

# 1 **Geology, geochemistry and geochronology of the** 2 **Songwe Hill carbonatite, Malawi**

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## 16 **Keywords:**

17 Songwe Hill; carbonatite; Chilwa Alkaline Province; REE; U-Pb dating; HREE; apatite

## 18 **Highlights:**

- 19
- Songwe Hill is a multi-phase intrusion, emplaced at a shallow depth in the crust
- 20
- Evidence for pervasive late (carbo)-hydrothermal overprinting
- 21
- Interaction with late fluids led to HREE mineralisation

- 22 • A large local heat source differentiates Songwe Hill from the other Chilwa Alkaline  
23 Province carbonatites

## 24 **Abstract**

25 Songwe Hill, Malawi, is one of the least studied carbonatites but has now become  
26 particularly important as it hosts a relatively large rare earth deposit. The results of new  
27 mapping, petrography, geochemistry and geochronology indicate that the 0.8 km diameter  
28 Songwe Hill is distinct from the other Chilwa Alkaline Province carbonatites in that it intruded  
29 the side of the much larger (4 x 6 km) and slightly older ( $134.6 \pm 4.4$  Ma) Mauze nepheline  
30 syenite and then evolved through three different carbonatite compositions (C1–C3). Early C1  
31 carbonatite is scarce and is composed of medium–coarse-grained calcite carbonatite  
32 containing zircons with a U–Pb age of  $132.9 \pm 6.7$  Ma. It is similar to magmatic carbonatite in  
33 other carbonatite complexes at Chilwa Island and Tundulu in the Chilwa Alkaline Province  
34 and others worldwide. The fine-grained calcite carbonatite (C2) is the most abundant stage  
35 at Songwe Hill, followed by a more REE- and Sr-rich ferroan calcite carbonatite (C3). Both  
36 stages C2 and C3 display evidence of extensive (carbo)-hydrothermal overprinting that has  
37 produced apatite enriched in HREE (<2000 ppm Y) and, in C3, synchysite-(Ce). The final  
38 stages comprise HREE-rich apatite fluorite veins and Mn-Fe-rich veins. Widespread  
39 brecciation and incorporation of fenite into carbonatite, brittle fracturing, rounded clasts and  
40 a fenite carapace at the top of the hill indicate a shallow level of emplacement into the crust.  
41 This shallow intrusion level acted as a reservoir for multiple stages of carbonatite-derived  
42 fluid and HREE-enriched apatite mineralisation as well as LREE-enriched synchysite-(Ce).  
43 The close proximity and similar age of the large Mauze nepheline syenite suggests it may  
44 have acted as a heat source driving a hydrothermal system that has differentiated Songwe  
45 Hill from other Chilwa carbonatites.

## 46 **1. Introduction**

47 Since the discovery of carbonatites in Africa (Dixey et al., 1937), many of the carbonatite  
48 complexes of the Chilwa Alkaline Province of Malawi have been extensively studied and  
49 documented, resulting in excellent memoirs on Chilwa Island (Garson and Campbell Smith,  
50 1958), Tundulu (Garson, 1962) and Kangankunde (Garson and Campbell Smith 1965).  
51 However, the Songwe Hill carbonatite, which is the fourth largest in the province, was not  
52 subject to the same extensive surveying and mapping as the other nearby complexes (e.g.  
53 Garson and Walshaw, 1969) and only short summaries exist in the literature (Garson, 1965,  
54 1966; Woolley, 2001; Harmer and Nex, 2016).

55 Interest in the Songwe Hill carbonatite has largely stemmed from exploration for rare earth  
56 elements (REE). Building on earlier descriptions of other occurrences in the area (e.g.  
57 Garson, 1962), a brief overview of the Songwe Hill carbonatite and related rocks was first  
58 provided by the Nyasaland Geological Survey who indicated the presence of REE minerals  
59 (Garson, 1965; Garson and Walshaw 1969). Further exploration for REE was carried out at  
60 Songwe Hill between 1986 and 1988 by the Japanese International Co-operation Agency  
61 (JICA) and the Metal Mining Agency of Japan (MMAJ). They concluded that Songwe Hill, as  
62 well as other occurrences within the Chilwa Alkaline Province, had “*high potentiality for a*  
63 *carbonatite deposit*” (JICA and MMAJ, 1989). Recent work by Mkango Resources has  
64 established a (NI 43-101 compliant) probable mineral reserve for the REE of 8.4 million  
65 tonnes at 1.6% TREO, with a higher proportion of HREE than many other carbonatite-related  
66 REE deposits (TREO = Total Rare Earth Oxides; Croll et al., 2014; Harmer and Nex, 2016).  
67 This contribution summarises many of the findings, including geological, mineralogical,  
68 geochemical and age data, from this most-recent exploration at Songwe. With these new  
69 data, we discuss the evolution of this intrusion in the context of other intrusions in the Chilwa  
70 Alkaline Province. Details on the REE mineralogy are presented in a separate contribution  
71 (Broom-Fendley et al., 2017).

## 72 **2. Geological setting**

73 Songwe Hill is located in the Phalombe district of south-eastern Malawi, an area where the  
74 regional basement comprises Neoproterozoic gneisses and granulites. These have  
75 undergone several stages of intrusion and deformation, including emplacement of a much  
76 earlier episode of late Precambrian alkaline magmatism in the north and south of the country  
77 (Bloomfield, 1968; 1970; Eby et al., 1998; Kröner et al., 2001; Ashwal et al., 2007). This  
78 earlier episode is suggested to be associated with an ancient continental rift zone (Burke et  
79 al., 2003). Regional Pan-African deformation is manifested through metamorphic  
80 overprinting (e.g. Woolley et al., 1996) and emplacement of granitic intrusions (Haslam et al.,  
81 1983). A subsequent Jurassic (Karoo) basic dyke swarm is prevalent throughout the south  
82 of the country (Woolley et al., 1979; Macdonald et al., 1983).

83 Songwe Hill is part of the Late Jurassic–Early Cretaceous Chilwa Alkaline Province: a region  
84 approximately 300–400 km in diameter in the south of Malawi and in Mozambique (Fig. 1).  
85 The province is made up of an essentially intrusive suite of carbonatite, nephelinite, ijolite,  
86 nepheline syenite, syenite, quartz syenite and granite (Woolley and Garson, 1970; Woolley,  
87 1991, 2001). The Chilwa Alkaline Province intrudes the basement in a variety of styles,  
88 including large plutons, carbonatite-bearing ring complexes, alkaline dyke swarms following  
89 NE/SW lines of crustal weakness, small nephelinite and phonolite plugs and small breccia  
90 vents (Woolley, 2001). Volcanic phonolites, nephelinites and blairmorites of the Lupata-  
91 Lebombo area in Mozambique are also considered to be associated with the Chilwa Alkaline  
92 Province as they are of similar composition and age (Woolley and Garson, 1970).  
93 Subsequent to the Chilwa Alkaline Province volcanism, the region has been eroded, possibly  
94 deeper in the east than in the west (Eby et al., 1995), such that many of the intrusions are  
95 now isolated inselbergs within a Cenozoic lacustrine peneplain.

96 Songwe Hill forms a steep-sided conical hill, approximately 800 m in diameter, rising about  
97 230 m from the surrounding plain. The hill abuts the north-western flank of Mauze, a large,

98 oval, nepheline syenite intrusion which is approximately 6x4 km in area and 800 m high. To  
99 the north-west and north-east are two, lower-lying, hills: Chenga and Pindani (Figure 2A).

### 100 **3. Geology of Songwe Hill**

101 Mapping and a two phase diamond drilling campaign, totalling approximately 6850 metres,  
102 were conducted by Mkango Resources between 2011 and 2012 (Croll et al., 2014). These  
103 activities focussed on the central and northern flanks of Songwe Hill, where most of the  
104 known REE mineralisation occurs (Figure 2B). Carbonatite outcrops broadly follow two  
105 lobate structures, trending SW-NE, intruding into the fenite host. To simplify the mapping  
106 process, however, small-scale heterogeneities such as fenite blocks and minor veins, were  
107 excluded. Drilling highlighted that these heterogeneities continue with depth and greatly  
108 complicate any structural interpretation. REE mineralisation occurs predominantly in  
109 carbonatite, but also in fenite, up to 350 m below the surface of the hill. No clear contact  
110 between Songwe and Mauze, or the basement, was encountered during drilling and surface  
111 contacts are obscured by cover (Croll et al., 2014).

112 Samples were collected over two field seasons in 2011 and 2012. Petrography was carried  
113 out using similar techniques to those described by Broom-Fendley et al (2016a). Cold  
114 cathodoluminescence (CL), using a CITL Mk3a electron source operated at 3–13 kV and  
115 ~350 nA, was primarily used for mineral identification, in combination with standard optical  
116 petrography. Selected samples were also analysed using BSE imagery and qualitative EDS  
117 analyses, using a JEOL JSM-5400LV SEM and a JEOL JXA-8200 electron microprobe, to  
118 confirm the identity of some minerals.

119 The main geological units comprise: variably fenitized basement granulite, gneiss and  
120 dolerite dykes; clast supported fenite breccia; fenite; carbonatite; and late-stage veins of  
121 apatite- and fluorite-rich rock, and Mn- and Fe-rich rock. New field observations,  
122 petrographic and geochemical data are presented for these units in the following sections.

### 123 **3.1 Clast-supported fenite breccia**

124 The outermost unit at Songwe comprises a clast-supported breccia formed of large angular  
125 slabs of country rock gneiss and dolerite. This unit is most prominent on Chenga Hill, north-  
126 west of the main Songwe Hill intrusion (Fig. 2A). The exposure of this unit is limited.  
127 However, based on comparison with other carbonatites in the Chilwa Alkaline Province, it  
128 presumably extends around the edge of the intrusion (c.f. Chilwa Island, Nkalonje, Tundulu;  
129 Garson and Smith, 1958, Garson, 1965). This rock is only fenitised to a low degree, with K-  
130 feldspar replacement most-prominent at the edges of the gneiss clasts while the centre  
131 retains the original mineralogy and banding (Figure 3A). As such, it differs considerably from  
132 the majority of fenite at Songwe.

### 133 **3.2 Fenite**

134 Songwe Hill is predominantly composed of fenite, which forms the entirety of the lower flanks  
135 of the hill and extends at least 500 m away from the intrusion, although the full extent is not  
136 known as the contact with low-grade fenite breccia is obscured by sediment. Major  
137 carbonatite outcrops are restricted to the upper parts of the hill (mapped in Fig. 2B). Within  
138 the carbonatite, fenite also occurs as large blocks/boulders (>100 m) down to small, angular,  
139 clasts (<1 mm) (Figure 3B). Fenite blocks and clasts are found at the greatest depths  
140 sampled in most drill-holes (e.g. 302 m, hole PX001). The large blocks of fenite appear to be  
141 in-situ, or transported a very short distance and are interpreted as fractured blocks from the  
142 margins, or the roof, of the carbonatite. At the summit of the hill, fenite is continuous with  
143 only rare carbonatite veinlets, suggesting that the roof zone of the carbonatite is exposed  
144 and, therefore, that the carbonatite never reached the surface.

145 In many fenite samples a coarse-grained equigranular igneous texture is preserved, strongly  
146 suggesting an igneous protolith, highly likely to be nepheline syenite from the neighbouring  
147 Mauze intrusion. Other clasts are fine grained and are highly likely to be altered phonolite.

148 Little evidence of banding, as seen in the country rock around Songwe, occurs in the fenite  
149 clasts.

150 Fenitization is predominantly potassic, and where rocks have been completely fenitized they  
151 are composed almost entirely of orthoclase with minor aegirine. Other minor phases include  
152 fluorapatite, zircon, rutile, ilmenite and hematite (commonly altered to goethite). In small  
153 clasts within the carbonatite, fenite commonly contains a proportion of carbonate, likely to be  
154 derived from carbonatite emplacement.

### 155 **3.3 Carbonatite**

156 Songwe Hill comprises three main stages of carbonatite intrusion: coarse grained calcite  
157 carbonatite (sövite); fine-grained calcite carbonatite (alvikite); and Fe-rich ferroan calcite  
158 carbonatite. The terms C1–C3 are used as shorthand to distinguish the different carbonatite  
159 types (Table 1). While the terminology broadly overlaps with the terminology used by Le Bas  
160 (1977, 1987, 1999), it is important to distinguish that the C3 'ferrocarbonatite' is not, in the  
161 strict sense, accurate, as the Fe in these rocks is hosted in oxide minerals (Gittins and  
162 Harmer, 1997).

#### 163 **3.3.1 Calcite carbonatite (C1 and C2)**

164 Coarse-grained calcite carbonatite (C1) is rare at Songwe and only occurs as rounded  
165 clasts, of up to a few cm, in other carbonatite types (Figure 3C). It is composed of medium-  
166 grained anhedral calcite (90 vol. %), minor anhedral ankerite (5 vol. %) and an assemblage  
167 of ovoid apatite, subhedral zircon, euhedral pyrite, anhedral pyrochlore and anhedral K-  
168 feldspar (Figure 4A). Zircon grains are euhedral to subhedral, and range from small  
169 (approximately 50 µm; Figure 5A–B) to relatively large grains (up to 0.5 mm), the latter of  
170 which can exhibit well developed zoning (Figure 5D–E). Based on their habit, formation at  
171 calcite grain boundaries and association with ovoid apatite, zircons in coarse-grained  
172 carbonatite are interpreted as a carbonatite magmatic phase.

173 Fine grained calcite carbonatite (C2) is the most abundant carbonatite type at Songwe, and  
174 the term includes all fine-grained white–grey calcite carbonatites. It ranges from calcite-rich  
175 rocks through to complex, multi-phase samples but most commonly comprises fine-grained  
176 calcite with minor interstitial ankerite (Figure 4B). Anhedra apatite stringers cross-cut earlier  
177 calcite (e.g. Figure 4B). Calcite in association with these apatite-stringers luminesces  
178 brighter in CL images than earlier-crystallising calcite, associated with increasing Mn relative  
179 to Fe. In some samples xenocrysts of zircon, pyrochlore, pyrite and K-feldspar are present  
180 and are rounded and altered to varying degrees. Pyrite, zircon and K-feldspar show little  
181 rounding and are found in bands while pyrochlore is commonly rounded and partially  
182 fractured. Zircons in the fine-grained calcite carbonatite are small, subhedral and appear out  
183 of equilibrium with the host rock, as indicated by edges that are embayed and cores that are  
184 pitted and corroded (Figure 5C). Localised assemblages of euhedral synchysite-(Ce), baryte,  
185 and anhedra strontianite are disseminated throughout the carbonatite (Figure 4C).

### 186 **3.3.2 Ferroan calcite carbonatite (C3)**

187 Ferroan calcite carbonatite (C3) is fine-grained and occurs as veins in calcite carbonatite, as  
188 breccia clasts and as large discrete masses. It is found throughout the intrusion but is most  
189 abundant towards the centre (Figure 2B). It commonly contains clasts of fenite and, less  
190 commonly, clasts of both coarse- (C1) and fine-grained calcite carbonatite (C2). It weathers  
191 dark brown–black and on a fresh surface it is dark, fine-grained and commonly banded  
192 (Figure 3D). The bands include (A) a ferroan calcite carbonatite groundmass, (B) pale  
193 apatite-bearing layers and (C) similarly pale LREE-fluorcarbonate-bearing layers.

194 The (A) ferroan calcite carbonatite groundmass is predominantly (95 vol. %) composed of an  
195 opaque carbonate which luminesces dark orange in CL images. This phase is likely to be an  
196 altered Fe-bearing calcite which has exsolved Fe from its structure. Minor hematite (4 vol.  
197 %) and zircon (1 vol. %) are also present in the groundmass. Zircon is subhedral and shows  
198 resorption textures indicating that it is out of equilibrium with the host rock. Apatite bands (B)



199 are monomineralic. The apatite is anhedral, very fine-grained and forms mm–cm bands,  
200 similar to those in fine-grained calcite carbonatite (Figure 4B). They are commonly aligned  
201 with each other and with the later LREE-fluorcarbonate-bearing assemblage (C). This  
202 assemblage is similar to the LREE-bearing assemblage found in vugs in the C2 calcite  
203 carbonate, and is composed of syntaxial intergrowths of euhedral synchysite-(Ce) and  
204 parisite-(Ce), euhedral baryte and anhedral strontianite or calcite (Figure 4D). Cross-cutting  
205 relationships between the various stages are evident from the termination of an apatite  
206 stringer by a REE-fluorcarbonate-bearing assemblage (Figure 4D), and from the presence of  
207 partially fragmented apatite in a matrix of strontianite and fluorcarbonates (Figure 4E).

### 208 **3.4 Late-stage veins**

209 Throughout the carbonatite several stages of overprinting and late cross-cutting veins are  
210 evident. These are subdivided into fluorite/apatite–fluorite enriched rocks and Mn-Fe-veins.

#### 211 **3.4.1 Apatite-fluorite veins**

212 Small veins, rich in fluorite and apatite, form a volumetrically minor part of the carbonatite  
213 and are most conspicuous in the breccia at Chenga Hill (Figure 3A). Similar fluorite-rich  
214 areas are also found in drill core, most prominently in core PX0016 (Croll et al., 2014). In  
215 addition, early descriptions of Songwe Hill identified similar apatite-fluorite rock in boulders  
216 approximately half a mile north-east of Songwe (Garson, 1965). In previous contributions  
217 (e.g. Broom-Fendley et al., 2016b) these veins have been termed 'C4'. However, this term  
218 has been discontinued as it suggests the veins are a variety of carbonatite.

219 The apatite-fluorite veins comprise major fluorite, apatite, calcite/ferroan-calcite, quartz and  
220 baryte (Figure 4F). Accessories include xenotime, zircon, rutile/anatase, Mn-oxides,  
221 hematite, and synchysite/parisite. Fluorite is sub–anhedral and typically occurs in elongate  
222 stringers and in discrete patches. Individual grains vary in size from 20–200 µm, with most  
223 typically around 20–50 µm. Apatite is subhedral and typically clumps together in veins

224 formed of equigranular crystals approximately 150–200 µm in size. Fragments of broken K-  
225 feldspar are common in the apatite-fluorite rock. The pieces are angular and show little  
226 evidence of alteration or reaction. Late-stage minerals include anhedral baryte and quartz.

### 227 **3.4.2 Mn-Fe-veins**

228 Mn-Fe-rich rocks form along centimetre to metre wide veins, following straight fractures  
229 cross-cutting carbonatite and fenite (Figure 3B). These are especially common at the edge,  
230 and towards the top, of the intrusion, cutting the fenite roof-zone. The rocks are composed of  
231 a mixture of carbonate rhombs and Fe- and Mn-oxides, commonly altered to limonite. In thin-  
232 section they exhibit distinct alteration halos, where Fe-bearing carbonates in the carbonatite  
233 are decomposed and replaced by Fe-oxides.

234 Mn-Fe-rocks can occur as areas of large-scale alteration and in these circumstances the  
235 relation with the host-rock is difficult to establish. These areas are deeply weathered and  
236 revealed by bulldozed road-cuts or as pockets at depth in drill core (Figure 3F). These rocks  
237 are typically carbonate-poor compared to their vein-hosted counterparts, and are  
238 predominantly composed of massive Mn- and Fe-oxides, with little evidence of other  
239 minerals.

## 240 **4. Whole-rock Geochemistry**

241 Whole-rock analyses were carried out on core samples (marked on Fig. 2B), drilled by  
242 Mkango Resources Ltd in 2011. Samples were taken between 0.3 and 1 m intervals, at the  
243 point of lithological change. Rock powders were analysed for major elements by ICP-OES  
244 and trace elements by ICP-MS at Intertek-Genalysis in Australia using technique FP6/MS33  
245 (Intertek-Genalysis internal catalogue number, see [www.genalysis.com.au/minerals/assay](http://www.genalysis.com.au/minerals/assay)).  
246 This technique involves digestion using a sodium peroxide fusion to dissolve refractory  
247 minerals. Unfortunately, however, this means no Na data are available for the whole-rock  
248 analyses. Fusion products are dissolved in HCl and diluted for analysis and the results are

249 corrected for dilution and the amount of flux used for digestion. Internal standards were used  
250 to correct for drift, viscosity effects and plasma fluctuations. Repeat blind analyses of  
251 commercial carbonatite reference standards AMIS0185 (n = 91) and GRE-04 (n = 105)  
252 indicate no significant departures from published major and trace element concentrations,  
253 with  $2\sigma$  values generally lower than the quoted uncertainty in the reference material  
254 (Supplementary Table 1). Owing to the fine-grained and heterogeneous nature of the  
255 samples, some carbonatite analyses incorporate a significant proportion of  $\text{Al}_2\text{O}_3$ ,  $\text{SiO}_2$ , and  
256  $\text{K}_2\text{O}$  derived from fenite (Supplementary Figure 1). Therefore, carbonatite analyses with  $>2$   
257 wt. %  $\text{Al}_2\text{O}_3$  are considered contaminated with fenite and are excluded from interpretation.

258 A summary of major and trace element whole-rock data is presented in Table 2 (full dataset  
259 Supplementary Table 2). Regardless of the absence of  $\text{Na}_2\text{O}$  values from the whole-rock  
260 analyses (owing to preparation with a Na-peroxide fusion), most of the fenite analyses give  
261 totals close to 100%, reflecting the K-feldspar-rich nature of these samples. Carbonatite  
262 contamination in these analyses is indicated by elevated average loss on ignition (LOI)  
263 values (15.5 Wt. %) and the correlation of LOI with SrO, MgO, CaO, and the REE. Given the  
264 dominant role of even small amounts of carbonatite for the REE concentration, the REE  
265 content of 'pure' fenite is undoubtedly very low.

266 Carbonatite analyses are split between calcite carbonatite (C2) and ferroan-calcite  
267 carbonatite (C3) (Table 2). The small clast size and scarcity of material from the coarse-  
268 grained C1 calcite carbonatite means that no data are available for this rock type. With the  
269 exception of  $\text{P}_2\text{O}_5$ , SrO, Th and REE, average concentrations for most elements are similar  
270 for both C2 and C3 carbonatite (Table 2). Using either of the ternary carbonatite  
271 classification diagrams (IUGS classification of Wt. % oxide [Woolley and Kempe, 1989; Le  
272 Maitre, 2002] or molar proportions [Gittins and Harmer, 1997]), calcite carbonatite plots  
273 between the calciocarbonatite and ferruginous calciocarbonatite fields while ferroan calcite  
274 carbonatite forms a small cluster, broadly within the ferruginous calciocarbonatite field

275 (Figure 6). Despite the assignation of C3 as a Fe-rich carbonatite, it should be stressed that  
276 the dominant mineralogical control on the Fe concentration is not a Fe carbonate, but rather  
277 Fe oxides or hydroxides. This is best evaluated on a plot of LOI against total MnO+FeO  
278 (Figure 7A), where LOI is assumed to be directly proportional to CO<sub>2</sub> concentration. There is  
279 a significant decrease in LOI with increasing FeO content for C2 and C3 carbonatites  
280 corresponding with mixing lines between Fe oxide minerals and calcite. While pyrite occurs  
281 as a minor mineral in these rocks, it is not abundant enough to account for the large  
282 variations in Fe content. A large contribution from siderite can also be excluded as this would  
283 result in a high FeO concentration whilst retaining a high LOI, a trend not shown in the data  
284 (Figure 7A). However, a minor amount of ankerite in the rocks is likely as indicated by the  
285 correlation between MgO and FeO (Figure 7B). This contribution from ankerite is highly likely  
286 to account for the scatter in LOI and Fe contents for C2 and C3 data (Figure 7A). The  
287 carbonatite analyses show a positive trend between MnO+FeO with SrO and REE (Figure  
288 7C–D), a common feature of more Fe-rich carbonatites and accounted for by the  
289 crystallisation of strontianite and REE fluorcarbonates in these samples (Le Bas, 1989).

290 Mn-Fe-veins have much higher average FeO and MnO concentrations and lower MgO, SrO  
291 and LOI values than both analysed carbonatite types (Table 2, Figure 7A–C). On a ternary  
292 plot (Figure 6), these rocks form a continuum between calcite carbonatite and  
293 ferrocarbonatite, as would be the case for mixtures of calcite and Fe-/Mn-oxides. Mixing is  
294 also supported by the trends in Figure 7A, where the Mn-Fe-vein data plot between  
295 carbonatite compositions and a hematite/goethite endmember, suggesting these samples  
296 are the result of carbonate removal and, therefore, residually enriched in Fe oxides. Mn-Fe-  
297 veins do not show any relationship between REE, SrO and MnO+FeO contents and have  
298 similar REE concentrations to both C2 and C3 carbonatites.

299 REE concentrations in the Songwe carbonatites range from approximately 5000–60,000  
300 ppm, with ferroan calcite carbonatite typically more REE-rich, averaging between 25,000 and

301 60,000 ppm (Figure 7C). All rock types at Songwe are LREE enriched (Figure 8A), a feature  
302 common to most carbonatites (e.g. Jones et al., 2013). The degree of LREE enrichment  
303 increases in later C3 carbonatites and, to a lesser extent, in the Mn-Fe-veins (Figure 8B).  
304 However, in the latest apatite-fluorite rocks, LREE contents are similar to those of C2  
305 carbonatite, while the HREE contents are relatively enriched. In these rocks, Y  
306 concentrations average 1135 ppm, approximately double the average for carbonatites at  
307 Songwe Hill, while having similar total REE contents. The elevated HREE contents  
308 correspond to higher P<sub>2</sub>O<sub>5</sub> concentrations, reaching >1500 ppm Y and >10.72 Wt. % P<sub>2</sub>O<sub>5</sub>  
309 (Figure 7F), and indicate that apatite is hosting the HREE at Songwe, as it is the most  
310 abundant phosphate mineral present (Broom-Fendley et al., 2017).

## 311 **5. U–Pb age determinations**

### 312 ***5.1 Analytical and data processing procedures***

313 Geochronological data from zircons in C1 and C2 carbonatites, as well as a sample from the  
314 Mauze nepheline syenite, were acquired over three sessions at NIGL (NERC Isotope  
315 Geosciences Laboratories). The methodology followed that described in Thomas et al (2010)  
316 with data reduction and reporting procedures following those described in Horstwood et al  
317 (2016).

318 While there is a significant fenite contribution to the carbonatite at Songwe, it is highly likely  
319 the zircons from C1 carbonatite crystallised from a carbonatite melt, rather than entrained as  
320 xenocrysts from the surrounding rocks. This is demonstrated by their formation at carbonate  
321 grain boundaries and association with rounded apatite grains, indicating crystallisation prior  
322 to carbonate (Figure 4A). Furthermore, the majority of these grains are relatively small (~0.2  
323 mm) and exhibit little zoning (e.g. Figure 5A–C). Comparing the carbonatite zircons with  
324 those from fenite and nepheline syenite (the two most-proximal potential contamination  
325 sources) with those from carbonatite indicates that the carbonatite zircons are texturally

326 distinct, exhibiting different luminescence colours crystal habit and zoning (Supplementary  
327 Figure 2).

328 The zircons were ablated in-situ using a New Wave Research UP193SS (193 nm), Nd:YAG  
329 laser ablation system fitted with a Large Format Cell (holding 6 standard-sized polished thin  
330 sections) coupled to a Nu Instruments Nu Plasma HR MC-ICP-MS. Operational parameters  
331 for laser ablation and mass spectrometry are detailed in Supplementary Table 3 with further  
332 methodology details in the Supplementary Information.

333 Data were normalised using '91500' as the primary zircon reference material with 'Mud Tank'  
334 and 'GJ1' used as validation materials to determine accuracy and the long-term uncertainty  
335 for propagation. The U and Pb concentration of all zircon analyses was generally low,  
336 averaging ca. 2 ppm Pb and ca. 100 ppm U with many analyses well below this. Combined  
337 with the required spatial constraints based on CL imaging, this necessitated using a 35  $\mu\text{m}$   
338 ablation spot size. At these low concentrations Mud Tank represents a useful validation  
339 material (ca. 2 ppm Pb, 20 ppm U), capturing the long term excess variance uncertainty  
340 component (2.1%  $2\sigma$  U–Pb) not reflected when analysing higher U reference materials such  
341 as GJ1 (no excess variance in U–Pb over the study, see Supplementary Table 4).

342 Propagating this systematic, long-term excess variance uncertainty into the weighted mean  
343 uncertainty of the sample result, follows community-derived guidelines recommended in  
344 Horstwood et al (2016), and accounts for systematic shifts between sessions for results from  
345 low U materials. This demonstrates that the results from the different sessions for these low  
346 U materials are within uncertainty of each other (N.B. the higher U concentration GJ1 did not  
347 show such session biases). U–Pb dates for each session, with and without propagation for  
348 their systematic uncertainty component, are listed in Table 3. Final ages are quantified as  
349 the average from all sessions  $\pm$  the maximum range of session results allowing for the  
350 propagated session uncertainties. This uncertainty range equates to 3.8–6.7 Ma on the  
351 average and therefore encompasses the systematic uncertainty level (typically 3–4 Ma  $2\sigma$ ).  
352 Using reference values from Horstwood et al (2016), validation results from all sessions for

353 Mud Tank and GJ1 (without propagation for systematic uncertainties) demonstrate accurate  
354 results within the defined uncertainties (i.e. bias < uncertainty) with those data for Mud Tank  
355 highlighting the need for an additional variance component ( $^{206}\text{Pb}/^{238}\text{U} = 0.11996 \pm 0.48\%$ ,  
356 MSWD = 4.3, n= 27, Bias = -0.21%, long term variance = 2.1%  $2\sigma$ ; see Supplementary  
357 Table 4 and Supplementary Figure 3).

## 358 **5.2 Results**

359 All data reflect the presence of small amounts of common-Pb however  $^{204}\text{Pb}$  is not  
360 resolvable by the LA-ICP-MS method used. As such, sample ages are interpreted using  
361 Tera-Wasserburg plots (e.g. Fig 9). Zircons from two calcite carbonatite (C1) samples  
362 (T0218 & T0206) were analysed in three different sessions over a period of 3 months  
363 returning ages between  $129.5 \pm 3.3/4.3$  and  $137.3 \pm 1.0/3.1$  ( $2\sigma$ , MSWD = 1.8, n=11), with  
364 both within-session and between-session uncertainties quoted following Horstwood et al  
365 (2016). The results are equivalent allowing for their systematic uncertainties and are  
366 combined in an average and maximum range of  $132.9 \pm 6.7$  Ma. Zircons from a single C2  
367 calcite carbonatite were dated in one session only, defining an age of  $135.6 \pm 2.5/3.8$  ( $2\sigma$ ,  
368 MSWD = 1.9, n=5) after systematic uncertainty propagation.

369 Zircons from Mauze (sample U4913) were analysed in two different sessions 3 months  
370 apart, returning ages between  $137.5 \pm 1.5/3.3$  Ma ( $2\sigma$ , MSWD = 3.6, n=18) and  $131.7 \pm$   
371  $0.9/2.9$  Ma ( $2\sigma$ , MSWD = 6.1, n=43). The results are within uncertainty after propagation and  
372 are combined in an average and maximum range of  $134.6 \pm 4.4$  Ma. As such, it is  
373 indistinguishable in age isotopically from the Songwe samples even though field  
374 relationships indicate that it is older (see below).

## 375 **6. Discussion**

### 376 **6.1 Order of emplacement and fluid evolution**

377 Field relationships indicate that the Songwe carbonatite intruded and fenitised the Mauze  
378 nepheline syenite and is, therefore, younger. The principal evidence for this is the  
379 equigranular texture of the fenite, incorporating large rectangular pseudomorphs of feldspar  
380 which is texturally reminiscent of the neighbouring nepheline syenite. Blocks of this material  
381 have been veined, brecciated and incorporated into the intruding carbonatite. These field  
382 relationships provide a better constraint on the relative age of the intrusion events than the  
383 radiometric ages of Songwe ( $132.9 \pm 6.7$  Ma) and Mauze ( $134.6 \pm 4.4$  Ma).

384 Carbonatites are theorised as being generated via one of two principal mechanisms: (1) as  
385 'segregations' from a carbonated silicate melt, either through (a) liquid immiscibility  
386 (Kjarsgaard and Hamilton, 1989; Lee and Wyllie 1997, 1998) or (b) as a residual melt  
387 fraction after silicate fractionation (Gittins, 1989; Lee and Wyllie, 1994). Alternatively (2), they  
388 could be generated from melting CO<sub>2</sub>-bearing peridotite directly from the mantle (Wallace  
389 and Green, 1988; Harmer and Gittins, 1998; Brey et al., 2008). Gittins and Harmer (2003)  
390 argue that even where silicate rocks accompany carbonatites, the two may not be  
391 genetically related, favouring a model where two separate melts utilise the same conduits to  
392 reach the crust from the mantle. Owing to the lack of analyses from the Mauze intrusion,  
393 there is insufficient geochemical data to robustly comment on the relationship between the  
394 Songwe carbonatite and Mauze nepheline syenite. However, the indistinguishable  
395 emplacement ages of the two intrusions provides circumstantial evidence of a genetic  
396 connection, suggesting direct emplacement of carbonatite from the mantle may not be an  
397 appropriate model (cf. Xu et al., 2015).

398 A common compositional trend during the evolution of many silicate-poor carbonatites is  
399 from calcium-rich to magnesium- and iron-rich compositions with decreasing P, Zr, Nb and



400 increasing LREE and F (e.g. Le Bas, 1987, 1989; Hamilton et al., 1989; Thompson et al.,  
401 2002). These trends represent a 'typical' evolution path for a Si-depleted carbonatite,  
402 dominated by fractional crystallisation of carbonates, as well as apatite and pyrochlore (Le  
403 Bas, 1989; Kjarsgaard and Hamilton, 1989; Hamilton et al., 1989). For instance, Xu et al.,  
404 (2010) showed that continued fractionation of carbonates at the Miaoya carbonatite, China,  
405 can lead to progressive enrichment in the light REE. This common chemical evolution is  
406 reflected by the carbonatite compositions at Songwe where Fe-rich carbonatites (C3) are  
407 more enriched in LREE and SrO than C2 calcite carbonatite (Figure 7C). C1 carbonatites,  
408 comprising relatively coarse-grained apatite, zircon and calcite, might, by analogy with more  
409 deeply eroded carbonatites (e.g. Le Bas, 1977, 1987), represent a cumulate developed from  
410 a lower level of the intrusion. Their presence at depth is evidenced by the occurrence of  
411 coarse calcite carbonatite clasts in the later carbonatite varieties (Figure 3E).

412 During the later stages of emplacement, carbonatites transition from a magmatic to a  
413 hydrothermal regime and fluids are expelled (Rankin, 2005). These fluids commonly lead to  
414 LREE, barite, strontianite and fluorite mineralisation, manifested in veins, or disseminated  
415 throughout the host carbonatite (e.g. Mariano, 1989; Ngwenya, 1994; Wall and Mariano,  
416 1996; Andrade et al., 1999; Wall and Zaitsev, 2004; Doroshkevich et al., 2009; Nadeau et  
417 al., 2015). Several features, such as crystallisation of LREE minerals in veins and in small  
418 cavities, were identified by Broom-Fendley et al., (2017) who ascribe a similar role for a  
419 hydrothermal component to the LREE mineralisation at Songwe.

## 420 **6.2 (Carbo)-hydrothermal alteration**

421 Despite the 'typical' evolution of the carbonatite at Songwe towards more Fe- and REE-rich  
422 compositions, some notable exceptions are apparent. One such example is the correlation  
423 between  $P_2O_5$  and the HREE (Fig. 7F), which has not been previously documented in  
424 carbonatites. Correlation between  $P_2O_5$  and the HREE is controlled by the uptake of the  
425 HREE in late-stage apatite at Songwe (Broom-Fendley et al., 2017). Late apatite forms

426 anhedral stringers and cross cuts many carbonate phases (Broom-Fendley et al., 2017; Figs  
427 4D–E). Late apatite is apparent in all carbonatite types at Songwe, and is independent of  
428 SrO, LREE and FeO contents. The disconnect between these common indicators of  
429 carbonatite evolution from the P<sub>2</sub>O<sub>5</sub> and HREE contents suggests HREE-enrichment and  
430 P<sub>2</sub>O<sub>5</sub>-bearing mineral crystallisation is an artefact of a late-stage fluid at Songwe, cross-  
431 cutting the principal carbonatite types. The products of such a fluid are locally manifested as  
432 apatite-fluorite-veins, which have notably higher HREE contents than the other carbonatite  
433 units at Songwe (Fig. 8B).

434 Further evidence for a hydrothermal contribution to the evolution of Songwe is indicated by  
435 the presence of a Y/Ho anomaly in the whole-rock REE distribution of all the carbonatite  
436 types (Fig. 8). While the magnitude of this Y/Ho anomaly is quite small, repeat analyses of  
437 the AMIS0185 and GRE-04 carbonatite standards indicate the data are accurate and the Y  
438 anomaly is related to natural processes and the anomaly is, therefore, significant to  
439 carbonatite evolution (Supplementary Table 1). This process is likely to be a hydrothermal  
440 overprint as fractionation of Y is generally considered to be caused by the preferential  
441 transport of Y, over the other HREE, in F-bearing aqueous fluids (Bau and Dulski, 1995;  
442 Bau, 1996; Loges et al., 2013). Regardless of the likelihood that the fluid is H<sub>2</sub>O-rich, a  
443 significant carbonate component cannot be discounted and the late fluids at Songwe are  
444 therefore termed (carbo)-hydrothermal. The presence of the Y/Ho anomaly throughout all  
445 analysed carbonatite types indicates these (carbo)-hydrothermal fluids overprinted all of the  
446 carbonatites (C1–C3; Bühn, 2008; Broom-Fendley et al., 2017).

447 Cross-cutting relationships indicate that Mn-Fe veins are the last stage to occur at Songwe,  
448 predominantly following brittle fractures. Mn-Fe-veins are composed of oxide minerals which  
449 are likely to be the result of the de-carbonation of previous carbonatites. This genesis is  
450 supported by the erratic composition of the Mn-Fe-veins, which contain a variable CaO and  
451 FeO content suggestive of partial CaCO<sub>3</sub> dissolution (Fig. 6). This is also supported by

452 petrographic observations, where samples from the centre of a vein are more calcite-poor  
453 than those from the edge of a vein. Low SrO and MgO concentrations in these rocks,  
454 compared with C2 and C3 carbonatites, indicates these elements have been removed during  
455 dissolution. Similar rock types to the Mn-Fe-veins have been described from other locations,  
456 summarised as hematite carbonatites by Andersen (1987a, b). Examples include: Fen,  
457 Norway; Lueshe, DR-Congo; and several locations in Kenya (Andersen, 1984, 1987b).  
458 These rocks are interpreted as the products of post-magmatic dissolution of Fe-rich  
459 carbonatite under oxidising conditions, where Fe is immobile (Andersen, 1987b). Based on  
460 O isotope analyses, Andersen (1987a) interprets the alteration to have been caused by  
461 meteoric water, after the last stages of intrusion.

### 462 **6.3 A shallow carbonatite**

463 While a hydrothermal component is generally required to generate ore grades of LREE,  
464 extensive remobilisation of the HREE is much less common. Extensive hydrothermal  
465 overprinting of the HREE does not occur in other carbonatites of the Chilwa Alkaline  
466 Province, suggesting Songwe is unique in some aspect. One such aspect could be the  
467 mineralising fluid source. Stable (C and O) data for apatite and calcite at Songwe indicate a  
468 likely meteoric contribution to the apatite mineralisation and, potentially, to calcite  
469 crystallisation. However, a contribution from magma degassing and carbonatite-derived fluid  
470 cannot be entirely discounted (Broom-Fendley et al., 2016b). A contribution from meteoric  
471 water suggests a shallow intrusion level and is consistent with the strong evidence from the  
472 abundance of fenite and lack of carbonatite at the top of the hill, probably indicating that the  
473 carbonatite has not intruded above the current erosion depth. A shallow intrusion level is  
474 also supported by the presence of brittle deformation, occurring both in fenite around the  
475 carbonatite (e.g. Figure 3A) and in breccia clasts and stoped blocks within the carbonatite.  
476 Such extensive brecciation indicates a change from lithostatic to hydrostatic pressure, which  
477 occurs at shallow crustal levels. This local field evidence is also supported by apatite fission-

478 track ages from other intrusions in the Chilwa Province. These ages indicate the other  
479 intrusions cooled rapidly and were therefore emplaced at a shallow level (Eby et al., 1995).

480 Given the features discussed above, we suggest that Songwe Hill, at its current level of  
481 erosion, is the highest level of a shallow, subvolcanic magma chamber. It is likely that a  
482 'head' of volatiles or late magmatic pulses has successively pooled in these upper levels,  
483 interacting with meteoric water, and resulting in multiple stages of hydrothermal overprinting  
484 and mineralisation. Rounded clasts of the carbonate-rich breccias and pervasive  
485 hydrothermal overprinting suggests widespread degassing and volatile loss from a  
486 carbonatite magma.

#### 487 **6.4 Comparison with other Chilwa intrusions**

488 The new U-Pb zircon ages indicate that Songwe is a similar age to the other major  
489 carbonatite complexes in the Chilwa Alkaline Province (Figure 1). Indeed, many parallels  
490 can be drawn between Songwe and these other large Chilwa carbonatite complexes.  
491 Carbonatites at Tundulu and Chilwa Island progressively evolve to increasing Fe and REE  
492 concentrations (Garson and Smith, 1958; Garson, 1962) while there is evidence for late  
493 HREE-enrichment in apatite at Tundulu and in minor rocks at Kangankunde (Broom-Fendley  
494 et al., 2016a). Furthermore, brecciation of country rock around these intrusions is common,  
495 with contact breccias and fenite breccia occurring at all localities. Cooling rates for other  
496 intrusions in the Chilwa Alkaline Province also suggest a shallow intrusion level (Eby et al.,  
497 1995). However, there are also significant differences between Songwe and other Chilwa  
498 Alkaline Province carbonatites. Whereas some carbonatites in the province are  
499 geographically, and in some cases temporally, accompanied by silicate rocks, the volume of  
500 silicate rock exposed is much lower, relative to the volume of nepheline syenite at Mauze  
501 adjacent to Songwe. This difference could be important in differentiating the HREE  
502 occurrences at Songwe Hill from the other intrusions of the Chilwa Alkaline Province. Given  
503 that the U–Pb age of Mauze is within the uncertainty of the age of Songwe, it is likely that

504 Mauze was still hot when Songwe intruded. Hydrothermal activity, with temperatures  
505 calculated by Ti in zircon thermometry of approximately 500–600°C, was estimated to  
506 continue at Zomba (another Chilwa intrusion, larger than Mauze; Fig. 1) for up to 3 Ma  
507 subsequent to emplacement (Soman et al., 2010). Furthermore, for the same intrusion, Eby  
508 et al. (1995) calculated, from K-Ar amphibole and fission-track zircon ages, that the cooling  
509 rate of Zomba was approximately 23°C/Ma. These characteristics suggest that, although  
510 Mauze is volumetrically less substantial, it could provide a source of sustained heat for many  
511 years after emplacement. Such a heat source would drive continued interaction with  
512 surrounding groundwater during the emplacement of Songwe. This, in turn, could explain the  
513 presence of extensive (carbo)-hydrothermal overprinting and HREE mineralisation at  
514 Songwe, which is essentially absent from other Chilwa carbonatites.

## 515 **7. Conclusions**

516 In this contribution we have summarised the age, geology and geochemistry of the Songwe  
517 Hill carbonatite. The earliest carbonatite type (C1) calcite carbonatite ( $132.9 \pm 6.7$  Ma)  
518 intruded the older Mauze nepheline syenite ( $134.6 \pm 4.4$ ), although the overlapping  
519 uncertainty between the absolute ages indicates these intrusions occurred over a short  
520 timeframe. The carbonatite evolved through increasingly Fe-rich contents, with an  
521 associated increase in REE and Sr, typical of many carbonatites. However, Songwe differs  
522 from other carbonatites, exhibiting a strong relationship between the HREE and  $P_2O_5$ , with  
523 apatite as the main HREE host. Cross-cutting relationships and anomalous Y/Ho ratios in all  
524 carbonatite types indicates that the HREE mineralisation was caused by the influx of a  
525 (carbo)-hydrothermal fluid, potentially sourced from interaction with surrounding  
526 groundwater. A high degree of brecciation and a large, contemporaneous, neighbouring heat  
527 source (Mauze nepheline syenite) contributed to the extensive development of groundwater  
528 circulation which differentiates Songwe from other Chilwa carbonatites in which HREE  
529 enrichment is absent or only minor.

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725 **List of Tables**

726 **Table 1.** *Nomenclature of the carbonatite types at Songwe Hill. Use of the field ID and*  
 727 *grouping is maintained throughout the manuscript*

Group	Field ID	Geochemical name	
		(Gittins & Harmer, 1997)	Other names
C1	Calcite carbonatite	Calciocarbonatite	Sövite
C2	Calcite carbonatite	Calciocarbonatite	Alvikite
	Ferroan calcite	Ferruginous	
C3	carbonatite	calciocarbonatite	Ferrocronatite
N/A	Apatite-fluorite veins		
N/A	Mn-Fe-veins		Rødberg

728

729 **Table 2.** Average whole-rock compositions for the different rock types at the Songwe Hill  
 730 carbonatite, plus two outlier apatite-fluorite-vein samples from Chenga Hill

Sample	Fenite	Calcite carbonatite	Ferroan calcite carbonatite	Mn-Fe- veins	Ap-FI-veins	Chenga Hill	
(Wt. %)	Avg. (n=421)	Avg. (n=145)	Avg. (n=30)	Avg. (n=34)	Avg. (n=20)	T0178	T0134
SiO <sub>2</sub>	29.92	1.53	1.03	1.98	1.47	22.68	47.70
TiO <sub>2</sub>	0.66	0.19	0.15	0.17	0.43	0.82	1.63
Al <sub>2</sub> O <sub>3</sub>	10.89	0.70	0.73	0.98	1.46	4.29	14.19
FeO	11.2	10.81	15.58	22.95	17.44	7.35	8.71
MnO	1.48	1.93	3.35	4.69	2.61	1.29	1.03
MgO	1.51	1.96	2.73	0.39	4.12	1.13	0.17
CaO	14.69	37.15	31.10	30.52	28.52	28.4	3.64
K <sub>2</sub> O	6.65	0.43	0.19	0.38	0.34	2.75	9.2
P <sub>2</sub> O <sub>5</sub>	1.19	2.43	3.85	2.95	6.83	9.17	3.05
S	0.42	0.61	0.73	0.28	0.77	0.13	bd
SrO	0.75	2.07	2.78	0.55	0.54	0.48	0.13
Nb <sub>2</sub> O <sub>5</sub>	0.14	0.19	0.21	0.13	0.10	0.04	0.04
LOI (1000°C)	15.49	34.87	28.73	25.28	na	na	na
Total	94.99	94.85	91.21	91.09	63.93	77.99	89.33
(ppm)							
Be	13	2.7	4.1	4.4	na	na	na
Cr	5.5	0.7	5.6	3.6	na	na	na
Ga	5.8	12	0.3	9.3	bd	10	38
Hf	5.8	0.8	2.0	1.7	4.1	24	15
Li	8.5	3.4	5.4	4.2	na	na	na
Sc	0.2	2.0	20	14	na	na	na
Sn	6.5	2.7	3.9	2.6	na	na	na
Ta	19	1.3	0.8	1.4	na	na	na
Th	270	280	370	420	370	380	350
U	19	11	7.7	11	9.8	28	11
V	110	140	140	200	na	na	na
W	14	8.6	15	9.1	na	na	na
Zr	361.1	37.3	101.1	87.8	223.6	1293	819



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La	1600	3500	9200	5300	3200	860	1300
Ce	2800	6700	13400	9100	6600	1300	1900
Pr	290	720	1400	940	760	120	190
Nd	970	2400	4100	2900	2700	420	620
Sm	150	340	480	380	400	100	100
Eu	44	95	120	100	120	55	37
Gd	110	220	280	240	280	250	130
Tb	14	29	34	31	39	79	26
Dy	62	140	150	150	210	560	150
Ho	9.9	24	24	24	39	100	28
Er	23	57	54	54	110	260	66
Tm	2.8	7.3	6.6	6.7	15	31	8.0
Yb	17	41	35	38	85	170	45
Lu	2.2	5.2	4.5	5.0	11	21	5.5
Y	250	610	590	590	1000	3000	740

bd: below detection limits, na: not analysed For full dataset see **Supplementary Table 2.**

NB: Ba and Na not analysed owing to preparation as Na-peroxide fusion

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731

732 **Table 3.** Laser-ablation U–Pb ages

Sample	Rock type	Regression intercept age (Ma), $2\sigma$	MSWD, n	Regression uncertainty with $\sigma_{\text{sys}}$ ( $2\sigma$ Ma)
T0208 (Session 1)	C1	$129.5 \pm 3.3$	1.9, n= 7	4.3
T0206 (Session 1)	C1	$131.8 \pm 2.5$	4.4, n= 11	3.7
T0206 (Session 2)	C1	$132.9 \pm 1.1$	7.5, n= 34	3.0
T0206 (Session 3)	C1	$137.3 \pm 1.0$	1.8, n= 11	3.1
U4913 (Session 1)	Nepheline syenite	$137.5 \pm 1.5$	3.6, n= 18	3.3
U4913 (Session 3)	Nepheline syenite	$131.7 \pm 0.9$	6.1, n= 43	2.9
T0225 (Session 3)*	C2	$135.6 \pm 2.5$	1.9, n= 5	3.8

733 \*Excluding one point from regression

734

## 735 **List of Figures**

736 **Figure 1:** Map of the Chilwa Alkaline Province, adapted from Woolley (2001). Included for  
737 reference are the ages of the major carbonatites, from: Snelling (1966) [Kangankunde,  
738 Chilwa Island, Tundulu], Wall et al., (1994) [Kangankunde], and Eby et al. (1995) [Chilwa  
739 Island].

740 **Figure 2:** Geological map of Mauze and Songwe (A) and of carbonatite types at Songwe  
741 (B). Drill holes from which samples were taken are indicated. For the complete geological  
742 map, and all drill hole collars, see Croll et al. (2014). Grid system is UTM 36S, WGS1984  
743 datum. Maps redrawn after Garson and Walshaw (1969) and Croll et al. (2014).

744 **Figure 3:** Field photos of the major lithologies at Songwe (A) contact breccia on Chenga Hill  
745 displaying decreasing degrees of fenitization between the edges and cores of clasts; (B)  
746 Grey C2 calcite carbonatite incorporating clasts of fenite and cross-cut by small Mn-Fe-  
747 veins; (C) Carbonate-rich breccia, incorporating two different types of calcite carbonatite:  
748 earlier C1 calcite carbonatite, and later C2 calcite carbonatite; (D) Contact between (C3)  
749 Ferroan calcite carbonatite and C2 calcite carbonate in drill core (PX003, 16 m depth); (E)  
750 Ferroan calcite carbonatite breccia with clasts of C1 calcite carbonatite and fenite; (F)  
751 Example of a road-cut with extensive alteration of Fe-rich rock to hematite, goethite, wad  
752 and limonite. Hammer head is approximately 10 cm.

753 **Figure 4:** Cathodoluminescence (B–D, H) and back-scattered electron (E–G) images of  
754 Songwe carbonatites. (A) Large euhedral apatite, zircon and pyrite in C1 calcite carbonatite;  
755 (B) apatite bands in C2 calcite carbonatite; (C) vugs of synchysite, baryte and strontianite in  
756 C2 calcite carbonatites; (D) truncation of an apatite stringer by a REE-fluorcarbonate-bearing  
757 assemblage in C3 carbonatites; (E) partially fragmented apatite in a matrix of strontianite  
758 and fluorcarbonates in C3 carbonatites; and, (F) apatite and fluorite in an apatite-fluorite vein  
759 sample.

760 **Figure 5:** Back-scattered electron images of zircon from Songwe carbonatite. (A–B)  
761 Unzoned subhedral zircons in C1 carbonatite; (C) corroded zircon from C2 carbonatite; (D–  
762 E) zoned fractured zircon in C1 carbonatite. Circles approximate ablation pit locations. Labels  
763 correspond to results in Supplementary Table 5.

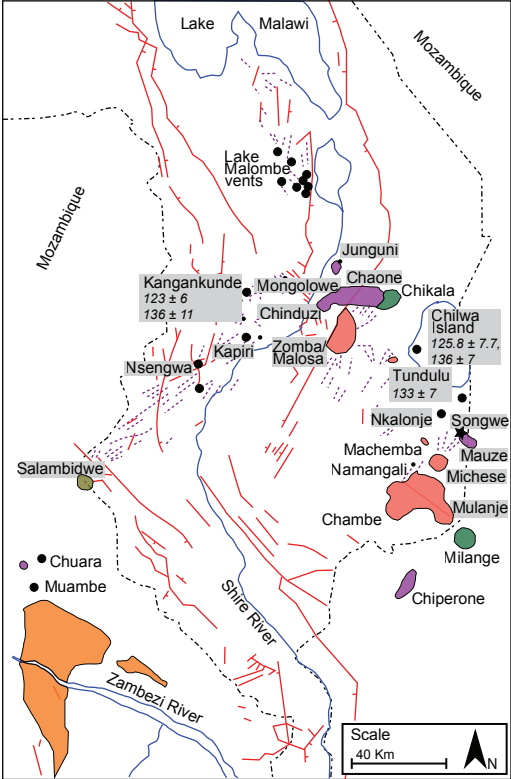
764 **Figure 6:** Ternary carbonatite classification diagrams after Gittins and Harmer (1997), and  
765 Woolley and Kempe (1989). The trends caused by increasing contents of minerals that can  
766 affect the carbonatite composition on this diagram are indicated.

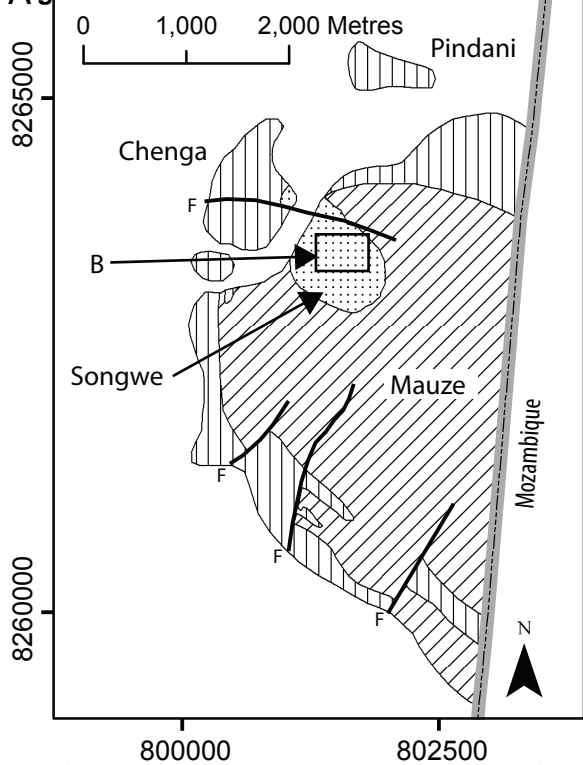
767 **Figure 7:** Whole-rock data from carbonatite, Mn-Fe- veins and apatite-fluorite veins from  
768 Songwe. (A) LOI represents a close approximation of CO<sub>2</sub>. Grey lines equate to mixing lines  
769 between pure calcite (bottom-right) and various Fe-phases indicated on the graph. These  
770 are simple plots of binary mixing and are not intended to be wholly representative of the  
771 composition of the samples. Lines in all other figures are to indicate the main interpreted  
772 trends in the data. NB, C1 samples were too scarce for whole-rock analysis.

773 **Figure 8:** Average distributions of the REE in Songwe carbonatites, Mn-Fe veins and  
774 apatite-fluorite veins normalised to (A) chondrite and (B) C2 carbonatite. Symbols same as  
775 for Figure 7.

776 **Figure 9:** Example Tera-Wasserburg plot of Songwe sample T0206 (session 3). See Table  
777 3 for other U–Pb ages and supplementary Table 5 for dates.

778



**Figure 2****Legend (A)**

- National Border
- Basement
- Nepheline Syenite
- Carbonatite complex
- Cenozoic sediments
- Faults

**Legend (B)**

- Selected drill collars
- Drill hole traces
- Breccia
- Ferroan calcite carbonatite
- Calcite carbonatite
- Fenite breccia

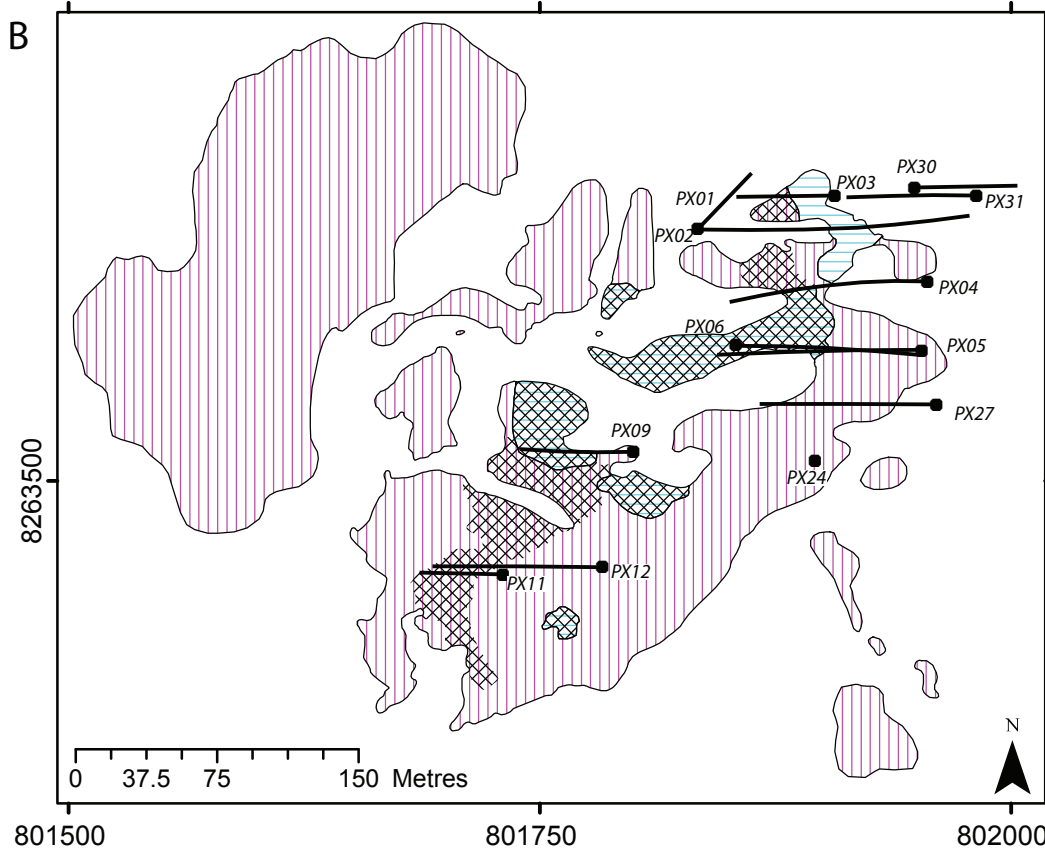
**B**

Figure 3  
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Figure 4  
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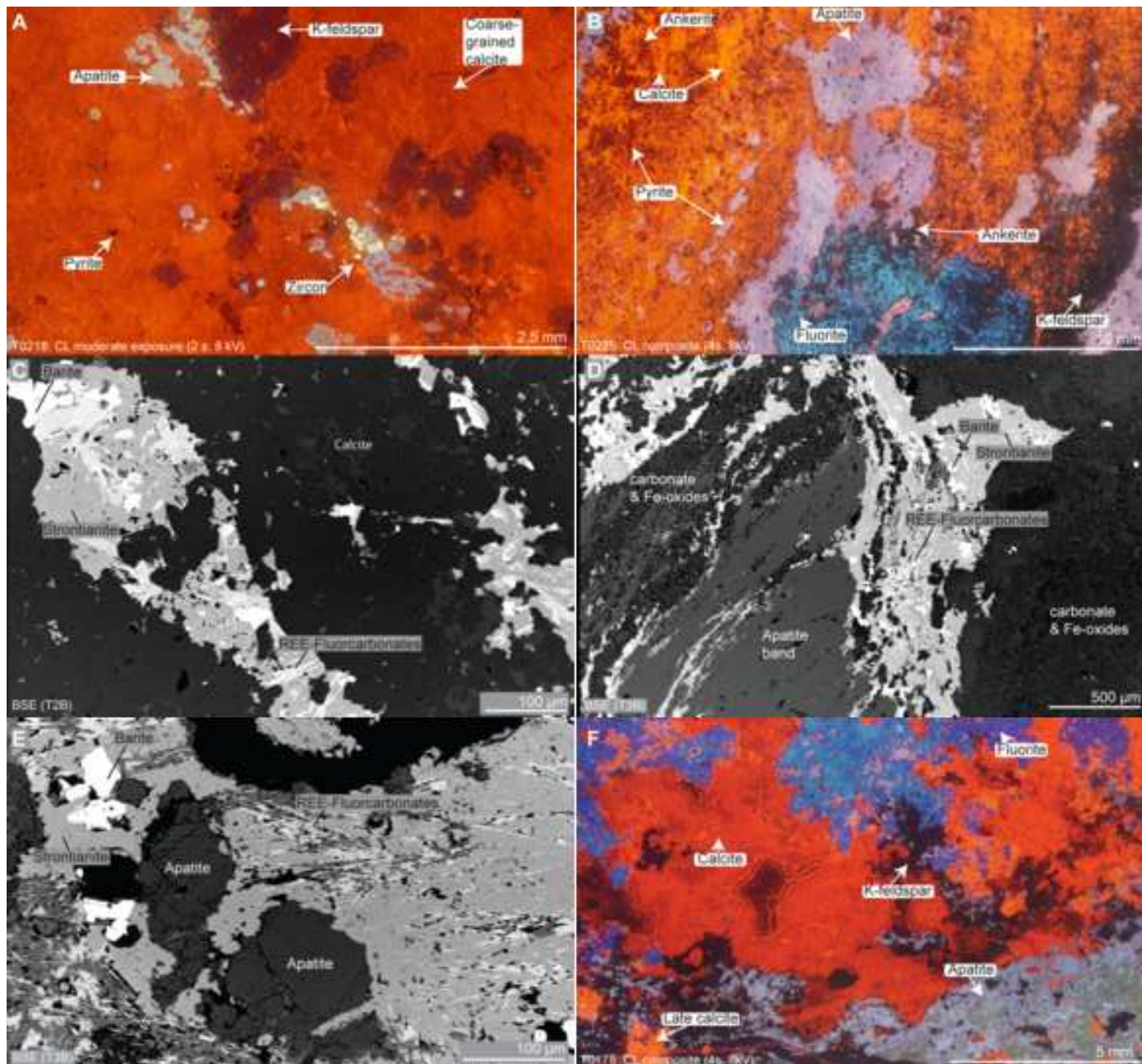
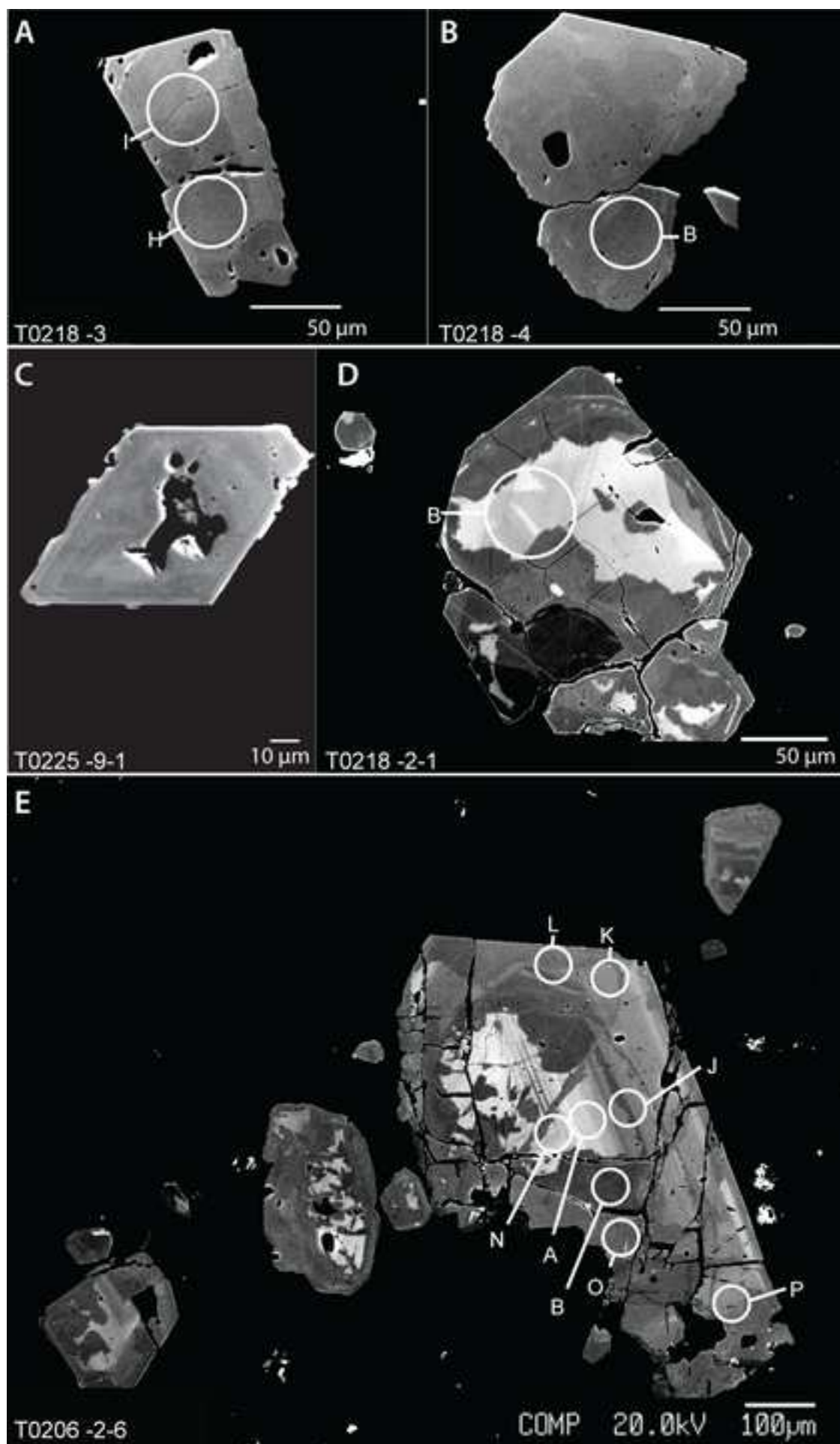
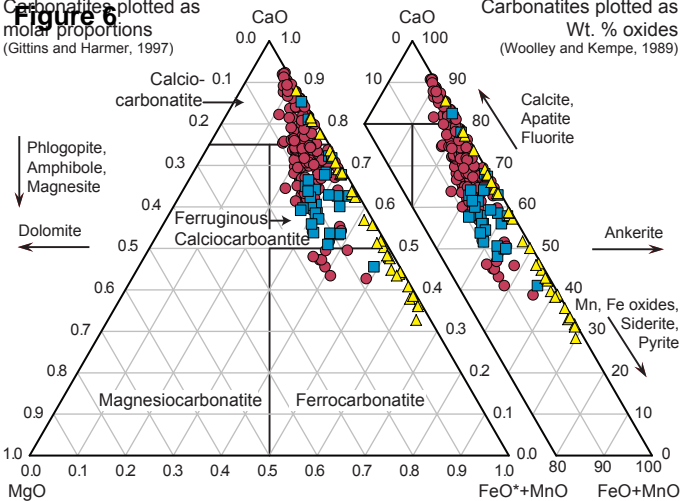




Figure 5  
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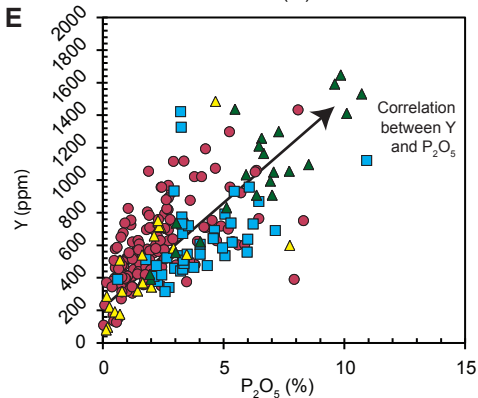
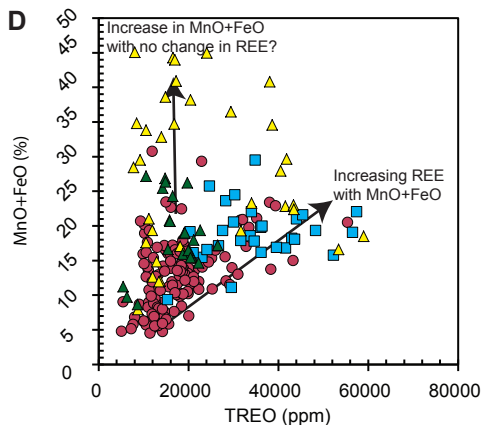
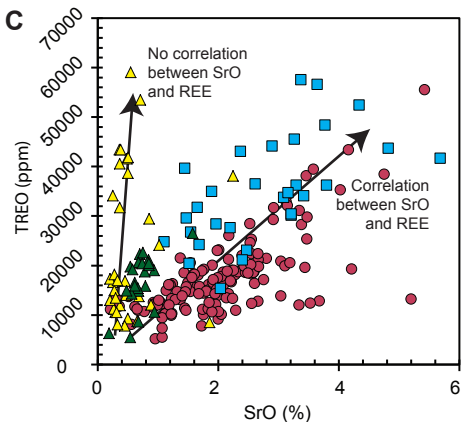
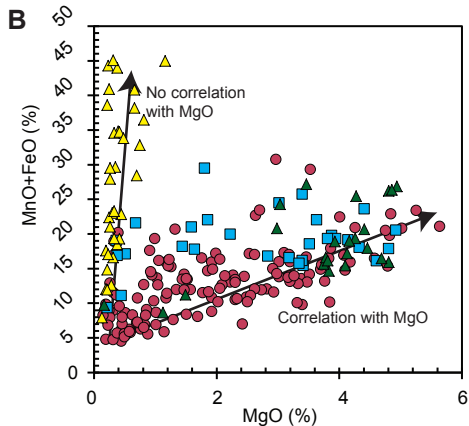
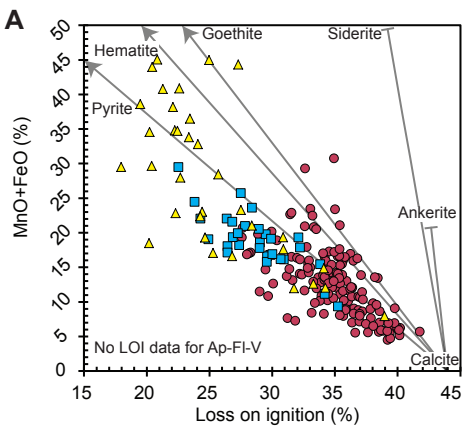


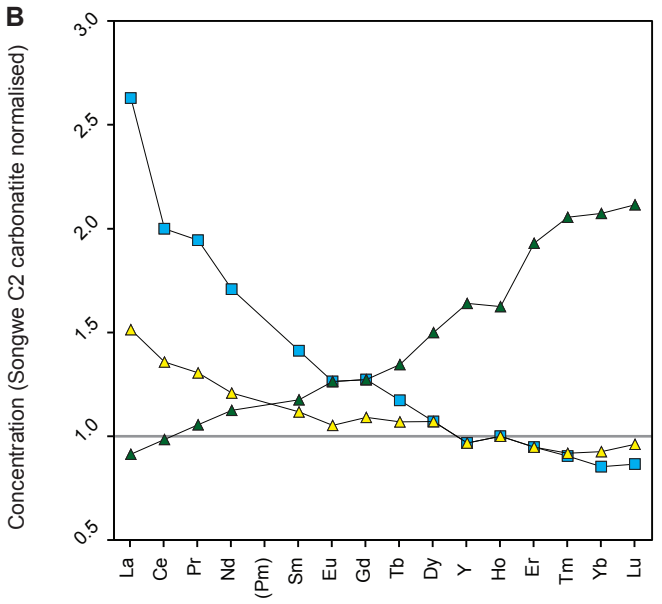
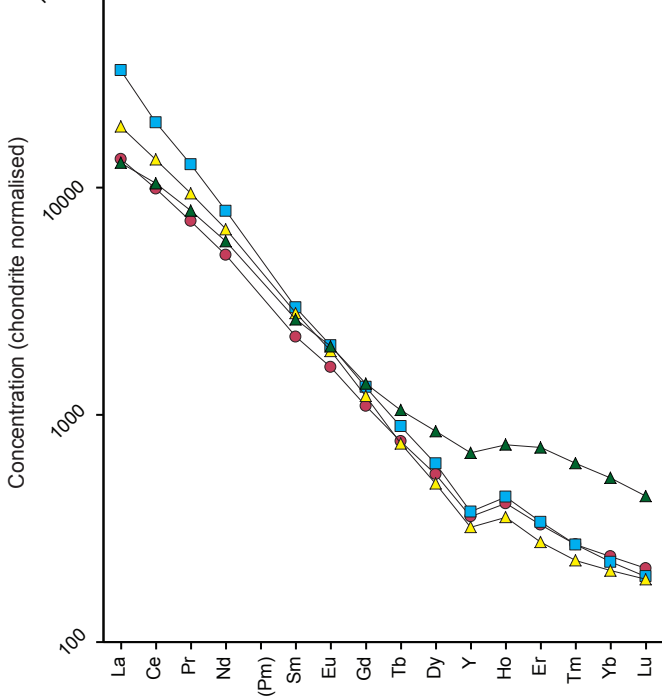
**Figure 6:** Carbonatites plotted as molar proportions (Gittins and Harmer, 1997)



● Calcite-carbonatite (C2) ■ Ferroan-calcite carbonatite (C3) ▲ Mn-Fe- vein

**Figure 7** Carbonatite (C2) ■ Ferroan calcite carbonatite (C3) ▲ Apatite-fluorite- veins ▲ Mn-Fe- vein



**Figure 8**

**Figure 9**data-point error symbols are  $2\sigma$ 