Supplementary Material

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FIELD AND PETROGRAPHIC EXPRESSION

The sampling of fenite involved performing of several traverses on km-scale away from the intrusion contact in the NNE-SSW direction towards the unaltered Eriksfjord exposure, which was compounded with samples collected in the N-S direction within the proximal aureole. Several sampling traverses were also done on m-scale in the NNW-SSE orientation across the Igaliku dykes (i.e. at 1 and ~2 km distance, see Fig. 1B) to evaluate the local impact of a couple major dyke swarm bodies (IDS in Fig. 1B).

The Illerfissalik-related fenite after Eriksfjord arenite at Narsaarsuk is typified by the loss of most primary sedimentary textures and thermal induration. The Eriksfjord arenite is typically white or reddish with reduction spots (e.g. Sillisit, S Qassiarsuk; Tirsgaard & Øxnevad, 1998; see also GEUS, 2013 for colour photography) but in the fenite, secondary hues of green, blue, red and pale yellow replace the common original white or red pigment (Fig. 1C) due to growth of perthitic feldspar and pyroxenes and amphiboles (Fig. 1E). The mafic minerals are found as podiform clusters, in veins and lining along the relict sedimentary foresets (Fig. 1D). Ca-pyroxene-rich selvages commonly develop separating feldspar and titanite-rich vein cores from the country rocks, while feldspar is more broadly disseminated throughout the fenite matrix. Pseudofoliation fabric develops locally at m-scale (resorption of solids also visible in thin-section), suggesting country rock rheomorphism (e.g. Goodspeed, 1952; Mehnert, 1968) and relatively high-T conditions in the aureole driving textural change in specific parts of stratigraphy.

Curved interlobate and near-ameboidal fenite matrix microstructures and quartz grain boundary attitudes (Fig. DR1.1b-g) suggest a relatively hot aureole environment (>600°C) with quartz recrystallizing by grain-boundary migration at high-termperature and low strain rate (cf. Passchier & Trouw, 2005). Individual mineral microstructures (i.e. in metasomatic mesoperthite, classified according to Parsons et al., 2015; Fig. DR1.1h, i) suggest an accordingly hot magmatic fluid medium. The metasomatic feldspar is found throughout the proximal zone filling interstices irrespective of investigated position in succession providing evidence for widespread pervasive alteration. Rocks show no significant overprint by retrograde processes and comprise a calc-silicate assemblage reminiscent of relatively high-T (~500-800°C) skarns (Einaudi et al., 1981; Meinert, 1992; Meinert et al., 2005), here free of often characteristic garnet, epidote, scapolite etc. and forming upon a limestone-free protolith.

Fenite and other alteration styles are usually classified based on the 'intensity' of textural or mineralogical change. As no consistent scheme yet exists for products of alkali metasomatism in carbonatitic-alkaline settings, the qualitative 'grade' classification of Morogan (1994) is preferred:

- Low-grade fenites (LGF) textures and mineralogy inherited from the protolith present and largely unaltered
- Medium-grade fenites (MGF) relicts rare, new rock textures develop from fluid injection and by replacement reactions or recrystallisation
- High-grade fenites (HGF) primary mineralogy almost fully consumed, flow-banding and igneous-like appearance common attesting to recrystallisation of remobilised solids by the fenitising fluid

The relict sedimentary structures present at N Narsaarsuk (Fig. 1D) determine the overall maximum grade to be no higher than 'medium' in this scheme and no syenites of metasomatic origin were noted within the transition across the Illerfissalik-Eriksfjord contact.

Table DR1.1 – Mineralogy summary and sample information for the fenite in the Narsaarsuk aureole and its protolith in Fig. DR1.1

	Sample	Easting	Northing	Main minerals (>5% mode, in order of abundance)	Accessory (<5% mode)
a	KSL-10- 2b	61.03417 (477608.9)	-45.41444 (6766664)	$Qz \pm Hem*$	Afs, Hem, Bt, Lm
b, c	KSL-2-3c	61.04028 (478783.2)	-45.39278 (6767337)	Qz, Afs (occasionally poikilitic with Aeg), Di, Bt	Ri, Ttn, Ap \pm Zrn
d, f	KSL-5-4a	61.03528 (478779.8)	-45.39278 (6766781)	Afs, Di, Qz, Ri, Ktp	Bt, Ap, Ttn, Zrn, Cal, Hem \pm Wo
e, g, i	KSL-14- 3a	61.02750 (479119.9)	-45.38639 (6765912)	Qz, Afs (invariably poikilitic with Aeg), Ri	Bt, Ttn, Nyb, Hem, CGM, Mzt, Ap, Bzr
h	KS19-5-1	61.01144 (480144.2)	-45.36725 (6764117)	Qz, Afs, Ri	Bt, Hem

GPS coordinates provided in decimal degree format and UTM Zone 23N to cross-reference with Fig. 1B in the main text Modal abundances determined by point counting (usually 500 counts, except for exceptionally homogeneous quartzites), see also Table DR1.3. Statistics available from the author on request.

Mineral abbreviations following IUGS Subcommission recommendations for metamorphic rocks (Siivola & Schmid, 2007): quartz (Qz), hematite (Hem), alkali feldspar (Afs), biotite (Bt), limonite (Lm), aegirine (Aeg), diopside (Di), richterite (Na-Ca amphibole, Ri), titanite (Ttn), apatite (Ap), zircon (Zrn), katophorite (Na-Ca amphibole, Ktp), calcite (Cal), wollastonite (Wo), nybøite (Na-amphibole, Nyb), chevkinite-group-minerals (CGM), monazite (Mzt), bazirite (Bzr)

Sample 5-1 collected further ~1 km south of S Narsaarsuk region along the contact towards Iganaq between Illerfissalik summit and the tip of the Igaliku Fjord (see also accounts of Emeleus & Harry, 1970; Pearce, 1988 referenced in the main text), however fenite textures and the Eriksfjord protolith are analogous.

Samples 2-3c and 14-3a correspond to fenite shown in Fig. 1D-E of the main text

^{*} Hematite listed twice for 10-2b due to relative variability in the Illiortarfik area where strongly hematised, BIF-like areas, or outcrops broadly similar to those at Sillisit, can occur

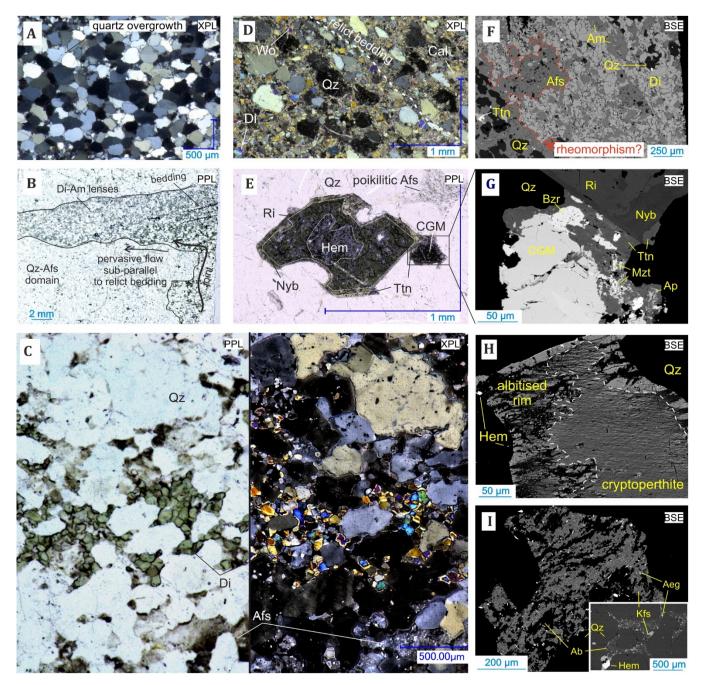


Figure DR1.1 – textural characteristics of the Eriksfjord arenite (Fig. 1A) and the subsequent Illerfissalik fenite (Fig. 1B, 1C) determined by optical (a-e) and backscatter electron microscopy (f-i); a) the quartz arenite protolith; b-i) Illerfissalik fenite; the figure showcases evidence for:

- pervasive (along grain boundaries) and structure-hosted (i.e. along joints and bedding) fenitizing fluid flow (b-c)
- significant interaction with a Ca-rich high-T fluid based on the secondary calc-silicate mineralogy, remobilisation of primary silica and the embayed mineral terminations (d, f)
- multiple generations of fenitizing fluid causing formation of reaction rims and outer Na-rich zones upon pre-existing Na-Ca amphiboles (e-g) and feldspars (h)
- the presence of secondary HFSE-hosting minerals i.e. CGM (e-g)
- deuteric recrystallisation of secondary high-T crypto- (h), meso- (i) and the often coexisting antiperthite (inset in i), often in a flame-perthite habit

Further modal mineralogy detail and the corresponding sample information in Table DR1.1. Amphibole end members determined from EPMA data using the Locock (2014) spreadsheet. Mineral abbreviations follow the IUGS Subcommission recommendations for metamorphic rocks (Siivola & Schmid, 2007), several uncommon minerals: CGM = chevkinite-group minerals (Macdonald et al., 2019), bzr = bazirite (Ba-variant of wadeite Zr-silicate)

WHOLE-ROCK METHODOLOGY

All samples were split and pulverised at the University of St Andrews using a conventional jaw crusher and tungsten carbide ring mill (2 minutes of milling, or longer in the case of syenite) to a uniform particle size <150 μm. The assay data were obtained for major elements at Bureau Veritas labs in Vancouver, Canada (BVC) by LiBO₂/Li₂B₄O₇ fusion using inductively coupled plasma atomic-emission spectrometry (ICP-AES; BVC analytical code LF300). Trace elements for the I4 intrusion samples were analysed for at the University of St Andrews (STA) by X-ray fluorescence (XRF) on pellets steel-pressed with FLUXANA CEREOX binding agent. Fenite and protolith were analysed by ICP-AES and mass spectrometry (ICP-MS) for major and trace elements by LiBO₂ fusion prior to acid dissolution at ALS Geochemistry, Loughrea, Ireland (analytical codes ME-ICP06 and ME-MS81d).

Accuracy and precision were tested using several internal (SO-19 @ BVC) and certified reference materials (GSP2 granodiorite @ STA; SY-4 syenite, REE-1, OREAS 102a, AMIS0304 @ ALS) - see Table DR1.2 and the source data within the Supplementary Dataset DR2.1. Replicates were analysed at BVC and ALS – data were within the 2σ error of the results from all labs confirming no systematic differences between data from each lab. Blanks from BVC were ≤0.03 wt% oxide for major elements and below detection for trace elements, and from ALS these were <0.05 wt% oxide and <1 ppm, respectively, indicating no contamination issues.

Table DR1.2 – standardisation summary

	Absolute accuracy (wt% ox. or ppm)	Accuracy relative to certified value (%)	Precision (% RSD)
BVC (ICP-AES, major elements)	<1	<2.6 (<1.5 for components > 1 wt%)	<1 (2.55 for P ₂ O ₅ only)
STA (XRF, trace elements)	<15	>8 for Sm and U only	>11 for Sm and U only
ALS (ICP-AES, majors)	<0.6	typically <1 (>10 for Na ₂ O, MnO & Cr ₂ O ₃)	>1 for Na ₂ O, MnO & Cr ₂ O ₃ only
ALS (ICP-MS, traces)	>3 for Nd, Y and Ba only	typically <8	>5 for Tm only

ISOCON METHOD CALIBRATION AND DISCUSSION OF LIMITATIONS

The raw compositional data (see Table DR1) for the fenite compared to unaltered rocks show an increase in all elements except silica ≤400 m of the Illerfissalik contact. Significant increase (~10 wt%) occurs in Al₂O₃, alkalis and ~2-3 wt% in CaO and MgO, and ≤400 ppm Sr. Past studies transformed the raw data to a 'standard cell' of 100 anions (typically O²⁻; McKie, 1966) or developed similar geochemical indices (i.e. IFQ – the

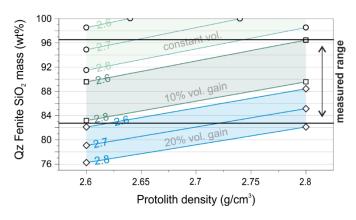


Figure DR1.2 – the effect of volume change and altered rock density (each diagonal) on the measured whole-rock composition in fenite using SiO₂ example

chemical fenitization index, as in Rodrigues et al., 2010), which however did not account for several critical physical parameters (Appleyard & Woolley, 1979). To account fully for element mass-transfer considering the physical mass and volume change, statistical analysis is traditionally performed by comparing compositions of the altered and unaltered rocks using the isocon method adapted from the original Gresens (1967) study. The equations (after Grant, 1986) account for rock specific gravity (p) and volume (f_v) changes, which minimise the closure effect (whole-rock data must add up to 100%) arising from standard bulk-rock data analysis (Ague & van Haren, 1996). The change in concentration of a component is calculated using $[\Delta C_i = ((\frac{m^a}{m^o})(\frac{C_i^a}{C_i^o}) - 1) C_i^o]$; where m and C refer to the mass and concentration of component i in altered and original sample (superscript a and o). Once calculated, the change is often divided by the original concentration ($\Delta C_i/C_i^o$, as in Grant, 1986) to represent best the degree of alteration, although in some cases only the gains experienced by the wall-rock were deemed pertinent in measuring the impact of metasomatic process (fenitisation index – F.I.; Kresten, 1988). The isocon model requires one of three assumptions be made to define the isocon reference line: (1) no mass change, (2) no volume change, or (3) one or several elements are immobile.

While the task of determining immobile elements is trivial in most silicate crustal systems, qualitative studies of alkaline magmatism commonly show mobility of HFSE and Al₂O₃ - components that are ordinarily considered immobile (MacLean & Barrett, 1993). Nevertheless, immobile

components can be defined iteratively (i.e. Arzamastsev et al., 2011; Korchak et al., 2011), or by proxy of the Gresens (1967) f_v curve clustering method where multiple elements with identical f_v values at 0 gain/loss, indicate both level of volume change and immobility (see Cooper et al., 2016). In the present study, HFSE mobility is evident from the presence of chevkinite-group minerals (CGM; Fig. S1e, g) and Nb-LREE-rich secondary titanite disseminated in the fenitized quartz arenite at Narsaarsuk. Examining the different isocons across the fenitized area also shows HFSE i.e. Zr, Y, Ta, Tm are variably mobile, suggesting they are not the ideal reference species.

Due to lack of an immobile reference element, we examined the alternative approaches. We choose to apply the constancy of mass scenario where "one may assume that the sums of components supplied and depleted during fenitization roughly balance, i.e. that the total mass change of the rock during fenitization will be at minimum" (Kresten, 1988; p. 349). It should be noted that in this case SiO_2 appears to define the CM-isocon (Fig. 2B-2D) implying immobility. Brief examination of the f_v parameter under these conditions shows the preservation of SiO_2 mass could be satisfied through volume increase up to ~20% (Fig. DR1.2) in the likely range of fenite and protolith density (2.6-2.8 g/cm³). Contact breccias resulting from expansion do occur at Illerfissalik (best exposed towards midto-S Narsaarsuk, Fig. 1B), but outcrops are small (m scale) and discontinuous, leading us to infer volume gain is not widespread. Some leaching must have occurred (based on textural lack of SiO_2 cement) and we therefore conclude volume loss in this system. We stress however that the measured values cannot be explained by volume change alone and metasomatic exchange is significant.

Table DR1.3 – modal mineralogy and the corresponding calculated bulk density (weighted based on mineral composition) of the selected fenite and protolith samples

Category		Fenite	Fenite	Fenite	Fenite	Fenite	Fenite	Fenite	Fenite	Fenite	Fenite	Fenite	Protolith	Protolith	Protolith
Туре		Matrix	Matrix	Matrix (+fine veins)	Matrix	Matrix	Matrix	Vein	Matrix	Matrix	Matrix (+fine veins)	Vein	Matrix	Matrix	Matrix
Sample #		14-3a	2-3a	2-3c	5-1	6-6	6-6	2v	14-3a	2-3a	2-3c	5-4a	10-5c	10-4a	10-4b
Method		Pt Count	Pt Count	Pt Count	Pt Count	Pt Count	Estimate	Estimate	Estimate	Estimate	Estimate	Estimate	Pt Count	Pt Count	Pt Count
<u>Mineral</u>	<u>Density</u> (g/cm³)														
Quartz	2.65	85.20	66.80	51.50	82.20	65.60	37.00	60.00	40.00	40.00	35.00	30.00	90.80	84.50	97.30
Alkali Feldspar (Or)	2.55	10.80	22.20	20.10	11.40	30.00	45.00	14.00	30.00	40.00	45.00	20.00		7.20	2.00
Pyroxene (Di)	3.4	0.20	7.00	9.00			7.00	3.00	10.00	7.00	6.00	25.00			
Amphibole (Ri)	3.06	1.80	1.40	4.80	4.80		2.00	10.00	4.00	2.00	2.00	5.00	4.20		
Biotite (Annite)	3.26			8.20			2.00		5.00	5.00	7.00	5.00	2.00		
Titanite	3.47		1.80	1.40		3.20	5.00	6.00	5.00	5.00	4.00	3.00			0.20
Zircon	4.65		0.40	0.60		1.00	2.00	1.00	1.00		0.00				0.20
Epidote	3.45							1.00	1.00		1.00	2.00			
Apatite	3.15			0.40		0.20		2.00	1.00			2.00	0.40		
Calcite	2.71							1.00	2.00			5.00			
Fe-Ti oxides (Hem)	5.3	2.00	0.40	4.00	1.60				1.00	1.00		3.00	2.60	8.30	0.30
Others								2.00							
Total		100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00	100.00
Arithmetic bulk density		2.70	2.72	2.90	2.70	2.67	2.76	2.73	2.84	2.77	2.74	3.00	2.75	2.86	2.66
Subcategory			Fenite (pt counting)					Fenite	(estimates)			Protolith	
Mean density				2.74						2.81				2.76	

Mineral abbreviations as in Table DR1.1. Mineral densities as reported on http://webmineral.com, assumed solid-solution end-member cited where appropriate.

The final uncertainty in isocon models is the choice of the unaltered reference sample. This is often difficult in cratonic regions where natural protolith variation and taking any arbitrary sample as a reference introduces large $\Delta C_i/C_i^0$ errors, frequently several orders of magnitude, into the calculations. The Kresten (1988) study attempted to reduce this effect by calculating an average wall-rock composition (AWR) of five samples in the Fen area. Subsequent studies highlighted that this approach is prone to problems as it is often difficult to measure the extent of the aureole and several specimens can be classed as low-grade fenite leading to underestimation of elemental transfer (Verschure & Maijer, 2005). The Eriksfjord quartz arenite, however, we find to be exceptionally well suited for this approach, as it shows no significant lateral variation in facies at the stratigraphic level (Stewart, 1964) and therefore inherent heterogeneity is assumed to be negligible. No protolith samples show >5% modal feldspar or clay, except within ~5 m of regional dyke contacts at >0.4 km distance (Fig. 2D) or where the protolith is strongly hematised (Table DR1.3). The mean whole-rock composition for the protolith population of 15 samples is used here as a reference composition rather than an arbitrary sample due to wide availability of sedimentary exposures, which represent the same formation and likely the same stratigraphic member, across a ~6 km² area, which decreases the likelihood of using an extreme composition or that the low-grade alteration signature would be imparted on the result. Density values used in this isocon study are 2.6 and 2.8 g/cm³ for the protolith and fenite respectively. As a guide, the generalized densities for the protolith and its fenite were calculated from the observed modal compositions of the selected samples (Table DR1.3). Although some relatively denser samples do occur especially where veins (Fig. 1D; pyroxenite 5-4a – Fig. DR1.1D, DR1.1F) or thin veinlets 10 μm to 2 mm wide (i.e. 2v, 2-3c – Fig. DR1.1B; Table DR1.3) cut the fenite otherwise comprised by uniformly dispersed metasomatic mineralogy, or locally in hematite-rich sandstone (i.e. 10-4a; Table DR1.3), the excursions are small and majority of the wall-rock is represented by compositions described in Table DR1.1 and the impact of changing between the values in the stated density range on the output was found to be insignificant.

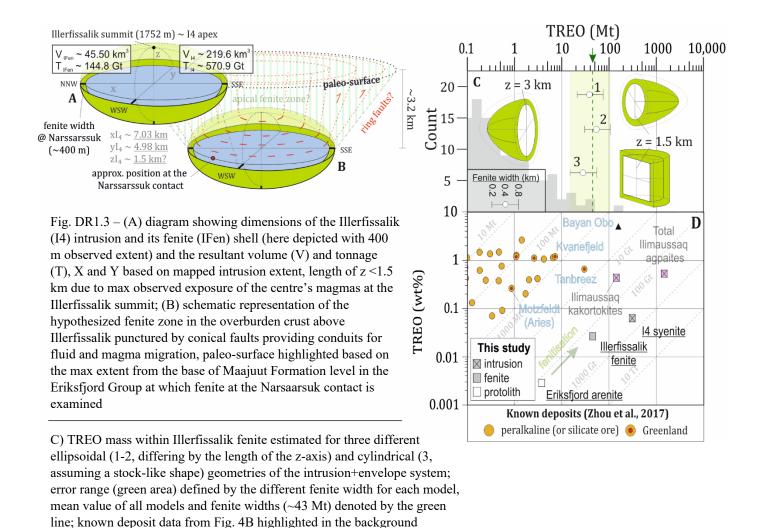
MODELLING BULK HFSE-MASS – GEOMETRIES AND PARAMETERS

The impact of fenitization on ore genesis in a large, open magmatic system can be shown through attributing compositional data to a scaled 3D geometric model. Our volumetric model was designed for three different geometries of the Illerfissalik centre and three fenite widths (the observed 400 m among those; Fig. DR1.3a-c) to obtain mean HFSE and TREO values representative of the cargo introduced into the wall-rock by the fenitizing fluids. The Data Repository tables DR2.3-2.5 contain calculations for our model, the results of which are summarized in Table DR2.2. For benchmarking purposes, here we also present the same calculation applied to a structurally better constrained Ilímaussaq Complex where it is often assumed that no significant HFSE or volatile loss and isotopic mixing have occurred, and the centre is a world-class magmatic HFSE ore deposit (Larsen & Sørensen, 1987; Marks & Markl, 2015). At Ilímaussaq a narrow (10-200 m; Ferguson, 1964) fenite of similar character (Derrey, 2012) is described and for comparative purposes it is here presented as a relatively 'closed' system.

The main models (those interpreted to best represent the true geometry) assume both centres are ellipsoids (Illerfissalik = $2x \sim 14$ km, $2y \sim 7.6$ km laterally and $2z \sim 3-6$ km from base to apex; Ilímaussaq = ~ 17 , 9.95 and ~ 1.8 km respectively, unit thicknesses based on Andersen et al., 1981; Bailey et al., 2001; incl. Black Madonna unit after Schønwandt et al., 2015; Tables DR2.3-2.5) and use only the reported cumulate unit compositions (weighted by unit thickness where appropriate i.e. for Ilímaussaq kakortokite and lujavrite sequences, see Table DR2.5). X and Y dimensions are inferred from the geological map data found in various sources i.e. Upton (2013), Marks & Markl (2015) or the GEUS Greenland Portal.

We model our data to three variants of the Illerfissalik system – 1) an ellipsoid with z = 1.5 km; 2) an ellipsoid with z = 3 km and 3) a cylinder with z = 1.5 km to represent the three most broadly realistic geometries. The fenite shell is calculated for each of these geometries with a 200, 400 and 800 metres strip added (Table DR2.3) to represent the actual observed fenite (400 m) and the plausible min-max range within which the average fenite concentration would still faithfully represent the true, appropriately scaled, gradationally weakening, fenitizing fluid signal (Fig. DR1.3). The volumetric calculation relationship can be described as follows: $V_{Fenite} = V_{I4+Fenite} - V_{I4}$ and for simplicity it is also

assumed that $V_{\text{Fenite}} = V_{\text{Unaltered Protolith}}$ (Table DR2.3, DR2.4). For Ilímaussaq, similar subtractive calculations were made to calculate the stratified chamber volumes for kakortokite, lujavrite, basal saucer-shaped Black Madonna unit and the naujaite dome sequences. Density of the igneous nepheline syenitic units for both intrusions is set at 2.7, while that of the fenite is kept at 2.6 g/cm³.



D) TREO tonnage vs grade comparison of the estimates in this study (Eriksfjord arenite, Illerfissalik I4 syenite and its fenite), simulated comparative Ilímaussaq volumes (pink) and the known deposit data (circles & triangle; from Zhou et al., 2017); green vector indicating the effect of fenitization on bulk TREO grade and tonnage of the modelled Eriksfjord arenite volume

After calculating the rock (or ore) volume we attributed compositional data for the average fenite, protolith, I4 Illerfissalik and Ilímaussaq units to the corresponding ellipsoidal or cylindrical volumes to determine the mass of selected HFSE oxides and TREO within. For this purpose, raw trace element data (in ppm) were converted to oxide form with all REE assumed to be trivalent. Only limited I4 intrusion trace element data by XRF are available, therefore in this study we assume LREO~TREO and

whenever comparing the cargo movement, we omit elements such as Ta or Hf which were not detected in the I4 syenite samples. Results are found in Table DR2.4 for Illerfissalik and DR2.5 for Ilimaussaq respectively. Table DR2.6 contains HFSEO data presented in Fig. 4A from both preceding tables arranged by ionic potential. The Ilimaussaq data (Fig. DR1.3d) show that our model estimates the total tonnage in the basal kakortokite unit, which host the Kringlerne deposit in the SE region towards (4.7 Gt @ 0.65 wt% TREO = ~31 Mt; Zhou et al., 2017), at 36.44 Gt @ 0.43 wt% TREO = ~155.7 Mt, indicating the unexposed kakortokite rock volume is ~4x larger – a result we find to be geologically consistent with the known deposit statistics assuming lopolith-like geometry of the intrusion base. We note that Figure 1.3d should not be confused for the conventional tonnage vs resource grade relationship (i.e. Fig. 3 in Paulick & Machacek, 2017) – it is intended to visualize how fenitization fertilizes the wall-rocks by increasing TREO proportion in the same rock mass.

It should be noted that the simple geometric models applied in this work do not account for many complications expected and unknowns due to lack of public data in such chemically complex magmatic-hydrothermal systems located in an exploration frontier i.e. 3D intrusion and wall-rock compositional heterogeneities, lateral and vertical discontinuity of various units, changing redox conditions, structural traps and several other variables. The volumes and tonnages presented here should not therefore be mistaken for JORC compliant resource estimates. Future workers are therefore recommended to exercise caution when applying a similar approach in geologically complex areas.

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