 1496 (C, G) AFT ages are colored by eCl (same scale for each sample) and plotted against α -dose. There is 1497 complete compositional overlap between populations. (D, H) The same plots as C and G only here the 1498 age populations are instead colored by their respective age populations identified in the radial plot mixture models. Note that grains of similar age (in D) fall above and below the 238 U α -recoil percolation 1499 1500 threshold (dashed black line).

1501

1502 Table 1. Summary of apatite fission track results from across the Canadian Shield (see fig. 1 for 1503 locations). All U measurements used for age calculations were obtained by LA-ICP-MS analysis. All of our samples fail the X^2 test. Percent difference between the central and pooled age is shown. Single-grain 1504 age span between the oldest/youngest grains (not including error) shows the AFT heterogeneity within 1505 1506 samples and eU range is given with the mean eU in brackets. Sample 03-GRS-013 has a single 1507 anomalously old grain, which refers to the age span value in brackets. Refer to text for discussion of 1508 results. Note: some single grains within samples were unable to be probed or do not have chemical data, 1509 therefore an effective Cl/r_{mr0} value was unable to be calculated for those grains, in these instances the 1510 average respective kinetic parameter value for the entire dataset was used for plotting purposes. See SI for 1511 full AFT dataset.

1512

Table 2. Pearson correlation coefficients for all Canadian Shield samples where eU correlated with other
 elemental data. All samples are shown in the SI with the ranked r-values for the top 20 elemental pairs for
 each sample.

Table 1. Apatite fission track results for the Canadian Shield

					no.	grain age	Pooled	1σ	Central	1σ	%	MTL	1σ	track	avg.	avg.	avg.	avg.	Eff.	U-Pb age	2σ
Sample	GSC Lab #	Rock Type	Long. DD	Lat. DD	age gr.	span (My)	Age (Ma)	(Ma)	Age (Ma)	(Ma)	diff	(µm)	(µm)	count	D _{par} (µm)	eU (ppm)	r _{mr0}	Cl (apfu)	Cl (apfu)	pop. (Ma)	(Ma)
Western Superior	Province, C	Intario																			
PBA-98-743	5479	tonalite	-90.12120	49.92996	40	1233	572	16	597	37	4	12.07	1.94	130	1.89	1-19 [7]	0.84	0.008	0.002	2971	174
03-GRS-013	8105	tonalite qtz. diorite	-87.89872	53.66566	40	715 [2119]	414	14	447	34	8	12.51	1.63	131	1.91	1-241 [33]	0.84	0.003	-0.005	3009	94
Slave Province, No	orthwest Te	erritories																			
BNB97-035	8460	biotite granitoid	-112.9215	63.23258	40	495	229	22	272	16	17	12.75	1.80	116	1.72	0.2-98 [21]	0.85	0.006	-0.041	2858	124
12-DRA03-001	8313	plag. porph. intrusive	-114.8500	63.33333	40	378	336	11	344	14	2	12.04	2.39	131	1.87	2-55 [16]	0.83	0.019	0.023	2628	96
12NK-L18A3	10889	granitic gneiss	-102.0765	66.14816	26	236	392	9	397	13	1	11.91	2.06	78	1.85	139-655 [339]	0.79	0.012	0.135	1788	40
Churchill-Rae Pro	ovince, Mel	ville Peninsula regio	n, Nunavut																		
SNB-01-M2055	7271	foliated tonalite	-91.11273	66.87706	30	482	349	10	356	18	2	12.19	2.30	130	2.00	0.2-46 [14]	0.84	0.003	0.002	1940	68
10CXAD-086A	10313	gabbro- anorthosite	-82.05779	67.82549	40	1832	363	28	460	44	24	12.60	1.79	132	2.12	0.2-73 [13]	0.82	0.029	0.059	1803	73
11CXAN017	10697	Bt Kfs monzogranite	-84.79986	67.75710	18	578	418	11	452	31	8	12.67	1.92	120	1.95	3-43 [21]	0.85	0.002	-0.022	1676	59
13LVA04	11068	gabbro	-101.7240	62.69358	40	739	546	34	557	31	2	12.44	2.06	121	1.92	3-42 [7]	0.80	0.012	0.097	2224	125
09SZ-23-01*	N/A	granitic gneiss	-82.84220	69.49660	19	406	463	30	486	27	5	12.18	1.58	100	1.67	11-20 [14]**	0.85	0.016	-0.043	-	-
Churchill-Rae Province, Southampton Island, Nunavut																					
07CYA-M38B	9422	gabbroic anorthosite	-83.60701	64.84525	40	595	342	10	384	21	12	12.22	2.00	132	1.90	1-71 [22]	0.84	0.024	0.007	1834	60
07CYA-M133A	9558	diorite	-81.90056	64.54453	15	357	364	11	375	17	3	11.57	2.16	101	2.26	7-258 [74]	0.80	0.086	0.098	1812	79
Rae craton, southern Baffin Island, Nunavut																					
09SRB-M100	10840	semipelite	-64.48663	65.46074	35	222	97	4	102	8	5	12.94	2.23	41	2.14	1-69 [22]	0.83	0.023	0.041	1674	35
14SUB-H43A	11261	gt-bt psammite	-66.91792	64.56119	25	256	440	11	442	12	0.5	12.46	1.72	130	2.29	16-35 [26]	0.82	0.022	0.045	1782	38
Grenville Province	e, Quebec																				
02NKL-871	7626	granitic orthogneiss	-62.64094	50.26647	35	540	161	5	177	13	9	12.27	1.80	135	1.83	2-160 [31]	0.85	0.008	-0.018	951	38
13JP10*	N/A	Grenv. basement	see Powell	et al., 2018	40	135	123	4	126	4	2	12.37	1.64	171	2.01	5-68 [29]	0.83	0.174	0.038	931	38
*Samples from Pin	net et al. (20	16) and Powell et al.	, 2018a, respect	tively																	

**Calculated by uniformly applying associated single-grain (U-Th)/He average Th and Sm concentrations with each AFT ²³⁸U measurement to give a more representative value for the eU content.

1516

Table 2: Pearson correlation coefficients (r-values) for eU with respect to other EPMA data

P	PBA-98-74	3	03GRS013		4S013 BNB97-035				12-DRA03-001			12NK-L18A3			SNB-01-M2055			10CXAD-086A		11CXAN017			07CYA-M38B			07CYA-M133A			09SRBM100				14SUB-H	143A	02-NKL-871			
eU	Ce	0.68	eU	Ce	0.71	eU	CI	0.30	eU	Na	-0.34	eU	La	0.50	eU	Mg	-0.43	eU	Y	0.41	eU	Y	0.69	eU	Na	0.44	eU	¥	0.58	eU	Fe	0.68	eU	Ce	0.40	eU	Y	0.63
eU	La	0.56	eU	Y	0.51							eU	Ce	0.48	eU	F	-0.37				eU	Mg	0.39	eU	Fe	-0.37	eU	Na	-0.57	eU	Mn	0.61				eU	Ce	0.36
eU	Cl	0.50	eU	s	0.48							eU	Fe	0.41	eU	он	0.36				eU	s	0.34	eU	Y	0.26	eU	La	0.48							eU	s	-0.34
eU	Y	0.46	eU	F	-0.42							•			eU	Fe	0.30				eU	Ce	0.33				•			•						eU	Sr	0.29
eU	Mn	0.45	eU	он	0.42													•			eU	Sr	0.33															
eU	F	-0.41	eU	La	0.39																eU	CI	0.31															
eU	он	0.40	eU	Mg	-0.33																			•														



Figure1





















eU (ppm)

est. alpha dose (α /g)



