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Revision 2

Word Count: 15,597 (excluding References)

**FLUORINE-RICH MAFIC LOWER CRUST IN THE SOUTHERN ROCKY  
MOUNTAINS: THE ROLE OF PRE-ENRICHMENT IN GENERATING FLUORINE-  
RICH SILICIC MAGMAS AND PORPHYRY MO DEPOSITS**

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**ABSTRACT**

Fluorine-rich granites and rhyolites occur throughout the southern Rocky Mountains, but  
the origin of F-enrichment has remained unclear. We test if F-enrichment could be inherited  
from ancient mafic lower crust by: (1) measuring amphibole compositions, including F and Cl  
contents, of lower crustal mafic granulite xenoliths from northern Colorado to determine if they  
are unusually enriched in halogens; (2) analyzing whole-rock elemental and Sr, Nd, and Pb  
isotopic compositions for upper crustal Cretaceous to Oligocene igneous rocks in Colorado to

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22 evaluate their sources; and (3) comparing batch melting models of mafic lower crustal source  
23 rocks to melt F and Cl abundances derived from biotite data from the F-rich silicic Never  
24 Summer batholith. This approach allows us to better determine if the mafic lower crust was pre-  
25 enriched in F, if it is concentrated enough to generate F-rich anatectic melts, and if geochemical  
26 data support an ancient lower crustal origin for the F-rich rocks in the southern Rocky  
27 Mountains.

28         Electron microprobe analyses of amphibole in lower crustal mafic granulite xenoliths  
29 show they contain 0.56–1.38 wt% F and 0.45–0.73 wt% Cl. Titanium in calcium amphibole  
30 thermometry indicates that the amphiboles equilibrated at high to ultrahigh temperature  
31 conditions (805 to 940 °C), and semi-quantitative amphibole thermobarometry indicates the  
32 amphiboles equilibrated at 0.5 to 1.0 GPa prior to entrainment in magmas during the Devonian.  
33 Mass balance calculations, based on these new measurements, indicate parts of the mafic lower  
34 crust in Colorado are at least 3.5 times more enriched in F than average mafic lower crust.  
35 Intrusions coeval with the Laramide Orogeny (75 to 38 Ma) pre-date F-rich magmatism in  
36 Colorado and have Sr and Nd isotopic compositions consistent with mafic lower crust ± mantle  
37 sources, but many of these intrusions contain elevated Sr/Y compositions (>40) that suggest  
38 amphibole was a stable phase during magma generation. The F-rich igneous rocks from the  
39 Never Summer igneous complex and Colorado Mineral Belt also have Sr and Nd isotopic  
40 compositions that overlap with the lower crustal mafic granulite xenoliths, but they have lower  
41 Sr/Y, higher Nb and Y abundances, and distinctly less radiogenic  $^{206}\text{Pb}/^{204}\text{Pb}_i$  compositions than  
42 preceding Laramide magmatism. Batch melt modeling indicates low-degree partial melts derived  
43 from rocks similar to the mafic lower crustal xenoliths we analyzed can yield silicic melts with

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44 >2000 ppm F, similar to estimated F melt concentrations for silicic melts that are interpreted to  
45 be parental to evolved leucogranites.

46 We suggest that F-rich silicic melts in the southern Rocky Mountains were sourced from  
47 garnet-free mafic lower crust, and that fluid-absent breakdown of amphibole in ultrahigh  
48 temperature metamorphic rocks was a key process in their generation. Based on the composition  
49 of high-F amphibole measured from lower crustal xenoliths, the temperature of amphibole  
50 breakdown and melt generation for these F-enriched source rocks is likely >100 °C greater than  
51 similar lower crust with low or average F abundances. As such, these source rocks only melted  
52 during periods of unusually high heat flow into the lower crust, such as during an influx of  
53 mantle-derived magmas related to rifting or the post-Laramide ignimbrite flare-up in the region.  
54 These data have direct implications for the genesis of porphyry Mo mineralization, because they  
55 indicate that pre-enrichment of F in the deep crust could be a necessary condition for later  
56 anatexis and generation of F-rich magmas.

57 Keywords: fluorine, amphibole, Climax-type Mo deposits, topaz rhyolite, xenolith, southern  
58 Rocky Mountains, ultrahigh temperature metamorphism, granulite

59 **INTRODUCTION**

60 Fluorine-rich granites and rhyolites are often associated with economic mineral deposits  
61 of Sn, W, Be, U, F, Mn, Y, and REEs (e.g., Burt et al. 1982; Carten et al. 1993; Lüders et al.  
62 2008; Dostal et al. 2016; Liu et al. 2016; O'Neill et al. 2017; Dailey et al. 2018; Girei et al. 2020;  
63 Hofstra and Kreiner 2020). Numerous Mo-producing mines in the western United States are also  
64 associated with F-rich granites (“Climax-type” deposits; Ludington and Plumlee 2009), and  
65 therefore there is considerable interest in exploring the origin and distribution of F-rich granitic

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66 systems. The mineralizing intrusions in Climax-type deposits are often highly silicic (>75 wt%  
67 SiO<sub>2</sub>), slightly peraluminous, enriched in incompatible elements (e.g., Nb, Y, Rb), and depleted  
68 in elements compatible in feldspar (Sr, Ba, Eu<sup>2+</sup>; Carten et al. 1993; Ludington and Plumlee  
69 2009). These mineralizing intrusions are also chemically similar to topaz rhyolites and topaz  
70 granites (Christiansen et al. 2007; Jacob et al. 2015). At least three Climax-type deposits,  
71 including the eponymous Climax mine (Colorado), Henderson deposit (Colorado), and the Pine  
72 Grove deposit (Utah), are directly associated with topaz rhyolite eruptions (Keith et al. 1986;  
73 Bookstrom 1989; Mercer et al. 2015). Topaz rhyolites, unmineralized topaz granites, and the  
74 mineralizing intrusions in Climax-type deposits all form in tectono-magmatic environments in  
75 which ferroan silicic magmas are commonly generated (e.g., Christiansen et al. 2007; Frost and  
76 Frost 2011; Jacob et al. 2015).

77         The origins of F-rich melts in porphyry Mo deposits and related systems are unclear  
78 despite decades of research (e.g., White et al. 1981; Christiansen et al. 1983; Farmer and  
79 DePaolo 1984; Clemens et al. 1986; Carten et al. 1988; Stein and Crock 1990; Christiansen et al.  
80 2007; Audétat and Li 2017). Audétat (2015) suggested F-rich granites and rhyolites could form  
81 by low-degree partial melting of F-rich sources. However, it is not clear which, if any, specific  
82 source rocks are ultimately responsible for generating F-rich mineralizing intrusions. Some  
83 models suggest that F-rich silicic melts are produced by anatexis of felsic or mafic granulites in  
84 the lower crust (Stein and Crock 1990; Skjerlie and Johnston 1993; Jacob et al. 2015), whereas  
85 others suggest that silicic melts associated with large porphyry Mo deposits are derived by  
86 differentiation of melts extracted from metasomatized lithospheric mantle (Pettke et al. 2010; Liu  
87 et al. 2019). It is also unclear whether the high F abundances (>2000 ppm) in these silicic  
88 igneous rocks are inherited from their melt source or are introduced externally. For example,

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89 topaz rhyolites and Climax-type intrusions are often part of bimodal magma suites (Christiansen  
90 et al. 2007; Ludington and Plumlee 2009), and therefore it is possible that the halogens are  
91 introduced into silicic magmas by fluids exsolved from crystallization of mafic magmas that  
92 intrude the bases of upper crustal silicic magma bodies, or during deep crustal hybridization  
93 related to intrusion of mantle-derived magmas (e.g., Rosera et al. 2013; Dailey et al. 2018).

94 Disentangling the origins of the silicic melts and whether F and Cl are introduced  
95 externally is challenging because the hypothesized processes responsible for their origin extend  
96 from deep to shallow crust. Trace element geochemistry and Sr, Nd, and Pb isotopic  
97 compositions are one method to explore sources of magmatism, but they still require some  
98 knowledge about the composition of deep crust or the lithospheric mantle. Early isotopic studies  
99 that explored the origin of F-rich granites and rhyolites in the western United States (e.g., Farmer  
100 and DePaolo 1984; Stein and Crock 1990) were conducted before isotopic compositions of lower  
101 crustal xenolith suites in the region were fully characterized (e.g., Selverstone et al. 1999; Farmer  
102 et al. 2005). Furthermore, early studies suggested basaltic end members to magma systems were  
103 sourced from depleted mantle, whereas other studies document evidence that large portions of  
104 the western United States contain enriched lithospheric mantle (e.g., Coleman et al. 1992; Pettke  
105 et al. 2010; Sun et al. 2021). A more comprehensive examination of lower crustal xenolith data,  
106 as well as trace element and isotope geochemistry, is required to characterize sources of F-rich  
107 granites and rhyolites in the western United States.

108 The Colorado Mineral Belt is a narrow region of Late Cretaceous and younger  
109 (approximately 75 to 5 Ma) intrusions and mineral deposits that trends southwest-northeast  
110 across the southern Rocky Mountains of Colorado (Fig. 1a). This area provides an exceptional  
111 opportunity to test hypotheses related to the origin of F-rich, highly silicic magmas because it

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112 hosts numerous Climax-type deposits and porphyry Mo prospects (Fig. 1a), as well as topaz  
113 rhyolites and fluorite-bearing leucogranites. This study tests inferences regarding the origins of  
114 F-rich magmas by several avenues. To determine whether mafic lower crust is a viable source for  
115 F-rich silicic melts, we measure halogen abundances of the mafic lower crust beneath the  
116 Colorado Mineral Belt region as sampled by local kimberlite-borne crustal xenoliths. We  
117 combine this information with estimated F and Cl concentrations in melt from an F-rich granite  
118 porphyry in the bimodal Never Summer batholith in northern Colorado, which is outside of the  
119 Colorado Mineral Belt as traditionally defined (e.g., Tweto and Sims 1963; Chapin, 2012), but  
120 contains granite that is temporally and chemically similar to Mo-mineralizing intrusions in the  
121 Colorado Mineral Belt. We also use whole-rock major, trace, and isotopic compositions (Sr, Nd,  
122 and Pb) from fifty-six new samples to assess the sources of F-rich silicic magmatism in the  
123 region and to place them within the greater context of Cenozoic magmatism in Colorado.  
124 Ultimately, these data provide a clearer picture of the processes that aligned to set the stage for  
125 generating Climax-type mineral systems.

## 126 **GEOLOGICAL SETTING**

### 127 **Cretaceous to Oligocene magmatism in central and northern Colorado**

128 Cretaceous to Oligocene igneous rocks in central and northern Colorado have been a  
129 topic of interest for over a century because they represent magmatism that developed deep within  
130 a continental interior, and because of their close spatial relationship to numerous mining districts  
131 throughout the state (Emmons 1886; Tweto and Sims 1963; Bookstrom 1990; Chapin 2012;  
132 Rosera et al. 2021). The igneous rocks in the region have been classified by numerous schemes,  
133 such as compositional or spatial groups, especially in the Colorado Mineral Belt (e.g., Simmons  
134 and Hedge 1978; Mutschler et al. 1987; Stein and Crock 1990). However, we have chosen to

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135 broadly group them into temporal tectono-magmatic suites: 1) Laramide: those broadly  
136 associated with Laramide-aged compression (approximately 75 to 38 Ma; Chapin 2012); 2)  
137 transitional: rocks that originated during a post-orogenic period when compression relaxed, but  
138 before major tectonic extension (ca. 38 to 30 Ma; Zimmerer and McIntosh 2012); and 3)  
139 extensional: rocks generated after initiation of significant extension in the region, including  
140 development of the Rio Grande Rift (after ca. 30 Ma; Table 1; Chapin 2012; Landman and  
141 Flowers 2013).

142 **Laramide compression.**

143 Shallow subduction of a thickened segment of the Farallon plate is suggested to have  
144 caused widespread deformation and uplift in southwestern North America during the Laramide  
145 orogeny (Erslev 2005; Jones et al. 2011). Continental magma systems were also active in Idaho,  
146 southern Arizona and southwestern New Mexico between approximately 75 to 40 Ma. The  
147 region extending from central New Mexico through Wyoming largely lacked significant  
148 magmatism, with the exception of the Colorado Mineral Belt, which occupied this broad  
149 magmatic gap (Fig. 1a; Jones et al. 2011). Most Laramide-age rocks exposed in the Colorado  
150 Mineral Belt are shallow intrusions and porphyries; penecontemporaneous volcanic rocks are  
151 volumetrically minor and mostly preserved in uplift-flanking basins (Tweto 1975). Intrusive  
152 rocks include alkaline plutons (“monzonite suite” of Simmons and Hedge 1978) and a much  
153 more voluminous suite of calc-alkaline rocks (quartz monzonites to granodiorite). The calc-  
154 alkaline suite has higher initial  $^{87}\text{Sr}/^{86}\text{Sr}$  ( $^{87}\text{Sr}/^{86}\text{Sr}_i$ , 0.706 to 0.7089) and slightly lower initial  $\epsilon\text{Nd}$   
155 ( $\epsilon\text{Nd}_i$ , -5 to -10) compared to alkaline rocks ( $^{87}\text{Sr}/^{86}\text{Sr}_i$  mostly <0.706,  $\epsilon\text{Nd}_i$  -1 to -9; Table 1) .  
156 Both suites have broadly been interpreted to be sourced from mafic lower crust (Simmons and  
157 Hedge 1978; Stein and Crock 1990). In the northeastern Colorado Mineral Belt, alkaline magmas

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158 are associated with Au, Ag, U, W, and Te mineralization in the form of stockworks and veins  
159 (Bookstrom 1990). Calc-alkaline magma systems that intruded between approximately 45 to 38  
160 Ma are genetically associated with intermediate sulfidation veins, stockwork, and carbonate-  
161 replacement deposits that were mined mainly for Pb-Zn-Ag in districts such as Montezuma,  
162 Breckenridge and Leadville (Fig. 1c; Bookstrom 1990; Rosera et al. 2021).

163 **Transitional.**

164 A shift in the tectono-magmatic framework for the region occurred near the end of the  
165 Eocene. We use the eruption of the 37.3 Ma Wall Mountain Tuff to mark the beginning of this  
166 transitional period (Rosera et al. 2021), as the supereruption of this tuff signaled the start of an  
167 ignimbrite flare-up and deposition of voluminous volcanic material in the Southern Rocky  
168 Mountain volcanic field (McIntosh and Chapin 2004; Lipman 2000; Lipman 2007; Farmer et al.  
169 2008). The Mount Princeton batholith, as well as the Mount Aetna and Grizzly Peak calderas,  
170 formed early in this time frame (Fig. 1c; Fridrich et al. 1998; Mills and Coleman 2013). Low-  
171 grade porphyry Mo deposits also formed during this interval, including those at Turquoise Lake,  
172 Middle Mountain, and Halfmoon Creek (Van Loenen et al. 1989; Rosera et al. 2021; Fig. 1c).  
173 The flare-up has been suggested to have been triggered by exposing relatively cold  
174 metasomatized lithospheric mantle of the North American lithosphere to hot asthenosphere  
175 following fragmentation and foundering of the Farallon plate near the end of the Laramide  
176 (Dumitru et al. 1991; Humphreys et al. 2003; Farmer et al. 2008, 2020).

177 **Extensional.**

178 The tectonic environment evolved towards continental extension and initial Rio Grande  
179 rift development around approximately 30 to 25 Ma (Tweto 1979; Landman and Flowers 2013;  
180 Abbey and Niemi 2020). Broadly north-south elongated rift-related basins terminate near



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181 Leadville in the central Colorado Mineral Belt, but major Neogene faults concurrent with rift  
182 subsidence extend from the central Colorado Mineral Belt through the Never Summer range near  
183 the Wyoming-Colorado border (Tweto 1979; Fig. 1). Magmatism associated with extension  
184 began as early as ca. 31 to 28 Ma, when F-rich leucogranites intruded at Mount Antero and in the  
185 Never Summer mountains (Fig. 1b; Zimmerer and McIntosh 2012; Jacob et al. 2015; Rosera et  
186 al. 2021). Large Mo-F deposits at Climax and Henderson were assembled after extension began  
187 (Carten et al. 1993; Ludington and Plumlee 2009), along with eruption of numerous topaz  
188 rhyolites (Christiansen et al. 2007). High-silica rocks in this “extensional suite” are often  
189 interpreted to have been sourced from intermediate to felsic crust, owing to their generally low  
190  $\epsilon\text{Nd}_i$  and slightly elevated  $^{87}\text{Sr}/^{86}\text{Sr}_i$  relative to older igneous rocks from the Laramide and post-  
191 Laramide transition (Farmer and DePaolo 1984; Stein and Crock 1990; Table 1), although some  
192 high-silica magmas may have been derived from mafic lower crust (e.g., the Never Summer  
193 batholith; Jacob et al. 2015).

194         The Never Summer batholith is the most voluminous bimodal intrusive center that  
195 formed during early stages of extension in Colorado and is composed of the Mount Richthofen  
196 and Mount Cumulus plutons (Fig. 1b). The oldest intrusive rocks in the system are 29.21 Ma  
197 rhyolite porphyry dikes that intruded dominantly Paleozoic-Mesozoic sedimentary rocks in the  
198 northern portion of the igneous complex (Fig. 1b; Rosera et al. 2021). These silicic dikes contain  
199 quartz, K-feldspar, and plagioclase phenocrysts, and mafic phenocrysts that are largely altered to  
200 chlorite. The 28.98 to 28.74 Ma Mount Richthofen pluton is a compositionally heterogenous  
201 granodiorite with  $\text{SiO}_2$  contents ranging from 55 to 67 wt%, and it is interpreted to have formed  
202 by mixing of mantle-derived mafic melts with crustally-derived silicic melt (Jacob et al. 2015).  
203 Quartz, orthoclase, plagioclase, amphibole, and biotite are present throughout the Mount

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204 Richthofen pluton. Biotite tends to be the more dominant mafic mineral phase in the silicic  
205 portions of the pluton, and amphibole ( $\pm$ clinopyroxene) is more abundant in mafic portions of  
206 the pluton (Jacob et al. 2015). The younger Mount Cumulus pluton (28.17 Ma; Rosera et al.  
207 2021) is composed of a fluorite-bearing granite porphyry with major and trace element  
208 abundances comparable to that of the topaz rhyolites and intrusions in Climax-type deposits  
209 (Jacob et al. 2015). Biotite is the dominant mafic mineral phase in the Mount Cumulus pluton.  
210 High-precision U/Pb zircon geochronology indicates that the Mount Cumulus pluton was  
211 assembled rapidly, over 10's to 100's of k.y (Rosera et al. 2021). The Never Summer batholith is  
212 not significantly mineralized, but an area west of Mount Cumulus was prospected for porphyry  
213 Mo mineralization (Pearson et al. 1981). Molybdenite was also reported to occur in miarolitic  
214 cavities in the Mount Cumulus pluton, and rock chip surveys identified anomalously high  
215 abundances of Mo and Ag (Pearson et al. 1981).

#### 216 **Lower crustal mafic granulite xenoliths**

217 The State Line district of xenolith-bearing kimberlite dikes and diatremes located near the  
218 Colorado-Wyoming border intruded in two episodes during the Neoproterozoic and in the  
219 Devonian (Lester and Farmer 1998; Fig. 1a). Thus, these xenoliths sampled northern Colorado  
220 lithosphere before inferred Laramide aqueous metasomatism and magmatism. Xenoliths are  
221 predominantly mafic granulites and broadly include garnet-bearing and garnet-free groups.  
222 Garnet-free samples contain locally abundant amphibole (up to 30 modal %), plagioclase,  
223 clinopyroxene and orthopyroxene (Fig. S1), as well as trace to minor quartz, alkali feldspar,  
224 biotite, rutile, ilmenite, zircon, and apatite (Bradley 1985). The variable mineralogy of the  
225 xenoliths has been interpreted to reflect equilibration of compositionally similar rocks over a  
226 range of mid- to lower-crustal temperature and pressure conditions (Bradley and McCallum

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227 1984; Bradley 1985). Whole-rock major and trace element abundances, as well as H<sub>2</sub>O  
228 concentrations in nominally anhydrous minerals, indicate that the protolith for the State Line  
229 granulite suite was a hydrous (>1 wt% H<sub>2</sub>O) mafic igneous rock that intruded the deep crust  
230 (Farmer et al. 2005; Chin et al. 2020). Zircon U/Pb geochronology indicates primary igneous  
231 crystallization occurred at approximately 1720 Ma, but a subset of approximately 1360 Ma  
232 zircons showed sector zoning in cathodoluminescence images were interpreted as metamorphic  
233 overprint (Farmer et al. 2005). Garnet-pyroxene-plagioclase equilibria indicate that the garnet-  
234 bearing assemblage of xenoliths range from 1.1–1.2 GPa and 700–800 °C (Farmer et al. 2005).  
235 Thermobarometry estimates for the garnet-free suite of State Line mafic granulite xenoliths have  
236 not been previously published.

## 237 RESULTS

238 We use electron microprobe analyses (EMPA) of amphibole in garnet-absent two-  
239 pyroxene granulite xenoliths from the Sloan and Nix diatreme pipes from the State Line district  
240 to derive a first-order estimate of halogen contents of the garnet-free mafic lower crust from  
241 which they were derived (Fig. 1a). Fluorine and Cl occur in limited to trace quantities in apatite  
242 and nominally anhydrous minerals in the garnet-bearing granulite xenoliths because amphibole is  
243 absent. Consequently, no garnet-bearing xenoliths were used in this study. We also analyzed  
244 biotite in five samples from the Never Summer batholith: two from the Mount Richthofen pluton  
245 and three from the fluorite-bearing Mount Cumulus pluton (all samples used for biotite EMPA  
246 are from Jacob et al. [2015], except for sample NS17-05; Fig. 1b). We focused on biotite because  
247 amphibole is absent in the Mount Cumulus pluton, and biotite occurs in much greater abundance  
248 than amphibole in the silicic portions of the Mount Richthofen pluton (Jacob et al. 2015). The  
249 mafic portion of the Mount Richthofen pluton contains both amphibole and biotite, but

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250 amphibole from this sample were analyzed previously by Jacob et al. (2015; sample 10-KJ-MR-  
251 109). We used the same sample to measure biotite compositions.

252 We measured major and trace element abundances, as well as isotopic compositions of  
253 Sr, Pb, and Nd, on fifty-six whole-rock samples (further whole rock sample details and analytical  
254 methods are presented in the Supporting Information, Text S1 and S2). Samples were selected to:  
255 1) cover the temporal (Cretaceous to Oligocene) and spatial (northern and central Colorado  
256 Mineral Belt, Never Summer batholith) range of calc-alkaline magmatism (Fig. 1), and 2) add  
257 sampling density to previously studied systems (e.g., Montezuma pluton; Twin Lakes pluton;  
258 Farmer and DePaolo 1984; Stein and Crock 1990). This approach allows us to detect spatial and  
259 temporal variations in the sources of magmatism in order to better place the origins of F-rich  
260 magmatism into the greater context of regional magmatism. Laramide-aged (75 to 38 Ma)  
261 intrusions were collected from the Montezuma, Breckenridge, Alma, Leadville, and the Twin  
262 Lakes areas (Fig. 1). Samples corresponding to a transitional period between Laramide  
263 compression and Rio Grande rift extension (38 Ma to about 31 Ma) were collected from the  
264 Sawatch Range, and include the Mount Princeton batholith, Grizzly Peak caldera, and  
265 porphyritic intrusions related to Mo prospects (Turquoise Lake and Middle Mountain). Finally,  
266 we collected numerous samples from units that formed after the start of Rio Grande rift  
267 extension, including the Chalk Mountain rhyolite that is directly related to the Climax porphyry  
268 Mo deposit (Bookstrom 1989; Audétat 2015), Mount Antero leucogranites, and the silicic end  
269 member of the Never Summer batholith (Jacob et al. 2015).

270 **Electron microprobe analysis**

271 **Normalization procedures.**

272 Normalization of electron microprobe data for biotite and amphibole can be complicated  
273 because: 1) analyses cannot detect the incorporation of anhydrous  $O^{2-}$  into W crystallographic  
274 sites that are usually occupied by OH, F, and Cl; and 2) it does not provide information about  
275 oxidation states, which is problematic for major elements with multiple common oxidation  
276 states, most notably Fe (i.e.,  $Fe^{3+}$  or  $Fe^{2+}$ ). Normalization schemes for EMPA data from biotite  
277 and amphibole therefore require defining a set of assumptions or normalization scenarios to  
278 estimate  $O^{2-}$  and  $Fe^{3+}/Fe_{total}$ . We note that the normalization procedures of EMPA data for  
279 estimating  $Fe^{3+}$  and  $O^{2-}$  in biotite and amphibole are not ideal, but they are considered more  
280 accurate than assuming: 1) that  $Fe^{3+}/Fe_{total} = 0$  (e.g., Hawthorne et al. 2012), or that 2) there is no  
281 anhydrous O in the W crystallographic site for these minerals, especially those that have  
282 abundant Ti (e.g., Hawthorne et al. 2012).

283 The incorporation of anhydrous O into the W sites of amphibole and biotite has been  
284 interpreted to be due to deprotonation related to Ti substitution, such as  $(Fe^{2+}, Mg) + 2OH^- = Ti^{4+}$   
285  $+ 2O^{2-}$  (Oberti et al. 1992; Henry et al. 2005). Consequently, the maximum abundance of  
286 anhydrous O can be calculated as  $O^{2-} = 2Ti^{4+}$  (e.g., Hawthorne et al. 2012; Henry and Daigle  
287 2018). In this study, we use these Ti substitution formulations to estimate  $O^{2-}$  occupancy in the  
288 W crystallographic sites for biotite and amphibole, and we normalize biotite to  $11O + 2^W(OH, F,$   
289  $Cl, O)$  and amphibole to  $22O + 2^W(OH, F, Cl, O)$ .

290 The  $Fe^{3+}/Fe_{total}$  value for biotite and amphibole can be estimated based on independent  
291 lines of evidence, or by using a cation normalization scheme and adjusting  $Fe^{3+}$  to maintain  
292 electroneutrality. For amphibole analyses, we averaged  $Fe^{3+}/Fe_{total}$  based on the 13-cation (i.e., Si

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293 through Mg) and 15-cation (Si through Ca) schemes using software from Locock (2014). For  
294 biotite from the Never Summer batholith, we assumed  $\text{Fe}^{3+}/\text{Fe}_{\text{total}} = 0.22$  based on the presence of  
295 magnetite throughout the system (Guidotti and Dyar 1991; Jacob et al. 2015; Fig. S2), which is  
296 within uncertainty of  $\text{Fe}^{3+}/\text{Fe}_{\text{total}}$  measured independently in biotite from the Mount Princeton  
297 quartz monzonite and Mount Antero leucogranite in the central Colorado Mineral Belt  
298 ( $\text{Fe}^{3+}/\text{Fe}_{\text{total}} = 0.18 \pm 0.06$ ,  $1\sigma$ ; Toulmin and Hammarstrom 1990).

299 **Amphibole compositions and thermobarometry.**

300 Amphibole in the State Line mafic granulite xenoliths are mainly potassic-pargasite to  
301 potassic-magnesio-hastingsite. Sample SD2-LC76 contains potassic-fluoro-pargasite (*i.e.*, F >  
302 OH; Hawthorne et al. 2012). Fluorine concentrations for all amphibole analyzed range from 0.56  
303 to 1.38 wt% F, corresponding to 0.27 and 0.67 F atoms per formula unit (apfu; Fig. 2 and Table  
304 S1). Chlorine concentrations range from 0.45 to 0.73 wt% Cl (0.12 to 0.19 apfu Cl) and broadly  
305 show a positive correlation with F (Fig 2b). Samples NX4-LC2 and SD2-LC76 contain Ti-rich  
306 amphibole ( $0.5 > \text{Ti} > 0.3$  apfu; Hawthorne et al. 2012). The entire amphibole data set has  
307 relatively limited ranges of Mg# (0.53 to 0.60), Si (5.97 to 6.28 apfu), Ca (1.79 to 1.97 apfu), and  
308  $^{\text{IV}}\text{Al}$  (1.72 to 2.03 apfu; Fig. 2; Table S1).

309 The presence of ilmenite and rutile in the two-pyroxene mafic granulite xenoliths permits  
310 use of a recently calibrated Ti in calcium amphibole thermometer (Liao et al. 2021). This  
311 thermometer has a reported precision of approximately  $\pm 35$  °C (Liao et al. 2021). We compare  
312 those data to a graphical, semiquantitative  $\text{Al}_2\text{O}_3$ - $\text{TiO}_2$  amphibole thermobarometer calibrated for  
313 basaltic protoliths by Ernst and Liu (1998). Hence, we estimate both the *T* and *P* equilibration  
314 conditions for amphibole in the xenolith samples, but it should be noted that the *P* estimates are  
315 derived from a semiquantitative method for which fully propagating errors is difficult.

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316 Amphibole in sample SD2-LC78 is compositionally distinct from amphibole in the other  
317 xenoliths (Fig. 2), and application of the Ti in calcium amphibole thermometer yields  
318 equilibration temperatures from 805 to 870 °C (Fig 3). This temperature estimate is slightly  
319 lower, but overlapping, temperature estimates from the semiquantitative thermobarometer of  
320 Ernst and Liu (1998), which yielded equilibration conditions between approximately 820 and  
321 880 °C and approximately 0.5–0.75 GPa (Fig. 3). The remaining samples all contain amphibole  
322 with greater abundances of F, K, Ti, and <sup>IV</sup>Al than those in SD2-LC78 (Fig. 2), and they yielded  
323 equilibration temperatures from 873 to 941 °C using the Ti in calcium amphibole thermometer.  
324 These temperature estimates overlap those from the semiquantitative method, which yielded  
325 values ranging from 870 to 940 °C, and an estimated equilibration pressure of 0.6 to ~1.0 GPa.  
326 These pressure estimates are lower than peak metamorphic conditions calculated for the garnet-  
327 pyroxene-plagioclase xenoliths from the State Line district, but the temperature estimate is up to  
328 140 °C hotter than peak conditions estimated by Farmer et al. (2005; Fig. 3).

329 **Biotite from the Never Summer batholith.**

330 Biotite was analyzed in five samples from the Oligocene Never Summer batholith.  
331 Biotite from two samples of the Mount Richthofen pluton were analyzed: one from the mafic end  
332 member (~55 wt% SiO<sub>2</sub>) and another from a more silicic sample (~64 wt% SiO<sub>2</sub>; Jacob et al.  
333 2015). Biotite from these two samples have total Al and  $\text{Fe}^{2+}/(\text{Fe}^{2+} + \text{Mg})$  values comparable to  
334 biotite from the Mount Princeton and Mount Antero system in the Colorado Mineral Belt  
335 (Toulmin and Hammarstrom 1990; Fig. 4a). Biotite from the mafic portion of the Mount  
336 Richthofen pluton has slightly lower  $X_{\text{phlogopite}}$  (0.46–0.48;  $X_{\text{phlogopite}}$  in this study is defined as  
337  $\text{Mg}/[\text{Mg} + \text{VIAl} + \text{Cr} + \text{Ti} + \text{Fe}^{2+} + \text{Fe}^{3+} + \text{Mn}]$ ) than those analyzed from the silicic portion of the  
338 pluton ( $X_{\text{phlogopite}}$ : 0.49–0.52; Fig. 4b). The more silicic sample yielded biotite with higher

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339 log(F/OH) values (-0.83 to 0.74) and lower log(Cl/OH) (-1.88 to -1.68) than biotite from the  
340 mafic sample (log(F/OH): -1.29 to -1.24; log(Cl/OH): -1.49 to -1.43; Fig. 4b,d). Biotite from the  
341 silicic portion of the Mount Richthofen pluton have higher Ti abundances than those in the more  
342 mafic sample (Fig. 4c). Biotite equilibration temperatures were estimated with the Ti in biotite  
343 geothermometer of Henry et al. (2005) and yielded equilibration temperatures of approximately  
344 730°C for the mafic sample, and 765 to 780°C for the silicic portion of the Mount Richthofen  
345 pluton (Fig. 4c, Table S2).

346 Mount Cumulus samples contain two petrographically and compositionally distinct types  
347 of biotite. The first group occurs in samples 10-KJ-MC-94 and 10-KJ-MC-91 from the eastern  
348 and central portion of the pluton, respectively (Fig. 1b) and is commonly associated with fluorite  
349 and zircon (Fig. S2c). Compositionally, these biotite have very low  $X_{\text{phlogopite}}$  components (< 0.03  
350 apfu), high siderophyllite components ( $Al_{\text{total}}$ : 1.59 to 1.67 apfu), high log(F/OH) (0.13 to 0.35)  
351 and correspondingly high F concentrations (3.9 to 4.8 wt%; Fig. 4a,d, Table S2). Log(Cl/OH)  
352 values for these biotite span a relatively large range (-1.34 to -1.02; Fig. 4b). The low Mg  
353 abundances for these biotite are outside of the calibration range for the Ti in biotite  
354 geothermometer of Henry et al. (2005), and the Ti abundances are the lowest we observed the  
355 Never Summer batholith (0.09 to 0.11 apfu; Fig. 4c).

356 Biotite in a sample from the western portion of the pluton near Baker Pass (NS17-05) are  
357 petrographically and compositionally distinct from Fe-rich biotite observed in the other two  
358 Mount Cumulus samples. Fluorite was not observed in sample NS17-05, and some biotite in this  
359 sample encloses and cuts Fe-Ti oxide clusters (Fig. S2d). Compositionally, biotite in this sample  
360 contain higher  $X_{\text{phlogopite}}$  (0.27 to 0.30), log(Cl/OH) (-1.27 to -1.10) and lower  $Al_{\text{total}}$  (1.13 to 1.24  
361 apfu) and log(F/OH) (-0.29 to 0.06) than the other Mount Cumulus samples (Fig. 4). Titanium



362 abundances in these biotite are intermediate between the other Mount Cumulus samples and  
363 biotite from the Mount Richthofen pluton, corresponding to estimated equilibration temperatures  
364 from 639 to 653 °C (Henry et al. 2005; Fig. 4c).

## 365 **Whole-rock geochemistry of Cretaceous to Oligocene rocks**

### 366 **Laramide compressional suite.**

367 Analyzed samples of intrusive rocks associated with Laramide compression contain 48 to  
368 78 wt% SiO<sub>2</sub>, are mostly slightly peraluminous, and range from magnesian to ferroan (Fig. 5).  
369 Strontium abundances broadly follow a flat log-linear relationship with SiO<sub>2</sub> (>400 ppm Sr),  
370 except in the most silicic samples that are Sr-depleted (>75 wt% SiO<sub>2</sub>; Fig. 6a). Rubidium, Nb,  
371 Y, and Rb/Sr values for the Laramide suite are low relative to younger rocks from transitional  
372 and extensional suites (Fig. 6b-e), except for a modally layered mafic rock from the Twin Lakes  
373 pluton (SiO<sub>2</sub> = 48 wt%), which contains greater abundances of Nb and Y. The Laramide suite  
374 includes samples with high Sr/Y (>40) values, most notably those from the Twin Lakes pluton  
375 and intermediate intrusions from the Breckenridge, Alma, and Leadville mining districts (Fig.  
376 6f).

377 The Laramide suite of rocks show little correlation between initial isotopic compositions  
378 of Pb, Sr, and Nd relative to silica content (Figs. 7-9). Samples from this suite plot near or below  
379 the Stacey and Kramers (1975) two-stage Pb growth curve in <sup>206</sup>Pb/<sup>204</sup>Pb-<sup>207</sup>Pb/<sup>204</sup>Pb space, and  
380 slightly above the <sup>206</sup>Pb/<sup>204</sup>Pb-<sup>208</sup>Pb/<sup>204</sup>Pb curve (Fig. 7b,d). Initial <sup>206</sup>Pb/<sup>204</sup>Pb compositions of  
381 the Twin Lakes pluton are similar to intermediate intrusions from the Breckenridge, Alma, and  
382 Leadville areas, whereas samples from Montezuma are more enriched in <sup>206</sup>Pb/<sup>204</sup>Pb (Fig. 7;  
383 Table S5). Initial <sup>87</sup>Sr/<sup>86</sup>Sr is not correlated with SiO<sub>2</sub> and generally increases from <0.707 in the  
384 southwest (Twin Lakes pluton) to 0.708 to 0.712 in the northeast (Montezuma mining district;

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385 Fig. 8; Table S6). Initial  $\epsilon\text{Nd}_i$  values for the Twin lakes pluton and intrusions from the  
386 Montezuma district are similar and generally yield  $\epsilon\text{Nd}_i$  near -8, except for a late trachyandesite  
387 dike that cuts the Montezuma pluton, which has  $\epsilon\text{Nd}_i = -6.4$  (Fig. 9; Table S6). Samples from  
388 intrusions around the Breckenridge, Alma, and Leadville mining districts yielded  $\epsilon\text{Nd}_i$  values  
389 between -11.1 to -5.9, with the lowest value being from a xenolith entrained in the younger  
390 Chalk Mountain topaz rhyolite (CMR16-01x; Table S6), and the highest from the Swan  
391 Mountain sill complex near Breckenridge.

392 **Transitional suite.**

393 Igneous rocks from the transitional suite are metaluminous to slightly peraluminous and  
394 generally contain higher total alkali contents than those from the older Laramide suite (Fig. 5).  
395 Values of Rb/Sr, Nb, Y, and Nb for samples from the transitional suite are similar to the  
396 Laramide suite. Strontium and Sr/Y values are mostly lower than those from the Laramide suite  
397 (i.e., lower than those from the Twin Lakes pluton and Breckenridge, Alma, and Leadville areas;  
398 Fig. 6a, f).

399 Samples from the transitional suite generally yield wider ranges in initial isotopic  
400 compositions of Pb, Sr, and Nd than the Laramide suite. Samples related to porphyry Mo  
401 prospects at Turquoise Lake and Middle Mountain yield relatively high  $^{208}\text{Pb}/^{204}\text{Pb}_i$  values  
402 relative to  $^{206}\text{Pb}/^{204}\text{Pb}_i$  and plot near the continental lithospheric mantle (CLM) Pb growth curve  
403 for the western United States proposed by Pettke et al. 2010 (Fig. 7d). Initial Pb and Sr isotope  
404 compositions do not vary relative to silica content, but they do vary between individual magma  
405 systems (i.e., Grizzly Peak caldera system relative to the Mount Princeton batholith; Figs. 6, 7).  
406 Samples from the Grizzly Peak caldera complex yield a slight negative correlation between  $\epsilon\text{Nd}_i$   
407 and  $\text{SiO}_2$ , and contain the lowest  $\epsilon\text{Nd}_i$  in the central Colorado Mineral Belt (-14.6 to -8.5; Fig. 9;

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408 Table S6). Samples from the Mount Princeton batholith and porphyry Mo prospects at Turquoise  
409 Lake and Middle Mountain yield more radiogenic  $\epsilon\text{Nd}_i$  values (-10.5 to -8.7; Fig. 9).

410 **Extensional suite.**

411 Ten samples analyzed belong to the <31 Ma extension suite. One sample is a mafic  
412 volcanic rock located in the Rio Grande Rift ( $\text{SiO}_2 = 59$  wt%), and the other samples are all  
413 rhyolites or granites ( $\text{SiO}_2 > 71$  wt%). The silicic samples from the extensional suite are slightly  
414 peraluminous and ferroan (Fig. 5) and yield higher values of Nb, Y, and Rb/Sr than the majority  
415 of samples from the transitional or Laramide suites (Fig. 6). Strontium abundances are mostly  
416 <200 ppm, and Sr/Y values are correspondingly low (<10; Fig. 6a, f).

417 Samples from the extensional suite yielded initial widely variable Pb, Sr, and Nd isotopic  
418 compositions. The samples from the Never Summer igneous complex are more radiogenic in  
419 terms of Pb, Sr, and Nd (Figs. 7-9). Extensional suite rocks from the Colorado Mineral Belt have  
420 relatively unradiogenic Pb isotopic compositions (Fig. 7) and intermediate Sr and Nd isotopic  
421 compositions relative to the transitional and Laramide suites (Figs. 8, 9). The Chalk Mountain  
422 rhyolite erupted from the magma system that formed the Climax Mo deposit (Bookstrom et al.  
423 1987), and the sample we analyzed yielded Pb, Sr, and Nd compositions comparable to  
424 mineralizing intrusions from the mine (Figs. 7-9; Stein 1985; Stein and Crock 1990).

425 **DISCUSSION**

426 **Interpretation of amphibole data from mafic granulite xenoliths**

427 **Characterization of amphibole in the State Line xenoliths.**

428 Bradley (1985) concluded that amphibole in the two-pyroxene granulites from the State  
429 Line district was in equilibrium with surrounding minerals. We observed numerous K-feldspar

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430 and barite rims and veinlets in the State Line xenoliths but found no significant core-to-rim  
431 variations in K abundances across amphibole crystals. Petrography and backscattered electron  
432 imagery indicate that plagioclase is variably altered to sericite in some of the samples we  
433 analyzed (Fig. S1), but amphibole appears unaffected (Fig. 10). There is no obvious correlation  
434 between the alteration intensity of feldspar and amphibole compositions within the samples  
435 studied. Therefore, we conclude the amphibole crystals we analyzed are unaffected by alteration  
436 from the kimberlite host rock.

437         There is a weak positive correlation between Cl and  $\text{Fe}^{2+}/(\text{Fe}^{2+} + \text{Mg})$  in amphibole from  
438 the xenoliths (Fig. 2c). This is consistent with the Mg-Cl avoidance principle (or Fe-F avoidance)  
439 in amphiboles and micas (Rosenberg and Foit 1977; Munoz 1984). The avoidance of Mg-Cl and  
440 Fe-F is potentially related to variations in bonding strength, wherein Fe cations create weaker  
441 bonds with F in amphibole than Mg cations (Rosenberg and Foit 1977). As a result, the ability of  
442 amphibole or biotite to incorporate F and Cl into their crystal structure is, in part, a function of  
443 Mg and Fe concentrations (e.g., Munoz 1984). Although we observe slight negative correlation  
444 between Cl and Fe in amphibole from the State Line xenoliths, the relatively poor correlation  
445 suggests other factors beyond amphibole composition also influenced halogen abundances (e.g.,  
446 Morrison 1991; Aranovich and Safonov 2018).

447         **P-T equilibration conditions.**

448         Mafic lower crustal xenoliths from the Leucite Hills (Wyoming) and the Four Corners  
449 area of the Colorado plateau (Fig. 1a) yield equilibrium pressure estimates similar to the State  
450 Line xenoliths (0.8–1.2 GPa; Selverstone et al. 1999; Farmer et al. 2005). However, amphiboles  
451 from most State Line mafic granulite xenoliths have higher concentrations of K, Ti and <sup>IV</sup>Al than  
452 those observed in otherwise similar western U.S. xenoliths, corresponding to higher equilibration

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453 temperatures at a given pressure (Spear 1981; Fig. 2). The high Ti and <sup>IV</sup>Al abundances in  
454 amphibole from the State Line xenoliths are similar to fluorian pargasite in the ultrahigh  
455 temperature (UHT) Highland Complex in Sri Lanka (Fig. 2e; Sajeev et al. 2009), and Ti in  
456 calcium amphibole thermometry also indicate that amphibole in some of the State Line mafic  
457 granulite xenolith samples equilibrated at temperatures greater than 900 °C (Fig. 3). Fluorine-  
458 rich pargasite is stable at more extreme *P-T* conditions than its F-poor counterparts because  
459 substitution of F into the hydroxyl site expands pargasite's thermal stability field (up to 950 °C at  
460 1.0 GPa; Holloway and Ford 1975; Tsunogae et al. 2003; Sajeev et al. 2009). The high F and Cl  
461 concentrations observed in State Line amphibole could therefore stabilize mafic lower crust and  
462 allow it to equilibrate at temperatures in excess of 800°C without significant dehydration or  
463 partial melting.

464       Altogether, the observations of granulite textures in the mafic lower crustal xenoliths  
465 (Bradley 1985; Farmer et al. 2005; Chin et al. 2020; Fig. S1), as well as amphibole compositions  
466 and Ti in calcium amphibole data presented here, strongly suggest that portions of the mafic  
467 lower crust of northern Colorado were subjected to high or UHT (>900 °C) peak metamorphic  
468 conditions. The temperature estimates for the garnet-absent mafic crust are greater than those  
469 from the deeper-sourced garnet-bearing xenoliths measured by Farmer et al. (2005; Fig. 3),  
470 suggesting that either some high-T process overprinted the shallower garnet-absent lower crust,  
471 or perhaps the lowermost garnet-bearing crust formed by a retrograde reaction beginning with  
472 amphibole-bearing garnet-absent granulite. The latter model seems unlikely, because  
473 disequilibrium textures observed in the garnet-bearing granulite xenoliths (Bradley and  
474 McCallum 1984; Farmer et al. 2005) document the growth of garnet at the expense of  
475 plagioclase and orthopyroxene, and not amphibole. Furthermore, the presence or absence of

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476 garnet in the State Line mafic lower crustal xenoliths has been interpreted to be a magmatic  
477 feature related to crystallization of basaltic melts at varying crustal depths (Bradley and  
478 McCallum 1984; Bradley 1985). Chin et al. (2020) suggested that the garnet-bearing mafic lower  
479 crust in northern Colorado represent cumulates that fractionated from primitive hydrous melts  
480 during the Proterozoic. Hence, the higher T recorded in shallower rocks garnet-absent mafic  
481 granulite xenoliths could be due to a different UHT metamorphism event that did not affect the  
482 lowermost, garnet-bearing mafic lower crust. These observations have important implications for  
483 origin of F-rich amphibole, and the apparent UHT conditions of their formation.

#### 484 **Origin of F-rich amphibole and ultrahigh temperature metamorphism**

485 The F- and Cl-rich composition of the State Line amphiboles could originate from  
486 halogen enrichment processes, such as: 1) concentration of halogens in residuum following  
487 partial melting, 2) a F- and Cl-rich protolith, and/or 3) an external addition of F- and Cl-rich  
488 fluid. Partial melting experiments demonstrate that F abundance in phlogopite will increase when  
489 it is in equilibrium with a melt phase (Dooley and Patiño Douce 1996). This predicts that as the  
490 modal abundance of hydrous phases decreases during melting, residual phases will become  
491 enriched in F. In the case of the State Line xenoliths, this model predicts that the modal  
492 abundance of amphibole would decrease with partial melting, and that the remaining amphibole  
493 would re-equilibrate to greater F abundances. However, such a trend is not apparent in the State  
494 Line xenoliths; the sample with the highest modal abundance of amphibole (20 modal %) has F-  
495 rich amphibole that contains approximately 1 wt.% F (Fig. S3). Furthermore, Cl has a slightly  
496 larger ionic radius than F and should behave more incompatibly during partial melting, creating a  
497 negative relationship between F and Cl concentrations; in contrast, our data show a clear positive

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498 trend (Fig. 2b). We conclude that a halogen-enriched residuum is the least likely explanation for  
499 the origin of F-rich amphibole in the State Line xenoliths.

500 The State Line xenoliths are interpreted to have been Paleoproterozoic arc-derived  
501 basaltic melts that intruded into the deep crust and underwent fractional crystallization (Bradley  
502 and McCallum 1984; Farmer et al. 2005; Chin et al. 2020) and therefore could have retained  
503 their igneous compositions. However, experiments run at deep crustal  $P$ - $T$  conditions yield  
504 amphibole-melt partition coefficients that would require the State Line amphiboles to have  
505 equilibrated with a melt with  $>1.1$  wt% Cl (Hauri et al. 2006; Dalou et al. 2012). The high F and  
506 Cl contents could be related to some combination of fractional crystallization or inheritance from  
507 the melt source. Arc-related basalts with relatively high F ( $>500$  ppm) and Cl (1000 to 10000  
508 ppm) abundances in olivine-hosted melt inclusions have been observed, and numerical modeling  
509 indicates the highest halogen abundances are derived from partial melting of eclogitic facies of  
510 altered ocean crust in the subducting slab (Van den Bleeken and Koga 2015). Therefore, the  
511 most likely scenario for generating arc basalts with  $>1$  wt% Cl involves slab melts derived from  
512 a mixture of altered oceanic crust and metasedimentary sources (Van den Bleeken and Koga  
513 2015). Farmer et al. (2005) noted the presence of inherited Archean (3.2 Ga) zircon in the State  
514 Line lower crust xenoliths, and they could be evidence for sediment derived from Archean craton  
515 during the Paleoproterozoic assembly of continental crust in Colorado, but more work is needed  
516 to test this interpretation.

517 An alternative explanation for the presence of F-rich amphibole that equilibrated at UHT  
518 grades is that the amphibole in the State Line xenoliths formed by interaction with an externally  
519 derived fluid. Hot, F-rich fluids are hypothesized to be the source for F-rich pargasite in mafic  
520 granulites in east Antarctica (Tsunogae et al. 2003). Amphibole from the State Line xenoliths

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521 have lower F/Cl ratios, as well as higher Cl and  $\text{Fe}^{2+}/(\text{Fe}^{2+} + \text{Mg})$  ratios, than those described by  
522 Tsunogae et al. (2003). These observations suggest that hot, externally derived fluids with high  
523 abundances of Fe, F, and Cl could have equilibrated with the State Line xenoliths. The source of  
524 the fluids is unclear, but they may have been exsolved from deeper crystallizing mafic magmas  
525 during prolonged episodic intrusion of mafic magma into the lower crust in the Paleoproterozoic  
526 (1.8 to 1.6 Ga), or during a later mafic underplating or intraplating event (e.g., 1.4 Ga; Farmer et  
527 al. 2005; Keller et al. 2005; Jones et al. 2010). Deep mafic intraplating and fluid exsolution  
528 might also explain why the xenolith sample SD2-LC78 has lower halogen concentrations and  $T$   
529 estimates, because this xenolith originated from a shallower portion of the crust than the other  
530 samples (Fig. 3), and potentially away from hot, F rich fluids that could have equilibrated with  
531 deeper xenoliths. Likewise, intraplating mafic magmas may have pooled near the transition from  
532 the garnet-bearing lowermost crust to the structurally higher garnet-absent mafic lower crust due  
533 to the density contrast between the two rheologies. This model could explain why UHT  
534 metamorphism is not observed in the xenolith samples from the lowermost crust (Farmer et al.  
535 2005). However, more detailed work is required to fully explore if external fluids are a viable  
536 origin for the fluorian pargasite from the State Line xenoliths.

### 537 **Estimation of F and Cl abundances in garnet-free mafic lower crust.**

538 By combining our amphibole EMPA data with modal abundance data for the same  
539 xenolith samples, we can derive a first-order estimate for F and Cl abundances in the mafic lower  
540 crust of northern Colorado. Our estimates are minima, as we do not account for halogens in  
541 nominally anhydrous minerals, nor did we analyze the trace apatite (average 2 modal %) present  
542 in the State Line mafic granulites (Bradley 1985). Assuming apatite in the xenoliths have 2 wt%  
543 F would add 0.04 wt% F to our whole-rock estimates, but because we did not analyze apatite we



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544 use a more conservative value based only on the amphibole compositions. Using this approach,  
545 we estimate the analyzed xenoliths contain at least 0.04–0.20 wt% F and 0.02–0.10 wt% Cl.  
546 Average mafic lower crust contains 0.057 and 0.025 wt% F and Cl, respectively (Rudnick and  
547 Gao 2003). Our estimates indicate the garnet-free mafic lower crust in northern Colorado could  
548 be enriched in F and Cl by 3.5 to 4 times that of average mafic lower crust. Sample SD2-LC78,  
549 which we interpret as coming from a shallower source than the other xenoliths, yields the lowest  
550 F estimate (0.04 wt%), which is comparable to average mafic lower crust (Rudnick and Gao  
551 2003). We tentatively interpret these data to reflect variable F and Cl abundances within the  
552 garnet-free mafic lower crust, potentially with lower values in shallower rocks.

553 **The origin of Cretaceous to Oligocene magmas: Insights from geochemical data**

554 The observations above demonstrate that portions of garnet-free mafic lower crust in the  
555 southern Rocky Mountains were enriched in F and Cl relative to average crustal abundances, that  
556 this enrichment occurred before the xenoliths were entrained in Devonian kimberlites, and that  
557 the deep crust in the region may have undergone high- to ultrahigh temperature granulite  
558 metamorphism. Seismic refraction studies suggest that mafic metamorphic rocks are present in  
559 the lower crust throughout the southern Rocky Mountains (e.g., Snelson et al. 2005), and  
560 therefore the mafic lower crust could be a source for silicic melts generated during Laramide  
561 compression, post-Laramide transition, and younger continental extension. In the following, we  
562 use geochemical data to revisit the discussion of silicic magma sources in the Never Summer  
563 batholith and Colorado Mineral Belt, and we put the generation of F-rich silicic magmas into a  
564 broader regional context. First, we examine biotite geochemical data from the Never Summer  
565 batholith, then we consider new trace element geochemistry, Pb, Sr, and Nd isotopic  
566 compositions, as well as previously published data, including information from Climax-type Mo

567 deposits (Farmer and DePaolo 1984; Stein 1985; Stein and Crock 1990) and recent Ca isotopic  
568 investigations (Mills et al. 2018).

569 **Interpretation of Never Summer batholith biotite data.**

570 Biotite across the Never Summer batholith have a large range of  $X_{\text{phlogopite}}$ , but there is  
571 little variation within individual samples (Fig. 4). There is negative correlation between Mg/(Mg  
572 + Fe<sub>total</sub>) and Ti abundances that could, in part, correspond to different biotite equilibration  
573 temperatures across the batholith (Fig. 4c). Titanium in biotite temperature estimates for the  
574 Mount Richthofen pluton overlap temperature estimates calculated by Jacob et al. (2015) with  
575 the two-feldspar thermometer (Putirka 2008; approximately 730 to 780 °C in this study versus  
576 700 to 780 °C from Jacob et al. 2015). Both log(F/OH) and log(Cl/OH) are negatively correlated  
577 with  $X_{\text{phlogopite}}$  across the entire dataset (Fig. 4b,d). A negative correlation between F abundances  
578 and  $X_{\text{phlogopite}}$  goes against commonly observed Mg-F association (or Fe-F avoidance) in biotite  
579 (e.g., Munoz 1984). Biotite from two Mount Cumulus samples have extremely low  $X_{\text{phlogopite}}$   
580 components and high F relative to the sample from the western portion of the pluton (Fig. 4d).  
581 The whole rock geochemistry of these two samples also yielded comparably high FeO<sub>tot</sub>/(FeO<sub>tot</sub>  
582 + MgO) and distinctly low abundances of Sr (Fig. 5, 6a; Jacob et al. 2015).

583 The Mount Cumulus granite has been interpreted as an intrusive equivalent to topaz  
584 rhyolites found throughout the western United States (Christiansen et al. 2007; Jacob et al.  
585 2015). Biotite in the low Sr, high Fe/Mg Mount Cumulus samples are comparable to those from  
586 the Honeycomb Hills topaz rhyolite in Utah, with similar Fe/Mg, total Al, log(F/OH) and  
587 log(Cl/OH) values (Fig. 4c; Congdon and Nash 1991; Byrd and Nash 1993). Although the  
588 Honeycomb Hills biotite are depleted in Mg, they were erupted in a melt with 1–3 wt% F, which  
589 also goes against the concept of the Fe-F avoidance principle (Byrd and Nash 1993). These Fe-

590 rich biotite in Mount Cumulus pluton have relatively low Ti abundances similar to biotite from  
591 the Mount Antero leucogranite, which could indicate they formed at relatively cool temperatures  
592 (Fig. 4c), but their low  $X_{\text{phlogopite}}$  values are below the Ti in biotite calibrations presented by  
593 Henry et al. (2005). Biotite from the western Mount Cumulus sample (NS17-05) yield  
594 equilibration temperatures between 639 and 653 °C. This range is comparable to Ti in biotite  
595 temperature estimates for the Hideaway Park and Chalk Mountain topaz rhyolites (Fig. 4c), both  
596 of which are located south of the Never Summer complex in the Colorado Mineral Belt and are  
597 associated with the Henderson and Climax Mo deposits, respectively (Fig. 1; e.g., Mercer et al.  
598 2015; Audétat 2015).

#### 599 **Trace element signature for anatexis of garnet-absent lower crust**

600 Biotite from the Mount Cumulus pluton suggest that it formed from high silica magmas  
601 comparable to those that result in topaz rhyolites or F-rich porphyry Mo deposits. This is  
602 intriguing because trace element modeling of partial batch melting indicates that silicic magmas  
603 in the Never Summer batholith are sourced from garnet-absent mafic lower crust (Jacob et al.  
604 2015), which challenges previous suggestions that the Cenozoic F-rich melts in the study area  
605 are from ancient felsic sources (Farmer and DePaolo 1984; Stein 1985; Stein and Crock 1990).  
606 Hence, geochemical data from the Never Summer batholith provide insight about deep mafic  
607 crustal anatexis and are a good starting point to reassess the role of the mafic lower crust in  
608 generating melts to the south in the Colorado Mineral Belt.

609 The silicic melts in the Never Summer magma system have low Sr/Y (<20) and high  
610 Rb/Sr, Nb, and Y (Fig. 6). The protolith for mafic granulite lower crust xenoliths is interpreted to  
611 be hydrous (>1 wt% H<sub>2</sub>O) arc-related basaltic melts (Farmer et al. 2005; Chin et al., 2020), and  
612 therefore the Sr/Y ratio of silicic melts derived from deep mafic crustal anatexis is likely

*For submission to American Mineralogist*

613 controlled by the stability of amphibole ( $\pm$  garnet) in the source (Jacob et al., 2015). The low  
614 Sr/Y, high Nb and Y trace element signature is also observed in F-rich leucogranites from Mount  
615 Antero, as well as high silica rocks related to Mo mineralization at Climax and Henderson (Fig.  
616 6). These observations indicate that amphibole in garnet-absent mafic lower crust could be  
617 controlling Sr/Y values in silicic magma systems throughout the area (i.e., Never Summer  
618 batholith and the Colorado Mineral Belt), and that breakdown of amphibole-bearing mafic lower  
619 crust might be a critical process for generating F-rich leucogranites.

620 Previous geochemical investigations in the Colorado Mineral Belt suggested that the  
621 early, F-poor Laramide suite was derived in part by mafic crustal anatexis (Stein and Crock  
622 1990). This suite has notably higher Sr/Y values, and in some magma systems Sr/Y is greater  
623 than 40 (e.g., Twin Lakes pluton, Breckenridge-Alma-Leadville area; Fig. 6). These samples  
624 have correspondingly low Nb, Y, and Rb/Sr. We hypothesize that the high Sr/Y compositions are  
625 the product of partial melts generated at deep crustal conditions (i.e., high pressure) where  
626 amphibole (or garnet) are stable phases, but plagioclase was unstable. Thus, if the Laramide suite  
627 involved anatexis of deep mafic crust, then we suggest the F-rich amphibole such as those we  
628 observed in the State Line xenoliths remained stable.

629 The trace element data support secular changes in the style of melting from Laramide to  
630 Oligocene magmatism. However, Sr/Y values can be influenced by involvement from felsic  
631 material (Moyen 2009), or by differentiation of deep hydrous mafic melts beneath thick crust  
632 (Chiaradia 2015; Lee and Tang 2020). That is, the secular changes in trace element abundances  
633 or their ratios is also a function of the starting source composition, and previous work in the  
634 Colorado Mineral Belt has argued for contributions from ancient felsic crust (Farmer and  
635 DePaolo 1984; Stein and Crock 1990), or ancient metasomatized lithospheric mantle (Pettke et

636 al. 2010). Disentangling what sources may have contributed to these magmas is further  
637 considered by evaluating spatial and secular patterns in Pb, Sr, and Nd isotopic data from the  
638 region.

639 **Regional characteristics of Pb, Sr, and Nd isotopic data.**

640 The initial Pb isotope data from the Colorado Mineral Belt and Never Summer batholith  
641 mostly plot between growth curves for average crust (Stacey and Kramers 1975) and the  
642 continental lithospheric mantle (CLM) model for the western United States (Pettke et al. 2010;  
643 Fig. 7). Some samples contain Pb isotope compositions that plot above growth models in  
644  $^{208}\text{Pb}/^{204}\text{Pb} - ^{206}\text{Pb}/^{204}\text{Pb}$  space, most notably intrusions from the Henderson porphyry Mo  
645 deposit (Stein 1985). The Henderson intrusions likely incorporated a small fraction (<5%) of Pb  
646 from unusually Th-rich host rock with high  $^{208}\text{Pb}/^{204}\text{Pb}$  (Silver Plume Granite; Stein 1985).  
647 Available Pb isotopic data for other upper crustal Proterozoic rocks in the region generally have  
648 variable and high  $^{208}\text{Pb}/^{204}\text{Pb}_{35\text{ Ma}}$ , which plot above the CLM growth curve (Frazer 2017; Fig.  
649 7d). If upper crustal contamination was significant, we would expect to see higher, and more  
650 variable,  $^{208}\text{Pb}/^{204}\text{Pb}_i$  values in the samples we analyzed. The Pb isotopic data therefore do not  
651 support significant upper crustal contamination. Instead, most of the magmatism in the area can  
652 be described either as: 1) a mixture between a low U/Th end member and another end member  
653 with average crustal Pb (i.e., Stacey and Kramers two-stage Pb), or 2) a product of reservoirs  
654 where Th/Pb and U/Pb were variably fractionated in the Proterozoic (approximately 1.8 to 1.7  
655 Ga; Stein 1985; Pettke et al. 2010).

656 A compilation of new and existing Sr and Nd isotopic data for Cretaceous and younger  
657 igneous rocks and Proterozoic country rocks indicates that ancient mafic lower crust has isotopic  
658 compositions similar to many of the magma systems analyzed in this study (Fig. 11). Cretaceous

*For submission to American Mineralogist*

659 basalts from the Windy Gap formation are inferred to have been derived from enriched  
660 lithospheric mantle and also have Sr-Nd isotopic compositions that overlap with the mafic lower  
661 crust (Bailey 2010; Fig. 11). Based on these Sr-Nd isotopic data, the samples we analyzed from  
662 the Colorado Mineral Belt could have either: (1) large contributions from ancient mafic lower  
663 crust, or (2) a two-component mixture of enriched mantle and ancient felsic crust sources.  
664 However, deciphering the exact contributions of mantle and crustal sources with these data alone  
665 can be ambiguous (e.g., Pettke et al. 2010; Mills et al. 2018). For example, the Mount Princeton  
666 batholith and Mount Antero leucogranite have Nd isotopic compositions that are comparable to  
667 mafic xenoliths in the region (Fig. 11), but radiogenic Ca isotopes indicate that only the Mount  
668 Princeton batholith contains significant ancient felsic crustal material (Mills et al. 2018). The F-  
669 rich Mount Antero leucogranite has relatively unradiogenic Ca isotopes, consistent with  
670 derivation from a low K/Ca (e.g., mafic lower crust) or juvenile mantle-derived source (Mills et  
671 al., 2018). Porphyries related to Mo mineralization in the Colorado Mineral Belt have a narrow  
672 range of  $\epsilon\text{Nd}_i$  (approximately -8 to -10) that overlaps the Mount Antero leucogranite (Fig. 11),  
673 which is consistent with derivation from a similar mafic crustal source. The silicic intrusions  
674 from the Never Summer complex have  $\epsilon\text{Nd}_i$  values much higher than those of ancient felsic  
675 country rocks (Fig. 11), and therefore likely incorporated only minor amounts of ancient felsic  
676 crustal material (Jacob et al. 2015). The Grizzly Peak caldera complex has the lowest  $\epsilon\text{Nd}_i$  values  
677 ( $< -12$ ), and some of the post-caldera intrusions overlap ancient felsic country rocks (Fig. 11a).  
678 Calcium isotope data corroborate incorporation of significant ancient felsic material into the  
679 Grizzly Peak Tuff (Mills et al. 2018). The Grizzly Peak caldera therefore has the strongest  
680 evidence for incorporation of large amounts of ancient felsic crust relative to many other  
681 Laramide and younger magma systems in the central Colorado Mineral Belt.

*For submission to American Mineralogist*

682 Magma systems in the northern Colorado Mineral Belt and Never Summer mountains  
683 generally have more radiogenic Pb and less radiogenic Nd compositions than those in the central  
684 Colorado Mineral Belt (Figs. 7, 9; Stein 1985; Stein and Crock 1990). There is a lack of  
685 correlation between silica content and the isotopic composition of Nd and Pb across the entire  
686 dataset (i.e., Never Summers, northern and central Colorado Mineral Belt), so the spatial  
687 variations are not solely due to variable contributions from laterally homogenous isotopic  
688 reservoirs of ancient crust and mantle end members (Figs. 7, 9). Instead, these data support  
689 inferences for isotopically heterogeneous source material in central and northern Colorado,  
690 perhaps with varying degrees of ancient granulitization of the deep crust that variably  
691 fractionated parent-daughter elements (Stein and Crock 1990) or with multiple episodes of  
692 metasomatism of the upper mantle during (i.e., during Proterozoic subduction and again during  
693 Laramide flat slab subduction; Pettke et al. 2010; Sun et al. 2021). Each of these processes can  
694 modify the parent-daughter ratios of isotopic systems or modify the isotopic compositions  
695 without significantly changing the major element composition of the reservoir. The lateral  
696 heterogeneities observed here and elsewhere (e.g., Stein and Crock 1990) are due to variations in  
697 Rb-Sr, Sm-Nd, and U-Th-Pb isotopic systems depending on their metamorphic and metasomatic  
698 histories.

699 **Secular variations in magmatism.**

700 Secular variations in Pb isotopic compositions support the hypothesis that F-rich  
701 leucogranites and topaz rhyolites are derived from different sources than their precursor  
702 intrusions. For example,  $^{206}\text{Pb}/^{204}\text{Pb}_i$  is observed to decrease through time in a given area,  
703 including (Fig. 12): 1) the 39.7 Ma pre-Mo white dikes from the Alma district relative to the 33  
704 to 25 Ma intrusions from the Climax Mo mine and the 26.2 Ma Chalk Mountain rhyolite

*For submission to American Mineralogist*

705 (Bookstrom 1989; Rosera et al. 2021); 2) the Paleocene phase of the Twin Lakes pluton relative  
706 to the Eocene phase and rhyolite porphyry from the Middle Mountain porphyry Mo deposit; 3)  
707 the Mount Princeton quartz monzonites that are intruded by the Mount Antero leucogranites; and  
708 4) early silicic dikes from the Never Summer complex contain more radiogenic Pb than the  
709 younger Mount Cumulus leucogranite. These data corroborate previous observations that young  
710 leucogranites and topaz rhyolites tend to have the least radiogenic Pb isotope compositions in  
711 any given area (Stein 1985).

712         The progression of magmatism from Laramide compression through extension/Rio  
713 Grande rifting can therefore be described as deriving from isotopically variable reservoirs in both  
714 space and time. Leucogranites across Colorado that are compositionally similar to each other in  
715 terms of trace elements were generated from isotopically distinct lithospheric domains, yet they  
716 follow a similar temporal-isotopic pattern relative to their precursor intrusions. For example, the  
717 Pb (and Nd) isotopic compositions of the Mount Cumulus leucogranites do not overlap with the  
718 Mount Antero leucogranites or Chalk Mountain rhyolite, but all of these high-silica rocks have  
719 slightly lower  $^{206}\text{Pb}/^{204}\text{Pb}_i$  than the intrusions that precede them. A source with low time-  
720 integrated U/Pb (and U/Th) is thus the likeliest one for F-rich silicic melts, but the degree of  
721 retardation of uranogenic Pb varies regionally. It is unclear if the spatial variation was inherited  
722 from ancient crust, possibly due to variability in proposed UHT metamorphism and F-  
723 enrichment, or if the spatial variation is related to long-term modification of the lower crust by  
724 an influx of melts derived from an enriched mantle source with laterally heterogeneous Pb  
725 isotope compositions.



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726           **The Grizzly Peak caldera and the role of ancient felsic crust.**

727           Radiogenic isotopic data, including analyses from Mo-mineralizing intrusions in the  
728 Colorado Mineral Belt, have been used to argue that melting of ancient felsic crust is essential  
729 for generating the F-rich high silica magmas that formed topaz rhyolites and porphyry Mo  
730 systems (Farmer and DePaolo 1984; Stein and Crock 1990), but data presented here and  
731 elsewhere (Mills et al. 2018) suggest magmas with the clearest evidence for significant  
732 contributions from felsic crust are not necessarily F-rich rocks. For example, the ~34.3 Ma  
733 Grizzly Peak Tuff (McIntosh and Chapin 2004) and resurgent plutons in its caldera have Nd  
734 isotopic compositions that overlap with felsic country rocks (Fig. 11), and a significant  
735 component of felsic Proterozoic crust in the tuff is supported by excess  $^{40}\text{Ca}$  (Mills et al. 2018).  
736 Fridrich et al. (1991, 1998) observed discontinuous rings of hydrothermally altered dikes and  
737 stocks outside of the Grizzly Peak caldera, some of which have been prospected for porphyry  
738 Mo, but our new data suggest these intrusions are not directly related to the Grizzly Peak system.  
739 A 34.0 Ma hydrothermally altered dike east of the caldera yields  $\epsilon\text{Nd}_i = -8.7$ , whereas the  
740 Grizzly Peak tuff and intrusions have  $\epsilon\text{Nd}_i \approx -12.5$  (Table S6; Figs. 9, 11; Johnson and Fridrich  
741 1990). Likewise, the 36.45 Ma low-grade Mo-F porphyry system at Middle Mountain, southeast  
742 of the caldera, predates the eruption of the tuff (Rosera et al. 2021), and the Middle Mountain  
743 porphyry has more radiogenic Nd than the Grizzly Peak system (Fig. 11b). Therefore, the  
744 Grizzly Peak caldera contains the rocks in the Colorado Mineral Belt with the lowest  $\epsilon\text{Nd}_i$  and  
745 perhaps the greatest contribution of ancient felsic sources, but the caldera rocks do not appear to  
746 be similar to topaz rhyolites, nor has deep exploration identified any significant Mo mineralizing  
747 intrusions related to the caldera-forming magma system. It is possible that such a system could  
748 occur at depth, perhaps below volcanic cover within the caldera, but there is currently no

749 evidence that the ancient felsic crustal-derived Grizzly Peak magmas generated any topaz  
750 rhyolites or Mo mineralization.

### 751 **Tracing F and Cl from mafic lower crustal anatexis to upper crustal silicic magma systems**

752 The biotite data presented in this study corroborate interpretations that the Mount  
753 Cumulus pluton is an intrusive analogue to topaz rhyolites (e.g., Jacob et al. 2015), including  
754 those erupted from the Climax-type deposits in the Colorado Mineral Belt. Melt inclusions and  
755 analysis of volcanic glass indicate that topaz rhyolites typically have  $>2000$  ppm F and  $F/Cl > 3$   
756 (Christiansen et al., 2007; Audéat 2015; Mercer et al. 2015), but reliable F and Cl estimates for  
757 their intrusive equivalents are scarce because whole-rock data cannot account for volatile loss. In  
758 the following section, we use recently calibrated biotite-melt exchange equations to estimate  
759 melt abundances of F and Cl in the Never Summer batholith, and we compare those data to batch  
760 melting models derived from analyses of garnet-free mafic lower crustal xenoliths. These models  
761 require various assumptions about: (1) how F/OH and Cl/OH partition between biotite and silicic  
762 melt, (2) the saturation concentration and speciation of water in melts based on whole rock data,  
763 (3) the behavior of volatiles (i.e., F and Cl) during open-system crystallization, and (4) the  
764 degree of post-solidification re-equilibration. We emphasize that these models are first-order  
765 approximations of the magmatic compositions of F and Cl in silicic intrusions, but by  
766 approaching the question from the source (i.e., xenolith data) to the sink (i.e., Never Summer  
767 batholith), we hope to gain further perspective about how deep crustal anatexis might lead to  
768 assembly of F-rich magma systems. To better validate these models, we compare our estimated F  
769 and Cl abundances in the Never Summer batholith to melt inclusion data from the Colorado  
770 Mineral Belt, as well as F and Cl estimates from average whole-rock and biotite data from the  
771 Mount Princeton batholith and Mount Antero leucogranites (Toulmin and Hammarstrom 1990).

*For submission to American Mineralogist*

772 **Estimating F and Cl in silicic melt from biotite and whole rock data.**

773 Melt F and Cl abundances were estimated following the recent formulations of Zhang et  
774 al. (2022). Their models calculate biotite-melt partition coefficients for F/OH and Cl/OH  
775 ( $Kd_{F/OH}^{\text{biotite-melt}}$  and  $Kd_{Cl/OH}^{\text{biotite-melt}}$ , respectively; Table S2). The exchange partition coefficients  
776 for each are function of  $X_{\text{phlogopite}}$  (which they refer to as  $X_{Mg}$ ) and Ti in biotite and melt F or Cl  
777 can be calculated by assuming water saturation and speciation models. Zhang et al. (2022) use  
778 the whole rock major element data and formulations of Moore et al. (1998) to model  $H_2O_{\text{melt}}$ , and  
779 the Behrens (2020) water speciation model. Modeled  $H_2O_{\text{melt}}$  values are sensitive to estimated  
780 pressure, and to a lesser degree, temperature. Aluminum in amphibole geobarometry from the  
781 Mount Richthofen pluton yielded an average pressure estimate of 100 MPa (Jacob et al. 2015),  
782 which overlaps pressure estimates derived from melt inclusion data for the Hideaway Park topaz  
783 rhyolite in the Colorado Mineral Belt (approximately 60 to 70 MPa; Mercer et al. 2015). We  
784 calculated  $H_2O_{\text{melt}}$  abundances at 50 and 100 MPa with an input temperature of 750 °C. These  
785 estimates yield a range from approximately 3 to 4.5 wt%  $H_2O_{\text{melt}}$  at 50 and 100 MPa,  
786 respectively. The calculated  $H_2O_{\text{melt}}$  abundances for the Never Summer batholith overlap those  
787 reported from quartz-hosted melt inclusions from the Hideaway Park and Chalk Mountain topaz  
788 rhyolites in the Colorado Mineral Belt (approximately 2 to 5 wt%  $H_2O$ ; Audétat 2015; Mercer et  
789 al. 2015). Using these data, we estimated F and Cl in melt using biotite and whole rock data from  
790 the Never Summer batholith as well as Mount Princeton batholith and Mount Antero  
791 leucogranite (Toulmin and Hammarstrom 1990; Table 2). Estimated F melt concentrations in  
792 equilibrium with biotite from the Mount Cumulus pluton at 50 MPa have a large range, from  
793 0.88 to 6.8 wt%. At 100 MPa, the F concentrations reach 9 wt%. It is possible that these  
794 unusually high estimates are affected by re-equilibration with late-stage, F-rich fluids after

795 crystallization, but whole-rock geochemical data still support that these samples are more  
796 evolved in terms of their trace element abundances (Fig. 6). Estimated Cl abundances in the  
797 Mount Cumulus pluton are much lower than F in the same samples (0.11 to 0.45 wt% Cl). The  
798 Mount Richthofen pluton has lower estimated F (0.1 to 0.55 wt%) and higher Cl (0.54 to 3.2  
799 wt%) than the Mount Cumulus pluton.

#### 800 **Comparison to garnet-free mafic lower crust batch melting models.**

801 We compare the estimated F and Cl melt abundances to partial batch melting models  
802 using data from garnet-free mafic lower crustal xenoliths (Fig. 13). The partial melting models  
803 were constructed using the minimum F and Cl whole rock concentrations discussed above (gray  
804 stars, Fig. 13) and they indicate that 1 to 5% melting would yield a melt with approximately  
805 1000 to 5000 ppm F, and 2000 to over 10000 ppm Cl. Batch melting models for the xenoliths  
806 richest in F and Cl are in good agreement with estimated F and Cl melt abundances for the silicic  
807 member of the Mount Richthofen pluton (purple triangles; Fig. 13). This observation  
808 corroborates previous batch melting models with Sr/Y and La/Yb that suggested that the silicic  
809 member of the Mount Richthofen pluton is a reasonable analogue to low-degree partial melts  
810 from garnet-free mafic lower crust (Jacob et al. 2015). These data, along with trace element and  
811 isotopic data presented in this study (Fig. 11) and Jacob et al. (2015), demonstrate that F-rich  
812 (i.e., >2000 ppm F) silicic melts in the Never Summer batholith formed from mafic lower crustal  
813 anatexis.

814 Estimated melt F and Cl abundances for the mafic member of the Mount Richthofen  
815 granodiorite and all three samples from the Mount Cumulus pluton are not near any of the partial  
816 melting models we generated (Fig. 13). The Mount Cumulus samples have higher F abundances  
817 that what is predicted for partial melts based on the xenolith data, but their deviation could be

*For submission to American Mineralogist*

818 explained by fractionation of F/Cl during crystallization with open-system degassing. The blue  
819 arrows in Figure 13 show H<sub>2</sub>O-saturated crystallization models after Candela (1986) for magmas  
820 at 4.5 and 3 wt% H<sub>2</sub>O; the black arrow shows how F and Cl increase with H<sub>2</sub>O-undersaturated  
821 crystallization. Chlorine is preferentially partitioned into the fluid phase relative to the melt  
822 during degassing, but it also behaves incompatibly during crystallization. Fluorine also behaves  
823 incompatibly during crystallization but tends to stay in the melt phase during open-system  
824 degassing. As such, H<sub>2</sub>O-saturated crystallization of melts with more than ~2.5 wt% H<sub>2</sub>O have a  
825 negative slope on Figure 13 (e.g., more Cl in the melt is lost to the fluid phase than what is  
826 gained from crystallization; Candela 1986). Finally, the H<sub>2</sub>O concentration at saturation might  
827 not be constant, because F, an incompatible element during crystallization, increases H<sub>2</sub>O  
828 saturation in melt (Holtz et al. 1993). Hence, H<sub>2</sub>O-saturated crystallization may not necessarily  
829 follow a linear path on Figure 13.

830         The models presented in Figure 13 suggest that low-degree partial melts from the F-rich  
831 mafic lower crust, such as the silicic member of the Mount Richthofen pluton, could represent  
832 primitive melts that, upon crystallization and open-system degassing, become enriched in F and  
833 F/Cl. This corroborates conclusions from Jacob et al. (2015) that the Mount Cumulus  
834 leucogranite formed from melts that formed following differentiation of primitive silicic melts  
835 comparable to the silicic member of the Mount Richthofen pluton, but high-precision  
836 geochronological data preclude that the two plutons are directly related (Jacob et al. 2015;  
837 Rosera et al. 2021).

838         Fluid-saturated fractional crystallization modelling of primitive low silica rhyolite melts  
839 has been used to explain trace element variations and F enrichment in the Chalk Mountain and  
840 Hideaway Park topaz rhyolites from the Colorado Mineral Belt (Audétat 2015; Mercer et al.

*For submission to American Mineralogist*

841 2015). Low-degree batch melting models for F-rich garnet-free mafic lower crust, accompanied  
842 by H<sub>2</sub>O-saturated fractional crystallization, could also explain the F and Cl abundance measured  
843 in melt inclusions from these two topaz rhyolites (Fig. 13), thereby supporting a mafic crustal  
844 source for primary F enrichment in their primitive silicic melts. Estimated F abundances for  
845 averaged data from the Mount Antero leucogranite are lower than estimates from Mount  
846 Cumulus and measurements from melt inclusions from the Chalk Mountain and Hideaway Park  
847 rhyolites (Fig. 13). It is unclear if these differences are related to different source material (i.e.,  
848 lower F) or the degree of shallow crustal differentiation, but these data do suggest that the silicic  
849 melts that formed the Mount Antero leucogranite also had >2000 ppm F, similar to the silicic  
850 member of the Mount Richthofen pluton.

851         Estimated F and Cl abundances for the Mount Princeton batholith indicate it had a much  
852 lower F/Cl than other systems we considered. Likewise, quartz-hosted melt inclusions from the  
853 Alma rhyolite dikes do not follow the partial melting and crystallization models. We suggest that  
854 the Mount Princeton batholith and Alma rhyolites likely come from a source with significantly  
855 different F and Cl abundances than those in the mafic lower crustal xenoliths studied here. This  
856 observation is also supported by isotopic data; both the Alma rhyolites analyzed in this study and  
857 the Mount Princeton batholith contain more less radiogenic Nd, and more radiogenic Pb than  
858 younger F-rich magmas that intruded near them (Figs. 11,12). Furthermore, Ca isotope data  
859 indicate that the Mount Princeton batholith contained significant ancient felsic crust (Mills et al.  
860 2018), which further demonstrates that ancient felsic crust is not necessarily F-rich within the  
861 central Colorado Mineral Belt.

862         The mafic member of the Mount Richthofen pluton yielded the highest estimated Cl and  
863 lowest F abundances from the Never Summer batholith (Fig. 13). Jacob et al. (2015) suggested

*For submission to American Mineralogist*

864 that mantle-derived melts mixed with silicic melts derived by mafic crustal anatexis to form the  
865 Mount Richthofen pluton, and therefore it is not surprising that the mafic member of Mount  
866 Richthofen does not follow the lower crustal partial melting models shown in Figure 13.  
867 However, the estimated Cl contents range from 2 to 3 wt% for the 50 and 100 MPa models,  
868 respectively (Table S2; Fig. 13), which is greater than what has been reported for magmas with  
869 similar silica and total alkali contents (<1 wt% Cl; Webster et al. 2018). This suggests that either  
870 the mantle-derived melts in the Never Summer batholith could have been highly enriched in Cl,  
871 or that the biotite reequilibrated with Cl-rich fluids after crystallization. Evidence supporting  
872 subsolidus reequilibration for this sample include: (1) calcium amphibole compositions range  
873 from actinolite to magnesio-hornblende (Jacob et al., 2015, classified after Hawthorn et al.  
874 2012); (2) plagioclase-amphibole thermometry from the same sample yielded subsolidus  
875 temperatures, as low as 650 °C (Jacob et al. 2015); (3) Ti in biotite thermometry for this sample  
876 yields lower temperatures than the silicic member of Mount Richthofen (Fig. 4c); and (4) the F  
877 and Cl melt estimates for the Mount Cumulus pluton require loss of Cl during open-system  
878 degassing (Fig. 11), and the mafic sample we used is located near the contact with Mount  
879 Cumulus (Fig. 1). Given these observations, we tentatively suggest that amphibole and biotite in  
880 the mafic Mount Richthofen sample used in this study and by Jacob et al. (2015) are affected by  
881 post-crystallization reequilibration. More data from mafic, mantle-derived igneous rocks in the  
882 region are required to further test if they are unusually enriched in Cl.

### 883 **Integrated evidence for a F-rich mafic lower crust source**

884 Integrating data from a variety of sources leads us to conclude that F-enrichment of mafic  
885 lower crust was an essential process before later anatexis and generation of F-rich magmas in the  
886 southern Rocky Mountains. Although enriched lithospheric mantle and mafic lower crust overlap

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887 in their inferred Sr, Nd, and Pb isotopic compositions (e.g., Fig. 11), other lines of evidence  
888 preclude an exclusively juvenile mantle-derived source. Climax-type prospects in the Sawatch  
889 Range, as well as older, F-poor Laramide intrusions, contain numerous xenocrystic zircon with  
890  $^{207}\text{Pb}/^{206}\text{Pb}$  ages between 1.7 and 1.4 Ga (Feldman 2010; Rosera et al. 2021), consistent with  
891 zircon ages from mafic granulite xenoliths (Farmer et al. 2005). These data indicate that at least  
892 the early low-grade Mo prospects interacted with ancient crustal material and cannot solely be  
893 the product of juvenile mantle-derived melts.

894 We suggest that Oligocene F-rich melts in the study area were predominately sourced  
895 from ancient mafic crust, but there is enough ambiguity in the isotopic composition of the  
896 mantle-derived end member that calculating specific proportions is challenging. The bimodal  
897 Never Summer batholith is perhaps best suited to unraveling relationships between mantle-  
898 derived melts and anatexis of mafic crust (Jacob et al. 2015). Trace element abundances  
899 estimated from batch melting models presented here (Fig. 13) and elsewhere (Sr/Y and La/Yb;  
900 Jacob et al. 2015) as well as radiogenic isotopic data suggest that silicic melts involved in  
901 assembling the Never Summer batholith were derived from anatexis of garnet-free mafic lower  
902 crust. Importantly, these silicic melts may have been enriched in F (2000 to 3000 ppm) from  
903 their ancient crustal source (Fig. 13), and the elevated F/Cl ratios can be explained by H<sub>2</sub>O-  
904 saturated crystallization of these primitive melts to high-silica granites and rhyolites. Thus,  
905 genetic models for topaz rhyolites and F-rich leucogranites do not necessarily require that the  
906 elevated F/Cl ratios are inherited from melting juvenile, mantle-derived lower crust (e.g.,  
907 Christiansen et al. 2007), but that is not to say that mantle-derived melts do not play any role in  
908 generating the F-rich high silica magmas.

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910 Ultimately, these observations suggest that (1) ancient felsic crust is not a critical  
911 component for generating F-rich rhyolite and leucogranites, (2) a solely juvenile, mantle-derived  
912 origin for these F-rich systems is also unlikely, and that (3) a combination of ancient, F-rich  
913 mafic lower crust ± juvenile mantle-derived melts that supply heat (and potentially some  
914 material), such as in a deep crustal hot zone (Annen et al. 2006) or hybridized zone (Rosera et al.  
915 2013; Dailey et al. 2018), is most compatible with data presented here and elsewhere.

## 916 **IMPLICATIONS**

### 917 **Crustal Pre-Enrichment Leading up to Fluorine-Rich Silicic Magmatism**

918 High-F rocks have been hypothesized as a viable source for F-rich silicic magmas (e.g.,  
919 Audétat 2015), and our data allow us to narrow down where, when, and how much F enrichment  
920 occurred in the magma source. Parts of the garnet-absent mafic lower crust in northern Colorado  
921 contain up to 20 modal percent F-rich amphibole, resulting in whole-rock F concentrations at  
922 least 3.5 times greater than average mafic crust (Rudnick and Gao 2003). Trace element  
923 modeling using xenolith amphibole compositions reproduces estimated F contents for inferred  
924 primitive silicic melts in the Never Summer batholith (Fig. 13), and isotopic and geochemical  
925 data support a mafic lower crust source for those rocks (Fig. 11; Jacob et al. 2015). Fluorine  
926 enrichment must have occurred prior to Laramide hydration due to the age of the xenoliths (e.g.,  
927 Humphreys et al. 2003), and it could be an ancient feature of the mafic lower crust that extends  
928 throughout the southern Rocky Mountain region (Snelson et al. 2005). Our results suggest that  
929 ancient enrichment of F in the deep mafic lower crust, either by UHT granulite grade  
930 metamorphism or deep crustal magma processes (e.g., Chin et al. 2020), could be a necessary  
931 pre-condition for generating F-rich silicic magmas at a later stage.

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932           In light of xenolith compositions reported here, we suggest that F-rich leucogranites and  
933 rhyolites in the Colorado Mineral Belt and Never Summer complex inherited their initially high  
934 halogen abundances mainly from their lower crustal source. Amphiboles are the dominant  
935 hydrous phase in the garnet-free mafic granulite xenoliths of the State Line district (Bradley  
936 1985) and their breakdown ultimately drives variations in primary melt F and Cl concentrations.  
937 Experimental data and thermodynamic modeling demonstrate increasing the  $X_F$  of pargasite  
938 above approximately 0.4 corresponds to a thermal stability increase of 100–150 °C over F-poor  
939 pargasite (Holloway and Ford 1975; Tsunogae et al. 2003). We hypothesize that intrusion of hot,  
940 mantle-derived melts during extension heated the lower crust and induced anatexis.

941           This hypothesis has significant implications for models of the origin of F-rich porphyries  
942 and topaz rhyolites in the Colorado Mineral Belt and western North America. Laramide magmas  
943 have isotopic compositions consistent with derivation from mafic lower crust  $\pm$  mantle, as well  
944 as slightly elevated Sr/Y and low F contents that could represent melts formed below the stability  
945 limit of F-rich amphibole. Spatio-temporal analyses indicate F-rich silicic magmatism in the  
946 southern Rocky Mountains began within a million years of the start of a regional ignimbrite  
947 “flare-up” (Rosera et al. 2021), and major Mo deposits at Henderson, Climax, Mount Emmons,  
948 and Questa all formed during later rifting events (e.g., Carten et al. 1993). Thus, the deposits  
949 formed during periods of increased invasion of mantle-derived melts into the crust, which  
950 significantly elevated the temperature of the mafic lower crust (e.g., Farmer et al. 2008) and led  
951 to the breakdown of F-rich amphibole (Fig. 14). The high heat flow during the flare-up and  
952 initial rifting therefore represents a distinctly different style of melting than during the Laramide  
953 (e.g., before the ca. 38 Ma ignimbrite flare-up).

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954           This interpretation does not necessarily require that F-rich leucogranites are sourced from  
955 shallower crust than their precursors (e.g., Stein and Crock 1990), and we suggest that shallower  
956 crust is not likely to have been a major source for F-rich magmas. The xenolith we analyzed with  
957 the lowest estimated equilibrium P (sample SD2-LC78; approximately 0.5 GPa) also contains  
958 trace biotite and has the lowest F contents in amphibole; therefore, shallower mafic crust may not  
959 have been F-rich. In addition, seismic refraction data indicate crust in Colorado shallower than  
960 20–25 km is more likely to be felsic (Snelson et al. 2005). The fact that magmatic systems in the  
961 Colorado Mineral Belt strongly linked to deep melting of ancient felsic crust (Grizzly Peak and  
962 Mount Princeton; Mills et al. 2018) lack significant F-rich alteration and porphyry Mo  
963 mineralization further rules out the need to infer changing sources due to a shallower magma  
964 source. Instead, we suggest that the F/OH ratio of the mafic crust could be heterogenous  
965 vertically and laterally, with high F/OH rocks restricted to the deeper crust. Thus, high F/OH  
966 rocks only melt during periods of high heat flow or direct interaction with mantle-derived melts.  
967 In this model, rocks with average-to-low F/OH are likely to melt earlier.

968   **Implications for porphyry Mo mineralization**

969           In a recent synthesis of isotopic data for Mesozoic porphyry Mo deposits in China, Shu  
970 and Chiaradia (2021) suggested that the magma source rocks are not pre-enriched in Mo prior to  
971 mineralization, and that there is significant isotopic diversity across different systems where  
972 deposits can incorporate juvenile or ancient lower crust with varying proportions of mantle-  
973 derived melt. Our study suggests that pre-enrichment of F, rather than Mo, could be a critical  
974 prerequisite for the genesis of large porphyry Mo deposits, and that in the case of the southern  
975 Rocky Mountains it is the ancient mafic lower crust that hosts F-rich rocks. Consequently, the  
976 isotopic diversity of porphyry Mo deposits noted by Shu and Chiaradia (2021) could reflect

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977 varying degrees of F enrichment throughout heterogeneous lithosphere. Fluorine-rich silicic  
978 melts derived from these enriched sources will have higher H<sub>2</sub>O saturation (Holtz et al. 1993), as  
979 well as lower viscosity and solidi than their low-F counterparts (Manning 1981; Ouyang et al.  
980 2020). Consequently, melts with high initial F upon ascent into the upper crust can achieve high  
981 levels of magmatic differentiation and expel large volumes of water, both processes that favor  
982 the development of porphyry Mo systems with high F/Cl ratios (e.g., Audétat 2015).

983         Our results provide information regarding how much F-enrichment is needed to set the  
984 stage for generating silicic magmas capable of forming a porphyry Mo deposit. Batch melt  
985 modeling demonstrates that parts of the garnet-absent mafic lower crust in northern Colorado can  
986 generate primary silicic melts with >2000 ppm F (Fig. 13). Estimated F abundances from biotite-  
987 silicic melt formulations demonstrate that primitive silicic melts in the Never Summer batholith  
988 also contained >2000 ppm F, and fluid-saturated crystallization in the shallow crust can drive F  
989 contents to 1 to 5 wt% F (Fig. 13). The similarity of F and Cl estimates for the Mount Cumulus  
990 leucogranite to melt inclusions measurements for topaz rhyolites related to porphyry Mo  
991 mineralization in the Colorado Mineral Belt implies the silicic magmas that formed the Mount  
992 Cumulus pluton are comparable to mineralizing intrusions in porphyry Mo deposits. Importantly,  
993 they attained these extreme levels of fractionation because they formed from low-degree partial  
994 melts that had >2000 ppm F (and correspondingly a suppressed solidus and decreased viscosity)  
995 upon ascent into the shallow crust. It is possible to generate high F and F/Cl from primitive melts  
996 with low F (e.g., <1000 ppm) abundances through extensive (>80%) H<sub>2</sub>O-saturated fractional  
997 crystallization (blue arrows; Fig. 13), but this pathway to F enrichment is less probable because  
998 the higher melt viscosity relative a F-rich starting melt would inhibit crystal-liquid separation,  
999 and the higher solidus temperature leads to earlier rheological lock up. Thus, pre-enrichment of

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1000 deep crustal sources in F might not be a requirement for generating F-rich granites and rhyolites,  
1001 but higher initial F abundances make extreme levels of differentiation more likely, and in turn  
1002 could explain why the largest known Climax-type deposits are highly concentrated in Colorado  
1003 (Ludington and Plumlee 2009).

1004 High-precision U/Pb zircon geochronology on the same samples we used in this study  
1005 indicate the Mount Cumulus pluton was assembled over 10's to 100's of k.y. (Rosera et al.  
1006 2021). Hence, silicic melts with >2000 ppm F and F/Cl near unity can differentiate rapidly; over  
1007 shorter time frames than the full lifespan of porphyry Mo systems (100's to 1000's of k.y.;  
1008 Gaynor et al. 2019; Zhao et al. 2021). Existing models have hypothesized that S in porphyry Mo  
1009 is introduced by small volumes of mafic melts, rather than being from the causative silicic  
1010 magmas (Mercer et al. 2015). The evidence for rapid differentiation, and therefore rapid  
1011 fluctuations in F (and Cl), coupled with the required external introduction of S could explain the  
1012 paucity of F-rich porphyry Mo deposits in the geological record (e.g., Ludington and Plumlee  
1013 2009; Audétat and Li 2017), and we suggest it may have contributed to the lack of significant  
1014 mineralization in the Never Summer batholith. Furthermore, the textural complexity of major  
1015 mineralizing intrusions at the Henderson deposit are hypothesized to be linked to solidus  
1016 depression due to high concentrations of F, whereas less texturally complex intrusions are  
1017 associated with lower ore grades (Carten et al. 1988). Thus, rapid increase in F concentration and  
1018 F/Cl values can explain observations where texturally complex mineralizing intrusions are  
1019 punctuated by unmineralized, or low-grade intrusions, during assembly of Climax-type systems  
1020 (Carten et al. 1988; Gaynor et al. 2019).

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## ACKNOWLEDGMENTS

1022

This project was funded by the Martin Fund and Dissertation Completion Fellowship at

1023

the University of North Carolina at Chapel Hill. Further support was provided by the Geological

1024

Society of America and Sigma Xi. Special thanks to Aaron Bell for his help and advice with the

1025

electron microprobe and data analysis, as well as Kelsey Woody and Rebecca Finchman for

1026

helping in the field. We thank Amy Gilmer, Paul Giesting, and an anonymous reviewer for their

1027

thorough review that helped to greatly improve the manuscript. Any use of trade, firm, or

1028

product names is for descriptive purposes only and does not imply endorsement by the U.S.

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Government.

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**FIGURE CAPTIONS**

- 1387 **Figure 1.** Maps showing the location of study area, as well as simplified geology. **(a)**  
1388 Generalized map showing the location of the Colorado Mineral Belt, Cenozoic F-rich silicic  
1389 rocks, and the State Line diatreme district. Blue outlines mark approximate extent of maps  
1390 shown in panels (b) and (c). **(b)** Simplified geological map showing the Never Summer batholith,  
1391 silicic dikes, and sampling sites used in this study. Faults are dashed where they are inferred  
1392 and/or concealed. Map modified after O'Neill (1981). Triangles indicate sample locations, and  
1393 those with sample number labels were selected for biotite EMP analysis. **(c)** Simplified  
1394 geological map of the central Colorado Mineral Belt and sample sites. Thin dashed ovals show

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1395 approximate extent of historical and active mining districts in the area. Modified after Horton et  
1396 al. (2017). Colored point symbols represent sample locations (see Table S3 for sample  
1397 summaries and coordinates). BC – Brown’s Canyon district, C – Climax Mo deposit, GPC –  
1398 Grizzly Peak caldera, H – Henderson Mo deposit; MAC – Mount Aetna caldera, ME – Mount  
1399 Emmons Mo deposit, MP – Montezuma pluton, MPB – Mount Princeton batholith, Q – Questa  
1400 Mo deposit, TLP – Twin Lakes pluton.

1401 **Figure 2.** Select plots showing amphibole compositions from two-pyroxene mafic granulite  
1402 xenoliths of the State Line diatreme district. **(a)** Nomenclature for Ca amphibole after Hawthorne  
1403 et al. (2012) and Locock (2014). **(b)** F versus Cl in wt% showing a positive correlation between  
1404 abundance of Cl and F in amphibole. **(c)** Cl (in apfu) versus  $\text{Fe}^{2+}/(\text{Fe}^{2+} + \text{Mg})$  showing a weak  
1405 positive correlation. **(d)** F versus K (both in apfu). Note that sample SD2-LC78 is an outlier.  
1406 Dashed field shows amphibole compositions from the ultrahigh temperature (UHT) Highland  
1407 Complex in Sri Lanka (Sajeev et al. 2009). **(e)** Ti versus  $^{\text{IV}}\text{Al}$ . Dashed field as in panel (c). Lower  
1408 crust xenolith literature data from Selverstone et al. (1999) and Farmer et al. (2005); mafic high-  
1409 grade metamorphic rock data from Nijland et al. (1993), Tsunogae et al. (2003), and Sajeev et al.  
1410 (2009).

1411 **Figure 3.** Plots showing estimated temperature and pressure equilibration conditions for  
1412 amphibole from garnet-free mafic granulite xenoliths from the State Line District. Red color fill  
1413 in each panel includes all analyses other than those from SD2-LC78 (four samples; see Fig. 2).  
1414 Top panel: kernel density estimates of temperatures calculated from the Ti in calcium amphibole  
1415 thermometer (Liao et al. 2021). Bottom panel: plot showing isopleths of  $\text{TiO}_2$  (solid lines) and  
1416  $\text{Al}_2\text{O}_3$  (dashed lines) in  $P$ - $T$  space after Ernst and Liu (1998). Colored boxes correspond to range  
1417 of values measured in amphiboles from the State Line two-pyroxene mafic granulites. For



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1418 comparison, the  $P$ - $T$  range estimated for peak metamorphism of garnet-bearing lithologies from  
1419 the State Line District are overlain (gray box; Farmer et al. 2005).

1420 **Figure 4.** Select plots showing the composition of biotite from the Never Summer batholith. **(a)**  
1421 Biotite quadrilateral;  $\text{Fe} / (\text{Fe} + \text{Mg})$  versus total Al, all in apfu. Light blue fields with dashed  
1422 outlines correspond to other topaz rhyolites from the western United States (after Christiansen et  
1423 al. 2007). Italicized biotite end member names correspond to their closest corner on the  
1424 quadrilateral. **(b)**  $\log(\text{Cl}/\text{OH})$  versus  $X_{\text{phlogopite}}$ . **(c)** Ti versus  $\text{Mg}/(\text{Mg} + \text{Fe}_{\text{total}})$  modified after  
1425 Henry et al. (2005). Gray curves show Ti in biotite isotherms, which are only calibrated above  
1426  $\text{Mg}/(\text{Mg} + \text{Fe}_{\text{total}}) > 0.25$  (vertical dashed line; Henry et al. 2005) and have been modified to  $110$   
1427  $+ 2^{\text{W}}(\text{OH}, \text{F}, \text{Cl}, \text{O})$  biotite normalization. **(d)**  $\log(\text{F}/\text{OH})$  versus  $X_{\text{phlogopite}}$ . Note that  $\log(\text{F}/\text{OH})$  in  
1428 Mount Cumulus increases for decreasing  $X_{\text{phlogopite}}$  across samples, which is opposite of the Fe-F  
1429 avoidance principle. The Mount Cumulus biotite from samples 10-KJ-MC-94 and 10-KJ-MC-91  
1430 have unusually low  $X_{\text{phlogopite}}$  and are from whole-rock samples with  $< 5$  ppm Sr (see Fig. 6a).

1431 **Figure 5.** Select major element variation diagrams versus wt% silica for samples analyzed in this  
1432 study (large symbols) as well as other Cretaceous and younger rocks from northern Colorado  
1433 (small symbols). Samples are broadly color-coded by tectono-magmatic groups discussed in text.  
1434 **(a)** Molar  $\text{Al}_2\text{O}_3/(\text{Na}_2\text{O} + \text{K}_2\text{O})$  versus  $\text{SiO}_2$  (wt%) shows that most samples are peraluminous  
1435 (horizontal dashed line marks peraluminous/metaluminous boundary). **(b)**  $\text{FeO}_{\text{tot}}/(\text{FeO}_{\text{tot}} + \text{MgO})$   
1436 versus  $\text{SiO}_2$  scatterplot showing that many of pre-extensional rocks are magnesian (i.e., below  
1437 the grey line, after Frost et al. 2001). Rift-related leucogranites and rhyolites, as well as aplites  
1438 associated with the Twin Lakes and Montezuma plutons, are also ferroan. **(c)**  $\text{Na}_2\text{O} + \text{K}_2\text{O}$  (in  
1439 wt%) versus  $\text{SiO}_2$  showing silicic rocks associated with extension tend to have higher alkali  
1440 contents than older igneous rocks. Extrusive total alkali-silica classification shown for reference

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1441 only; note that many samples are intrusive. Literature data sources are: Simmons and Hedge  
1442 (1978), Farmer and DePaolo (1984), Stein (1985), Bailey (2010), and Jacob et al. (2015). CMB  
1443 – Colorado Mineral Belt.

1444 **Figure 6.** Trace element abundance and ratio variation diagrams. Symbols as in Figure 5. **(a)** Sr  
1445 vs. SiO<sub>2</sub> showing strongly depleted Sr abundances in rift-related leucogranites, rhyolites, older  
1446 aplites, and the Alma district rhyolites. Note that portions of Mount Cumulus contain < 5 ppm  
1447 Sr. **(b)** Rb vs. SiO<sub>2</sub> showing a weak positive correlation between log(Rb) and silica content. **(c)**  
1448 Nb vs. SiO<sub>2</sub> showing distinct contrast between extension-related rocks and early magma centers.  
1449 **(d)** Y vs. SiO<sub>2</sub> showing similar patterns similar to Nb versus SiO<sub>2</sub>. **(e)** Variation of Rb/Sr (note  
1450 log scale) versus SiO<sub>2</sub> showing a positive curvilinear relationship. Extension-related silicic rocks  
1451 have the highest Rb/Sr values. **(f)** Variation of Sr/Y versus SiO<sub>2</sub> showing relatively high Sr/Y  
1452 (>40) of Laramide suite and low Sr/Y for extension-related leucogranites and rhyolites.

1453 **Figure 7.** Lead isotopic variation diagrams for samples from this study as well as other  
1454 Cretaceous and younger igneous rocks from central and northern Colorado. Symbols as in Figure  
1455 5. **(a,c)** <sup>206</sup>Pb/<sup>204</sup>Pb and <sup>208</sup>Pb/<sup>204</sup>Pb variation in comparison to SiO<sub>2</sub>. **(b,d)** Initial <sup>206</sup>Pb/<sup>204</sup>Pb-  
1456 <sup>207</sup>Pb/<sup>204</sup>Pb and <sup>206</sup>Pb/<sup>204</sup>Pb-<sup>208</sup>Pb/<sup>204</sup>Pb plots showing central and northern Colorado igneous  
1457 rocks relative to growth curve after Stacey and Kramers (1975; SK, solid gray line), one of the  
1458 proposed curves for the subcontinental lithospheric mantle beneath much of the western United  
1459 States (CLM, dashed line; Pettke et al. 2010), and the northern hemisphere reference line  
1460 (NHRL; Hart 1984). Black numbers and boxes along growth curves correspond to age, in Ga.  
1461 Gray field outlines felsic Proterozoic rocks whose isotopic compositions are corrected to 35 Ma  
1462 (Frazer 2017). Most of the data fall between the SK and CLM model curves. Dark gray outline  
1463 corresponds to rocks located in the northern Colorado Mineral Belt (including the Montezuma

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1464 pluton, after Stein 1985) and Never Summer igneous complex. Dashed blue polygon highlights  
1465 samples from the central and southern Colorado Mineral Belt, which tend to have less radiogenic  
1466  $^{206}\text{Pb}/^{204}\text{Pb}_i$  compositions. Samples associated with F-rich porphyry Mo mineralization are  
1467 highlighted in pink in panel d (data from this study and Stein 1985). Note that two samples from  
1468 the Alma district with highly radiogenic Pb isotopic compositions are not included in these plots  
1469 (see Table S5).

1470 **Figure 8.** Strontium isotopic variation diagrams. **(a)**  $^{87}\text{Sr}/^{86}\text{Sr}_i$  versus  $\text{SiO}_2$  diagram showing that  
1471 the two Alma rhyolites, as well as the Turquoise Lake porphyry, have highly radiogenic Sr  
1472 ( $>0.715$ ). Most of the intermediate samples from Twin Lakes through Breckenridge have  
1473  $^{87}\text{Sr}/^{86}\text{Sr} < 0.708$  and samples from the Montezuma complex have slightly more radiogenic Sr.  
1474 Horizontal dashed line in panel a shows limit of ordinate axes for the same plot in panel **(b)**.  
1475 Symbols as in Figure 5. CMB – Colorado Mineral Belt.

1476 **Figure 9.** Initial  $\epsilon\text{Nd}$  versus  $\text{SiO}_2$  variation diagram, showing no strong correlation between Nd  
1477 isotopic composition and silica content. Symbols as in Figure 5.

1478 **Figure 10.** Back-scattered electron image of amphibole (analysis 4) from sample NX4-LC2. This  
1479 xenolith contains plagioclase that is variably altered to clay, but this effect showed no influence  
1480 on amphibole measurements (two examples of alteration are highlighted by dashed blue lines).  
1481 Bright minerals located around grain boundaries are mostly barite that was introduced by host  
1482 kimberlite (see text). Amphibole (amph) show no significant disequilibrium textures or obvious  
1483 zonations. opx – orthopyroxene, cpx – clinopyroxene, plag – plagioclase (black and gray areas).

1484 **Figure 11.** Histograms and variation diagrams showing Sr and Nd isotopic compositions in  
1485 comparison to Proterozoic country rocks (Proterozoic rocks corrected to 35 Ma, all other samples

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1486 corrected to their initial ages). **(a)**  $\epsilon\text{Nd}$  versus  $^{87}\text{Sr}/^{86}\text{Sr}$  for Cenozoic and younger igneous rocks  
1487 in the study area. Dashed box shows area of panel (b). Colored fields are simplified outlines of  
1488 country rocks, as represented by felsic and mafic xenoliths from the Four Corners area, felsic  
1489 rocks from the surface, and mafic granulite xenoliths from the State Line District. Histograms  
1490 above and to the right of panel a show distribution of country rock data for Sr (top) and Nd  
1491 (right). Notice that most of the felsic country rocks have  $\epsilon\text{Nd}_{35\text{ Ma}} < -10$ . **(b)** As in panel (a) but in  
1492 a smaller area to show more detail. Ellipse encloses 66 Ma Windy Gap basalts. for comparison to  
1493 mafic xenoliths from the region. CMB – Colorado Mineral Belt.

1494 **Figure 12.** Initial  $^{206}\text{Pb}/^{204}\text{Pb}$  isotopic compositions through time. **(a)**  $^{206}\text{Pb}/^{204}\text{Pb}_i$  through time  
1495 for samples from the Colorado Mineral Belt. Gray boxes and arrows highlight locations where  
1496 younger magmas shift towards less radiogenic Pb compositions. **(b)** Same as panel (a), but only  
1497 showing data for the Never Summer batholith. Two silicic dikes from the northern portion of the  
1498 magma system have slightly more radiogenic  $^{206}\text{Pb}/^{204}\text{Pb}_i$  than most of the samples from the  
1499 Mount Cumulus leucogranite. Symbols as in Figure 5. MA – Mount Antero, MP – Mount  
1500 Princeton.

1501 **Figure 13.** Plots showing estimated concentrations of F and Cl in equilibrium with biotite for the  
1502 Never Summer batholith samples at: **(a)** 50 MPa, and **(b)** 100 MPa. In both cases, the estimated  
1503 F and Cl from biotite-melt exchange formulations are compared to batch melting models (gray  
1504 curves) of garnet-free mafic lower crust using xenolith data. Labeled circles along batch melt  
1505 models represent fraction melted. Gray boxes show extent of F and Cl concentrations measured  
1506 in melt inclusions from Audétat (2015) and Mercer et al. (2015). Also shown for comparison are  
1507 estimated average F and Cl melt concentrations from Mount Antero leucogranites (using data  
1508 from Toulmin and Hammarstrom 1990: note they analyzed biotite from different samples than

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1509 the whole rock data). Gray stars denote minimum F and Cl concentrations in xenoliths that were  
1510 used as starting compositions for batch melt models. Black star shows average mafic lower crust,  
1511 for reference (Rudnick and Gao 2003). Arrows show fractional crystallization models for H<sub>2</sub>O-  
1512 saturated (blue arrows) and H<sub>2</sub>O-undersaturated conditions (black arrow). Length of arrow  
1513 corresponds to 0.2 melt fraction remaining. Partition coefficients for batch melting and H<sub>2</sub>O-  
1514 undersaturated fractional crystallization models are shown in Table S7. H<sub>2</sub>O-saturated models  
1515 are after Candela (1986).

1516 **Figure 14.** Cartoon model depicting how deep mafic crust potentially controlled F-enrichment of  
1517 silicic anatectic melts in central and northern Colorado from the Cretaceous through the  
1518 Oligocene. Left side of diagram depicts a lower flux of mantle derived melt and fluids (smaller  
1519 black arrow) and in turn lower heat flux into garnet-free mafic lower crust. Fluorine-rich  
1520 pargasite are likely to remain stable at a lower heat flux, hence the anatectic products will not be  
1521 enriched in F. Right side shows higher mantle melt and fluid fluxes, perhaps during extension or  
1522 caldera-forming magma events. The higher heat flux in the garnet-free mafic lower crust allows  
1523 for the breakdown of F-rich pargasite observed in xenoliths, thereby producing F-rich melts that  
1524 can differentiate towards leucogranitic compositions. Model modified after Mercer et al. (2015)  
1525 and Jacob et al. (2015).

TABLE 1. GENERAL SUMMARY OF CRETACEOUS TO OLIGOCENE IGNEOUS ROCKS IN CENTRAL COLORADO

Tectono-Magmatic Group	Major Lithology	$^{87}\text{Sr}/^{86}\text{Sr}_i$	$\epsilon\text{Nd}_i$	Examples
Laramide to start of flare-up (75 to 38Ma)	Monzonite	0.705 to 0.708 (mostly < 0.706)	-1 to -9	Empire stock
	Gd to Qtz Monz.	0.706 to 0.7089	-5 to -10	Twin Lakes pluton; Montezuma pluton
Transition (37.3 to 30 Ma)	Qtz Monz. to Gr	0.707 to 0.710	-7 to -10	<b>Middle Mountain porphyry;</b> Mount Princeton batholith
Extension and Rio Grande rifting < 30 Ma	Gr. (bimodal suite)	0.708 to 0.735	-8 to -14 (CMB)	Mount Antero leucogranites
			-2 to -5 (NS)	<b>Climax porphyry system</b> Never Summer complex

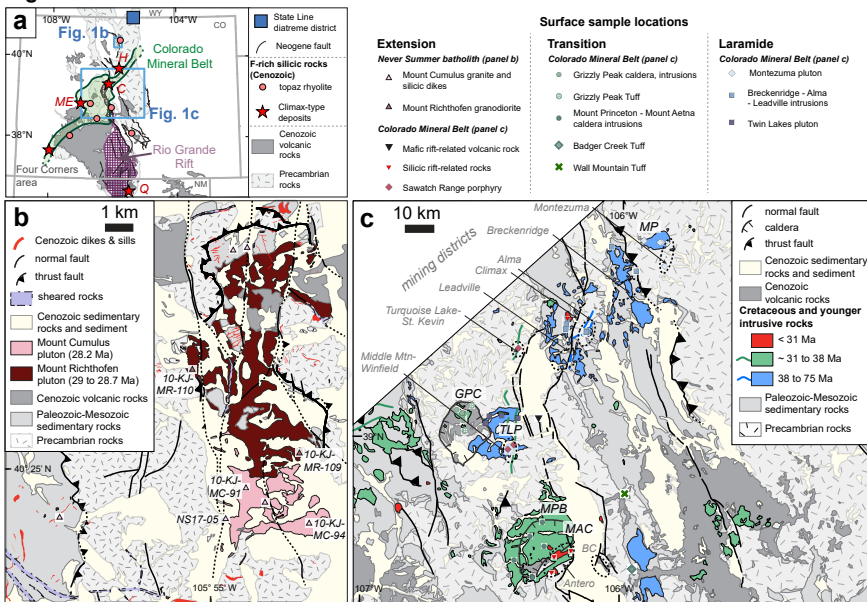
Examples in bold are associated with Climax-type porphyry Mo mineralization.

Sources: Simmons and Hedge (1978), Farmer and DePaolo (1984), Stein and Crock (1990), and Jacob et al. (2015)  
CMB – Colorado Mineral Belt; Gd – granodiorite; Qtz Monz. – quartz monzonite; Gr – granite; NS – Never Summer

TABLE 2. ESTIMATED RANGE OF F AND Cl CONCENTRATIONS OF MELT IN EQUILIBRIUM WITH BIOTITE

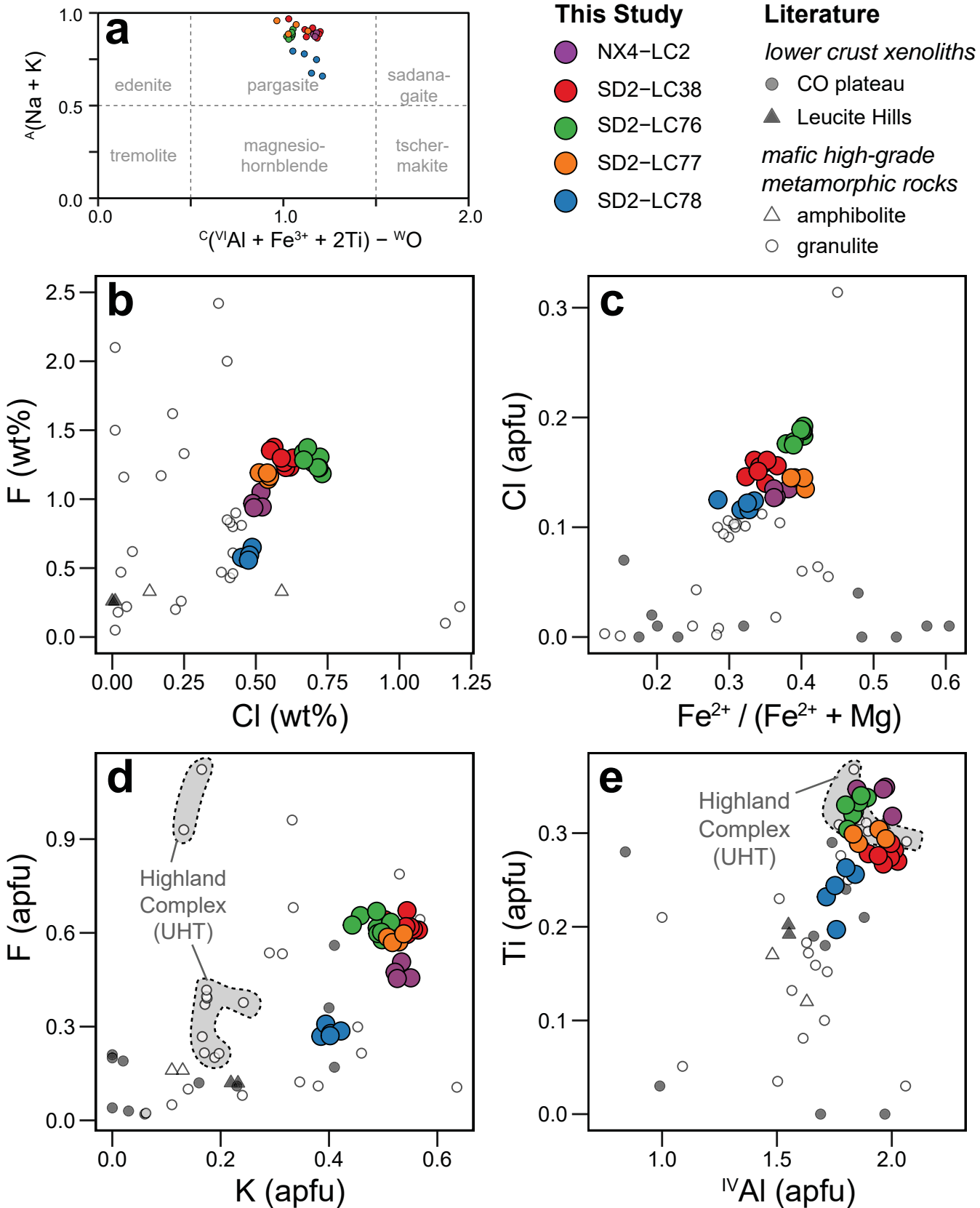
Sample	Description	50 MPa		100 MPa	
		F, ppm ( <i>mean</i> )	Cl, ppm ( <i>mean</i> )	F, ppm ( <i>mean</i> )	Cl, ppm ( <i>mean</i> )
10-KJ- MR-91	low-Sr Mount Cumulus granite	47000 to 65000 (55000)	1800 to 2600 (2200)	62000 to 87000 (73000)	2500 to 3400 (2900)
10-KJ- MR-94	low-Sr Mount Cumulus granite	37000 to 68000 (50000)	1100 to 1600 (1300)	50000 to 90000 (67000)	1400 to 2000 (1700)
NS17-05	Mount Cumulus	8800 to 20000 (14000)	2200 to 3400 (2800)	12000 to 26000 (19000)	3000 to 4500 (3700)
10-KJ- MR-109	Mafic Mount Richthofen	1000 to 1200 (1100)	19000 to 24000 (22000)	1400 to 1600 (1500)	26000 to 32000 (30000)
10-KJ- MR-110	Silicic Mount Richthofen	2500 to 4100 (3200)	5400 to 9500 (7900)	3500 to 5500 (4300)	7300 to 13000 (11000)

**Figure 1**

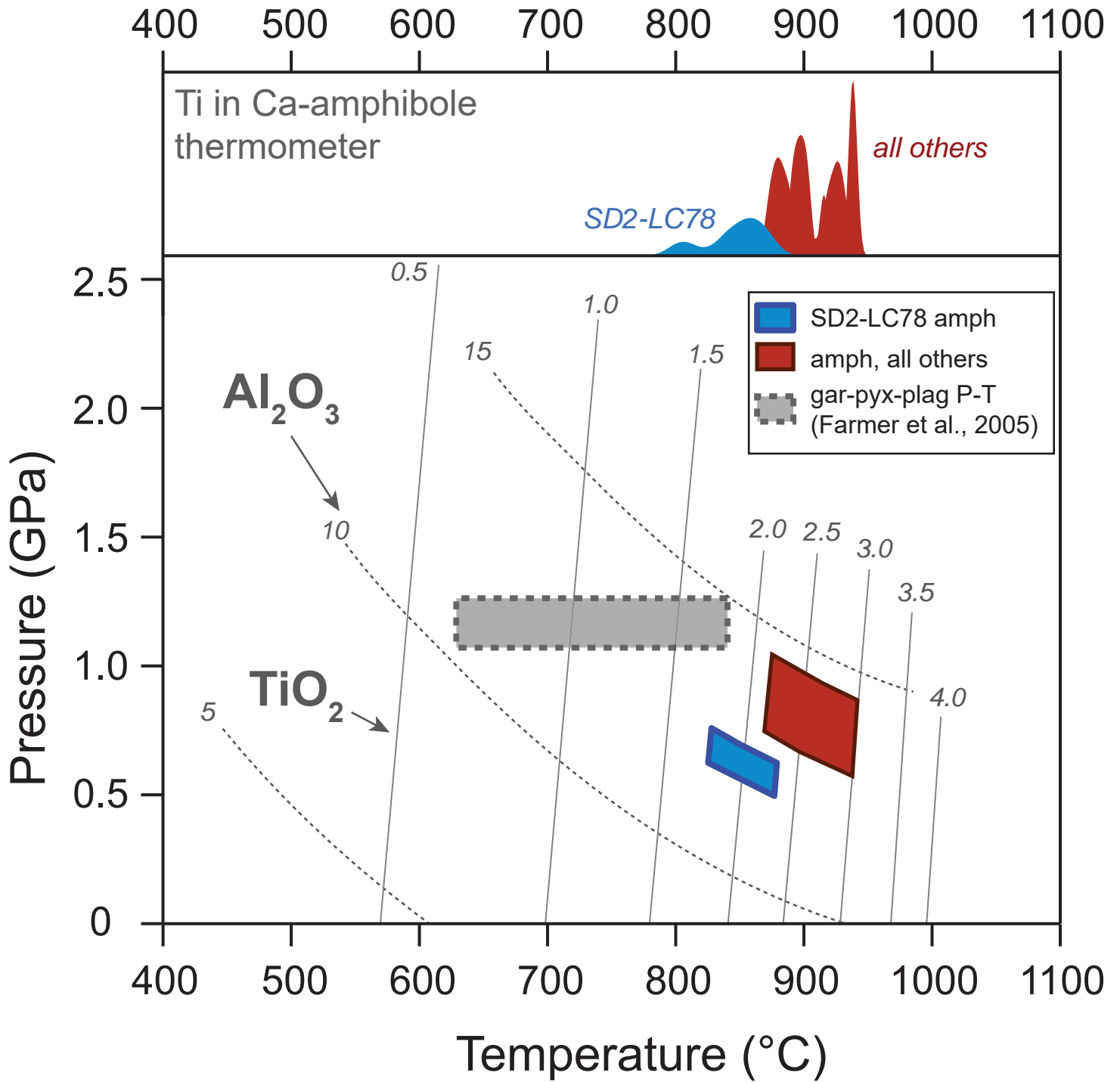




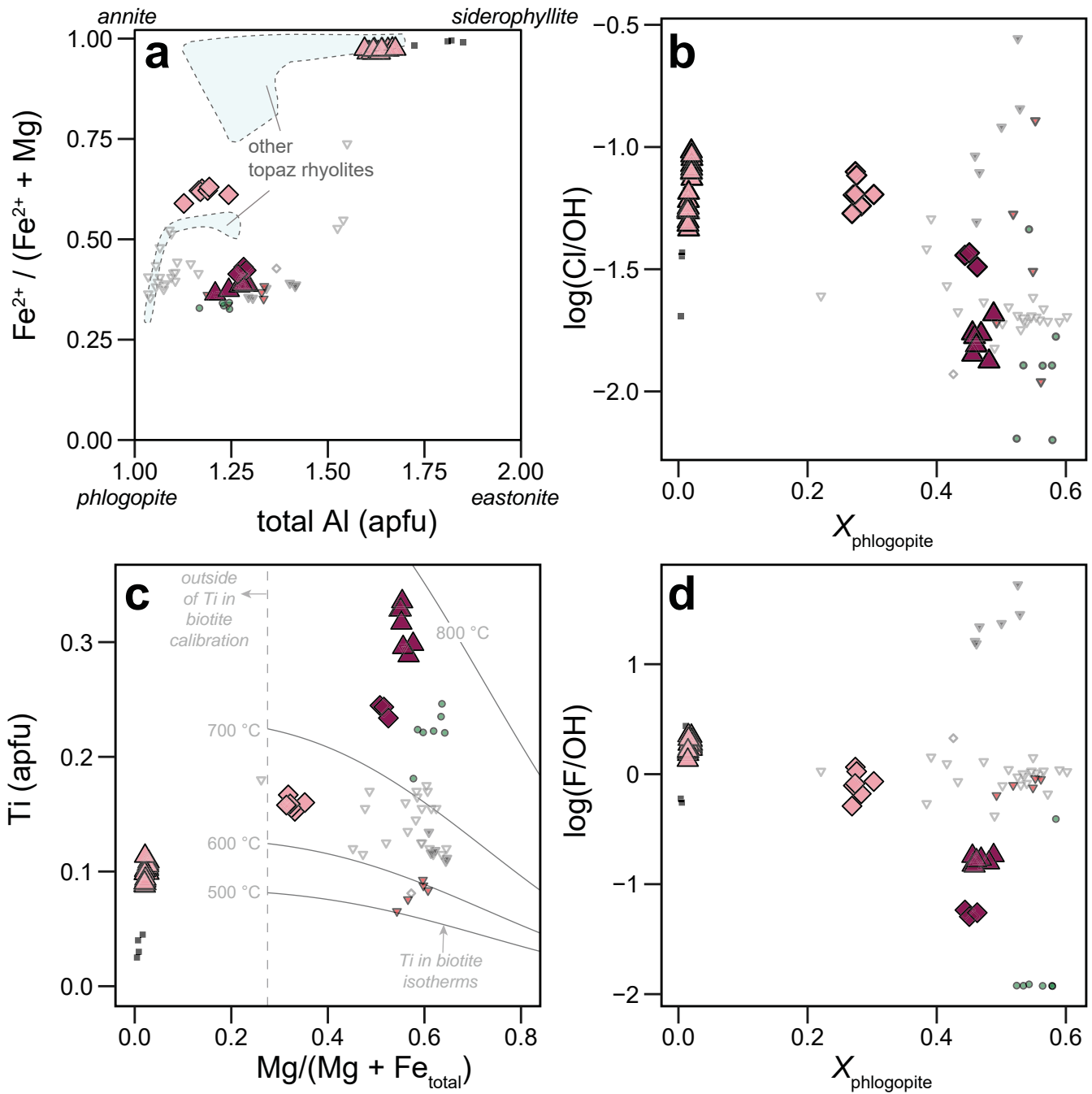
# Figure 2



**Figure 3**



# Figure 4



## This Study

### Mount Cumulus pluton

- ▲ Samples 10-KJ-MC-94 & 10-KJ-MC-91
- ◆ Baker Pass, western phase (NS17-05)

### Mount Richthofen pluton

- ▲ silicic member (10-KJ-MR-110)
- ◆ mafic member (10-KJ-MR-109)

## Literature

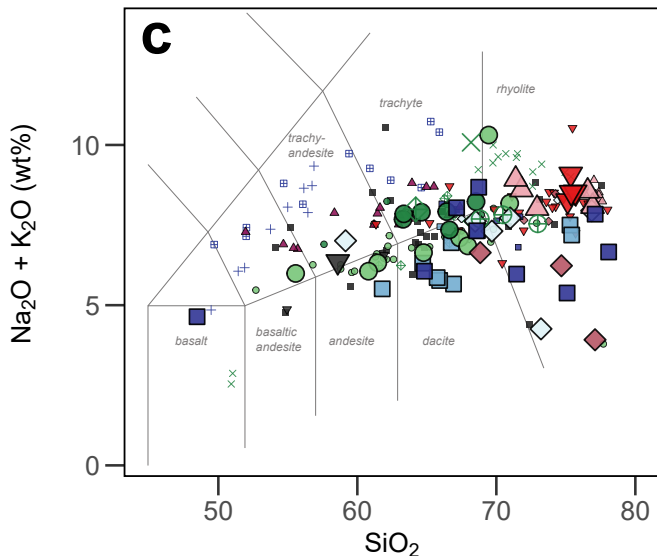
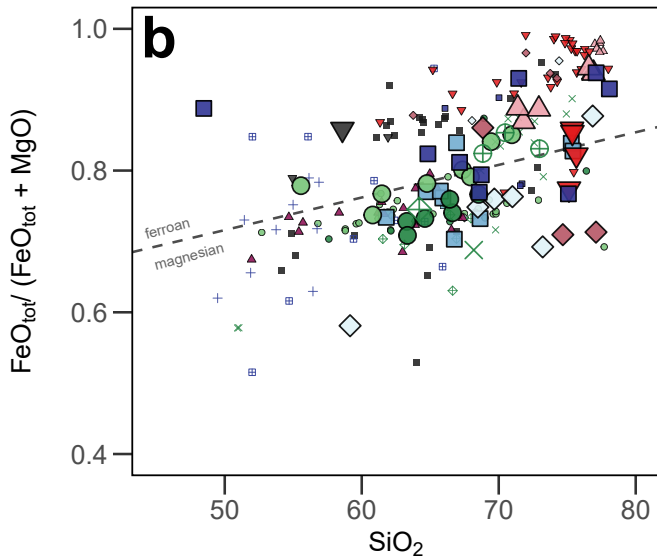
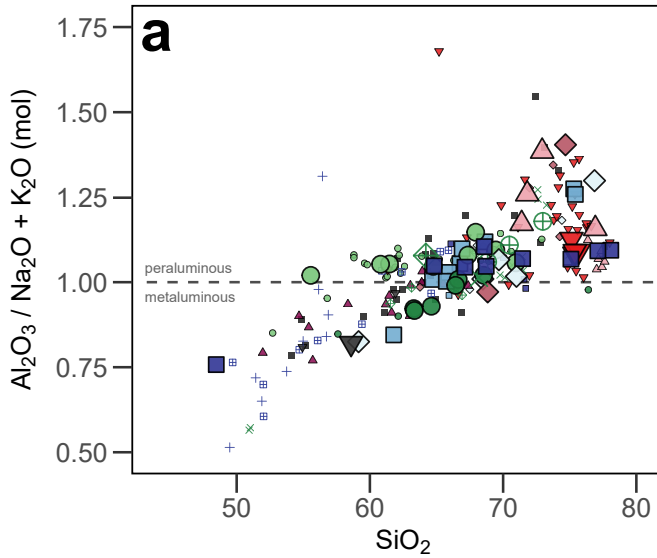
### Colorado Mineral Belt

- ▼ Chalk Mountain rhyolite
- ◇ Henderson (igneous biotite)
- ▽ Hideaway Park tuff
- ▼ Mount Antero leucogranite
- Mount Princeton batholith

### Other topaz rhyolite

- Honeycomb Hills (Utah)

# Figure 5



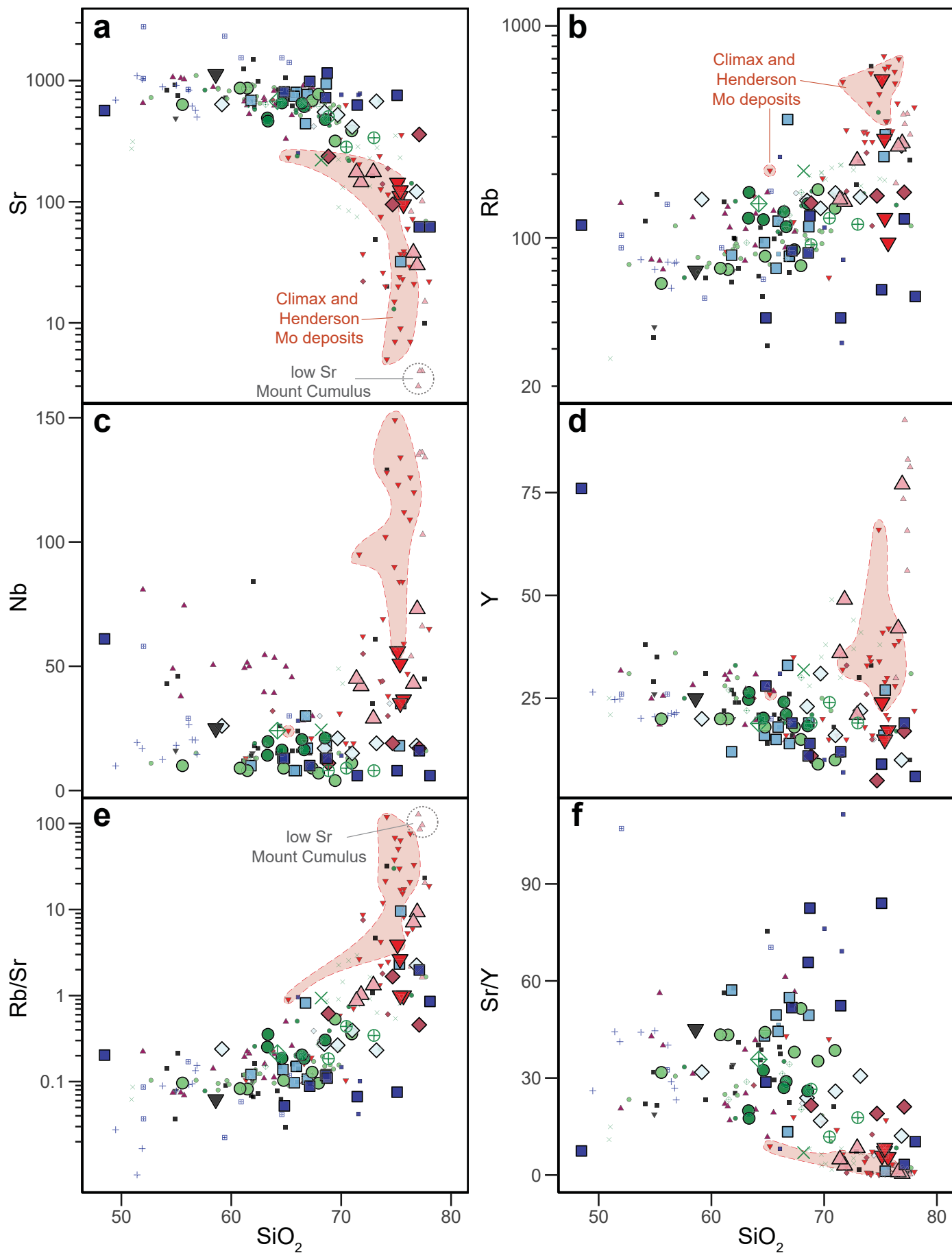
## This Study

- ▼ Mafic rift-related rocks in the CMB **Extension**
- ▲ Mount Cumulus granite and silicic dikes
- ▼ Silicic rift-related rocks in the CMB
- ◆ Sawatch Range porphyry **Transition**
- Grizzly Peak caldera intrusions
- ⊕ Grizzly Peak Tuff
- Mount Princeton – Mount Aetna caldera
- ◇ Badger Creek Tuff
- × Wall Mountain Tuff
- ◇ Montezuma pluton **Laramide**
- Breckenridge – Alma – Leadville intrusions
- Twin Lakes pluton

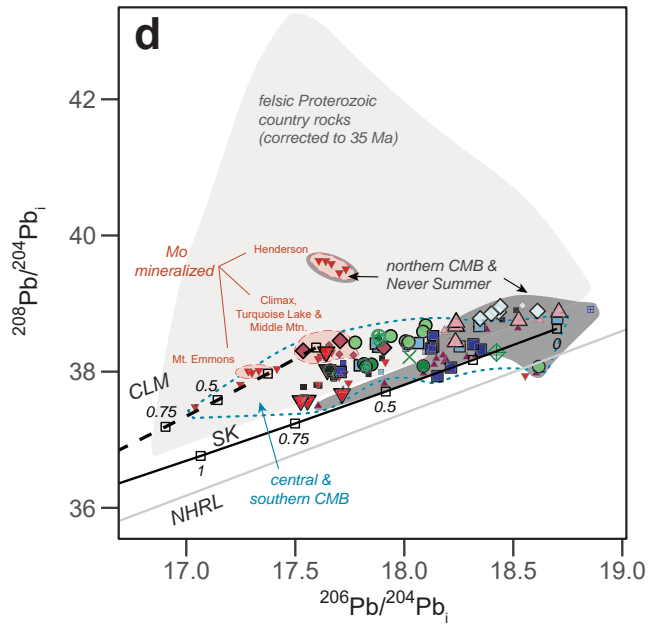
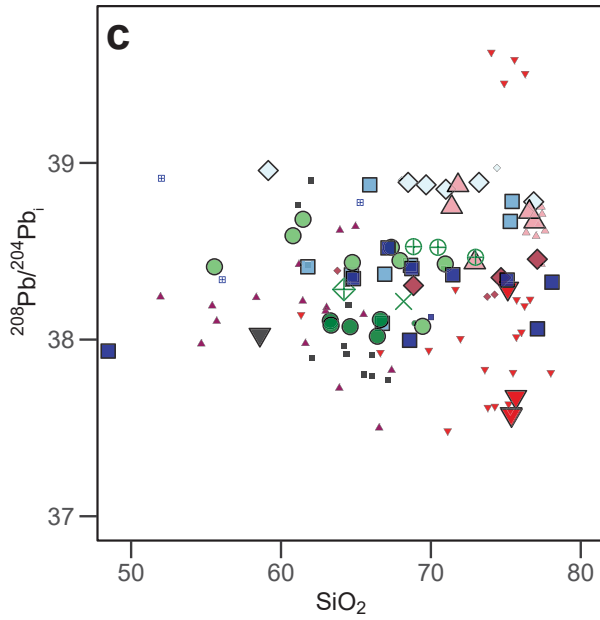
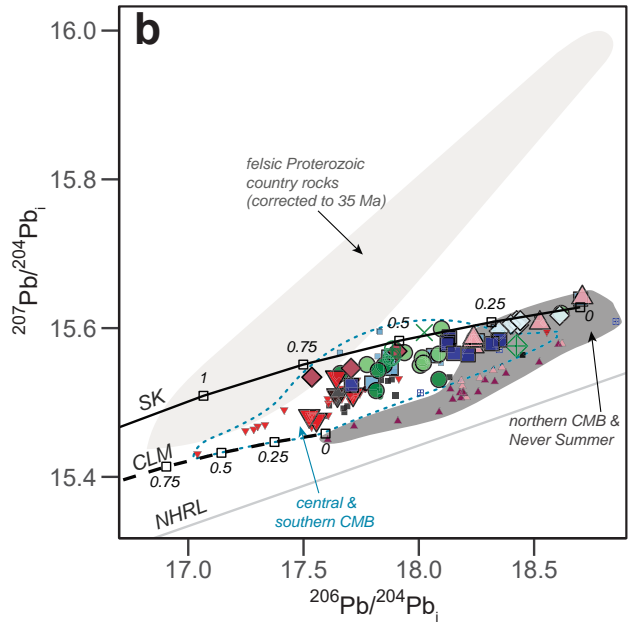
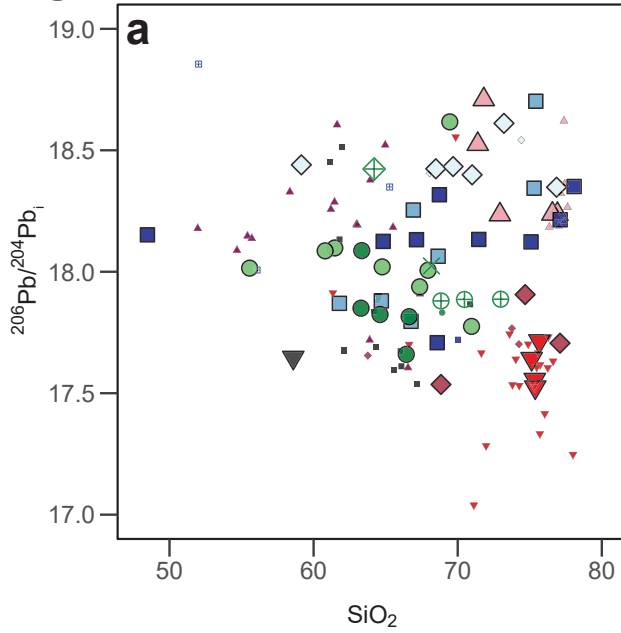
## Literature

- ▼ Mafic rift-related rocks in the CMB **Extension**
- ▲ Never Summer, Mount Richthofen
- ▲ Mount Cumulus granite and silicic dikes
- ▼ Silicic rift-related rocks in the CMB
- ◆ Sawatch Range porphyry **Transition**
- Grizzly Peak caldera intrusions
- Mount Princeton – Mount Aetna intrusions
- ◇ Badger Creek Tuff
- × Wall Mountain Tuff
- ◇ Montezuma pluton **Laramide**
- Breckenridge – Alma – Leadville intrusions
- Twin Lakes pluton
- + Windy Gap mafic volcanic rocks
- Monzonite suite
- Granodiorite suite, other (Laramide & Transition)

**Figure 6**



# Figure 7



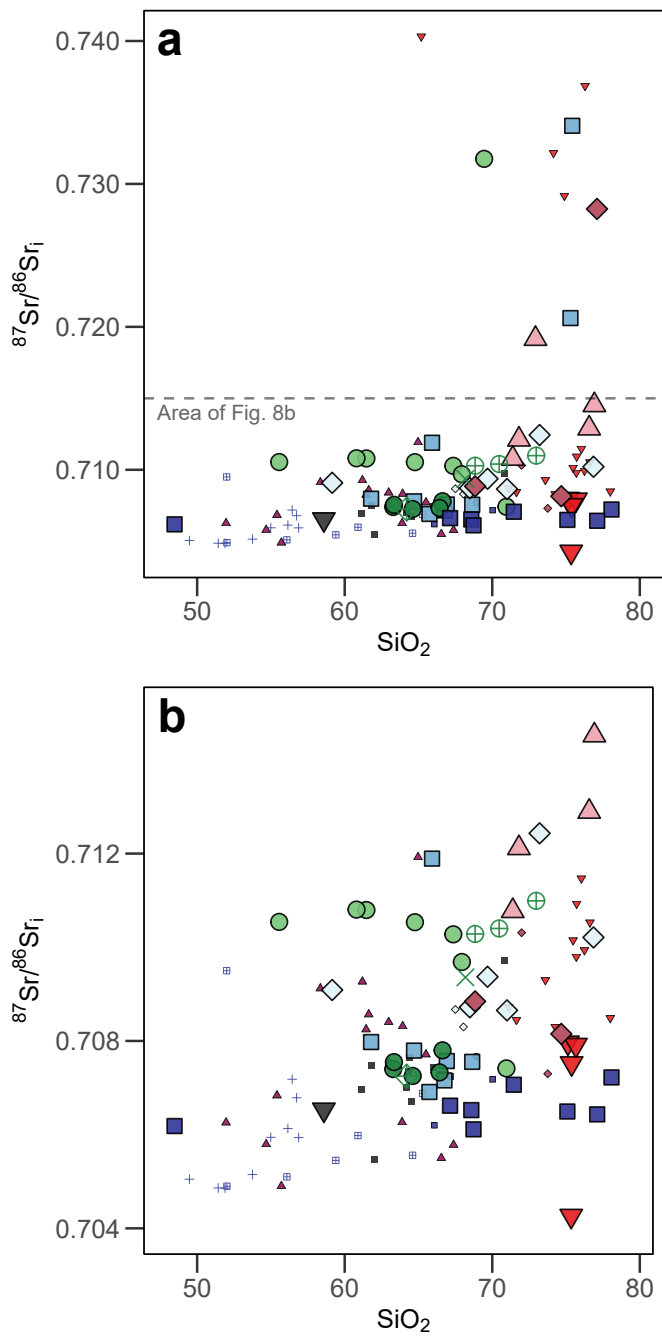
## This Study

- ▼ Mafic rift-related rocks in the CMB **Extension**
  - ▲ Mount Cumulus granite and silicic dikes
  - ▼ Silicic rift-related rocks in the CMB
  - ◆ Sawatch Range porphyry **Transition**
  - Grizzly Peak caldera intrusions
  - ⊕ Grizzly Peak Tuff
  - Mount Princeton – Mount Aetna caldera
  - ◆ Badger Creek Tuff
  - × Wall Mountain Tuff
  - ◇ Montezuma pluton **Laramide**
  - Breckenridge – Alma – Leadville intrusions
  - Twin Lakes pluton
- systems with F-rich magmas*

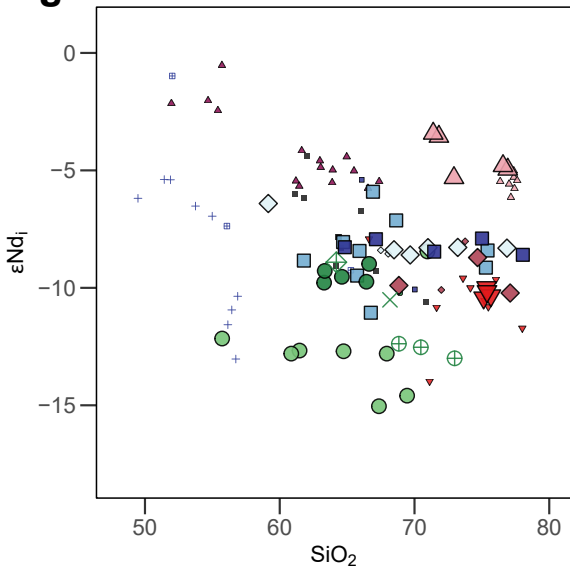
## Literature

- ▼ Mafic rift-related rocks in the CMB **Extension**
  - ▲ Never Summer, Mount Richthofen
  - ▲ Mount Cumulus granite and silicic dikes
  - ▼ Silicic rift-related rocks in the CMB
  - ◆ Sawatch Range porphyry **Transition**
  - Grizzly Peak caldera intrusions
  - Mount Princeton – Mount Aetna intrusions
  - ◆ Badger Creek Tuff
  - × Wall Mountain Tuff
  - ◇ Montezuma pluton **Laramide**
  - Breckenridge – Alma – Leadville intrusions
  - Twin Lakes pluton
  - Monzonite suite
  - Granodiorite suite, other
- systems with F-rich magmas*

# Figure 8

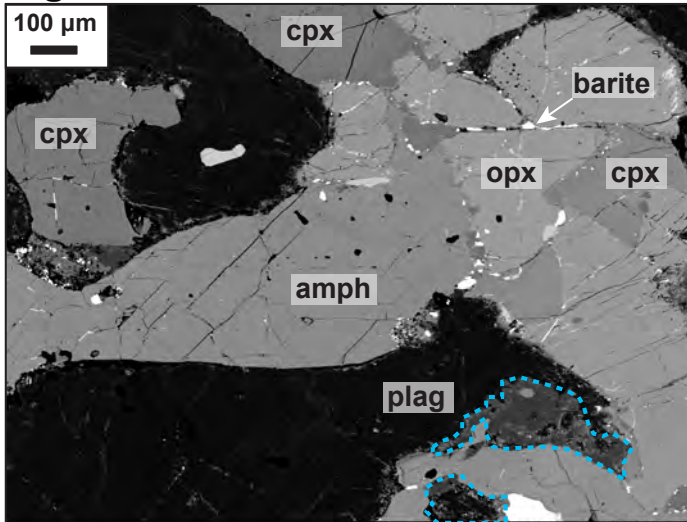


**Figure 9**

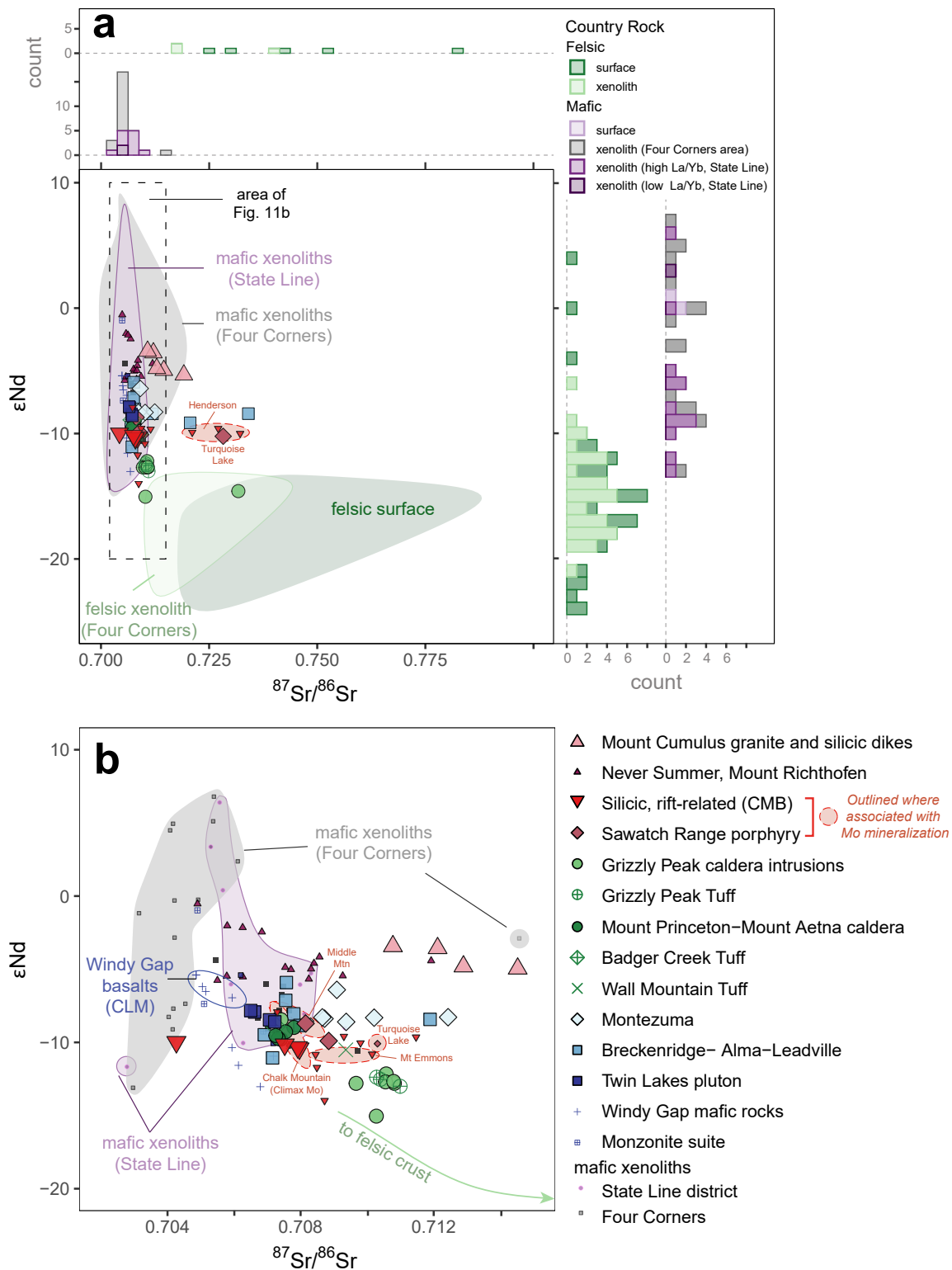




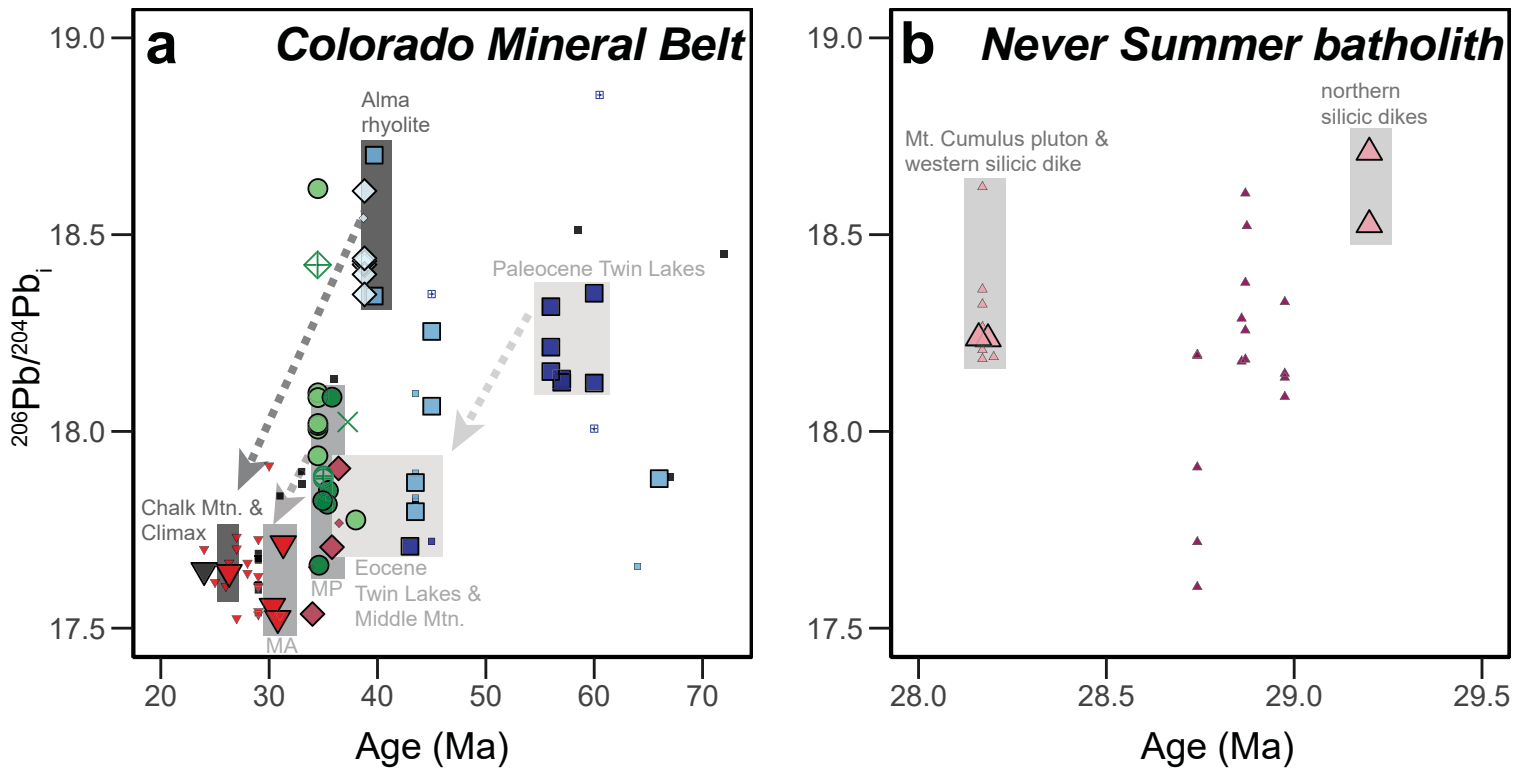
# Figure 10



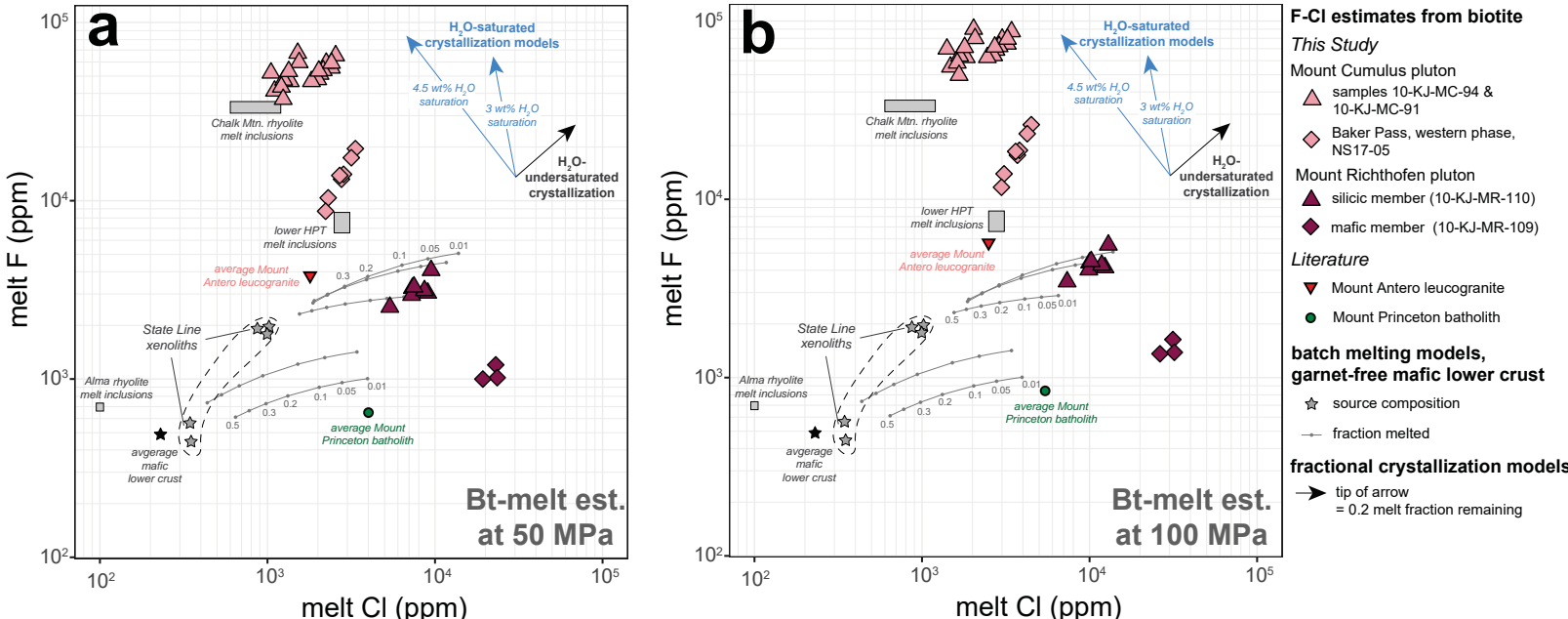
# Figure 11



**Figure 12**



**Figure 13**



**Figure 14**

