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Thorium distribution in the crust: Outcrop and grain-scale perspectives



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ABSTRACT

The spatial distribution of heat producing elements (K, U, and Th) in the continental crust has long-term implications for the thermal and physical evolution of orogens. Heat producing elements, in particular Th, are most abundant in metasedimentary rock types. As such, these rock types have a significant control on the spatial distribution of heat production in the crust. The major host of the heat producing element thorium in pelitic metasedimentary rocks is the REE–Th phosphate monazite. We present in-field gamma ray spectrometry (in-field GRS) data integrated with grain-scale electron probe microanalysis data to reveal grain to terrane scale links in thorium distribution. In-field GRS data shows that thorium is not depleted in granulite facies residual rocks that have lost melt with respect to their subsolidus counterparts. Concurrently, the bulk thorium budget of monazite is approximately uniform within samples and if anything increases with increasing metamorphic grade. Monazite average grain size increases with metamorphic grade and prograde cores are largely preserved in granulite facies samples. Thorium is preserved in residual metasediments after melting and melt loss implying that even when melting and melt extraction is efficient it does not strip Th from granulite facies rocks.

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1. Introduction

Monazite is the major Th-bearing mineral in high temperature (>500 °C) continental crust. Thus, the controls on monazite growth, dissolution and recrystallization control the spatial distribution of Th in the continental crust. It has been a tacit assumption that melting of continental crust will remove Th - a major heat producing element (HPE) from the residue and concentrate it in granitic rocks. However, it is now understood that Th will preferentially partition into monazite over silicate melt regardless of the temperature of crustal melting (Rapp et al., 1987; Skora and Blundy, 2010; Stepanov et al., 2012) and that monazite can remain stable well into the granulite facies (Yakymchuk, 2017; Yakymchuk and Brown, 2014). Recently, Alessio et al. (2018) showed that heat production rates, sampled at the outcrop scale, at least remain constant between sub- and supra-solidus rocks of similar bulk compositions. The implication of this is that Th can commonly remain in the residuum during crustal melting and melt extraction from that residuum.

Controls on the behaviour of Th as a function of progressive metamorphism in metasedimentary sequences are poorly quantified. As the major host of Th in crustal rocks, monazite is ideal for studying these controls. Numerous monazite forming reactions have been proposed (e.g. Corrie and Kohn, 2008; Kohn and Malloy, 2004; Pyle et al., 2001; Pyle and Spear, 2003; Spear and Pyle, 2010), but these studies do not systematically investigate Th behaviour within monazite. However, disparate empirical data suggests that monazite becomes more Th-rich in granulite facies rocks (e.g. Engi, 2017; Watt, 1995).

The aim of this study is to understand the behaviour of Th by presenting a database which quantifies the spatial distribution and chemistry of monazite for rock compositions that have undergone progressive metamorphism from below to above the solidus, with the expressed purpose of understanding the concentration and behaviour of Th in monazite. We track the textural and chemical evolution of monazite in pelitic and psammitic prograde sequences from Mt. Stafford, central Australia and show that metamorphic monazite from midamphibolite facies rocks is comparatively low in Th and that upperamphibolite and granulite facies monazite is elevated in Th. The abundance and overall Th budget of monazite increases with increasing metamorphic grade, in agreement with trends in outcrop-scale gamma ray spectrometer data. Moreover, our dataset lends support to the experimental and thermodynamic modelling findings of Stepanov et al. (2012) and Yakymchuk et al. (2018) in which monazite is found or calculated to increase in Th with increasing temperature.

2. Geological background

Mount Stafford, central Australia (Fig. 1) records an uninterrupted prograde metamorphic sequence in rocks of broadly homogeneous bulk rock composition (Greenfield et al., 1998). Metamorphism ranges from mid amphibolite to granulite facies and the rocks preserve evidence for low accumulated strain (Vernon et al., 1990). The metamorphic system is bounded to the north and east by two granite bodies, with the Northern Granite being emplaced *syn*-post-metamorphism,



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Fig. 1. Map of the Mt. Stafford area showing major metamorphic zones (see Table 1; after Anderson et al., 2013; Greenfield et al., 1996; White et al., 2003) and location of samples in this study. Inset: Location of Mt. Stafford within the Arunta Region and within Australia.

and the Eastern Granite being emplaced pre-syn-metamorphism (Greenfield et al., 1998; Rubatto et al., 2006; White et al., 2003). The sequence consists of interlayered pelites and psammites of the Mt. Stafford beds and their metamorphosed equivalents. The Mt. Stafford beds are interpreted to have been deposited as part of the

more extensive Lander Formation (Greenfield et al., 1998; White et al., 2003), which is constrained by detrital zircons and intrusive granites to be between 1820 and 1802 Ma (Rubatto et al., 2006). Metamorphism in the sequence occurred between 1805 and 1795 Ma (Rubatto et al., 2006).

Table 1

Metamorphic zones in the Mt. Stafford Terrane (after White et al., 2003). Mineral abbreviations after Holland and Powell (1998). Samples from this study are aluminous metapelites and metapsammites. Pressure-temperature constraints from White et al. (2003). Andalusite in the sequence is interpreted to be metastable (White et al., 2003).

Rock type	Mineral assemblages										
	Zone 1	Zone 2	Zone 3	Zone 4							
Aluminous metapelite	mu-bi-a-and	cd-ksp-bi-q-and/sill	cd-ksp-q-sill	cd-ksp-q-sill							
			cd-ksp-sp-bi-sill	cd-ksp-sp-sill							
			cd-ksp-sp-sill								
Subaluminous metapelite	mu–bi–q	cd-ksp-bi-q-and/sill	cd-ksp-bi-q	cd-ksp-bi-q							
	mu-bi-q-and		cd-ksp-sp-bi-sill	cd-ksp-q							
			cd-ksp-sp-bi	cd-ksp-sp							
			cd-ksp-bi-q-g	cd-ksp-bi-sp							
				cd-ksp-q-g							
Metapsammite	mu-bi-q	cd-ksp-bi-q-and/sill	cd-ksp-bi-q	cd-ksp-q-g-opx							
			cd-ksp-bi-q-g	cd-ksp-q-g							
				cd-ksp-q-opx							
Cordierite Granofels	mu-bi-q-cd-and	cd-bi-q-ksp	cd-bi-q-ksp	cd-q-ksp							
	bi-q-cd-ksp	cd-ksp-bi-and/sill	cd-ksp-q-sill	opx-cd-q-ksp							
	bi-q-cd-ksp-and	cd-ksp-bi-q-and/sill	cd-ksp-bi-sill-sp	g-cd-q-ks							
			cd-ksp-bi-q-sill	cd-ksp-q-sill							
				cd-ksp-bi-sill-sp							
P-T constraints	$<\!\!2.55\pm0.25$ kbar	$2.55 \pm 0.25 - 3.05 \pm 0.25$ kbar	$3.05 \pm 0.25 3.65 \pm 0.35$ kbar	$>$ 3.65 \pm 0.35 kbar							
	<620 °C	620-665 ± 15 °C	665 ± 15-780 ± 5 °C	>820 °C							

The sequence at Mt. Stafford preserves a near-isobaric section with a temperature increase of ~300 °C from mid amphibolite (~580 °C; historically called greenschist after Greenfield et al., 1996) to granulite facies rocks (~820 °C; Bartoli, 2017; White et al., 2003). The section has been separated into four zones, bound by mineral isograds, described in Table 1 (Greenfield et al., 1998; White et al., 2003). The abundance of biotite decreases with metamorphic grade, linked by White et al. (2003) to the melt-producing reactions (lowest to highest grade) at Mt. Stafford (mineral abbreviations after Holland and Powell, 2011):

$$bi + sill + q \rightarrow cd + ksp + liq \tag{1}$$

$$bi + sill \rightarrow cd + sp + ksp + liq$$
 (2)

$$bi + q + cd \rightarrow g + ksp + liq$$
 (3)

All of these reactions imply that the decrease in biotite abundance in the samples is linked to the production of melt. The maximum melt productivity of 22–23 wt% was predicted at the peak temperature of 820 °C in the Mt. Stafford area (Bartoli, 2017) and melt loss in the area has been documented by depletion of melt-mobile elements in granulite facies samples with respect to the protolith compositions (Palya et al., 2011; see also White et al., 2003).

3. Sample selection

Samples were selected to be representative of the prograde sequence of metamorphism across the solidus at Mt. Stafford (Fig. 1). There is inherent variability in the trace element concentration between samples due to natural variation in their sedimentary protoliths. This was minimized by careful sample selection on the basis of similarity in major element bulk composition. Metapelite and metapsammite lithologies (Table 2). were selected on the basis of their general propensity for forming monazite. At the outcrop and thin section scale, samples in Table 2 contain an even distribution of poikiloblasts of andalusite and/or cordierite. Only peritectic granulite facies migmatites with poikiloblasts of garnet are obviously more heterogeneous.

4. Methods

4.1. Whole-rock geochemistry

Whole-rock geochemical analyses of samples were undertaken to quantify bulk rock budgets of HPE as well as determine similarity of chemical composition between samples. Whole-rock geochemical analyses were undertaken by Wavelength Dispersive X-ray Fluorescence spectrometry at the Department of Earth and Environment, Franklin and Marshall College, Lancaster PA, USA. Major elements were analysed on fused disks prepared using a lithium tetraborate flux, calculated on a volatile-free basis. Element and oxide concentrations from whole-rock geochemistry will be denoted in the form Th_WR and CaO_WR respectively (i.e. trace and major elements).

4.2. In-field gamma-ray spectrometry (in-field GRS)

Handheld gamma-ray spectrometers (GRSs) were used in the field to provide a broad scale, bulk sampling of the concentrations of heat producing elements (HPEs; K, U, Th) of outcrops. Analyses were conducted using four Radiation Solutions RS-230 model handheld GRSs. The RS-230 model has a 103 cm³ bismuth germinate oxide crystal sensor, and the units were calibrated using concrete test pads, constructed by Radiation Solutions. The detectors were placed directly onto the outcrops and analyses were integrated over 120 s. The sample volume for the RS-230 model is approximately 0.5 m³, therefore only fresh outcrops larger than this were analysed. Machine errors are propagated for all analyses, as per the manufacturer's instructions (see Appendix S1 in Alessio et al., 2018). Analyses were collected from transects of Mt. Stafford from lowest to highest grade, with multiple analyses collected at each location to account for the natural heterogeneity of sequences in outcrop. Thorium concentrations obtained from in-field GRS data are denoted Th_GRS.

4.3. Mineral liberation analysis (MLA)

Entire thin sections of samples were mapped for monazite using a FEI Quanta600 Scanning Electron Microscope (SEM) with automated mineral liberation analysis (MLA) software at Adelaide Microscopy, The University of Adelaide. Imaging and mapping conditions were an accelerating voltage of 25 kV, beam current of 40 nA, spot size of 6.8, working distance of 10 mm and 250× magnification. Mineral

Table 2

Samples and locations. Samples listed in increasing grade order, refer to Fig. 1 (Map). Metamorphic zones after Greenfield et al. (1998); White et al. (2003). Mineral abbreviations after Holland and Powell (2011). Metm., metamorphic. Locations are in UTM coordinates, zone 53 K, using the WGS84 datum.

Sample	Grade	Metm. zone	Rock type	Mineralogy	Location
ST16-31 J	Mid amphibolite	1	Pelite	bi-q-and-mu-ilm	241,394 mE 7,567,785 mS
				apt-mnz-xtm	
STF33P	Mid amphibolite	1	Pelite	mu–bi–q–ksp–mt	247,199 mE 7,562,360 mS
				apt-mnz-xtm	
STF02B	Amphibolite	2	Pelite	ksp-bi-and-q-mu-ilm	251,197 mE 7,562,283 mS
				apt-mnz-xtm	
STF16A	Amphibolite	2	Pelite	ksp-crd-bi-q-and-sill-ilm	252,677 mE 7,564,418 mS
				apt-mnz-xtm	
ST16-09	Granulite	4	Pelite	ksp-sill-crd-bi-pl-q-ilm \pm tor	259,078 mE 7,566,925 mS
				apt-mnz-xtm-zrn	
ST16-19A	Granulite	4	Pelite	ksp-crd-sill-bi-q-pl-ilm \pm g	259,976 mE 7,564,102 mS
				apt-mnz-xtm-zrn	
SI16-31A	Mid amphibolite	1	Psammite	$q-b_1-ksp-mu \pm tor$	241,394 mE 7,567,785 mS
077700	N		D	apt-mnz-xtm-zrn	
STF33	Mid amphibolite	1	Psammite	$b_1-mu-1lm \pm tor apt-mnz-xtm-zrn$	247,199 mE 7,562,360 mS
STF04A	Amphibolite	2	Psammite	$mu-bi-ksp-ilm \pm tor$	251,292 mE 7,562,338 mS
CTE4 OD	A 1 11 11.	2	D	apt-mnz-xtm-zrn	
STF10B	Amphibolite	2	Psammite	ksp-crd-pl-bi-sill-mt	252,150 mE 7,563,754 mS
CTTC CA	Consulity	4	December 14	mnz–apt–xtm–zrn	260 260
51F26A	Granulite	4	Psammite	$ksp-ca-g-bi-sp-q-lim \pm pi$	260,269 mE 7,563,397 mS
CT1C 02C	Consulito	4	Deemarite	dpt=11112=Xtttt=2ftt	259 476 - 57 565 092 - 5
SI 16-03C	Granulite	4	Psammite	q -ksp-cra-bi-pi-siii-iim \pm tor \pm mu mnz-apt-xtm-zrn-ain	258,476 mE 7,565,983 mS



Fig. 2. Grain Average ThO₂ wt% Calculations. Example of calculation from STF16-03C, monazite 7. Grey value and corresponding point analysis ThO₂ wt% concentration are used to scale EPMA maps. Grey values of zones and whole grain are then assigned a ThO₂ wt% concentration based on this calibration. Circles labelled i–iv represent the locations of spot analyses, with the ThO₂ wt% concentrations from those spots given in the greyscale bar.

identification was undertaken by X-ray spot analysis with electron dispersive spectrometers and by mineral liberation analysis (MLA) mapping. MLA mapping is based on defining a searchable grey scale interval within the full range of 0 (black) to 255 (white), where the brightness of 255 is defined by the BSE response of an Au standard (for liberation of monazite, xenotime and zircon) and a rutile standard (for liberation of apatite and allanite). Wherever a mineral occurs within the map region within the searchable range an X-ray spot analysis is taken. Processing of X-ray data is performed by specifying a mineral list, whereby each mineral in the list has an X-ray spectra to be matched against the collected X-rays, to a matching threshold of 70%. Back scattered electron (BSE) scans of full thin sections were collected simultaneously with MLA maps. MLA maps were analysed using pixel counting techniques in Adobe software to calculate the proportion of monazite, apatite, xenotime and zircon in the samples, as well as abundance and grain size of these minerals.

4.4. Electron probe microanalysis (EPMA)

Qualitative mapping and quantitative point analyses of monazite chemistry were performed at Adelaide Microscopy, the University of Adelaide, using a Cameca SXFive electron microprobe. Qualitative compositional mapping used a beam current of 200 nA, accelerating voltage of 20 kV, a dwell time of 100 ms and a step size of 1 µm. For each sample, elements Ca, La, Nd, P, Si and Sm were mapped with wavelength dispersive spectrometers (WDS) and Th, Ce, U, Pb and Y were mapped with energy dispersive spectrometers (EDS). Quantitative point analyses used combined beam conditions, with a beam current of 20 nA and accelerating voltage of 15 kv used for the elements F, Cl, Si, Mg, Al, K, P and Ca, and a beam current of 100 nA and accelerating voltage of 15kv used for the elements Th, U, Pb, Gd, Sm, Nd, Pr, La, Ce and Y. Pb was not analysed for samples STF33, STF33P, STF04A and STF16A to reduce individual spot analysis time. Calibration was done on certified synthetic and natural mineral standards from Astimex Ltd. and P&H Associates. Data calibration and reduction was carried out in 'Probe for EPMA', distributed by Probe Software Inc. Monazite weight % oxide and cation data from Electron Probe Microanalysis will be denoted in the form ThO₂_mnz and Th⁴⁺_mnz respectively.

Average ThO₂ concentrations for individual grains and compositionally distinct parts of grains (monazite zones) were calculated by manually scaling qualitative maps using quantitative point analyses from the same grains (Fig. 2). For each individual grayscale map, the locations of each EPMA point was located, and a grey value for that point was calculated, averaging over 9 pixels (pixel size = $1 \times 1 \mu m$, spot size = 2×2 µm). The average point grey values were plotted against point ThO₂_mnz, and a regression line (and equation) was calculated for each grain. Average ThO₂ concentrations for the whole grain and monazite zones within were then calculated using this regression and the average grey values of each area respectively. Where regression equation slope could not be calculated for an individual grain, the average slope from other grains in the sample was used as the slopes were uniform for a given sample. Grain average and Zone average ThO₂ contents calculated by this method are denoted ThO2_GA and ThO2_ZA respectively.

5. Results

5.1. Whole-rock geochemistry

Whole-rock geochemistry of the 12 samples (6 metapelite, 6 metapsammite) selected for MLA and EPMA are provided in Table 3. A comparison of whole-rock geochemistry of the 12 investigated samples with other samples from the terrane is shown in Fig. 3. Th_WR concentrations for all samples taken at Mt. Stafford are shown in Fig. 4, adjacent to Th_GRS concentrations acquired at the same locations (Alessio et al., 2018). The whole-rock geochemistry for all samples in shown in Fig. 3 are provided in the Appendix. The selected metapelite and metapsammite samples cluster in two distinct compositional groups for most indicator elements (Fig. 3a, c, d); however, there are some elements for which there is cross over between the groups (e.g. Fe, Mg; Fig. 3b).

5.2. In-field GRS

In-field GRS data from Mt. Stafford shows no change in the average Th_GRS concentrations from subsolidus to suprasolidus amphibolite facies rocks (22.9 \pm 2.7 and 23.4 \pm 2.7 ppm Th_GRS respectively, Fig. 4). The range of concentrations in sub- and supra-solidus amphibolite facies rocks is similar (14.3-30.1 and 17.7-47.5 ppm Th_GRS respectively). Seven anomalously high analyses (of 259 total analyses) in the suprasolidus amphibolite facies rocks are in the range 33.1-47.5 ppm Th_GRS. When these analyses are considered outliers, the range of Th_GRS concentrations for suprasolidus amphibolite facies rocks is 17.7–31.9 ppm. This range is the same within error as the range of concentrations for subsolidus amphibolite facies rocks. Granulite facies rocks show increased scatter in Th_GRS concentrations (between 17.3 and 41.7 ppm Th_GRS). Clear distinction between Th_GRS concentrations for leucosome-rich and residuum-rich parts of the sequence is generally not seen in the granulite facies samples, except within the highest temperature migmatites ('schlieren migmatites'; Fig. 4; Greenfield et al., 1996; White et al., 2003). For these rocks, residual-

Table 3

Whole-rock geochemistry. Fe₂O₃ and FeO determined by titration.

	Mt Stafford metapelites							Mt Stafford metapsammites							
	ST16-31J	STF33P	STF02B	STF16A	ST16-09	ST16-19A	ST16-31A	STF33	STF04A	STF10B	STF26A	ST16-03C			
Major eler	ments (wt%)														
SiO ₂	56.90	58.92	55.39	55.53	56.38	54.78	82.18	76.73	83.20	75.16	73.69	73.92			
TiO ₂	0.58	0.54	0.61	0.59	0.52	0.74	0.31	0.37	0.32	0.47	0.55	0.49			
Al_2O_3	25.15	21.06	24.43	24.73	25.13	25.18	9.26	11.17	8.84	13.17	12.78	13.74			
Fe ₂ O ₃ T	7.68	7.13	8.06	7.83	6.90	9.55	2.63	4.20	1.77	4.48	6.61	4.57			
MnO	0.07	0.07	0.13	0.06	0.10	0.11	0.05	0.04	0.06	0.13	0.11	0.07			
MgO	2.33	1.89	2.60	2.03	2.31	3.20	0.75	1.32	0.48	1.35	1.83	1.48			
CaO	0.03	0.10	0.34	0.21	0.36	0.33	0.64	0.44	0.66	0.71	0.47	0.84			
Na ₂ O	0.68	0.44	0.62	1.23	1.37	1.17	1.24	0.98	1.05	0.74	0.59	1.49			
K ₂ O	6.26	6.04	6.64	6.62	6.66	4.58	2.76	2.97	2.41	3.25	2.36	3.63			
P_2O_5	0.05	0.10	0.13	0.13	0.15	0.20	0.10	0.09	0.11	0.11	0.10	0.07			
Total	99.73	96.29	98.95	98.96	99.88	99.84	99.92	98.31	98.90	99.57	99.09	100.30			
LOI	4.42	3.94	2.06	0.72	1.16	0.75	0.70	1.82	1.11	0.89	1.25	0.95			
Fe ₂ O ₃	2.19	-	-	-	0.50	0.50	0.46	-	0.27	-	-	0.57			
FeO	4.94	-	-	-	5.76	8.14	1.95	-	1.35	-	-	3.60			
Trace elen	nents (ppm)														
Rb	448	409	381	408	284	229	128	199	96	162	119	188			
Sr	74	61	106	49	134	148	105	85	57	121	57	117			
Y	25	14	34	29	32	23	20	22	25	24	24	20			
Zr	118	117	118	92	120	166	265	224	335	251	260	273			
V	90	81	92	69	135	113	40	43	27	60	64	57			
Ni	54	55	42	38	43	66	18	29	16	31	38	27			
Cr	86	78	70	60	75	94	56	54	25	59	81	78			
Nb	17	16	17	17	15	17	10	12	10	14	14	13			
Ga	35	32	34	35	36	35	15	17	13	20	20	21			
Cu	34	34	37	41	17	25	13	9	8	16	7	49			
Zn	107	118	111	115	93	117	45	73	27	73	134	65			
Со	17	19	17	18	17	29	2	9	1	11	21	8			
Ba	1011	884	913	1060	1014	956	587	567	487	521	516	852			
La	36	26	45	42	41	53	47	43	46	41	40	34			
Ce	78	163	108	109	93	132	80	75	80	84	83	74			
U	0	1	1	0	0	0	3	1	3	1	0	1			
Th	33	34	34	35	25	38	30	27	37	34	25	30			
Sc	14	15	14	13	12	2	2	5	2	6	17	4			
Pb	11	15	17	23	20	20	12	25	14	13	14	16			

rich layers have systematically higher heat production than leucocraticrich layers (28.7–41.7 and 19.0–24.8 ppm Th_GRS respectively).

5.3. MLA

Representative BSE images of the microstructural settings of monazite are presented in Fig. 5. Monazite grain size, abundance and chemical zones are summarised in Table 4 and monazite microstructural location is summarised in Table 5. A comparison of the proportions of accessory minerals is shown in Fig. 6.

In amphibolite facies samples (ST16-31J, STF33P, STF02B, STF16A, ST16-31A, STF33, STF04A and STF10B) monazite is predominantly hosted at grain boundaries between K-feldspar, biotite, quartz, apatite \pm cordierite, muscovite, ilmenite and andalusite within the matrix (Table 5). Monazite is evenly distributed throughout the samples and frequently occurs as clusters of small grains (<10 µm). Monazite is rarely found in contact with or adjacent to xenotime. Monazite is additionally found as inclusions in some porphyroblastic minerals as follows: ST16-31J, inclusion in porphyroblastic andalusite and within coarse-grained patches of biotite; STF02B and STF16A, included in K-feldspar, biotite and quartz; STF10B, included in cordierite, K-feldspar \pm and quartz. Notably, monazite is not included in K-feldspar porphyroblasts in the sample STF33P. Huttonite-rich grains in sample STF10B are found at grain boundaries between quartz, K-feldspar and cordierite.

In granulite facies samples (ST16-09, ST16-19A, ST16-03C and STF26A), monazite occurs as inclusions in coarse-grained minerals, at grain boundaries between matrix minerals and as elongate grains along the edges of larger apatite grains. Monazite is rarely found in contact with or adjacent to xenotime. In ST16-09, monazite grains are

evenly distributed throughout the sample, and occur at grain boundaries between K-feldspar, biotite, guartz, spinel, cordierite \pm ilmenite. In addition to monazite grains found on the edges of coarser grained apatite, micro-inclusions ($<1 \mu m$) of monazite are found within these coarse grained apatites. In sample ST16-19A, monazite occurs at grain boundaries between K-feldspar, cordierite, quartz, spinel and ilmenite, and additionally as inclusions in coarse grained K-feldspar, cordierite, biotite, ilmenite and spinel. In sample STF26A- which is a heterogeneous peritectic-mineral bearing migmatite- monazite occurs in a broad range of microstructural locations, but is notably absent from within peritetic garnet grains. In this sample monazite occur at grain boundaries between K-feldspar, cordierite, quartz, spinel, biotite, ilmenite \pm sillimanite and garnet. They are also included in coarse grained K-feldspar, apatite, ilmenite, spinel and biotite, as well as within fine grained aggregates of cordierite, and within fine grained spinel-quartz symplectites (adjacent to cordierite). Sample ST16-03A contains monazite hosted at grain boundaries between cordierite, K-feldspar, biotite and quartz, and as inclusions in biotite, K-feldspar, cordierite, quartz and ilmenite.

Xenotime is predominantly hosted at grain boundaries (>89.77% of xenotime) in the subsolidus samples and the two granulite facies psammite samples (ST16-31J, STF33P, STF02B, ST16-31A, STF33, STF04A, STF26A and ST16-03C), with the remainder being hosted as inclusions in K-feldspar. In the suprasolidus pelite samples (STF16A, ST16–09 and ST16-19A) approximately half of the xenotime is hosted at grain boundaries (59.70%, 49.14% and 43.13% respectively). The remainder of the xenotime in these samples is hosted as inclusions in K-feldspar (40.30%, 50.86% and 18.75% respectively) and, in ST16-19A, cordierite (34.38%) and coarse grained biotite (3.75%). The lowest proportion of grain boundary xenotime is found in sample STF10B



Fig. 3. Whole-rock geochemistry from Mt. Stafford. a. Al₂O₃-CaO + Na₂O-K₂O diagram after Janots et al. (2006). Average pelite composition after Shaw (1956); b. AFM diagram; c. Al₂O₃-WR vs Ce_WR + La_WR; d. Al₂O₃-WR vs SiO₂-WR; e. Zr_WR for investigated samples. Samples increase in metamorphic grade from left to right. Metapelite samples (blue) are labelled below, metapsammite samples (red) above. Sample prefixes (STF and ST16-) have been omitted, refer to Table 2 for full sample names.



Fig. 4. Th_GRS (open cirlces) and Th_WR (filled circles) concentrations for Mt. Stafford as a function of metamorphic grade (Th_GRS from Alessio et al., 2018): Solid red line indicates the infield location of the solidus, black solid lines indicate facies transitions, black dotted lines indicate significant boundaries within facies and gray shaded area indicates a break in the GRS transect (see Fig. 1). Error bars are 2 sigma, Th_GRS errors calculated from machine errors and Th_WR errors calculated from standard data. Pressure-temperature estimates from White et al. (2003). "Schlieren migmatites" refer to samples of segregated leucosome and residuum (see White et al., 2003).



Fig. 5. Representative BSE photomicrographs showing microstructural setting of monazite. White arrows indicate monazite grains. a. ST16-31J, monazite included in porphyroblastic andalusite and within matrix; b. ST16-31J, monazite included in biotite clusters and within matrix; c. STF10B, monazite and huttonite within matrix; d. ST16-09, monazite and xenotime at grain boundaries between coarse-grained apatite and matrix. Monazite included within coarse-grained apatite; e. ST16-09, monazite within matrix; f. ST16-19A, monazite included in biotite, K-feldspar, cordierite and interstitial quartz; g. STF26A, monazite and xenotime at grain boundaries between coarse-grained apatite and matrix. H. STF26A, monazite included in cordierite, quartz and spinel and within spinel-quartz symplectites; j. monazite included in coarse grained biotite. Scale bar = 200 µm. And, andalusite; ap, apatite; bi, biotite; crd, cordierite; gt, garnet; hut, huttonite; ilm, ilmenite; ksp, K-feldspar; mu, muscovite; qz, quartz; sill, sillimanite; sp, spinel; xtm, xenotime; zrn, zircon.

(17.46%) with the remainder of the xenotime hosted as inclusions in K-feldspar. In the two samples that contain zone D (apatite grain boundary) monazite (ST16–09 and STF26A), xenotime found at grain boundaries of coarse grained apatite account for 3.09% and 49.26% respectively of all grain boundary xenotime.

Monazite volume proportions at Mt. Stafford, calculated from point counting, are in the range 0.003–0.027% for pelite samples and 0.007–0.016% for metapsammite samples (Fig. 6).Monazite is present in all samples and, if anything, the proportion of monazite increases with metamorphic grade.

Monazite volume increase is accompanied by an increase in apatite volume at higher grades in both metapelite and metapsammite samples (Fig. 6). Zircon proportion in the metapelite samples decreases from mid to upper amphibolite, then increases to the granulite facies. In the metapsammite samples, zircon proportion is similar in all samples, but slightly higher in the upper amphibolite facies samples (STF04A, STF10B). Xenotime abundance decreases with metamorphic grade in the metapelite samples, and increases with metamorphic grade in the metapsammite samples. Allanite was not detected in the samples except as rare intergrowths with apatite on monazite rims in sample ST16-03C (see Fig. 7).

5.4. EPMA

Representative analyses of monazite composition are given in Table 6 and compositional ranges are summarised in Table 7. Representative EPMA maps of monazite and trends in ThO2_mnz with metamorphic grade are shown in Figs. 7 and 8 respectively. On the basis of EPMA

Table 4

Monazite grain characteristics. Number of monazite grains and average grainsize from MLA maps of thin sections, ThO₂_GA and mnz zones from EPMA determined from map and point analysis data (see text for details). Mnz, monazite; total incl. mnz, volume % of monazite included in all minerals.

Sample	Grade	n mnz grains	Average grainsize (µm²)	ThO ₂ _GA	mnz zones	total incl. mnz
Mt Stafford	metapelites					
ST16-31J	Mid amphibolite	628	87	1.70-3.81	А	0.122
STF33P	Mid amphibolite	56	95	2.10-3.58	А	0.044
STF02B	Amphibolite	84	201	2.70-3.77	A, B	0.335
STF16A	Amphibolite	48	415	5.10-5.70	A, B	0.411
ST16-09	Granulite	152	346	3.35-4.98	C*, D	0.451
ST16-19A	Granulite	9	4418	4.23-5.01	A, B, C	0.860
Mt Stafford	metapsammites					
ST16-31A	Mid amphibolite	902	20	0.21-4.73	А	0.015
STF33	Mid amphibolite	254	79	1.65-5.98	А	0.052
STF04A	Amphibolite	117	177	2.45-3.45	A, B	0.072
STF10B	Amphibolite	97	314	2.18-5.8	A, B, B*	0.185
STF26A	Granulite	22	1257	4.83-9.44	A, B, C, D	0.431
ST16-03C	Granulite	75	707	2.02-22.76	A, B, C	0.193

map and point data as well as the spatial distribution of compositional zones within monazite, monazite grains and parts of grains in the samples have been separated into six chemical zones as follows:

Zone A monazite occurs in all samples except ST16-09, as either whole grains or as cores of grains. Zone A monazite commonly occurs as clusters of small grains (<10 μ m each, Fig. 7), particularly in the amphibolite facies samples. In upper amphibolite and granulite facies samples, zone A monazite forms the cores of larger grains, with typically embayed zone edges (Fig. 7). This zone is rich in the Ce-monazite endmember, but shows a range of ThO₂_ZA contents (0.21–6.58 wt%; mean 2.76 wt%; Table 6), which define a sublinear negative correlation with Ce₂O₃_mnz contents. These grains have low Y₂O₃_mnz concentrations (0.75–2.84 wt%; Table 6).

Zone B monazite occurs in upper amphibolite and granulite facies samples. This zone occurs rim-ward of zone A and has higher Th_ZA (0.50–8.76 wt%; mean 5.49 wt%; Table 6) and lower Ce_2O_3 _mnz (25.66–30.04 wt%; Table 6) than Zone A.

Zone B* monazite is found exclusively within sample STF10B, occasionally adjacent to grains of huttonite. Zone B* monazite has very high Th_ZA contents in the range 9.06–21.14 wt% (Table 6). Zone B*



Fig. 6. Accessory mineral volume (as %), calculated by point counting of MLA maps of entire thin sections (approximately 25x50mm). Pixel size of maps is 1.03 μ m. Samples increase in metamorphic grade from left to right. Mnz = monazite, apt = apatite, zrn = zircon, xtm = xenotime.

monazite also occurs as grains with cores containing Zone A and B monazite (Fig. 7). Grains of zone B* monazite show no core-rim relationship with adjacent huttonite.

Zone C monazite occurs in granulite facies samples, rim-ward of zone B. Zone C monazite commonly surrounds cores with components of both zones A and B. Zone C monazite has high Th_ZA (3.71–6.99 wt %; mean 5.54 wt%) and comparable Ce₂O₃_mnz (25.23–29.03; Table 6) to Zone B.

Table 5

Summary of monazite microstructural locations. Microstrutural locations and proportions determined by point counting of MLA maps (see text for details). Mineral abbreviations after Holland and Powell (2011). Cg, coarse grained.

Sample	Grain b	oundary	/	Stable	e phases					React	ant phas	es		Product p	ohases					
	matrix	apt	total	and	bi (cg)	mu	Ksp core	ilm	total	bi	qz	crd	total	ksp rim	sill	pl	crd	sp	gt	total
Metapelite	S																			
ST16-31J	94.05		94.05	3.49	2.46				5.95				0.00							0.00
STF33P	87.24		87.24	8.16		2.04	1.53		11.73		1.02		1.02							0.00
STF02B	75.26		75.26		0.61		1.69		2.30	3.91	6.47		10.38	12.06						12.06
STF16A	68.61		68.61		0.89		7.59		8.48	0.71	4.88		5.60	17.32						17.32
ST16-09	76.42	0.39	76.81				1.17		1.17	2.09			2.09	19.16	0.78					19.94
ST16-19A	67.64		67.64		5.04		4.81		9.85	4.84		1.69	6.53	10.59	0.11	0.15	5.14			15.98
Metapsami	mites																			
ST16-31A	97.89		97.89						0.00		2.11		2.11							0.00
STF33	94.58		94.58		0.61	1.06			1.68		3.74		3.74							0.00
STF04A	93.75		93.75		2.12	1.06	3.07		6.25				0.00							0.00
STF10B	88.65		88.65				0.97		0.97		7.68	0.92	8.60	1.30			0.47			1.77
STF26A	65.78	6.15	71.93		1.26		8.83	0.15	10.24	0.87	5.88		6.74	6.67	0.27		1.34	1.53	1.27	11.08
ST16-03C	86.86		86.86		1.01		0.47		1.48		11.66		11.66							0.00



Fig. 7. Representative EPMA maps of monazite from Mt. Stafford. Images are composite qualitative maps of thorium and cerium with the two element maps overlain (thorium in green and cerium in blue). A–D, B^{*}, C^{*} refer to monazite zones described in text. Yellow areas represent thorium-rich minerals allanite and huttonite (labelled Aln and Hut respectively).

Zone C* monazite is found exclusively in sample ST16–09 and has the highest Y_2O_3 _mnz concentrations of the zones (2.04–5.10 wt%). It has lower Ce_2O_3 _mnz concentrations (24.56–27.61 wt%) and Th_ZA concentrations in the range 3.35–5.57 wt% (mean 4.17 wt%; Table 6). Zone C* monazite typically has higher HREE concentrations (Table 6).

Zone D monazite occurs as small, elongate grains at the grain boundaries of apatite (Fig. 5) in samples ST16-09 and STF26A. This zone has characteristically low Th_ZA contents (0.38-0.44), high Ce₂O₃_mnz (29.13–30.77) and (relatively) high Y₂O₃_mnz (2.37–3.06; Table 6). Zone D monazite typically has very low Si and Ca concentrations (Table 6).

Not all EPMA point analyses could be uniquely assigned to a monazite zone either due to mixing or zones being unresolvable at the scale of the EPMA maps. The zones A–D are described on the basis of analyses that could be unambiguously assigned to one zone. In both metapelitic and metapsammitic samples, ThO₂_GA concentrations (weighed average Th concentrations from individual grains) are higher in granulite facies rocks than subsolidus samples with similar bulk compositions (Fig. 8b,c).

Monazite grains for every unique microstructural location were analysed (EPMA points and EPMA maps) to allow textural analysis of monazite composition. This analysis revealed limited microstructural control on the distribution of all monazite zones except zone D, which is limited to the grain boundaries of large apatite grains (see above). In samples ST16-31J, STF33P, STF16A, ST16-09, ST16-31A, STF33, STF04A, STF10B and STF26A there is no correlation between microstructural location and composition for zones A. B. B^{*}. C and C^{*}. such that monazite found at grain boundaries and as inclusions show the same type of zonation. In sample STF02B, rare grains of monazite composed wholly of zone A occur as inclusions in K-feldspar, with monazites in all other microstructural locations composed of both zones A and B monazite. In sample ST16-03C, there is one monazite grain wholly composed of monazite zone B, which is found in contact with xenotime. There are two grains which show breakdown (interpreted as retrogression) to a mixture of finer-grained allanite and apatite, one of which is included in biotite and the other of which is at a grain boundary between quartz and K-feldspar. Sample ST16-19A contains one instance of monazite with only zones A and B included in K-feldspar. All other

Table 6	
Representative EPMA analyses of monazite.	Hut, Huttonite: N/A, not analysed.

mnz zone	А	В	B*	С	C*	D	Hut
Sample	ST16-31J	STF04A	STF10B	ST16-19A	ST16-09	STF26A	STF10B
Grain	mnz 19	mnz 4	mnz 4	mnz 8	mnz 17	mnz24	mnz 3
Analysis	59-1	48-1	7-5	29-2	119-3	82-1	6-4
6:0	0.22	0.27	1.50	0.42	0.22	0.17	2.20
SIU ₂	1.32	0.37	1.52	1.00	1.07	0.17	3.20
CaO V O	1.20	1.20	2.42	1.00	1.07	0.62	2.08
$1_{2}U_{3}$	1.20	1.20	2.54	1.57	4.04	12 65	1.74
Ld_2U_3	14.49	20.04	10.76	14.42	14.00	15.05	16.06
Ce_2O_3	20.31	2 27	22.55	20.77	20.40	2 21	10.90
PI2U3	2.05	5.27 11 17	2.29	2.99	2.09	5.21 12.44	1.70
Sm 0	10.22	1 60	0.40	1 72	5.72 1.72	12, 44 2.01	1.05
Cd-O-	1.70	1.05	1.44	1.75	1.75	1.60	1.24
	5.21	3.24	1.23	5.12	1.07	0.38	1.15 27.18
1102	0.55	0.12	0.47	0.13	1.00	0.38	0.41
PbO	0.53	0.12 N/Δ	1.28	0.15	0.61	0.42	2.10
PDO PO	22.27	20.42	20.55	21 22	21.79	22.06	2.10
Total	100.21	08 33	00 10	100.15	00.10	100.23	20.51
Total	100.21	50.52	55.15	100.15	55.15	100.25	55.50
Si ⁴⁺	0.012	0.014	0.059	0.016	0.012	0.007	0.129
Ca ²⁺	0.051	0.021	0.101	0.041	0.044	0.025	0.116
Y ³⁺	0.024	0.027	0.048	0.028	0.082	0.061	0.037
La ³⁺	0.202	0.217	0.155	0.203	0.197	0.190	0.117
Ce ³⁺	0.391	0.430	0.321	0.402	0.367	0.419	0.250
Pr ³⁺	0.039	0.047	0.032	0.042	0.037	0.044	0.026
Nd ³⁺	0.138	0.156	0.118	0.151	0.132	0.167	0.096
Sm ³⁺	0.023	0.023	0.019	0.023	0.023	0.026	0.017
Gd ³⁺	0.016	0.015	0.016	0.018	0.021	0.021	0.015
Th ⁴⁺	0.045	0.029	0.131	0.044	0.035	0.003	0.249
U^{4+}	0.005	0.001	0.004	0.001	0.008	0.004	0.004
Pb^{2+}	0.005	N/A	0.013	0.004	0.006	0.001	0.023
P ⁵⁺	1.030	1.008	0.973	1.013	1.021	1.022	0.904
Total	1.979	1.987	1.99	1.986	1.984	1.99	1.983
cations							
(S)							

monazite grains included in K-feldspar also contain zone C. In this sample, the only grain found to be wholly composed of zone C monazite occurs at the grain boundary between ilmenite, biotite and quartz, but other grains found in similar microstructural locations also contain monazite zone B.

6. Discussion

6.1. Melt loss and the preservation of monazite

Previous studies of the Mt. Stafford terrane have established that up to 23% melt was produced at the highest metamorphic grade (Bartoli, 2017) and that a portion of this melt was subsequently extracted from

the sequence (Palya et al., 2011; White et al., 2003) consistent with the decrease in biotite abundance across the terrane represented by the samples in this study. Monazite is present in all of the samples in this study and grains from the highest grade rocks preserve interpreted prograde zoning features (Fig.7). This indicates that monazite was stable up to the highest grades of metamorphism in the terrane.

Zr_WR from the studied samples (Fig. 3) increases with metamorphic grade in both metapelitic and metapsammitic lithologies, with this increase more pronounced in the metapelite samples. Typically Zr is highly compatible and partitions into zircon rather than melt (e.g. Rubatto and Hermann, 2007), and with increasing temperature, the mode of relatively Zr-rich minerals (e.g. garnet) also increases (Kohn et al., 2015). The concurrent increase in zircon proportion with metamorphic grade, particularly in the metapelite samples, suggests that in the Mt. Stafford rocks zircon did not participate in melting reactions to a large degree and that the Zr_WR concentration is a proxy for increasing melt loss from the residue. This trend supports the findings of Bartoli (2017) and Palya et al. (2011) that melt was progressively lost for the Mt. Stafford terrane along the prograde path. Given the role of bulk rock Zr content on relative changes to zircon abundance between the rock types we have studied (Kelsey et al., 2008; Yakymchuk and Brown, 2014), it is not possible to determine the relative abundance of melt loss in the pelite and psammite sequences uniquely from this dataset. That is, rocks with higher bulk Zr will record a proportionally smaller increase to zircon abundance with increasing temperature as a function of melt production and loss (Kelsey et al., 2008; Yakymchuk and Brown, 2014), as seen for the metapsammites in Fig. 3, that is not uniquely a function of more or less melt loss than the metapelites.

6.2. Bulk-rock trends in thorium distribution

GRS data from Mt. Stafford shows that there is an overall increase in Th_GRS concentration from amphibolite to granulite facies rocks (Fig. 4; Alessio et al., 2018). In the granulite facies, the scatter in Th_GRS and Th_WR data is larger than in the amphibolite facies rocks (Fig. 4). This probably suggests small-scale differentiation in the granulite facies, consistent with greater heterogeneity of grain size and mineralogy in these samples due to the presence of coarse peritectic mineral-bearing leucosomes. Residual rocks segregated from leucosomes ("schlieren migmatites"; Fig. 4; White et al., 2003) show an increase in Th_GRS concentrations relative to subsolidus rocks with similar compositions, as well as an increase compared to Th_GRS concentrations in the leucosome-rich rocks in the same part of the sequence. Together these observations suggest that during melting Th preferentially partitioned into the residuum, which in turn suggests that with increasing melt fraction the residuum becomes more enriched in Th. This finding supports experimental data of Stepanov et al. (2012) in which Th strongly

Table 7

Ranges of compositional variables for monazite zones. Th_ZA wt% concentrations calculated from EPMA point analyses and compositional maps. All other variables calculated from EPMA point analyses.

1							
	Zone A	Zone B	Zone B*	Zone C	Zone C*	Zone D	Huttonite
ThO2_ZA wt%							
Range	0.21-6.58	0.50-8.76	9.06-21.14	3.71-6.99	3.35-5.57	0.38-0.44	25.43-42.58
Mean	2.76	5.49	16.54	5.54	4.17	0.41	32.20
Point analyses							
ThO ₂ wt%	0.40-10.32	3.02-9.74	13.93-27.18	4.50-7.58	3.27-5.80	0.22-1.69	7.92-25.20
Ce ₂ O ₃ wt%	23.89-31.70	25.66-30.04	16.96-22.88	25.23-29.03	24.56-27.61	29.13-30.77	16.73-25.82
Y ₂ O ₃ wt%	0.75-2.84	0.93-3.77	1.71-2.30	1.85-3.61	2.04-5.10	2.37-3.06	1.64-2.80
Th wt%	0.00-9.07	2.65-8.56	12.25-23.89	3.95-6.66	2.87-5.10	0.19-1.49	6.96-22.14
Ce wt%	10.20-13.53	10.96-12.83	7.24-9.77	10.77-12.39	10.48-11.79	12.43-13.14	7.14-11.02
Y wt%	0.30-1.12	0.37-1.48	0.67-0.91	0.73-1.42	0.80-2.01	0.93-1.21	0.65-1.10
p(mnz)	0.81-1.01	0.85-0.95	0.04-0.15	0.87-0.93	0.89-0.93	0.97-1.00	0.01-0.09
p(cher)	0.01-0.19	0.04-0.14	0.18-0.29	0.06-0.13	0.07-0.11	0.01-0.03	0.12-0.29
p(hut)	-0.02 - 0.04	-0.03-0.05	0.60-0.78	-0.01 - 0.01	-0.01 - 0.01	-0.02 - 0.00	0.62-0.87

 $Th_ZA = zone average Th, p(mnz) = proportion of monazite end-member (\sum REE), p(cher) = proportion of cheralite end-member (2Ca^{2+}), p(hut) = proportion of huttonite end-member (Th^{4+}+U^{4+}-Ca^{2+}), p(hut) = proportion of huttonite end-member (Decay), p(hut) = proportion of huttonite end-member (Th^{4+}+U^{4+}-Ca^{2+}), p(hut) = proportion of huttonite end-member (Decay), p(hut) = proportion o$



Fig. 8. Zone average and grain average thorium concentrations from Mt. Stafford. a. Zone average ThO_2 wt% concentrations vs monazite zone. Monazite zones B* and C* are found exclusively within samples STF10B and ST16–09 respectively, see text for details; b. Grain average ThO_2 wt% concentrations for metapelite samples; c. Grain average ThO_2 wt% concentrations for metapelite samples.

partitioned into monazite rather than silicate melt, with monazite commonly being retained in the residuum (Watt, 1995; Watt and Harley, 1993).

Th_WR concentrations for all investigated samples from Mt. Stafford are the same within error, however 2σ errors on these analyses are very large (~20 ppm) and thus analytical error masks any systematic trend that may be present in the whole rock budget of thorium for these samples. Mass balance approximations of the abundance and chemistry of

monazite and other Th-bearing minerals (apatite, xenotime and huttonite) in the samples suggest that the whole rock concentration of thorium should increase with metamorphic grade. This is consistent with trends in Th_GRS which show that thorium concentration (albeit from larger sample volumes than whole rock XRF chemistry) is preserved through the prograde sequence at Mt. Stafford, and enriched in the residue with respect to the melt fraction (Fig. 4).

6.3. Monazite distribution at Mt. Stafford

Monazite volume proportions in metapelitic samples broadly increase with increasing metamorphic grade (Fig. 6). The two lower amphibolite facies metapelitic samples, ST16-31J and STF33, have significantly different proportions of monazite (0.020 and 0.003 vol% respectively). These samples also have markedly different proportions of apatite, with 0.001 and 0.072 vol% respectively, correlated with Ce_WR of 78 and 163 ppm, La_WR 36 ppm and 26 ppm and Ca_WR of 0.03 and 0.10 wt%, all respectively. This is interpreted to be a relic of source heterogeneity in the abundance of the phosphates or bulk-rock composition, with ST16-31J being a monazite-rich layer, and STF33P being a relatively monazite-poor and LREE-calcium-rich layer. Elevated calcium content of a rock has been reported to increase the stability of apatite and/or allanite at the expense of REE-monazite (discussed further below; Spear and Pyle, 2010; Yakymchuk, 2017). Monazite volume proportions increase consistently in the metapelitic samples from the upper amphibolite facies into the granulite faces, accompanied by a decrease in biotite abundance.

In the metapsammitic samples, monazite volume proportions increase through the amphibolite facies and then decrease slightly in the granulite facies rocks (Fig. 6), suggesting that monazite is less stable in metapsammitic compositions than metapelitic compositions in the granulite facies. This may be a function of lower total LREE whole rock concentration in metapsammites compared to metapelites, a known control on monazite stability (e.g. Kelsey et al., 2008; Rapp et al., 1987; Rapp and Watson, 1986; Yakymchuk, 2017). Furthermore, in granulite facies sample ST16-03C allanite occurs as rims on monazite in some cases (Fig. 7). Allanite is known to form by the breakdown of monazite in (sub-)greenschist facies rocks (e.g. Budzyń et al., 2017; Gieré and Sorensen, 2004; Janots et al., 2007; Smith and Barreiro, 1990; Wing et al., 2003), so these allanite rims likely represent breakdown of monazite on the retrograde path, and account for some of the decrease in monazite abundance in the granulite facies samples. The allanite in these rare cases is intergrown with apatite, and from raw EPMA map data the intergrowth has similar Th content to zone B monazite from the same grain.

In both lithologies, monazite grain size broadly increases with increasing metamorphic grade (Table 4). This is linked with decreasing numbers of monazite grains (Table 4), which suggests sample scale recrystallization of monazite and movement of thorium occurred, with increasing efficiency at higher grades, as monazite is the only major Th-bearing mineral in the samples. This may be a result of Ostwald ripening, where smaller grains are preferentially dissolved which facilitates the growth of new monazite on the crystal faces of larger grains (cf. Rapp and Watson, 1986; Yakymchuk and Brown, 2014). Wholesale recrystallization of all monazite grains is unlikely as monazite grains from granulite facies rocks preserve compositional zones (A and B), predominantly within cores of grains, (Fig. 7; see Taylor et al., 2016), which have consistent chemistry with monazite found in lower grade samples, interpreted as preservation of prograde zoning in these grains.

Monazite in all samples is evenly (but not entirely homogeneously) distributed at the thin section scale, occurring at grain boundaries and within porphyroblasts in all mineralogical domains in the granulite facies samples. The total amount of monazite hosted as inclusions is higher in the granulite facies samples than the subsolidus amphibolite facies samples (total incl. mnz, Table 4). This represents an important mechanism for retaining monazite in granulite facies rocks (e.g. Bea,

1996; Watson et al., 1989). In all samples the majority of monazite is hosted at grain boundaries (67.64–94.05% in pelite samples, 71.93–97.89% in psammite samples, Table 5), with slightly more monazite hosted at grain boundaries in the psammitic samples. The grainsize of monazite hosted at grain boundaries is also larger in the granulite facies samples relative to amphibolite facies samples, suggesting that monazite dissolution and reprecipitation was assisted by the mobility of monazite forming elements (REEs and Th) along grain boundaries. The grainsize of included monazite is typically <10 μ m and thus the internal variation of many included grains was irresolvable at the scale of the maps. Those grains for which internal zones were resolvable record both prograde and peak monazite compositions.

Monazite throughout each thin section has ThO₂_GA concentrations that show no systematic variation with microstructural setting, except for where grains are wholly composed of zone D, discussed further below. This suggests that dissolution/reprecipitation processes occurred in a somewhat efficient manner, as grains become more systematically zoned with increasing metamorphic grade. Zones A–D are found in multiple samples and suggest that similar reactions and/or diffusion processes were in operation within the samples during the growth of monazite. Zones B* and C* are only found in samples STF10B and ST16-09 respectively, suggesting that these samples have distinct chemistries that allowed alternative reactions/exchange mechanisms to dominate monazite growth.

6.4. Monazite-forming reactions

In the following we explore possible reaction mechanisms by which monazite may have grown, taking into account (a) volume proportion changes; (b) microstructural location of monazite; (c) chemistry of monazite zones A–D; and (d) possible sources of LREE and Th.

Most studies linking monazite growth to silicate mineral reactions have focussed on garnet-bearing rocks or sequences (e.g. Corrie and Kohn, 2008; Kohn and Malloy, 2004; Pyle et al., 2001; Pyle and Spear, 2003) and have linked the Y concentration of monazite to Y-in-garnet and Y-in-xenotime (e.g. Pyle et al., 2001; Spear and Pyle, 2010). However, almost all the Mt. Stafford rocks are garnet- and plagioclaseabsent (REE hosts), except in the leucosome (melt)-bearing granulite facies rocks (Greenfield et al., 1996; Rubatto et al., 2006). Therefore it is more difficult to link monazite/accessory mineral growth to major silicate mineral reactions for the Mt. Stafford samples, but K-feldspar is likely to play an important role as a LREE host/source (e.g. Rubatto et al., 2006), as is apatite (e.g. Yakymchuk, 2017).

Previously proposed monazite-forming reactions at amphibolite facies (e.g. Gasser et al., 2012; Janots et al., 2008; Tomkins and Pattison, 2007; Wing et al., 2003) generally contain allanite as a reactant and plagioclase as a product, neither of which is common in the Mt. Stafford samples. P–T equilibrium modelling of monazite and allanite (Spear, 2010; Spear and Pyle, 2010) shows that the bulk rock CaO composition is one factor which has a dramatic effect on the stability of allanite and monazite (cf. Yakymchuk et al., 2017). For a bulk rock CaO content of 1.08 wt%, the allanite-monazite phase transition occurs at ~450 °C at 3 kbar, whereas in a composition with double the CaO content, the same transition occurs at ~700 °C at the same pressure. This suggests that for the Mt. Stafford samples (all with CaO <1.08 wt%), any prograde/detrital allanite would have been removed from the rocks a temperatures below 450 °C, well below the minimum temperature estimate for the lowest grade samples (580 °C). However, as there is no direct evidence for the presence of allanite below the amphibolite facies at Mt. Stafford, we cannot prove that it existed during prograde metamorphism. If allanite was not the Ca source for apatite (and monazite) then the source may have been pre-existing/ detrital apatite, or small amounts liberated from silicate minerals such as feldspar. The potential role for xenotime in this reaction is not clear.

The concentration of Th in monazite is known to increase up temperature from both natural examples (e.g. Engi, 2017) and predictions from phase equilibria forward modelling (Yakymchuk et al., 2018). Zone A monazite is interpreted to have grown at temperature conditions lower than the recorded conditions in the lowest grade rocks exposed at Mt. Stafford (i.e. on the prograde path), since it has the lowest Th concentration of the three major zones (A-C) and is present in all samples except ST16-09. The heterogeneity of Th_ZA concentrations (and individual EPMA point analyses) in zone A monazite is large compared with other zones, and this may reflect a larger proportion of detrital monazite with highly variable composition. Much of the monazite in the mid-amphibolite facies rocks where zone A monazite is most prevalent was too small to be analysed, and thus the full extent of this heterogeneity cannot be assessed. Zone A monazite may have been grown according to a reaction that involved the breakdown of allanite at lower temperatures (i.e. greenschist or lower amphibolite) than those previously proposed (e.g. Gasser et al., 2012; Janots et al., 2007; Rasmussen et al., 2006; Tomkins and Pattison, 2007; Wing et al., 2003), although this cannot be assessed in these samples.

Growth of monazite zones B and C are similarly difficult to link to precise mineral reactions. The progression of low Th cores (zone A) to higher Th rims (zones B and C) is contrary to the findings of (Kohn and Malloy, 2004), where monazite evolved from high Th to low Th with prograde metamorphism. This was proposed to be due to stoichiometric constraints, with earlier monazite sequestering the majority of Th in the rock. Thus, later monazite (rims) was enriched in LREE because less thorium was available. This mechanism may explain the slight reduction in ThO₂_ZA between zones B and C (see also Engi, 2017). The increase in ThO₂_ZA between zones A and B is concurrent with an increase in apatite volume proportion from the mid to the upper amphibolite facies. Both these trends are more apparent in the metapsammite samples and may have a causal relationship, where the increase in the stability (and therefore growth) of apatite makes the LREEs less available for the growth of monazite, and thus zone B monazite is more Th-rich than zone A monazite. Simultaneously, melt has been lost from the system with metamorphic grade which likely results in an apparent increase in the volume of monazite with metamorphic grade, but limited extraction of thorium from the residue (see below). The slight decrease in Th between zone B and C monazite may reflect the Rayleigh-like stoichiometric mechanism proposed by Kohn and Malloy (2004), or that some Th is lost to melt as a consequence of partial monazite dissolution (e.g. Stepanov et al., 2012).

Zone D monazite occurs exclusively around the margins of larger metamorphic apatite grains in the granulite facies samples (ST16–09, STF26A), and is interpreted to have formed by breakdown of apatite during the retrograde metamorphic history (Fig. 5; e.g. Harlov et al., 2002; Harlov et al., 2005). These same apatite grains have microinclusions of monazite (Fig. 5) which were too small to be analysed, but are interpreted to have formed during recrystallization of the apatite. Similarly, these large apatite grains also have grain margin xenotime associated with them, interpreted to have formed by the same breakdown process.

6.5. Grain scale trends in thorium distribution

In the metapelites, there is an upward trend in ThO₂_GA until the upper amphibolite facies (sample STF16A), and the trend plateaus in the granulite facies (Fig. 8b). This coincides with the appearance of zones C and C* monazite in the granulite facies samples (ST16–09A and ST16-19A), both of which are lower in ThO₂_ZA than zone B monazite, which predominates amphibolite facies samples (e.g. STF02B). In the metapsammite samples a similar trend is seen, however the granulite facies samples (ST1F26A and ST16-03C) continue the upward trend of the lower grade samples, with STF10B the outlier (Fig. 8c). The ThO₂_GA concentrations in a given sample typically span \leq 5 wt%, except in sample STF10B where it spans ~20 wt% (Fig. 8b). STF10B has several grains with systematically higher ThO₂_GA contents than any other sample in the study. This sample contains a small population of

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monazites with compositions equivalent to zones A and B; however, the majority of the monazite analysed in the sample has significant proportions of the huttonite end-member (zone B* monazite) and coexists with huttonite (Fig. 7). The whole rock chemistry of STF10B is similar to that of other samples (Fig. 3). The sample fits within the metapsammitic trends of both monazite abundance and the abundance of the other phosphates (Fig. 6). Furthermore, the Th_WR of the sample is the same as the other metapsammite samples within error (albeit with large analytical uncertainty). It is therefore unclear why this sample contains monazites with such high Th_mnz concentrations. The P₂O₅_WR content of sample STF10B is amongst the highest of all the metapsammite samples, so a deficiency in P2O5 cannot explain why monazite in STF10B has such high huttonite component, or why huttonite is stable as a separate phase. A similar relationship between monazite and huttonite/thorite was reported by Bingen et al. (1996), which suggested that composite monazite/thorite grains were produced by the prograde breakdown of allanite. This may indicate that the protolith of STF10B contained allanite. However as there is no occurrence of allanite in the low grade samples in this study, and STF10B contains no relict allanite, this cannot be confirmed. Alternatively, the protolith of STF10B may have contained more Th and P due to the sedimentary concentration of heavy minerals in this layer.

In all other metapsammite samples the range of ThO₂_GA increases with metamorphic grade (Fig. 8b), but notably the spread of compositions within each monazite zone gets smaller (Fig. 8a). Thus the spread of ThO₂_GA in the mid amphibolite facies samples is representative of low grade and pre-metamorphic monazite of zone A, whereas the granulite facies samples represent within-grain averaging of zones A–C of various proportions, as well as zone C* grains.

By sampling monazites across metamorphic grade, we can identify that a considerable proportion of monazite present in granulite facies rocks in the Mt. Stafford system was grown on the prograde path (Fig. 7). This is demonstrated by the increase in the total amount of monazite found as inclusions in both the pelitic and psammitic granulite facies samples relative to chemically similar subsolidus samples (total incl. mnz, Table 4).The ThO₂_ZA concentrations of zone C monazite, interpreted by its presence exclusively within granulite facies rocks to have grown at granulite facies conditions, shows considerable overlap with monazite compositions from zones A and B. Combined with the large proportion of zone A and B monazite in granulite facies samples, this helps explain that Th is not depleted in the residual rocks in this study (as shown by trends in Th_GRS) due to the retention of considerable proportions of high-Th monazite from lower metamorphic grades in high grade (granulite facies) rocks.

Monazite-melt partitioning coefficients for Th (Stepanov et al., 2012) show that Th is approximately 30% more compatible in monazite than the LREE, resulting in an increasing Th/La ratio in monazite with increasing temperature in their melting experiments. The implication in melt-bearing systems is that monazite compositions with become increasingly Th-rich up to the point of total dissolution. Additionally, monazite has been shown to be stable to higher temperatures in open systems where melt extraction is efficient (Yakymchuk and Brown, 2014). When the relative proportions of monazite and melt are taken into account (typically 0.001–0.05% and 1–25% respectively), D_{mnz/liq} dictates that Th in the residue (i.e. monazite) is equal to or greater than Th in melt for typical pelitic compositions (ThO₂–in–mnz = 1–6%, Th_WR = 20–40 ppm), which provides a mechanism for the retention of Th in residual rocks.

6.6. Retention of thorium in granulite facies terranes

The Mt. Stafford sequence can be considered an endmember in terms of metasedimentary composition (low Ca) and geothermal gradient. However, as shown by Alessio et al. (2018) thorium is retained in the residuum well into the granulite facies in several terranes worldwide. Additionally, retention of monazite and thus thorium into the granulite facies was previously reported for several terranes (e.g. Bea and Montero, 1999; Bingen et al., 1996; Gasser et al., 2012; Rubatto et al., 2001) suggesting that the results presented here represent common mid-deep crustal processes. However, there are terranes that potentially have lower Th concentrations in granulite facies rocks with respect to their amphibolite facies counter parts (e.g. the Superior Province, Canadian Shield; Ashwal et al., 1987) which cannot be explained by the same partial melting processes that occurred at Mt. Stafford.

The destabilisation of monazite in granulite facies rocks may be caused by a range of factors including bulk-rock composition, P-T history, melting and melt extraction history, and fluid influx. Monazite is known to be most stable in rocks of metapelitic composition. High bulk-rock calcium contents can result in the destabilisation of monazite at lower temperatures, as can low bulk rock silica contents (e.g. Kelsey et al., 2008; Spear and Pyle, 2002). However, we show in this study that monazite and Th can be retained in rock of both pelitic and psammitic composition along steep geothermal gradients, both as inclusions within major phases and at grain boundaries. Modelling studies by Kelsey et al. (2008), Yakymchuk (2017) and Yakymchuk and Brown (2014) suggest that monazite solubility in silicate melt has limited pressure dependence. Therefore, despite the high geothermal gradient of the Mt. Stafford terrane it is likely that the results presented here are applicable to other low-Ca metapelites which attained higher pressure conditions.

The results presented here, in combination with experimental studies (e.g. Stepanov et al., 2012) suggest that partitioning of Th into monazite over melt sequesters the majority of bulk rock Th from the original source rocks while monazite remains stable. Modelling in Yakymchuk (2017) indicates that in an open system (with incremental melt loss) monazite may be stable well into UHT granulite facies conditions (>900 °C). The Mt. Stafford granulite facies rocks reached temperatures of 820 °C (Bartoli, 2017), which is below the predicted monazite 100% dissolution contours of Yakymchuk (2017) for rocks of pelitic composition with similar LREE and P budgets. Therefore, for the pelitic residue to be depleted in Th, the wholesale dissolution of monazite may be required and thus temperatures in excess of 900 °C.

Monazite is also known to be unstable in fluids of particular compositions (e.g. brines, aluminous melts; Budzyń et al., 2011; Hetherington et al., 2010). However, the deep continental crust is widely considered to be largely fluid absent (White et al., 2005; White and Powell, 2002). Additionally, fluid absent partial melting is interpreted to be the main process of melt generation, both in the examples of Th rich granulite facies above and in the mid–lower crust as a whole (Brown and Korhonen, 2009). Therefore regional depletion of Th in residual rocks by fluids would require pervasive fluid influx at a terrane scale.

7. Conclusions

Mt. Stafford is a low pressure, low-strain terrane with significant field and microtextural evidence of partial melting and some evidence for melt extraction. Whole-rock thorium concentration is preserved at even the highest grades of metamorphism both at the outcrop and micro-scales. This suggests that the process of partial melting of mid-lower crustal metapelites does not result in the broad-scale destabilisation of monazite, and thus bulk movement of thorium into silicate melt and out of residual crust (see also Alessio et al., 2018). Therefore, in terranes that contain thorium-depleted residual rocks, other factors must be involved, potentially including: differences in composition, resulting in the destabilisation of monazite at lower temperatures; attainment of higher peak temperature conditions; or influx of fluid and/or different melting styles.

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Appendix A. Supplementary data

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