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2	Crystallization order effects on inclusion assemblages in magmatic accessory minerals and
3	implications for the detrital record
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8	Abstract:
9	Mineral inclusion assemblages in zircon are almost certainly a complex function of crystallization
10	sequence, magma chemistry, and physical proximity to other crystallizing phases. While the latter may
11	be largely random, the former two factors should have more systematic controls. To better constrain the

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12 effects of crystallization sequence, we investigated a suite of granitoids from the Cretaceous arc of 13 southern California and from the Miocene Colorado River Extensional Corridor, ranging from tonalites to 14 leucogranites. In particular we examined inclusions in a variety of accessory minerals to elucidate 15 crystallization order effects. Apatite is almost universally overrepresented as an inclusion in the studied minerals relative to its volumetric abundance in the rock as a whole. Where crystallization order can be 16 17 inferred, earlier phases usually either have a higher proportion of apatite inclusions (11 out of 20 cases) or show no significant differences in apatite content from later phases (7 cases). An increase in the 18 19 proportion of apatite with progressive crystallization is observed in four out of the 20 samples with clear 20 crystallization orders (two rocks out of the 20 contain both an increasing and a decreasing case among 21 different mineral pairs and are included in both groups). We interpret these observations as showing the 22 widespread early crystallization of apatite inclusions - likely by pileup of the slow-diffusing phosphorus

23 complex along growing mineral grains. The lower apatite contents in late-crystallizing modal phases are 24 also reflected in the very low apatite contents within late-crystallizing zircon from some studied mafic-25 intermediate units, contrary to earlier suggestions that apatite decreases as a proportion of the inclusion 26 assemblage with increasing whole rock silica. Plagioclase inclusions in zircon are on average more albitic 27 than matrix plagioclase. In some samples, sequences of plagioclase crystallization can be identified by 28 albite content among 1) magmatic cores of matrix grains, 2) inclusions in mafic accessory minerals, 3) 29 inclusions in zircon, and 4) post-magmatic alteration veins and exsolution lamellae. However, the relative 30 proportions of quartz, K-feldspar, and plagioclase inclusions do not systematically vary in accessory 31 minerals with crystallization order, suggesting other origins of the often-recognized modal shifts toward 32 more quartz- and K-feldspar-rich inclusion assemblage compared to the matrix. Unlike Himalayan 33 leucogranites, when muscovite is present in the matrix in our sample suite, it is a rare inclusion phase in 34 later-crystallizing accessories and may, in some cases, even be absent in zircon even when present in other 35 phases. It is possible to mistake non-magmatic inclusions in detrital grains for primary magmatic 36 inclusions, and examination of internal host grain texture and geochronology (when applicable) is 37 especially important in a detrital setting. These results should allow an improved framework for 38 interpreting inclusion assemblages in detrital magmatic minerals and in interpreting the phase petrology 39 of the host igneous rock. Given the remaining ambiguities in linking a number of observations (i.e., apatite 40 contents, position on the quartz-alkali feldspar-plagioclase ternary, and the ratio of felsic to mafic silicate 41 phases) to specific magmatic provenance, the identification of rare but petrologically significant inclusion 42 phases such as muscovite, halides, Mn and Nb minerals, sulfides, or baddeleyite, may be more helpful in pointing to either highly evolved or more mafic provenances. 43

44 **1. Introduction**

Detrital zircon has long been exploited for the information it records about its protolith in the quest to reconstruct sedimentary basin provenance (e.g., Maas and McCulloch, 1991). The increasing

availability of rapid in situ zircon U-Th-Pb dating, first with the ion microprobe and more recently using 47 48 LA-ICPMS, has allowed for an explosion in detrital zircon age studies (Harrison et al., 2015). Increasingly, 49 other aspects of zircon geochemistry and geochronology, such as Lu-Hf isotopic fingerprinting (e.g., Fisher 50 et al., 2014), trace element inferences on source rock type (e.g., Belousova et al., 2002a; Burnham and 51 Berry, 2017), and low-temperature thermochronology (e.g., Reiners et al., 2005), have also been 52 employed in order to extract source rock information. Mineral inclusions in zircon represent an under-53 exploited source of information for these purposes. Although inclusions in metamorphic zircon have long 54 been utilized in studies of UHP metamorphism (e.g., Tabata et al., 1998), systematic studies of inclusion 55 assemblages in magmatic zircon are still in their early stages (e.g., Hopkins et al., 2008; Darling et al., 2009; 56 Rasmussen et al., 2011; Jennings et al., 2011; Bell et al., 2017, 2018; Faltys and Wielicki, 2020).

57 The relatively small literature on mineral inclusions in detrital zircon directly reflects the largely unknown relationship between granitoid petrogenesis and the mineral inclusion assemblages within their 58 59 zircons. The clearest such relationship is the near universal dominance of apatite inclusions in zircon (e.g., 60 Rasmussen et al., 2011; Bell et al., 2018), albeit with a weak magma compositional control (Bell et al., 61 2018). That a trace matrix phase dominates an included assemblage is clear evidence that incorporation 62 of phases into zircon is not a simple reflection of the coexisting mineral population in the host magma 63 during zircon growth. An additional question pertains to the abundances of specific phases: what controls 64 the presence of, for example, muscovite inclusions in magmatic zircon (muscovite being a dominant phase 65 in originally magmatic Hadean zircons from Jack Hills, Western Australia; Hopkins et al., 2008, 2010; Bell 66 et al., 2015). Muscovite has been identified in Tertiary S-type zircons (e.g., Harrison and Wielicki, 2016), 67 but is rare as an inclusion phase in zircon even where muscovite is abundant in the matrix (e.g., Bell et al., 68 2018).

Finally, inclusion assemblages in zircon appear more "evolved" than their host rock matrix with
 respect to modal minerals. The abundance ratios among quartz, alkali feldspar, and plagioclase (QAP)

71 inclusion assemblages in zircon appear almost universally more felsic than the matrices of their host 72 granitoids – i.e., the inclusion assemblage is richer in guartz over feldspars and alkali feldspar over 73 plagioclase (Darling et al., 2009; Bell et al., 2018; Jennings et al., 2011; Faltys and Wielicki, 2020). However, 74 there is no obvious systematic relationship that would permit simple conversion of zircon inclusion QAP 75 to matrix QAP (Bell et al., 2018). The compositions of feldspar inclusions in zircon are also more evolved 76 (plagioclase more albitic and alkali feldspar more potassic) than matrix grains, while the compositions of 77 more mafic phases like biotite are similar between matrix and zircon inclusions (Jennings et al., 2011). 78 Apatite also displays similar composition between matrix grains and inclusions in both zircon and sphene 79 (Bruand et al., 2016).

80 1.1 Questions and approach

81 Primary magmatic mineral inclusion assemblages in accessory phases are almost certainly a 82 complex function of magmatic history, including 1) crystallization sequence, 2) magma chemistry, and 3) 83 physical proximity to other crystallizing phases. The latter is likely to be largely random and thus we don't 84 attempt to further evaluate crystal proximity effects in this study (with the exception of the 'pileup' 85 effect). We hypothesize that crystallization sequence and magma chemistry are likely to have more 86 systematic controls on the inclusion assemblages trapped by various minerals. If so, then these effects 87 may permit interpretation of the detrital record in terms of host petrology and merit further study, both 88 in zircon and in other accessory phases relevant to detrital investigations, such as monazite and sphene.

- In order to better understand the significance that mineral inclusion assemblages in detrital zircon
 and other minerals may carry with regard to their source, in this study we addressed the questions:
- 91 1) Do primary magmatic inclusion assemblages in accessory minerals exhibit characteristic changes
 92 as magma differentiation progresses?

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2) How well and how consistently can crystallization order and consequent changes in the primary magmatic inclusion assemblages with progressive crystallization be determined?

95 In order to separate the effects of crystallization sequence from magma chemistry, we selected several granitoids which contain multiple abundant minor and accessory minerals that are relatively 96 97 robust during detrital transport (i.e., zircon, magnetite, ilmenite, hornblende, orthopyroxene, monazite, 98 and thorite). We determined the mineral inclusion assemblages in these accessory minerals and 99 petrographically estimated crystallization order within individual host granitoids by examination of 100 mutual inclusions among accessory minerals (e.g., earlier mineral X is found as an inclusion species in later 101 mineral Y but vice versa less frequently). We also compared feldspar compositions among inclusions in 102 zircon, inclusions in mafic accessory phases, matrix grains, and post-magmatic alteration phases to better 103 evaluate the relationship of inclusion chemistry with crystallization sequence.

104 2. Materials and Methods

105 We selected 27 granitoids from the southwestern US, ranging from tonalite to leucogranite. 106 Nineteen samples are from the Cretaceous Peninsular Ranges Batholith (PRB), made up of arc granitoids 107 that range from oceanic affinity in the western PRB to continental affinity in the eastern PRB (e.g., Todd 108 et al., 2003). In particular, we sampled all four units of the normally zoned, late-stage, high-Sr/Y La Posta 109 plutonic complex in the eastern PRB (e.g., Clinkenbeard and Walawender, 1989), ranging from 110 hornblende-biotite tonalite to 2-mica granodiorite. We also sampled several units of the Cretaceous Big 111 Bear Lake Intrusive Suite, including the Cactus Flat quartz diorite, granodiorite of Hanna Flat, and inner 2-112 mica granite of Butler Peak (e.g., Barth et al., 2016). Finally, we included several samples of the highly evolved leucogranite of the Spirit Mountain Batholith (e.g., Walker et al., 2007) along with intermediate 113 114 volcanic and plutonic units from the Searchlight, NV area. The Spirit Mountain and Searchlight rock units 115 occur in the Colorado River Extensional Corridor (CREC), a Miocene continental rift (e.g., Howard and John,

116 1987). The sample textures are summarized in Table 1, and further details about their petrology, sampling
117 location, and geologic setting are provided in the online supplemental text. Zircon inclusion assemblages
118 in 4 of these samples were previously reported in Bell et al. (2018).

119 Hand samples were crushed to <0.425 mm and separated by panning and magnetic separation. 120 Accessory mineral phases were picked from the various magnetic separates, mounted in epoxy, and 121 polished to expose an interior surface. This selection process likely produced bias against especially small 122 accessory grains, as well as bias towards euhedral, easily-identifiable examples of each mineral. Our 123 sample preparation technique also does not allow us to distinguish between grains of accessory minerals 124 that were hosted as inclusions in larger crystals of the major and minor minerals and grains of accessory 125 minerals hosted at grain boundaries. We judged that the loss of this textural context was justified by the 126 rapid identification of many crystals of the studied minerals possible due to this sample processing.

127 Epoxy mounts of mineral separates were searched for mineral inclusions using a Tescan Vega 3 128 XMU scanning electron microscope and the inclusions were identified using energy dispersive X-ray 129 analysis. Each inclusion was characterized based on its textural relationship with its host: isolated and 130 away from cracks in the host (likely primary), in contact with cracks in the host (ambiguous as to whether 131 it is primary or secondary), or filling cracks (secondary). Only those inclusions isolated from cracks were 132 considered further. We used cathodoluminescence (CL) imaging of internal zircon structures to determine 133 whether inclusions were hosted in inherited cores or neo-grown magmatic zones. While obvious cores 134 are rare among the studied zircon suites, inclusions hosted within them were not considered further. 135 Where multiple styles of magmatic zoning were present in a suite (sometimes in multiple domains of the 136 same individual grains), we compare inclusion assemblages between the different textural categories. All 137 cathodoluminescence observations are noted in supplemental table S2. We undertook transmitted light 138 investigation of 3-dimensional cracks beneath the polished grain surfaces of most samples for zircon 139 hosts, in order to determine more certainly that these inclusions were isolated from the outside geologic

140 environment. Most of the other investigated phases were too opaque and CL-inactive for these methods 141 to shed additional light on the primary or secondary nature of their hosted inclusions. Although we were 142 unable to use transmitted light microscopy on the Fe-Ti oxides, for instance, when considering 143 uncertainties in the primary nature of apparently isolated inclusions in the oxides, we make reference to 144 the results for zircon and the magnitude of adjustments these incurred compared to the original BSE-145 derived estimations for the primary inclusion assemblage. In addition to adjustments to inclusion 146 assemblages due to subsurface cracks or embayments, the existence of multiple age domains in oxide or 147 mafic silicate phases may be investigable by chemical or structural mapping and may have surface 148 expression in, for example, cracks between domains. Phases with generally abundant isolated inclusions 149 were zircon, ilmenite, and magnetite. Hornblende, monazite, apatite, and thorite also occasionally had 150 large numbers of isolated inclusions. Sphene and biotite were investigated for several samples but did 151 not contain many isolated inclusions and are not discussed in detail. Inclusion counts for all samples are 152 provided in the online supplement (Table S1).

153 2.1 Constraints on magmatic histories

154 Previous petrologic, trace element, and isotopic investigations of the magmatic units we sampled 155 for this study provide context for the crystallization histories we interpret.

Peninsular Ranges Batholith: The Cretaceous Peninsular Ranges Batholith consists of a western zone with oceanic affinity that generally lacks evidence for significant crustal assimilation (Gromet and Silver, 1984) and an eastern zone with significant crustal assimilation (Gromet and Silver, 1984), to the point that the interior granites of the zoned La Posta pluton series in the eastern zone display many characteristics of S-type granite (Walawender et al., 1990). A transition zone between the western and eastern zones records deeper magma origins with less evidence than the eastern zone for crustal assimilation (Gromet and Silver, 1984). Many of the PRB units in this study were analyzed for zircon trace elements by Bell and Kirkpatrick (2021), who found evidence for relatively simple fractional crystallization
histories in the Alpine and Las Bancas tonalites of the western zone and likely extensive magma mixing
with variable amounts of crustal assimilation in the eastern transition zone and eastern zone. The eastern
PRB San Jacinto and La Posta plutons analyzed here show minimal inheritance of zircon from the Paleozoic
framework rocks of the eastern PRB (Bell et al., 2018; Kirkpatrick et al., 2020, 2022).

168 Big Bear Lake Intrusive Suite: Barth et al. (2016) investigated the magmatic history and assembly 169 of the late Cretaceous arc-related Big Bear Lake Intrusive Suite (BBLIS) through U-Pb, Lu-Hf, and trace 170 element measurements on zircon from various units along with textural and geochemical investigations 171 of other accessory minerals. This discussion is based on their findings. The BBLIS consists of a main 172 intrusive mass of granite with marginal granodiorites; related satellite plutons contain both dioritic and 173 granitic components. Inherited zircon cores from lower crustal Proterozoic and earlier Mesozoic magmas 174 are common in the main mass but not the satellite plutons. Neo-grown magmatic zircon in the central 175 granites appears to contain hotter, early magmatic zircon likely formed in a magma more similar to that 176 expressed in the satellite diorites, followed by cooler magmatic zircon grown in a more homogenized 177 later-stage magma.

178 Spirit Mountain Batholith: The Spirit Mountain Batholith is a large and notably well-exposed 179 Miocene magmatic body in the northern Colorado River Extensional Corridor which is zoned from quartz 180 diorite in the paleo-lower portion of the batholith to highly felsic leucogranite at the paleo-top of the 181 batholith (Walker et al., 2007). Petrography and zircon U-Pb geochronology (Walker et al., 2007) suggest 182 a long formation history through incremental recharge of new granitoid magma, followed by expulsion of 183 highly fractionated melt which rose to form the upper leucogranite and is preserved as many separate 184 dikes and sills. Zircon trace element geochemistry of the interpreted cumulate units and the leucogranite 185 suggest zircon from the leucogranite were often first formed in lower regions of the batholith, with their 186 rims recording on average more evolved melt chemistry despite the large compositional overlap between

leucogranite and cumulate zircon (Claiborne et al., 2010). The younger Mirage Granite intruded the lower
part of the batholith when it was partially crystallized (Walker et al., 2007).

189 Other CREC Samples: We sampled both the upper quartz monzonite unit of the Searchlight Pluton 190 and an undated lava from the nearby Highland Range. The lavas of the Highland Range are hypothesized 191 to be the eruptive products of the magma chamber which is now preserved as the Searchlight Pluton (e.g., 192 Colombini et al., 2011; Eddy et al., 2022), and range in composition from dacite to rhyolite (Colombini et 193 al., 2011). The Searchlight Pluton is a well-exposed Miocene granitic pluton in the northern Colorado 194 River Extensional Corridor, and zircon U-Pb geochronology of the upper unit shows a relatively narrow 195 range in ages between 16.25 and 16.75 Ma, with individual samples showing distinct age clusters 196 suggesting the preservation of incremental small intrusions (Eddy et al., 2022). While crystal-liquid 197 segregation leading to the formation of a highly differentiated leucogranite cap (similar to the Spirit 198 Mountain Batholith) occurred in the middle unit of the Searchlight Pluton, the upper unit does not appear 199 to have undergone this process unless the resulting highly differentiated liquid was efficiently transported 200 away during eruption or was mixed progressively into later intrusions (Eddy et al., 2022).

201 2.2 Mineral Compositions

202 Six granitoids (four from the Spirit Mountain Batholith and two from the PRB) with abundant 203 feldspar inclusions in zircon (and in two cases, other phases) were selected for feldspar compositional 204 studies. These samples were crushed to <0.425 mm and panned for a heavy mineral concentrate. Zircons 205 were extracted by heavy liquid and magnetic separation methods. Other accessory phases were not 206 mounted for these samples. Zircons were mounted in 1" diameter epoxy disks and polished to expose an 207 interior surface. Inclusions were identified by SEM and EDS, and feldspar inclusions were targeted for 208 electron probe microanalysis (EPMA). Thick section billets of each of these granitoids were also prepared 209 for EPMA. Mineral inclusion assemblages for these six granitoids were reported in Bell et al. (2018).

Feldspar compositions for free, non-included matrix grains and feldspar inclusions in other minerals arefound in the online supplement (Table S3).

212 We measured the major element compositions of feldspar using a JEOL-8600 electron microprobe 213 at UCLA with a 15 nA beam and a focused beam (<1 μm) for inclusions and small vein and exsolution 214 features and an unfocused beam for large, non-included phases in order to limit alkali loss. We measured 215 feldspar inclusions in zircon from our grain mounts, while we used the thick sections to measure feldspar 216 inclusions in hornblende and biotite as well as the free, non-included feldspars. We monitored secondary 217 fluorescence during inclusion measurements by monitoring Zr, which we used to subtract any SiO₂ signal 218 contribution from the host zircon. All analyses were renormalized to 100 wt. %.

219 2.3 Whole Rock XRF

Eleven samples were analyzed for whole rock major element and trace element chemistry by Xray fluorescence (XRF), and several samples had previously been analyzed for whole rock major element and trace element chemistry by XRF (Bell et al., 2017, 2018). For both sample sets, this work was undertaken at Pomona College and involved preparation of glass beads by double fusion of a 2:1 lithium tetraborate:rock powder mixture in graphite crucibles (method described by Poletti et al., 2016). Whole rock chemical data for all samples is found in the online supplement (Table S4).

226 2.4 SIMS U-Pb dating

U-Th-Pb analyses were undertaken for samples from the Big Bear Lake Intrusive Suite, given the high proportion of inherited zircon cores noted in previous studies (Barth et al., 2016). Analyses were undertaken on the CAMECA *ims*1290 ion microprobe at UCLA with Hyperion-II RF frequency ion source (Liu et al., 2018). A ca. 3 nA primary O_3^- beam was focused to a ca. 6 µm spot and accelerated with a primary voltage of -13 kV to a sample stage held at +10 kV. Analyses were made at a mass-resolving power of ~5000 to resolve molecular interferences on the Pb isotopes, and the relative sensitivities of Pb and U were calibrated using the standard zircon AS3 (e.g., Schmitz et al., 2003). Inclusions hosted in
inherited Precambrian or older Mesozoic zircon (e.g., Barth et al., 2016) were excluded from further
analyses. U-Pb data are collected in table S5 of the online supplement.

236 **3. Results**

237 Of the 27 studied samples, we report inclusion assemblages in zircon for all 27, magnetite for 15, 238 ilmenite for 22, hornblende for 9, orthopyroxene for 2, monazite for 3, and thorite for 2. In most of these 239 suites the number of isolated inclusions exceeded 20, but we also interpret several magnetite and 240 hornblende suites with between 10 and 20 isolated inclusions. The La Posta 2-mica unit alone yielded 241 abundant inclusions in apatite (n = 49), although one Las Bancas sample yielded a handful of isolated 242 inclusions in apatite (n = 17). While BSE imagery was used to distinguish primary and secondary inclusions 243 in all phases, we used CL imagery and visible light microscopy in addition for inclusions in zircon. This 244 resulted in minor changes to the estimated primary inclusion assemblages, with the vast majority of 245 samples falling within error of the BSE-derived assemblages in proportions of apatite and other major 246 mineral groups (see supplemental figure S1). Significant changes were more likely for populations with 247 smaller total numbers of inclusions. Changes were most common for apatite and polymineralic quartz + 248 K-feldspar + plagioclase inclusions (hereafter "QAP structures") in zircon. The former is largely due to the 249 high aspect ratio of apatite making it more likely to intersect a subsurface crack or the edge of the grain 250 at depth. The QAP structures were found in many samples to be in contact with host grain surfaces or 251 cracks at depth. However, most QAP structures that appear to be isolated based on BSE imagery are also 252 isolated from cracks or grain surfaces at depth. One overarching observation from CL imaging is that QAP 253 structures along with biotite inclusions were more likely than other included phases to be surrounded by 254 a CL-bright rim or to be found within a larger CL-bright patch overprinting the host's internal magmatic 255 texture. In the case of larger patches we do not further consider these inclusions as they may have formed 256 following alteration. We interpret the thinner rims (which follow the shape of the inclusion) as either

arising as an artifact of topography or reflecting post-crystallization processes (perhaps diffusion of water)
not likely to alter the primary mineralogy of the inclusion. We note the presence of these rims in
supplemental table S2.

260 U-Pb dates for two units in the main intrusive mass of the Big Bear Lake suite were used to exclude 261 pre-magmatic grains (see supplemental table S5). Although 39% of dated zircon in the inner granite of 262 Butler Peak were of Precambrian or earlier Mesozoic age (similar to the findings of Barth et al., 2016), 263 these older ages were much more likely to be found among grains with patchy, chaotic, or absent CL 264 zoning which were unusually bright or dark compared to the rest of the zircon suite. 88% of dated 265 oscillatory zoned grains in the granite of Butler Peak were of magmatic age, with average age 76±2 Ma (2 266 s.e.) and a range from 62 to 85 Ma. The handful of younger zircons (<70 Ma) may represent Pb loss, but 267 the young age and moderate common Pb contamination complicates the accurate calculation of 268 discordance for this zircon suite. Sampled zircons from the granodiorite of Hanna Flat display oscillatory 269 zonation and all dated zircons were of magmatic age (78±2 Ma, 2 s.e.). For undated zircons, we accepted 270 inclusion counts from zircons with oscillatory zoning like that in the dated magmatic grains.

271 Among the 27 samples, the apparent relative order of crystallization was interpretable in 20 272 samples between at least two of the studied accessory minerals, and in several cases among 3 or 4. 273 Relative crystallization order was determined by comparison of mutual inclusion occurrence. In general, 274 in cases where the proportion of phase A as an inclusion in phase B exceeded the reverse case (i.e., phase 275 B as an inclusion in phase A), we recorded phase A as beginning to crystallize earlier than phase B. When 276 the inclusion suite proportions for phase A and phase B did not differ significantly within error (using 1 o 277 based on counting statistics), we consider the crystallization order ambiguous. When there are no 278 inclusions of phase A in phase B or vice versa we also consider the order ambiguous.

279 The proportions of apatite and various classes of silicate mineral inclusions in each mineral suite 280 are tallied in Table 1 (orthopyroxene and apatite not included) and representative cases are shown in 281 Figures 1 and 2. The class "late silicates" includes guartz, K-feldspar, and muscovite. The class "mafic 282 silicates" includes pyroxene, hornblende, sphene, biotite, and chlorite (the chlorite is likely an alteration 283 product of biotite and is thus counted as a primary mafic silicate). Mafic silicate inclusions are not tallied 284 for the mafic accessory minerals hornblende and orthopyroxene in Table 1 or Figs. 1 and 2. This is because 285 hornblende itself is a fairly widespread inclusion in the other accessory minerals and we wished to define 286 the categories "mafic silicate" and "late silicate" in the same manner for all reported host species. Older 287 hornblende cores included in younger hornblende crystals will be difficult to identify with BSE imaging 288 and may form under different influences compared to inclusions of species other than the host. All 289 inclusion data are presented in the online supplement (Table S1), and more details as to how the 290 crystallization sequence was determined for individual samples are also included in the online 291 supplemental text. All uncertainties on inclusion abundance are 1o based on counting statistics.

292 In general, apatite as an inclusion is overrepresented in most of the studied accessory minerals 293 compared to its volumetric abundance in the host rock. Apatite makes up >5% of inclusions in almost all 294 cases. The exceptions are thorite and one instance of apatite-free ilmenite in the Spirit Mountain 295 leucogranite, along with an instance each of apatite-free orthopyroxene and ilmenite in two mafic-296 intermediate Western PRB granitoids (Table S1). Ilmenite tends to contain the highest proportions of 297 mafic silicate inclusions, even when it appears to occur relatively late in the crystallization sequence (see 298 Fig. 1c,d). Free grains of apatite not included in other minerals were inclusion-poor in most samples, with 299 rare exceptions including one of the tonalites of Las Bancas and the inner two units (small-biotite and 300 muscovite-biotite) of the La Posta pluton (Eastern PRB), the latter for which we report inclusion totals 301 (Table S1).

302 Geochronologically useful inclusion phases are typically of low abundance in the inclusion suite. 303 Sphene, allanite, xenotime, and monazite are absent from many inclusion assemblages but might 304 constitute up to several percent in a few samples. Zircon is a rare inclusion phase in the other accessory 305 minerals and usually abundant at most at the few percent level. Several granitoids with relatively early-306 crystallizing zircon are exceptions: the oxides of the Butler Peak granite (Big Bear Lake Intrusive Suite) and 307 phosphates of the La Posta muscovite-biotite unit all contain >20% zircon in their inclusion assemblages, 308 with free apatite grains in the La Posta muscovite-biotite unit containing an inclusion assemblage that is 309 73% zircon. In the Las Bancas tonalite (LB2) and a mafic-intermediate Japatul Valley unit (JV2) of the 310 western PRB, early-crystallizing magnetite and ilmenite contain inclusions of baddeleyite (ZrO₂), but not 311 of zircon. We did not identify baddeleyite in mineral separates or as an inclusion phase in orthopyroxene 312 or zircon in this unit. Baddeleyite is also a minor phase included in and embaying ilmenite in the Alpine 313 Tonalite (AT1) of the western PRB, but is not observed elsewhere in our sample set. It is possible these 314 small enclosed baddeleyite grains result from exsolution (e.g., Naslund, 1987) instead of originating as 315 free baddeleyite grains trapped from the surrounding melt.

316 3.1 Textural studies

317 As shown in Table 2, in 11 out of the 20 samples in which relative crystallization order could be 318 established, the apatite proportion of the inclusion assemblage decreases as crystallization progresses 319 ('Group A' in Table 2). Figure 1 shows four such examples with a variety of crystallization sequences. Ten 320 out of the 23 samples with a relatively clear crystallization order have equal apatite contents within error 321 among the various minerals (examples in Fig. 2a,b; Group C in Table 2). Only four of these 20 samples 322 show the case of increasing apatite with progressive crystallization (Group B in Table 2): one granodiorite 323 from the San Jacinto pluton (RG1; Fig. 2c) contains earlier-crystallizing ilmenite which is apatite-poor 324 compared to later-crystallizing zircon. Similarly, an intermediate lava from the Colorado River Extensional 325 Corridor (HR1) contains later-crystallizing but apatite-richer zircon compared to magnetite. Two samples 326 from the Spirit Mountain leucogranite in the Colorado River Extensional Corridor contain both increasing 327 and decreasing apatite relationships. Based on mutual inclusions, in sample SpLa ilmenite appears to 328 begin crystallizing first, followed by magnetite, and later monazite. Observations in thick section suggest 329 that ilmenite may have also continued crystallizing after magnetite - in addition to euhedral ilmenite in 330 the matrix, ilmenite is also commonly observed rimming magnetite in a manner suggestive of late growth. 331 Other samples of the leucogranite have texturally ambiguous chronological relationship between 332 magnetite and ilmenite based on mutual inclusions. Apatite inclusion contents increase from ilmenite to 333 magnetite and then decrease in monazite. The leucogranite samples are consequently included in both 334 Groups A and B in Table 2.

335 Abundances of mafic and late-crystallizing silicates also display large variations in the inclusion 336 suites of various accessory minerals (Figs. 3-4). Zircon inclusion suites in this study follow the general 337 rough negative correlation between apatite and late silicate (i.e., quartz, K-feldspar, and muscovite) 338 abundances (Bell et al., 2018), while ilmenite inclusion assemblages show a similar trend but are offset to 339 lower late-silicate abundances for similar apatite contents. For both, the correlation is approximate, with 340 $r^2 = 0.77$ for zircon and 0.65 for ilmenite (Fig. 3). Magnetite inclusion assemblages do not convincingly 341 show a relationship between apatite and late-crystallizing silicates (Fig. 3), and there are too few examples 342 of other host phases to assess their behavior. On the other hand, most minerals show no relationship 343 between mafic silicate and apatite abundance, while ilmenite shows only a weak positive correlation between the two (Fig. 3b; $r^2 = 0.3$). These results contrast with the lack of relationship between late-344 345 crystallizing silicate inclusion phases and whole rock SiO₂ in any of the studied minerals (Fig. 4a). Mafic 346 inclusion phases show a negative relationship with whole rock SiO₂ in ilmenite, but there is little 347 discernible relationship for magnetite. Zircon inclusion assemblages only become more mafic-rich with 348 SiO_2 below 65%, showing no relationship with SiO_2 for more felsic granitoids (Fig. 4). When mafic silicate 349 and Fe-Ti oxide (including magnetite, ilmenite, and rutile) inclusions are tallied together, this category shows a similar relationship with whole rock SiO_2 (Fig. 5). However, the higher mafic + oxide tally for zircon from intermediate magmas does help to distinguish them from other low-apatite suites derived from highly felsic magmas (Bell et al., 2018), which have resolvably lower mafic + oxide totals (Fig. 5).

353 Several previous studies have noted that inclusion suites in zircon tend to be proportionally richer 354 in quartz and alkali feldspar relative to plagioclase than the whole rock (Darling et al., 2009; Jennings et 355 al., 2011; Bell et al., 2018; Faltys and Wielicki, 2020). Because zircon is a relatively late-crystallizing phase 356 in most magmas (e.g., Boehnke et al., 2013), its capture of a more evolved major mineral inclusion 357 assemblage might be mainly a function of its place in the crystallization order, so we compare it with the 358 other studied host minerals. The ratio of quartz to total quartz + felspar (Q/QAP) and the ratio of K-359 feldspar to total feldspar were measured for the various accessory minerals and vary widely, but there is 360 little systematic relationship between inclusion Q/QAP and either crystallization order or apatite 361 abundance between samples and only locally within a small number of samples (notably the Spirit 362 Mountain leucogranite; see figures in online supplemental text). However, for the majority of the accessory minerals, the inclusion assemblage is proportionally more quartz- and K-feldspar-rich than the 363 364 whole rock (Fig. 6). Orthopyroxene is a notable exception, with both examples (AT2, LB2) being relatively 365 poorer in quartz and K-feldspar compared to their whole rock CIPW norms (see Fig. 6).

366 Apart from the major minerals, muscovite and sphene inclusions may also yield significant 367 information in a detrital context. Muscovite inclusions can serve as an indicator for peraluminous host 368 rocks, while sphene is characteristic of calcic metaluminous rocks, can be dated by U-Pb, and can provide 369 P-T conditions of formation (Hayden et al. 2008). In our muscovite-bearing granitoids, muscovite is a rare 370 inclusion phase (cf., Harrison and Wielicki, 2016), typically found only in the later-crystallizing accessories. 371 It is found in ilmenite but not zircon in the Butler Peak granite (BB14), La Posta 2-mica unit (LP6), and in 372 one felsic sample from the western PRB (JV1). Sphene occurs as a separate free grain in several samples 373 and often constitutes a minor part of the inclusion assemblage. Aside from primary inclusions, however,

374 sphene is commonly found embaying other minerals or as rims on oxides. This may suggest its relatively
375 late position relative to these other phases in the crystallization sequences of these granitoids.

376 *3.2 Feldspar chemistry*

377 Matrix feldspars range widely in composition across the 6 analyzed samples (Fig. 7). In the San 378 Jacinto samples, inclusions in zircon and other minerals range to more evolved chemistry (higher 379 orthoclase content for K-feldspar; higher albite content for plagioclase) than the cores of matrix feldspar 380 grains. However, in the Spirit Mountain zircons, K-feldspar inclusions range widely compared to the 381 matrix grains and are not systematically more potassic. A handful of alkali feldspar inclusions in both the 382 San Jacinto pegmatite (RG6) and Mirage Granite samples range to much more albitic compositions (45-383 60% Ab) than matrix grains. It is unclear if these represent true K-feldspar compositions or the analysis of 384 mixed phases, so they are not included in Figure 7. Feldspar inclusions are similar in composition to post-385 magmatic alteration phases (veins and plagioclase exsolution lamellae in K-feldspar), although these 386 alteration phases range to nearly pure albite and orthoclase even when inclusions do not (Fig. 7). 387 Plagioclase inclusions in zircon are more albitic on average than inclusions in either hornblende (sample 388 IG1) or biotite (sample SpLb).

389 *3.3 Oxide textures and peculiarities*

Magnetite and ilmenite are in high enough abundance and rich enough in inclusions to routinely yield useful inclusion counts. Some exceptions exist: magnetite is absent in most eastern PRB samples, consistent with previous observations (e.g., Gastil et al., 1990) and ilmenite is sparse in some of our oxidized granitoids. Both magnetite and ilmenite show several varieties, either texturally or chemically distinct. First, magnetite in many (although not all) samples displays parallel linear bands of an Mn-Ti-Fe phase. These are morphologically suggestive of exsolution lamellae (see Fig. 9), but given the structural differences between magnetite and ilmenite they are unlikely to be directly exsolved ilmenite. They 397 instead probably represent an ulvospinel component incorporated into the magnetite under magmatic 398 conditions, which may have undergone oxidation to magnetite + ilmenite during subsolidus cooling 399 (Buddington and Lindsley, 1964). While it is also possible that some apparent magnetite grains with Mn-400 Ti-Fe lamellae, such as in Fig. 9c, may instead be originally grains of ilmenite that exsolved structurally 401 similar hematite (e.g., Haggerty, 1991), the vast majority of our Fe oxide host grains were picked from the 402 portion of the heavy mineral separate that responded to a hand magnet, suggesting that the vast majority 403 is in fact magnetite. In all cases, given the EDS similarity of the Fe-Ti-Mn lamellae to the Mn-rich free 404 grains of ilmenite observed in most of the rocks, an inclusion of ilmenite in magnetite is only tallied when 405 it appears to have a morphology distinct from the lamellae in its host magnetite. Ilmenite occurs in most 406 samples with a noticeable Mn peak in its EDS spectrum. In the La Posta 2-mica unit, there appear to be 407 multiple generations of ilmenite, with Mn-rich cores often surrounded by a Mn-poor rim. In many 408 samples, regions within ilmenite grains contain fine-grained intergrowths of separate Fe and Ti oxides (Fig. 409 9a). The irregular textures of these regions and their frequent occurrence on or near cracks or grain edges 410 suggest that they are alteration features.

411 3.4 Rare mineral inclusion phases by rock type

412 As mentioned above, abundant baddeleyite inclusions in relatively early-crystallized oxide phases 413 appears to be characteristic of the mafic-intermediate samples. Among the more highly evolved units 414 including the Spirit Mountain leucogranite and the granite of Butler Peak, there are inclusions of Mn and 415 Zn oxides, Nb-rich phases, anhydrite, and halides within several minerals. The Nb, Mn, and Zn-rich phases 416 tend to be concentrated in the later-crystallizing accessory minerals: ilmenite and monazite in the granite 417 of Butler Peak, and monazite and thorite in the Spirit Mountain leucogranite. These phases are also 418 widespread in ilmenite of the Spirit Mountain leucogranite, which is suspected on the basis of mutual 419 inclusions to have begun crystallizing early but based on thick section textural information probably 420 persisted late into the crystallization history. Halide inclusions are common in earlier-crystallizing zircon

and magnetite of the Spirit Mountain leucogranite. Sulfide minerals are minor or absent in most inclusion
assemblages, and their presence does not appear to correlate with the whole rock chemistry. Where they
are present as inclusions, they are included in most of the studied accessory minerals. EDS spectra suggest
the presence of Fe, Cu (with and without Fe), and Zn sulfides in various samples, with Fe sulfides being
most common.

426 4. Discussion

427 The high frequency of apatite inclusions in most studied accessory phases is similar to its common 428 observation as an inclusion phase in major minerals, such as plagioclase. The abundance of apatite 429 inclusions in plagioclase and other magmatic phenocrysts is often attributed to the local saturation of 430 apatite against growing mineral surfaces due to the slow diffusion of the phosphate complex in silicate 431 melts and resulting 'pileup effect' (e.g., Gill, 1978; Woodruff et al., 1979; Rose et al., 1980; Green and 432 Watson, 1982; Harrison and Watson, 1984; Bacon, 1989). As such, the variations in apatite proportions 433 across the inclusion suites of multiple minerals within a single magma should yield insight into the extent 434 to which crystallization order or other factors such as magma chemistry or crystal growth rate exert 435 dominant control over the apatite inclusion abundance. Rare but petrologically useful inclusion phases 436 such as muscovite, sulfides, Nb or REE phases, and halides, can also be better studied in the context of 437 multiple mineral suites. For example, muscovite's more common occurrence in late-crystallizing phases 438 may help to explain its rarity in detrital zircon compared to other markers of peraluminous magmatic 439 origins.

In addition to assessing inclusion assemblage behavior with progressive differentiation, identifying mineral-specific effects and apparent preferences for certain inclusion phases will help to better interpret inclusion suites in detrital samples of other accessory minerals. For example, regardless of its position in the crystallization sequence, ilmenite tends to contain a higher proportion of mafic phases

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(see Figs. 1, 2) and usually a higher Q/QAP (see Fig. 6) than other studied minerals from the same magma.
This mineral-specific affinity for mafic phases could be due to ilmenite being more likely to saturate in the
vicinity of other Fe- and Ti-bearing phases.
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447 4.1 Interpretation of polymineralic inclusions

448 Both monomineralic and polymineralic inclusions are common in the various hosts, and the 449 pattern of mineral occurrences may have implications for inclusion genesis. Monomineralic inclusions are 450 most common. As an example, 60% of inclusion-bearing zircons in this study contain only a single inclusion 451 species exposed at the polished surface (see supplemental table S2). Zircon suites range widely in this 452 measure, with the rate of single-species zircons between 15 and 89% across the 27 studied rocks. For 453 zircon grains (and other accessory mineral grains) containing more than one inclusion species, 454 polymineralic inclusions are common. As mentioned in the preceding section, globular inclusions 455 containing quartz, K-feldspar, and plagioclase ("QAP structures") – less commonly with volumetrically 456 minor mafic silicates or oxides – are the most common polymineralic inclusions in all host species. When 457 quartz is not present, K-feldspar and plagioclase are often found in contact within the same inclusion. In 458 zircon hosts, magnetite and ilmenite are often found in contact. By contrast, apatite inclusions in all host 459 species are mostly monomineralic.

Globular QAP structures can be imaged below the grain surface in most zircons. These structures may be self-contained and isolated from cracks or grain surfaces, but in some cases they are part of larger embaying structures in contact with a grain surface at depth. This is likely to be the case also for the opaque accessory minerals. We interpret these QAP structures as crystallized melt pockets, either inclusions captured by the zircon during its growth or pockets in which melt remained in contact with the magma potentially throughout host crystallization. QAP embayments could potentially also result from re-melting (and subsequent re-crystallization) of glass or late-crystallizing magma components during later reheating, potentially accompanied by local zircon dissolution. QAP structures associated with disturbed CL zoning (e.g., bright patches as seen surrounding some inclusions) are particularly suspect for this. Quartz, K-feldspar, and plagioclase are also present as monomineralic inclusions. Monomineralic inclusions may arise by capture of pre-existing crystals from the melt during host grain growth. For species such as apatite, monomineralic inclusions may also arise as a result of host grain growth, by local saturation in the inclusion phase due to piling up of slow-diffusing chemical species against the growing host grain surfaces (e.g., Green and Watson, 1982; Harrison and Watson, 1984).

474 *4.2 Apatite dominance and crystallization order*

475 Samples with clear crystallization sequence relationships between at least two minerals are about 476 equally likely to show no significant differences in apatite contents among the studied minerals and to 477 show decreasing proportions of apatite in the inclusion suite with progressive crystallization, with only 478 four out of 27 samples showing apatite increase. The relatively early crystallization of abundant small 479 apatite crystals is expected in the presence of growing modal crystals due to the remarkably slow 480 diffusivity of the large P complex (likely PO₄M⁺³; Ryerson and Hess, 1980; Ryerson, 1985) compared to 481 most other components in silicate magmas (e.g., Harrison and Watson, 1984; Zhang et al., 2010). This 482 causes a local "pileup" of P at the surfaces of growing modal, P-free, crystals leading to a local saturation 483 in apatite, even when most parts of the magma may remain apatite-undersaturated (e.g., Green and Watson, 1982; Harrison and Watson, 1984). 484

For an example calculation to demonstrate the pileup effect, we use the Si diffusion data of Baker (1991) for a 70% SiO₂ melt with 3% H₂O and Arrhenius data for Zr and P (Watson and Harrison, 1983; Harrison and Watson, 1984, respectively) scaled to a viscosity equivalent of 70% SiO₂ and 3% H₂O (Mungall, 2002). We further assume an Al/Si diffusivity ratio of 2 and that the Baker (1991) data approximates effective binary diffusion rates for the range of expected chemical gradients in the magma (Zhang et al., 2010). Calculated diffusivities of Si and Al at temperatures approaching the onset of zircon
saturation in the magmas under consideration (ca. 800°C) are in the range 2-4 x 10⁻¹⁰ cm²/sec with Zr and
P two and seven orders of magnitude lower, respectively. Diffusivities of alkalis and Ca are all several
order of magnitude higher than Si under these conditions (Mungall, 2002).

Modal phase growth rates in the neighborhood of 10⁻¹² cm/sec (Brandeis and Jaupart, 1987) 494 495 would result in 5 mm crystals forming in ca. 10^4 years – roughly the timescale for a 10-km diameter 496 hydrous pluton to cool from liquidus to solidus with no additional magma input (Spera, 1980). The above 497 kinetic data are consistent with Si and Al maintaining diffusive equilibrium over that lengthscale in a few 498 10s of years and a few thousands of years for Zr. Thus local kinetic effects are unlikely to result in 499 disequilibrium crystallization of feldpar, quartz or even zircon in their growth boundaries that could then 500 be incorporated as inclusions. However, the very low phosphorous diffusivity will likely lead to P "pileup" 501 adjacent the growth surfaces of early crystallizing modal phases (e.g., plagioclase) leading to local apatite 502 saturation (Green and Watson, 1982).

503 If most apatite inclusions result from the pileup effect, then the pattern of decreasing apatite 504 inclusion abundance with progressive crystallization is the expected result of continual local saturation 505 and sequestering of apatite within other minerals and the consequent depletion of P in the remaining 506 melt. Efficient small-scale stirring of the magma will enhance the appearance of this systematic phosphate 507 depletion by better homogenizing the melt outside the growing crystal face-adjacent boundary layers, 508 while more viscous magmas (e.g., more siliceous) will be more likely to develop substantial local 509 heterogeneities. Notably, there are regional variations in the frequency of the "decreasing apatite 510 inclusions" vs "indistinguishable apatite frequency" patterns. Within the PRB, the generally hotter and 511 less silicic (e.g., Morton et al., 2014; Bell and Kirkpatrick, 2021) magmas from the Western Zone show 512 many examples of clear decrease in the proportions of apatite inclusions in accessory phases during 513 progressive crystallization. Eastern Zone plutons are generally more silicic (e.g., Morton et al., 2014), 514 contain lower-temperature zircon (Bell and Kirkpatrick, 2021) and mostly lack significant changes in 515 apatite inclusion content with progressive crystallization. These regional differences also extend to the 516 average apatite inclusion content of all studied minerals (Fig. 10): while there is a large range in apatite 517 inclusion contents in all regions, the Western PRB and CREC samples tend to have lower apatite inclusion 518 contents in all studied minerals compared to the Eastern PRB and Big Bear Lake samples. Whether this is 519 also due to regional chemical or petrogenetic variations is an interesting path for future study.

520 Notably, all of the granitoids in this study are intermediate to felsic and have whole rock 521 compositions at least 59% SiO₂, so it is unclear whether viscosity differences would be sufficient to explain 522 the differences in apatite behavior. Instead, it may be helpful to refer to known magmatic histories from 523 each region. Most rocks showing clear apatite decreases are either from the western PRB or are highly silicic, late-stage differentiates within a larger magmatic complex with a range of compositions (Spirit 524 525 Mountain Leucogranite, granite of Butler Peak, La Posta 2-mica unit). Western PRB and CREC magmas 526 probably underwent fractional crystallization without much crustal assimilation or magma mixing (Gromet 527 and Silver, 1984; Todd et al., 2003; Kistler et al., 2014; Bell and Kirkpatrick, 2021; Claiborne et al., 2010). 528 Although magma mixing probably contributed to the origins of the Big Bear and La Posta suites (Barth et 529 al., 2016; Bell and Kirkpatrick, 2021), zircon from the inner granite of Butler Peak appear to record the 530 evolution of a more isotopically homogenized melt compared to other components of the batholith (Barth 531 et al., 2016). Similarly, shallow crystal fractionation processes appear to dominate the zircon trace 532 element history of the La Posta 2-mica inner unit, in contrast to zircon from the outer units of the pluton 533 (Bell and Kirkpatrick, 2021). By contrast, the less evolved units of the eastern PRB, the PRB transition zone, and the BBLIS appear to record substantial magma mingling without homogenization (Bell and 534 535 Kirkpatrick, 2021; Barth et al., 2016). These units are also more likely to lack clear inclusion evidence for 536 crystallization order. Where clear orders can be determined, apatite is usually a similar proportion of the 537 inclusion assemblage in all phases.

538 An important aspect of the pileup effect with regards to apatite inclusion abundance is 539 dependence of local saturation on the diffusivity of P vs the rate of growth of the crystal in question (e.g., 540 Smith et al., 1955). Faster growth rates relative to diffusivity will lead to sharper gradients in phosphate 541 concentration against the growing crystal face and to ultimately higher P concentrations at the interface 542 (Smith et al., 1955). Accessory minerals often have lower growth rates (or dissolution rates) than modal 543 minerals in magmas (Bacon, 1989; Watson, 1996), owing to the relatively slow diffusion of many trace 544 constituents that compose essential structural components of the accessory minerals compared to major 545 elements (e.g., Zhang et al., 2010). Therefore, inclusion suites among accessory minerals and major minerals may differ substantially and directly comparing them in the detrital record requires further study. 546 547 Compositionally moderated changes in the relative diffusivities of various elements may also play a role 548 in setting inclusion suites and particularly the proportion of apatite inclusions. Notably, although the 549 diffusivities of most elements increase with increasing water contents, this effect is enhanced for slower-550 diffusing species, with a good example being the marked increase in P diffusivity in hydrous compared to 551 anhydrous melts (Harrison and Watson, 1984).

552 The dependence of local saturation on the diffusivity of P versus the growth rate of the 553 prospective host crystal introduces the possibility that co-crystallizing phases with different growth rates 554 might record different apatite contents despite crystallizing simultaneously. Additionally, the lower 555 apatite inclusion abundance in later-crystallizing phases means that magmas which saturate zircon late in 556 their crystallization histories should tend to yield zircon with lower apatite inclusion abundances, as illustrated here by the Alpine and Las Bancas tonalites whose zircons contain 5-23% apatite. This would 557 558 be expected to produce a positive relationship between apatite inclusion content and whole rock SiO₂, 559 since zircon will tend to crystallize relatively later in mafic-intermediate magmas and relatively earlier in 560 felsic magmas (e.g., Boehnke et al., 2013). This expected positive trend is opposite to the rough negative 561 relationship identified by Bell et al. (2018): in that previous study, apatite inclusion abundances ~20% and

562	lower were only observed from highly silicic magmas while more intermediate granitoids yielded zircon
563	with higher apatite inclusion abundances. Consideration of an expanded dataset for inclusion suites and
564	whole rock chemistry will be needed to better constrain which of these trends is dominant.

565 *4.3 Other effects of crystallization order*

Several other aspects of zircon inclusion suites compared to whole rock mineralogy have been hypothesized as due to zircon's relatively late position in the crystallization order, including the higher Q/QAP and A/QAP proportions of most inclusion suites and the more evolved feldspar chemistry among inclusions (Darling et al., 2009; Jennings et al., 2011; Bell et al., 2018). The consideration of other minerals which crystallize mostly before or after zircon allows us to evaluate several of these hypotheses more closely. The behavior of the minerals apatite and muscovite are also of interest, especially in interpreting the earliest terrestrial detrital records (Hopkins et al., 2008, 2010; Rasmussen et al., 2011; Bell et al., 2015).

573 We observe more evolved plagioclase chemistry moving from (least evolved) the cores of larger 574 non-included, free plagioclase grains to inclusions in mafic phases (hornblende, biotite) to inclusions in 575 zircon to late-stage veins and exsolution in the free grains (most evolved). Hornblende in IG1 appears in 576 thin section as roughly euhedral to intergrown with plagioclase and quartz, and has been partially 577 chloritized in places (see Fig. 8). We were only able to identify 8 isolated inclusions in IG1 hornblende, 578 none of which were zircon, and we found only one primary hornblende inclusion out of 134 in IG1 zircon. 579 This makes a textural determination of their relative crystallization order difficult; other methods such as 580 comparison of trace elements between the two phases might be required for a more certain determination (e.g., Barnes et al., 2017, 2019); Ti-in-zircon crystallization temperatures as low as 651°C 581 582 were reported for the granite by Bell et al. (2019) demonstrating some late-stage zircon growth. IG1 583 hornblende's primary inclusion suite (n = 8) contains $38\pm22\%$ apatite, indistinguishable from zircon's 584 30±5% apatite. Biotite in SpLb is mostly euhedral in thin section (see Fig. 8). It contains inclusions of apatite, oxides, and some zircon in addition to plagioclase, so there is some overlap with zircon growth during crystallization. Co-crystallization is consistent with the minor biotite content in zircon's inclusion assemblage (ranging 0-2%; see table S2). The persistence of zircon to very low crystallization temperatures (730°C in SpLb of Bell et al., 2019; ~700°C in other samples of the leucogranite analyzed by Claiborne et al., 2010) leads us to infer that zircon crystallization probably occurred on average later than biotite.

591 Another potential effect of crystallization order may be on the proportion of late-crystallizing and 592 mafic silicates. In particular, late-crystallizing silicates show a negative relationship with apatite inclusions 593 in zircon, hornblende, and ilmenite. In zircon, apatite and late silicates together make up the majority of 594 the inclusion assemblage, although they rarely sum to 100%, and mafic silicates typically exceed late-595 crystallizing silicates by share in the inclusion assemblage of ilmenite (Figs. 3,4). Mafic silicates increase 596 in ilmenite with decreasing SiO₂, while in zircon this effect is only evident below 65 wt. %. Mafic silicates 597 show little relationship with apatite contents except among ilmenite, where the two are positively 598 correlated (Figs. 3, 4). Overall, crystallization order correlates reasonably well with the proportion of late-599 crystallizing silicate inclusions, with the caveat that the (generally early crystallizing) ilmenite-hosted 600 inclusion suites will be offset to even lower proportions of late-crystallizing silicate inclusions compared 601 to other early-crystallizing host phases in the magma (i.e., magnetite).

The observed feldspar chemistry fits a logical crystallization order with more evolved feldspars occurring apparently later in the crystallization sequence of accessory minerals. This is especially clear for plagioclase. Compared to the cores of free plagioclase grains in the rock, inclusions in accessory minerals are more albitic (Fig. 7), inclusions in these mafic phases are less albitic than inclusions in zircon in the same rocks. As expected, late to post-magmatic veins and exsolution features in feldspar are more albitic than primary plagioclase in the matrix, but are also more albitic on average than the inclusions in zircon. These patterns hold true to a lesser extent for K-feldspar, where there is more overlap among 609 compositions for matrix grains, inclusions, and late veins and exsolution features. Our inferences about 610 relative crystallization order can be tested by two-feldspar thermometry on samples with coexisting 611 plagioclase and alkali feldspar inclusions in multiple host phases, as in the older granite sample of San 612 Jacinto (IG1; ~110 Ma, Bell et al., 2018) which was later intruded by the San Jacinto Pluton. We calculate 613 example temperatures for feldspar pairs identified by host phase (see supplemental table S3), using an 614 estimated pressure of 6 kbars (based on estimates for eastern PRB wallrocks from Rothstein and Manning, 615 2003). The highest estimated temperatures are found in matrix grains (avg. 607°C), followed by inclusions 616 in hornblende (avg. 529°C) and inclusions in zircon isolated from cracks (avg. 422°C). The mostly 617 subsolidus temperatures point to either a lack of equilibrium between the plagioclase and K-feldspar or 618 to post-crystallization re-equilibration. It is possible this is due to reheating during later intrusion of the 619 San Jacinto Pluton.

620 Muscovite is typically one of the later-crystallizing minerals in evolving igneous complexes (e.g., 621 Scaillet et al., 1995) similar to the muscovite-bearing lithologies in our study. It is a rare inclusion phase 622 in our muscovite-bearing granitoids (Butler Peak granite; Spirit Mountain leucogranite; La Posta 2-mica 623 unit) but is present at the several percent level in some later-crystallizing accessory minerals. For the 624 muscovite-bearing granitoids in our sample suite, magnetite and ilmenite typically occur later in the 625 crystallization sequence than zircon and contain muscovite which we interpret as primary (i.e., not in 626 contact with cracks and embayments). Despite the late appearance of monazite in our samples, we do 627 not observe it to contain muscovite inclusions. It will be useful to determine the generality of this 628 observation that monazite, and perhaps zircon, are unlikely carriers of muscovite inclusions in 629 metaluminous to weakly peraluminous rocks. The abundant primary muscovite inclusions in >4 Ga zircons 630 from Jack Hills, Western Australia (Hopkins et al., 2008, 2010; Bell et al., 2015; cf. Rasmussen et al., 2011) 631 have been interpreted as evidence for felsic peraluminous Hadean magmas. In light of the results above 632 regarding muscovite inclusion occurrence, an explanation for the very muscovite-rich inclusion

633 assemblages in Hadean Jack Hills zircons is required. Jack Hills zircons display unusually low Ti-in-zircon 634 crystallization temperatures compared to most terrestrial magmas (Watson and Harrison, 2005), which 635 potentially points to a relatively late onset of crystallization for these zircons in their host magmas 636 compared to the zircon suites in this study. Consequently, a larger share of inclusions in the Jack Hills 637 zircons would be the very late-crystallizing silicates such as muscovite compared to zircon suites that 638 began to crystallize at higher temperatures and thus include a larger proportion of earlier phases. By 639 contrast, the weakly peraluminous granites studied here may not have become peraluminous, much less 640 saturated in muscovite, until relatively late in zircon's crystallization history and after the crystallization 641 of the vast majority of the zircon. A notable exception to the generally higher temperatures of igneous 642 zircon as seen here is zircon found in certain low-temperature Himalayan leucogranites in which trace 643 element chemistry indicates vapor-present anatectic melting (Harrison and Wielicki, 2016). Zircons from 644 these leucogranites contain abundant primary muscovite inclusions. It is possible that vapor-present 645 anatectic melting is a scenario uniquely suited to creating zircon suites with muscovite-rich inclusion 646 assemblages.

647 In some cases, our inferred crystallization orders based on mutual inclusions can be compared to earlier work on crystallization sequence using different methods. Barth et al. (2016) calculated 648 649 crystallization temperatures for sphene and zircon in the main intrusive suite of the Big Bear Lake Intrusive 650 Suite, using the methods of Hayden and Watson (2008) and Ferry and Watson (2006), respectively, and 651 determined that on average primary magmatic sphene – which appears texturally to overgrow earlier 652 magnetite and ilmenite – crystallized after the majority of zircon had crystallized. We find that in both 653 the granodiorite of Hanna Flat and the granite of Butler Peak, zircon appears to begin crystallizing before 654 magnetite, and also, in the granite of Butler Peak, before ilmenite. Both sequences suggest a relatively 655 early crystallization for zircon consistent with the relatively high zircon saturation temperatures derived 656 from whole rock geochemistry. The paucity of sphene inclusions in zircon (similar to magnetite and

ilmenite) is consistent with the later crystallization of all three phases relative to zircon, while the paucity
of sphene inclusions in magnetite and ilmenite is also consistent with sphene crystallizing on average later
than the oxides, as observed more generally in some hydrous ilmenite-bearing magmas (e.g., Frost et al.,
2001).

661 4.4 Quartz, K-feldspar, and plagioclase (QAP shifts)

662 Neither the expected crystallization order in an intermediate or granitic magma (e.g., Whitney, 663 1975) nor the observed crystallization order among studied accessory minerals appears to predict the 664 proportions of the major phases – quartz, K-feldspar, and plagioclase – as inclusions (Darling et al., 2009; 665 Jennings et al., 2011; Bell et al., 2018). The general observation of higher A/P and to a lesser extent Q/AP in included phases relative to the whole rock could simply reflect, in the case of a late appearing accessory 666 667 phase such as zircon, the more felsic melt composition expected in the later stages of an evolving system. 668 In this case earlier-crystallizing phases could reasonably be expected to show lower Q/AP and A/P shifts 669 than later-crystallizing phases forming in more evolved melt. However, although most of the accessory 670 minerals in this study do indeed have inclusion assemblages richer in A/P (and, as noted, Q/AP to a lesser 671 extent) than their respective whole rocks, there is no overall clear relationship among A/P, Q/AP and 672 crystallization order, nor between Q/AP and apatite content (e.g., Fig. S2). There are individual 673 exceptions, such as the Spirit Mountain leucogranite, where overall earlier crystallizing phases such as 674 magnetite have lower Q/AP than later phases ilmenite and latest phase monazite (Fig. S2) – however, this 675 is not the case for most samples. Ilmenite may in fact be more likely to show high Q/AP regardless of 676 position in the crystallization sequence (Figs. 6, S3). Similar to ilmenite's affinity for capturing mafic silicate 677 inclusions, ilmenite may be less likely to capture feldspar. A more general process not dependent on 678 degree of magma differentiation may instead lead to the higher Q/AP and A/P among inclusion 679 assemblages. As discussed in a previous section, many inclusions identified in this study intersect the 680 polished grain surface with circular or elliptical geometry and contain guartz with two feldspars, often

without other phases present or only small crystals of additional phases. It is likely that these represent melt inclusions that subsequently crystallized (e.g., Thomas et al., 2003). However, isolated single-phase inclusions of all three minerals also occur and these monomineralic inclusions are responsible for the Q/AP and A/P shifts between the whole rock and the inclusion suite. Although most samples in this study are tonalitic to granodioritic (see Fig. 6), a lesser A/P shift for more silicic granitoids is demonstrated by Bell et al.'s (2018, Figs. 5, 7) survey of zircon inclusion assemblages in a variety of granitoids spanning a wide range in composition.

688 It is possible that differing nucleation and growth rates for quartz, alkali feldspar, and plagioclase, 689 independent of the host phase's position in the magma crystallization history, drive their differing 690 proportions between the matrix and as inclusions in various accessory minerals. For example, Swanson's 691 (1977) experiments with hydrous granitic and granodioritic melts revealed differing nucleation and 692 growth rates for quartz, alkali feldspar, and plagioclase as a function of bulk composition, degree of water 693 saturation, and degree of undercooling. While high growth rate coupled with low nucleation density should lead to visible matrix crystals, lower growth rate and possibly higher nucleation density might yield 694 695 the small isolated crystals that would be captured as inclusions. While high undercoolings can produce 696 these conditions for all three phases (Swanson, 1977), at low undercoolings granodioritic melts show high 697 plagioclase growth rates that may be especially incompatible with capture as inclusions compared to 698 quartz and K-feldspar. The lack of a high plagioclase growth rate in Swanson's (1977) granitic melt would 699 then suggest that more felsic magmas should yield accessory mineral inclusion suites with less of an A/P 700 shift compared to the whole rock.

There are many caveats to applying these experimental results to the plutonic record. For example, although plagioclase appears to nucleate at relatively low degrees of undercooling with quartz and alkali feldspar appearing later, this order simply reflects the governing phase equilibria given Swanson's (1977) choice of bulk compositions and quenching from an initial equilibrium temperature of 1000°C. Variations in pressure will often be correlated with cooling rates given that lower-pressure granitoids will reflect shallower intrusion, with accompanying faster cooling rates. Importantly, multiple competing mechanisms influence the outcome of such experiments. For example, as SiO₂ increases with initial plagioclase crystallization, so does melt viscosity, thus depressing diffusive transport of essential structural constituents. Melt viscosity further increases with degree of undercooling and decreasing water content. However, so does disequilibrium which can overcome kinetics to induce rapid crystallization (Couch, 2003).

712 Given uncertainty regarding the relative contributions of crystallization kinetics to crystallization order and to the availability of specific phases as small inclusions for accessory host minerals, it is 713 714 premature to predict host magma composition from inclusion QAP. A more focused study of QAP modal 715 shift vs. zircon crystallization temperature for different bulk compositions and pressures might better 716 elucidate whether these inclusion proportions can yield any specific petrologic information about the host 717 magma. As it stands, an inclusion suite's A/P ratio can be reasonably inferred to be higher than the whole 718 rock A/P, but a more specific relationship governing the extent of the shift is not yet clear, except that 719 shifts appear to be greater for inclusion suites from less silicic magmas.

720 4.5 Integrity of igneous inclusion record during later alteration

Following magma crystallization, the solid components will undergo a number of processes that may result in chemical or mechanical alteration, and it is worth considering the effects of such processes on the inclusion record. Late-stage magmatic and deuteric fluid circulation may chemically alter the accessory mineral host grains. Several oxide grains in this study appear to have undergone subsolidus exsolution, for instance (see Fig. 9), and hornblende investigated in thin section in this study had undergone varying degrees of chloritization (see Fig. 8). Deuteric alteration may also affect the chemistry of igneous zircon (e.g., Bell et al., 2019). However, Bell (2016) demonstrated that metamorphic zircon can 728 armor its feldspar and biotite inclusions against chemical re-equilibration with the surrounding, 729 metamorphic feldspar and biotite during amphibolite-grade regional metamorphism. Apparently 730 metamorphic phases (epidote, muscovite, chlorite) in the Bell (2016) samples were present and isolated 731 from cracks only in host zircon with disturbed U-Pb systems pointing to chemical alteration. Given the 732 relative temperatures involved, these results suggest that inclusions armored away from cracks in 733 chemically undisturbed zircon probably maintain their mineralogy and major element compositions 734 during deuteric alteration as well as regional metamorphism. Whether this is the case for other accessory 735 minerals will depend on their chemical and mechanical response to metamorphism. Another caveat is that Bell's (2016) analysis depended on identifying inclusions that remained isolated from cracks in the 736 737 host zircon following regional metamorphism and had no comparison from an unmetamorphosed 738 example of the protolith which could be used to assess, for example, the potential for preferential loss of 739 certain inclusions.

740 There is reason to believe that high-temperature metamorphism may alter the inclusion records 741 of some granitoid minerals, particularly the occurrence of accessory minerals in the major phases due to 742 the minimization of interfacial energies during grain boundary migration (Watson et al., 1989). Although 743 interfacial energies between quartz and feldspar are relatively similar (Hiraga et al., 2002), they differ from 744 many accessory minerals, meaning that in quartzofeldspathic aggregates under stress, accessory mineral 745 inclusions will tend to migrate relative to and become pinned at the boundaries of major phases, with a 746 driving force proportional to the square of the accessory grain's radius (Watson et al., 1989). Examination 747 of natural migmatite samples by Watson et al. (1989) revealed that while some accessory grains (in this 748 case zircon and rutile) remained as inclusions, the larger accessories tended to be found at grain 749 boundaries in the major phases. Notably most of the accessory inclusions in their study were also found 750 in mafic rather than felsic minerals. This suggests that significant caution is in order when comparing the 751 inclusion records of different minerals in metamorphic rocks, especially for mineral phases with

significantly differing interfacial energies, such as olivine and quartz or feldspar (e.g., Hiraga et al., 2002).
In the case of our samples there is little evidence for regional metamorphism following their final
solidification. However, for many detrital samples this may become a significant issue.

755 *4.6 Trace minerals of interest*

756 A number of trace phases found as inclusions in magnetite, ilmenite, zircon, hornblende, and 757 monazite are either absent from host rock matrices as free grains, or are, at least, best found by searching 758 the accessory minerals. These phases exist both as primary inclusions and as embaying or intergrown 759 phases. These include various sulfides, sulfates, rare oxides, and the occasional native metal. A handful 760 of occurrences of apparent native Ni in various accessories from the Peninsular Ranges Batholith are 761 documented in this study. Inclusions of various Zn phases (sulfide and ZnAl oxide) appear to be 762 characteristic of several accessory minerals in the evolved granite of Butler Peak, along with scattered 763 occurrences in the Peninsular Ranges Batholith. Nb minerals such as columbite and aeschynite are seen 764 as inclusions in the later-crystallizing phases (ilmenite, monazite, thorite) from the highly evolved granite 765 of Butler Peak and the Spirit Mountain Leucogranite. Mn oxide, anhydrite, halite, and fluorite inclusions 766 are also observed in magnetite and zircon of the Spirit Mountain Leucogranite.

767 Based on the above observations, highly evolved granites with high concentrations of 768 incompatible trace elements and abundant sulfate or halide minerals appear to yield characteristic 769 inclusion phases that might be useful for linking the provenances of the different host phases both to each 770 other in a detrital setting and to ultimate origins in a highly evolved granite. A good example is ilmenite, 771 thorite, and monazite from the Spirit Mountain Leucogranite, which we have shown are rich in Nb phases. 772 Downstream of the leucogranite these minerals could be linked to a common provenance using their 773 shared Nb cargo. This may be possible to a smaller extent with inclusions characteristic of mafic to 774 intermediate units, such as baddeleyite (seen as an inclusion in the oxides of the Alpine and Las Bancas

775	tonalites in this study) – although it is only observed in relatively early-crystallizing oxides in this study.
776	Linking sulfide chemistry across the various inclusion phases may also be a viable strategy for establishing
777	the similar provenance of different detrital accessory mineral suites.

778 *4.7 Implications for detrital studies*

779 Determining the primary vs secondary nature of inclusions is as important in a detrital context as 780 in the original host magma – if not more. Inclusion phases in contact with cracks and grain surfaces will 781 be subjected to weathering agents such as surface water and groundwater, and may be partially to fully 782 dissolved and replaced during the detrital grain's residence in the sedimentary system. A similar process has been suggested for preferential dissolution of apatite inclusions in detrital zircon and their 783 784 replacement by clays (Rasmussen et al., 2011; Nutman et al., 2014). Indeed, we find likely aluminosilicate phases ("AI-Si," chlorite, "Mg-AI-Si," and "Fe-AI-Si") preferentially associated with cracks in many of our 785 786 samples even though these rocks are largely unweathered. For host grains that are readily weatherable, 787 residence in altered regions of the host must also be assessed if this may have resulted in changes to the 788 inclusion assemblage. Relatively rapid two-dimensional scanning with backscattered electron imagery 789 and EDS spectroscopy, as in this study, can efficiently identify inclusion phases intersecting the polished 790 surface of the sample and provides some screening for exposure to alteration agents or potential 791 secondary origins by exposure to the host magma or later metamorphic/weathering fluids along cracks or 792 embayments in the host grain.

For host grains like zircon, which are often translucent and CL-active, additional imaging can provide evidence against exposure along subsurface cracks or embayments. Residence in chemically disturbed regions or pre-magmatic inherited cores can also be assessed both by CL imaging and U-Pb dating of detrital zircon. Primary mineral inclusions are remarkably hardy when not in contact with avenues of fluid ingress: as evidenced, for example, by the preservation of mineral inclusion assemblages 798 and chemistry in originally magmatic zircon from a Grenville-age orthogneiss that had undergone both 799 upper amphibolite facies prograde metamorphism and later retrograde greenschist facies metamorphism 800 inside isotopically undisturbed zircon (Bell, 2016). For monazite, Th-Pb dating and chemical zoning may 801 help to supplement the BSE determinations. For detrital oxide and hornblende hosts, a more certain 802 distinction of the primary inclusion phase may be more difficult and may need to involve detailed 803 structural and chemical mapping by EBSD and EPMA, respectively. The relatively minor adjustments to 804 mineral inclusion assemblages in zircon derived from only BSE imaging (as used for all host grains in this 805 study) after accounting for CL imagery and subsurface cracks/embayments also yields some lessons 806 applicable to opaque or CL-inactive hosts. Predictably, the most significant changes to the inferred 807 primary inclusion assemblages occurred among the zircon suites with the lowest numbers of identified 808 inclusions. Although apatite and QAP structures were both likely to be associated with subsurface cracks 809 and embayments (owing to their high aspect ratio and typically large size, respectively) their relative 810 proportions changed little with the additional screening. For opaque hosts, identifying a large number of 811 inclusions is advisable. Large, near-edge inclusions (particularly of QAP and apatite) are more likely to be 812 in contact with the grain edge or cracks.

813 Unlike in a detrital setting, pre-magmatic domains of some accessory minerals can be better 814 anticipated in well-studied magmatic settings with constrained magmatic histories, as with most of the 815 units in this study. Further constraints on individual host grain samples can be made using CL imagery and 816 U-Pb dating (in the case of zircon) or potentially EBSD to identify different structural domains. Among our 817 samples, magmatic evolution dominated by fractional crystallization rather than magma mixing or 818 mingling yielded the most readily interpretable crystallization histories among magnetite, ilmenite, 819 hornblende, zircon, and in some cases monazite, thorite, or orthopyroxene. The other samples with less 820 clear histories may represent zircon, ilmenite, and hornblende potentially grown in different magmas that 821 were later mingled – and either grains of mixed domains or separate grains deriving from the different
magmas cannot by our methods be distinguished. Some eastern and transition zone PRB magmas contain zircon with a variety of CL zoning styles that might derive from magmas of differing composition (e.g., Corfu et al., 2003), but these typically did not yield statistically significant differences in inclusion assemblage (see table S2). While individual grains from separate mingled magmas will simply look like distinct provenances in a detrital setting, distinguishing premagmatic and secondary altered domains in a detrital setting will be more crucial for provenance interpretations.

828 One of the important implications of our results for the detrital record is that there is no reason 829 to suspect that co-genetic accessory minerals in the detrital record will have similar inclusion suites, at 830 least with regards to major minerals and apatite contents. The various accessory minerals in a single 831 magma may capture distinct inclusion suites based on their position in the crystallization history. The rare 832 REE, Nb, and Mn inclusion minerals characteristic of highly evolved granites, along with the baddeleyite 833 inclusions seen in oxides from mafic-intermediate granitoids, are probably the most specific indicators 834 (both for host mineral provenance in highly evolved magmas and for linking different detrital minerals to 835 other minerals in the same sediment), absent geochronological methods for doing so. Other rare, 836 geochronologically useful inclusion phases include allanite, sphene, and monazite. Monazite and allanite 837 inclusions are somewhat more likely to be found in accessory minerals from more evolved granitoids and 838 sphene from more intermediate-composition metaluminous magmas. Sphene is also found more 839 consistently as a minor inclusion phase in ilmenite compared to the other studied minerals.

The evidence for proportions of mafic vs late-crystallizing silicate inclusions changing systematically in the studied accessory minerals through a magma's crystallization history is currently ambiguous. Late-crystallizing silicates correlate with apatite abundance in the inclusion suite for several accessory minerals, but ilmenite's special affinity for mafic inclusions complicate its interpretation relative to other accessory minerals grown from the same magma. On the other hand, estimates of source magma SiO₂ may be possible using the proportion of mafic silicate inclusions in ilmenite. Linking the ilmenite grains to a similar origin may prove difficult absent an inclusion phase that can be dated but may be possible through morphological and trace element discrimination schemes for ilmenite from different origins (e.g., Basu and Molinaroli, 1991; Grigsby, 1992). Zircon suites rich in mafic silicate and Fe-Ti oxide inclusions are more likely to be low in silica. Although samples in this study >65 wt. % SiO₂ show no relationship between mafic inclusion content and SiO₂, mafic and oxide inclusions may be useful in distinguishing low- and high-silica origins of low-apatite zircon suites.

852 The ubiquity of apatite as an inclusion phase in a variety of accessory minerals that are potentially 853 hardy in detrital environments also highlights the usefulness of apatite geochemistry as a marker for 854 sediment provenance. Various studies have linked apatite inclusions' geochemistry to host magma 855 genesis (Jennings et al., 2011; Bruand et al., 2016; Boehnke et al., 2018). The chemical similarity of apatite 856 inclusions in zircon to free apatite in the host magma (Bruand et al., 2016; Jennings et al., 2011) lends 857 itself to investigating apatite inclusions in detrital minerals for their likely origins based on, for example, 858 trace element geochemistry (Belousova et al., 2002b) or Sr isotope geochemistry (e.g., Boehnke et al., 859 2018). Linking apatite inclusion chemistry to morphological and trace element discriminant schemes in 860 the host ilmenite (e.g., Basu and Molinaroli, 1991; Grigsby, 1992) or zircon (e.g., Grimes et al., 2015) could 861 assist in constraining their provenance with more specificity.

862 **5. Conclusions**

A survey of inclusion assemblages in several accessory minerals across 27 granitoids clarifies some of the effects of crystallization order vs. magma chemistry in creating the observed distributions. Apatite is a ubiquitous inclusion phase, almost universally overrepresented in inclusion assemblages compared to its often trace presence in the rock as a whole. In most samples in which the relative crystallization order of the accessory minerals can be established, apatite inclusion abundances either decrease as crystallization progresses or remain similar. This is likely due to the early and continued local saturation 869 of apatite along growing accessory phase boundaries due to pileup of phosphorous. The abundance of 870 late-crystallizing silicate inclusions may be partially controlled by crystallization order given their 871 correlation with apatite abundance, but the proportion of mafic silicate inclusions shows similar behavior 872 only in ilmenite. Interpretations of late-crystallizing silicate inclusion abundance across mineral phases in 873 the same granitoid are stymied by the overabundance of mafic silicate inclusions in ilmenite regardless of 874 its position in the crystallization sequence. Magma chemistry also correlates well with the proportion of 875 mafic silicates included in ilmenite suites. The more evolved chemistry of feldspar inclusions in zircon 876 compared to mafic accessory minerals and free grains in the host rock corroborate control by late position 877 in the crystallization order. However, the higher proportion of quartz compared to feldspars among 878 inclusion assemblages relative to host rock does not track systematically with crystallization order among 879 the studied host minerals. A number of open questions related to mineral inclusion assemblages require 880 further study: this includes both the question of whether crystallization kinetics control quartz/feldspar 881 ratio in inclusion assemblages as well as a clarification of the apatite inclusion abundance vs whole rock 882 silica content for zircons, since this work predicts lower apatite inclusion abundances in most later-883 crystallizing phases, which would stand in contrast to the predicted higher apatite abundance suggested 884 for zircon from mafic-intermediate granitoids (almost universally later-crystallizing) by a previous survey 885 of zircon-hosted inclusion assemblages (Bell et al., 2018).

Overall, linking multiple detrital accessory mineral suites from the same rock to each other is very difficult and in many cases impossible using the proportions of major inclusion phases like apatite and mafic or late-crystallizing silicates due to their variations with crystallization order and other effects. Instead, identification of rare oxide, sulfide, and halide phases may provide a better fingerprint for both shared provenance of different detrital mineral suites and for origins in either highly evolved (halides, Nb or Mn phases) or mafic-intermediate (baddeleyite) granitoids. Geochronologically useful inclusion phases in oxides and hornblende include zircon, rutile, allanite, sphene, and baddeleyite, and may also be usefulin linking mineral provenance by age.

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1096 Figure Captions:



Figure 1: Examples of the plurality pattern (11/20) of decreasing apatite in the inclusion assemblage with 1098 1099 progressive crystallization (one sample contains both apatite-increasing and apatite-decreasing 1100 relationships between different minerals). This includes samples where zircon appears late in the 1101 crystallization order (A, B) and where zircon appears early compared to the other studied accessory 1102 minerals (C, D). Inclusions of other mafic phases are not shown for hornblende or orthopyroxene. Late 1103 silicates: quartz, K-feldspar, muscovite; mafic silicates: pyroxene, hornblende, biotite, chlorite, sphene. 1104 Parentheses denote two phases with uncertain relative crystallization order. Opx: orthopyroxene. Error 1105 bars are 1σ from counting statistics.



Figure 2: Examples where crystallization order can be inferred but apatite contents either do not differ significantly with progressive crystallization (A, B; reflecting 10/20 samples) or increase with progressive crystallization (C; 4/20 samples; two of these also contain an apatite-decreasing relationship). Inclusions of other mafic phases are not shown for hornblende. Late silicates: quartz, K-feldspar, muscovite; mafic silicates: pyroxene, hornblende, biotite, chlorite, sphene. Parentheses denote two phases with uncertain relative crystallization order. Error bars are 1σ from counting statistics.



Figure 3: Inclusion counts in all samples and all phases plotted as A) late-crystallizing silicates vs apatite
and B) mafic silicates vs apatite. The mafic accessory minerals hornblende and orthopyroxene are
omitted from B). There is a systematic increase in late silicates with decreasing apatite contents among
zircon, hornblende, and ilmenite, with ilmenite offset to lower late silicate contents. Zircon and
magnetite show no systematic relationship between mafic silicate and apatite inclusion abundances, but
ilmenite shows a rough positive relationship between mafic silicate and apatite.



Figure 4: Late-crystallizing (A) and mafic (B) silicate inclusion phases plotted vs whole-rock weight %
SiO₂. There is little relationship between SiO₂ and the proportion of late-crystallizing silicates in the

1123 studied accessory minerals. Ilmenite shows increasing mafic silicate inclusions with decreasing SiO₂, and



1124 zircon shows a slight increase in mafic inclusions below 65 weight % SiO₂.

Figure 5: Zircon inclusion suites from this study and Bell et al. (2018), with the sum of all mafic silicate
and Fe-Ti oxide (magnetite, ilmenite, rutile) inclusions plotted against a) apatite content and b) whole
rock SiO₂.



Figure 6: A) Quartz and feldspar inclusion compositions plotted for various accessory minerals. Most
samples have associated whole rock compositional data, for which CIPW norms are shown. Shifts in B)
Q/QAP and C) A/AP for zircon and the Fe-Ti oxides also shown. Shifts toward higher Q/QAP and A/QAP
are common among most of the studied accessory minerals.

1134



Figure 7: Chemistry of matrix feldspars compared with inclusions in zircon. For several samples, postmagmatic veins or exsolution features in the matrix are also shown (A, C, D). Several samples also
contain inclusions in hornblende (A) or biotite (D). Inclusions range from similar to more evolved (higher
% or in K-feldspar, higher % ab in plagioclase) than magmatic matrix grains and similar to less evolved

- 1140 than post-magmatic features. Inclusions in zircon are slightly more evolved than inclusions in biotite
- and hornblende.



- 1143 *Figure 8:* Mafic phases hosting feldspar inclusions for comparative chemistry: A) a biotite in the Spirit
- 1144 Mountain leucogranite; B) a hornblende in granite IG1 of the San Jacinto Mountains.





- 1146 *Figure 9:* representative images of exsolution features in ilmenite and magnetite. A) An ilmenite grain
- 1147 from the Alpine tonalite showing a region with separated Fe and Ti oxides. B, C) Magnetite grains (from
- 1148 Alpine tonalite and Cactus Flats quartz diorite, respectively) with exsolution lamellae of Mn-rich
- 1149 ilmenite. In C), one of the ilmenite lamellae is exsolved to separate Fe and Ti oxide phases.



1151 *Figure 10:* Apatite contents in all studied accessory minerals grouped by geographic region. BB: Big Bear

1152 Lake Intrusive Suite; CREC: Colorado River Extensional Corridor; EPRB: Eastern Peninsular Ranges

1153 Batholith; WPRB: Western Peninsular Ranges Batholith.

1154

Table One: Textural Observations of Inclusions

Sample Order clear?		Mag.	Ilm.	Zrc.	Hbl.	Орх	Mon.	Thr.	
Western Peninsular Ranges Batholith									
AT1: Alpine	: Alpine -zrc after oxides % apatite		64±14	57±11	20±4	21±5			
Tonalite	onalite -zrc after hbl % late silicate		6±4	6±4	33±5	23±5			
	-Hbl after mag % mafic sil		15±7	31±8	14±3				
AT2: Alpine	-opx after oxides	% apatite	50±13	59±19	23±9		0		
Tonalite		% late silicate	13±6	0	31±11		7±5		
		% mafic silicate	22±8	24±12	15±8				
LB1: Las Bancas	-ilm after mag	% apatite	53±19	34±9	5±2				
pluton	-zrc after oxides	% late silicate	27±13	12±5	41±7				
		% mafic silicate	7±7	24±8	13±4				
LB2: Las Bancas	-zrc after oxides	% apatite	57±16	48±13	19±6		12±5		
pluton	-opx after ilm	% late silicate	10±7	10±6	31±8		2±2		
		% mafic silicate	14±8	28±10	21±7				
JV1: Japatul	-zrc & ilm after mag	% apatite	32±10	38±11	29±5				
Valley granite		% late silicate	18±7	18±7	21±4				
(Corte Madera		% mafic silicate	15±7	3±3	2±1				
monzogranite)									
JV2: Japatul	-hbl after ilm	% apatite		0	16±7	13±5			
Valley	-zrc after hbl	% late silicate		19±7	35±10	29±7			
intermediate		% mafic silicate		51±11	14±6				
JV6: Japtul Valley	-ilm after mag	% apatite	67±13	32±8	36±7				
intermediate		% late silicate	0	0	32±6				
		% mafic silicate	3±3	30±7	4±2				
	1	Eastern Penins	ular Range	es Batholi	th	1	T	1	1
LP3: La Posta hbl-	-zrc after ilm	% apatite		49±5	55±5	57±16			
bt unit (outer)		% late silicate		13±2	21±3	10±7			
		% mafic silicate		25±4	4±1				
LP1: La Posta	-zrc after ilm	% apatite		58±8	51±6	80±23			
large biotite unit		% late silicate		7±3	15±3	0			
(middle outer)		% mafic silicate		19±5	5±2				
LP2: La Posta	-order unclear	% apatite		63±7	77±10				
small biotite unit		% late silicate		14±3	3±3				
(middle inner)		% mafic silicate	-	10±3	1±1	-		-	
LP5: La Posta	-order unclear	% apatite		54±12	78±15				
small biotite unit		% late silicate		3±3	3±3				
(middle inner)		% matic silicate		19±7	6±4				
LP6: La Posta	-ilm & mon after zrc	% apatite		35±9	53±10			18±5	
musc-bt unit % late silicate			14±6	22±7			15±5		
(inner)	(inner) % mafic silicate			14±6	0			0	
IG3: San Jacinto	-order unclear	% apatite		33±/	84±8	5/±11			
	unit iii % late silicate		16±5	6±2	11±5				
granodiorite #1	granodiorite #1 % mafic silicate		42±8	1±1			 		
IG4: San Jacinto	-order unclear	% apatite			85±15	69±16			
		% late silicate			5±4	U			
granodiorite #2	ana aftan ilar	% matic silicate		4610	3±3				
KG1: San Jacinto	-zrc atter ilm	% apatite		40±8	10+2	29±12			
		% late silicate		15±5	10±3	22±9			
granodiorite		% matic silicate		28‡6	1±1				

MCG1: San	-zrc after ilm	% apatite		36±6	41±7				
Jacinto unit i		% late silicate		8±3	27±6				
granodiorite #1		% mafic silicate		41±7	1±1				
MCG3: San	-zrc after ilm	% apatite		68±11	40±5				
Jacinto unit i		% late silicate		0	31±5				
Granodiorite #2		% mafic silicate		23±7	4±2				
ML1: Granite	-zrc after oxides	% apatite	46±14	54±20	61±8				
Mountain		% late silicate	13±7	0	13±4				
tonalite		% mafic silicate	17±8	31±15	9±3				
GM1: Granite	-order unclear	% apatite		62±11	43±8				
Mountain		% late silicate		4±3	28±6				
tonalite		% mafic silicate		22±6	12±4				
		Big Bear La	ke Intrusi	ve Suite	•		•	•	
BB6: Hanna Flat	-mag after zrc	% apatite	57±9		51±7	60±20			
granodiorite		% late silicate	12±4		27±5	7±7			
		% mafic silicate	4±2		2±2				
BB12: Cactus Flat	-order unclear	% apatite	59±16		73±11	67±13			
quartz diorite		% late silicate	14±8		18±6	15±6			
		% mafic silicate	19±9		5±3				
BB14: Butler Peak	-oxides after zrc	% apatite	44±5	29±5	40±7			18±7	
granite	-mon after oxides	% late silicate	13±3	13±4	34±6			15±7	
		% mafic silicate	4±2	10±3	5±2			6±4	
Colorado River Extensional Corridor					or				
SpLa: coarse-	-ilm & mon after mag	% apatite	30±6	13±6	38±7			6±4	
grained lower		% late silicate	4±2	28±9	26±6			24±8	
unit		% mafic silicate	3±2	5±4	1±1			9±5	
SLT1: fine-grained	-order unclear	% apatite	17±12	4±3	28±6				0
upper unit		% late silicate	25±14	38±9	33±7				45±15
		% mafic silicate	0	2±2	1±1				0
SLT2: coarse-	-zrc after oxides	% apatite	19±8	0	25±5				0
grained upper		% late silicate	7±5	59±13	32±5				20±9
unit		% mafic silicate	0	3±3	4±2				0
HR1:	-zrc after mag	% apatite	34±8		62±8				
intermediate lava		% late silicate	9±4		14±4				
		% mafic silicate	30±9		3±2				
SLP1: Searchlight	-order unclear	% apatite	52±8		23±5				
pluton quartz		% late silicate	6±3		36±6				
monzonite		% mafic silicate	23±5		7±3				
Sample	Order clear?		Mag.	Ilm.	Zrc.	Hbl.	Орх.	Mon.	Thr.

Table 1: textural observations of inclusions in various accessory phases, sorted by geographic region. Only inclusions isolated from cracks or edges in the host phase are considered. All error bars are 1σ based on counting statistics. The determinations of crystallization order are based on comparing proportions of mutual inclusions between two phases and in some cases textural analysis of thick sections; more detail for individual samples are included in the online supplemental text. When no inclusions are reported for a particular phase in a particular sample, that host phase was not identified in the sample.

Sample	Location	Crystallization order	Apatite behavior					
Group A: Contains evidence for decreasing apatite (n=11)								
AT1: Alpine Tonalite	WPRB	a) Oxides $ ightarrow$ hbl	a) decreases					
		b) hbl → zrc	b) no change					
AT2: Alpine Tonalite	WPRB	a) Mag \rightarrow zrc	a) decreases					
		b) oxides \rightarrow opx	b) decreases					
LB1: Las Bancas	WPRB	a) Mag \rightarrow ilm	a) no change					
pluton		b) ilm → zrc	b) decreases					
LB2: Las Bancas	WPRB	a) oxides → zrc	a) decreases					
pluton		b) oxides \rightarrow opx	b) decreases					
JV2: Japatul Valley	WPRB	a) hbl → ilm	a) decreases					
intermediate		b) hbl \rightarrow zrc	b) no change					
JV6: Japtul Valley	WPRB	a) mag \rightarrow ilm	a) decreases					
intermediate								
LP6: La Posta musc-	EPRB	a) zrc → ilm & mon	a) decreases					
bt unit (inner)		b) ilm & mon (??)	b)					
MCG3: San Jacinto	EPRB	a) ilm → zrc	a) decreases					
unit i								
Granodiorite #2								
BB14: Butler Peak	Big Bear	a) zrc → mag	a) no change					
granite		b) zrc → ilm	b) decreases					
		c) mag&zrc → mon	c) decreases					
**SpLa: coarse-	CREC	a) mag → mon	a) decreases					
grained lower unit	SMB	b) ilm → mag	b) increases					
** in Groups A & B								
**SLT2: coarse-	CREC	a) mag → zrc	a) no change					
grained upper unit	SMB	b) mag \rightarrow thor	b) decreases					
**in Groups A & B		c) ilm \rightarrow zrc	c) increases					
Group B	: Contains e	evidence for increasing apatit	e (n=4)					
RG1: San Jacinto	EPRB	a) ilm → zrc	a) increases					
unit ii granodiorite								
**SpLa: coarse-	CREC	a) mag → mon	a) decreases					
grained lower unit	SMB	b) ilm → mag	b) increases					
** in Groups A & B								
**SLT2: coarse-	CREC	a) mag → zrc	a) no change					
grained upper unit	SMB	b) mag → thor	b) decreases					
**in Groups A & B		c) ilm \rightarrow zrc	c) increases					
HR1: intermediate	CREC HR	a) mag → zrc	a) increases					
lava								
G	roup C: All a	patite contents similar (n=11)					
JV1: Corte Madera	WPRB	a) mag → zrc	a) no change					
monzogranite								

 Table 2: Apatite inclusion behavior versus crystallization order

LP1: La Posta large	EPRB	a) ilm → zrc	a) no change
biotite unit (middle			
outer)			
LP2: La Posta small	EPRB		
biotite unit (middle			
inner)			
LP3: La Posta hbl-bt	EPRB	a) ilm → zrc	a) no change
unit (outer)			
LP5: La Posta small	EPRB		
biotite unit (middle			
inner)			
IG4: San Jacinto unit	EPRB		
iii granodiorite #2			
MCG1: San Jacinto	EPRB	a) ilm → zrc	a) no change
unit i granodiorite			
#1			
ML1: Granite	PRB ETZ	a) oxides → zrc	a) no change
Mountain tonalite			
BB6: Hanna Flat	Big Bear	a) zrc → mag	a) no change
granodiorite			
BB12: Cactus Flat	Big Bear		
quartz diorite			
SLT1: fine-grained	CREC	a) mag → zrc	a) no change
upper unit	SMB		
Group D: Di	ffering apat	ite contents but ambiguous c	order (n=3)
IG3: San Jacinto unit	EPRB		
iii granodiorite #1			
GM1: Granite	PRB ETZ		
Mountain tonalite			
SLP1: Searchlight	CREC SLP		
pluton quartz			
monzonite			

Table 2: Studied granitoids characterized based on the behavior of apatite during progressive crystallization. Group A (n=11) shows robust evidence for decreasing apatite inclusion contents during some stages of crystallization (with some evidence of similar apatite contents at other intervals); Group B (n=4) shows robust evidence for increasing apatite. Two samples fall into both Group A and Group B. Group C (n=11) shows similar apatite contents among the minerals with robust crystallization order constraints. Group D (n=3) contains differing apatite contents among the studied minerals but not robust crystallization order constraints.

<u>Supplemental Text and Figures for "Crystallization order effects on inclusion assemblages in magmatic</u> <u>accessory minerals and implications for the detrital record"</u>

Hand Sample Descriptions

Descriptions for BB12, BB14, IG1, IG4m, RG1, RG6, SpLb, SpMb, MG3a, and MG3b are from Bell et al. (2018).

Peninsular Ranges Batholith:

AT1: Alpine tonalite sampled in a roadcut off Victoria Drive in the town of Alpine. N 32° 50' 8.77726'', W 116° 45' 2.43414''.

AT2: Alpine tonalite sampled in a bank off Puetz Valley Road near the town of Alpine. N 32.85003°, W 116.80425°.

LB1: Intermediate granitoid of the Las Bancas suite sampled off Sunrise Highway. 32° 50' 40.65562'' N, 116° 26' 35.78813'' W.

LB2: Intermediate granitoid of the Las Bancas suite sampled off Sunrise Highway. N 32° 51' 22.99957", W 116° 27' 59.70362".

JV1: Slightly foliated granitoid sampled on Japatul Valley Road south of I-8. N 32.80679°, W 116.63818°.

JV2: Melanocratic granitoid sampled on Lyons-Japatul Road south of the Horsethief Canyon trailhead. N 32.73377°, W 116.69292°.

JV6: Slightly foliated granitoid sampled at the intersection of Japatul Valley Road and Lyons-Japatul Road. N 32.76378°, W 116.67491°.

LP3: Medium-grained granitoid sampled from the hornblende-biotite unit of the La Posta pluton as mapped by Clinkenbeard and Walawender (1989). N 32.72239°, W 116.44599°.

LP1: Granitoid sampled from road cut on old highway 80; in the large biotite unit of the La Posta pluton as mapped by Clinkenbeard and Walawender (1989). N 32° 43' 16.41277'', W 116° 24' 31.03502''.

LP2: Granitoid sampled from the small biotite unit of the La Posta pluton as mapped by Clinkenbeard and Walawender (1989). N 32° 42' 28.16248'', W 116° 22' 51.91813''.

LP5: Granitoid sampled from the small biotite unit of the La Posta pluton as mapped by Clinkenbeard and Walawender (1989). N 32.67657°, W 116.33081°.

LP6: Muscovite- and biotite-bearing granitoid sampled from the muscovite-biotite unit of the La Posta pluton as mapped by Clinkenbeard and Walawender (1989). N 32.67405°, W 116.09682°.

GM1: White-gray Granite Mountain tonalite with weak foliation sampled off old highway 80. N 32.73152°, W 116.48302°.

ML1: White-gray, medium-grained granitoid with weak foliation sampled off Corral Canyon road. Abundant mafic enclaves, not included in sample. Granite Mountain tonalite (e.g., Morton et al., 2014). N 32.70856°, W 116.50724°.

IG1 (feldspar chemistry only): A medium- to fine-grained, biotite- and hornblende-bearing granitoid sampled on CA highway 243 south of Idyllwild in the San Jacinto Mountains of southern California. Collected at N 33° 43' 42.18," W 116° 43' 15.12."

IG3: Medium-grained granitoid sampled from a roadcut on CA route 243 north of Pine Cove. Collected at N 33° 44' 55.409", W 16° 44' 14.657".

IG4m: A medium-grained biotite-bearing granitoid sampled from a road cut on CA highway 243 (same location as IG4c) northwest of Idyllwild in the San Jacinto Mountains, southern California. Collected at N 33° 46' 33.745", W 116° 44' 11.318".

RG1: Medium-grained biotite-bearing granitoid collected on Ramona Trail south of CA highway 74 in the San Jacinto Mountains, southern California. Collected at N 33° 36′ 50.4″, W 116° 39′ 28.0″.

RG6 (feldspar chemistry only): Leucocratic medium-grained granitoid collected on Ramona Trail south of CA highway 74 in the San Jacinto Mountains, southern California. Collected at N 33° 36′ 50.4,″ W 116° 9′ 1.44.″

MCG1: Medium-grained hornblende- and biotite-bearing white-gray granitoid collected on CA highway 74 west of Mountain Center, CA in the San Jacinto Mountains. N 33° 42′ 9.697″, W 116° 46′ 8.842″.

MCG3: Coarse-grained biotite-bearing white-gray granitoid collected on CA highway 74 west of Mountain Center, CA in the San Jacinto Mountains. N 33° 42' 1.025", W 116° 44' 54.162".

Colorado River Extensional Corridor:

MG3a (feldspar chemistry only): A medium-grained porphyritic (~1cm K-feldspar phenocrysts), biotite-bearing granitoid sampled in the Mirage pluton (e.g., Walker et al., 2007) several hundred meters west of Needles Highway south of Laughlin, Nevada. Collected at N 35° 07' 31.02," W 114° 38' 27.42."

MG3b (feldspar chemistry only): Pods of foundered coarse-grained biotite-bearing Spirit Mountain granite within the Mirage Pluton sampled several hundred meters west of Needles Highway south of Laughlin, Nevada. Collected at N 35° 07' 31.02," W 114° 38' 27.42."

SpLb (feldspar chemistry only): Leucocratic, medium-grained, biotite-bearing granitoid with white feldspars of the Spirit Mountain leucogranite (Walker et al., 2007) sampled south of highway 163 west of Laughlin, Nevada at N 35° 10′ 9.96," W 114° 45′ 39.54."

SpLa (leucogranite): medium- to coarse-grained granitoid with white feldpsars sampled south of highway 163 at N 35° 10.166', W 114° 45.777'.

SLT1 (leucogranite): Fine-grained leucogranite with white feldspars sampled on highway 163. N 35.16954°, W 114.76512°.

SLT2 (leucogranite): Coarse-grained leucogranite with white feldspars sampled on highway 163. N 35.16972388°, W 114.76456792°.

SpMb (feldspar chemistry only): Medium-grained, porphyritic (~1cm K-feldspar phenocrysts) biotite-bearing granitoid of the Spirit Mountain granite (Walker et al., 2007) sampled ~1m from a later diabase dike on a roadcut in highway 163 west of Laughlin, Nevada. Collected at N 35° 10' 16.56," W 114° 42' 20.58."

SLP1: Fine-grained white-gray granitoid from the upper quartz monzonite unit of the Searchlight Pluton, sampled in the wall of a creek valley south of Cottonwood Cove Road. N 35.47169°, W 114.82578°.

HR1: Melanocratic, brown-purple volcanic unit of the Highland Range with plagioclase phenocrysts. N 35.50256°, W 114.98038°.

Big Bear:

BB6: A coarse biotite granitoid from CA highway 38 near Grout Bay. Mapped as Hanna Flat Granodiorite (Barth et al., 2016). N 34° 15' 31.587", W 116° 56' 39.283".

BB12: A dark gray, medium-grained biotite granitoid from CA highway 18 near Cactus Flat in the San Bernardino Mountains, southern California. Collected at N 34° 20′ 6.681″, W 116° 50′ 2.309″.

BB14: A leucocratic, fine-grained, biotite- and muscovite-bearing granitoid from CA highway 18 near Big Bear Lake in the San Bernardino Mountains, southern California. Mapped as Butler Peak Granite (Barth et al., 2016). Collected at N 34° 14′ 34.583″, W 117° 1′ 47.735″.

Crystallization Order Details:

These interpretations only consider inclusions that are isolated from cracks and host grain edges.

" \rightarrow " = "began crystallizing before"

Western Peninsular Ranges Batholith:

AT1: magnetite & ilmenite \rightarrow hornblende \rightarrow zircon

Magnetite and ilmenite have mutual inclusions at a similar proportion so their order is texturally ambiguous.

Magnetite contains 6% hornblende inclusions but hornblende has 30% magnetite inclusions, so magnetite started crytallizing earlier than hornblende.

Ilmenite contains 16% hornblende inclusions and hornblende has 14% ilmenite inclusions, which are indistinguishable within error making their order texturally ambiguous.

Magnetite and ilmenite contain no zircon inclusions but zircon contains 7% and 6% magnetite and ilmenite respectively, meaning zircon began crystallizing later.

Zircon contains 3% hornblende inclusions but hornblende has no zircon inclusions, so hornblende began crystallizing before zircon.

AT2: magnetite \rightarrow zircon; oxides \rightarrow opx

Magnetite contains no zircon inclusions but zircon has 7% magnetite inclusions, so magnetite began crystallizing earlier than zircon.

Ilmenite contains one zircon inclusion but zircon has no ilmenite inclusions. This remains somewhat ambiguous because the counting statistics on 1 are ±1, but ilmenite probably started crystallizing after zircon.

Orthopyroxene contains 28% ilmenite inclusions and 28% magnetite inclusions, but magnetite contains no orthopyroxene inclusions and ilmenite contains a single orthopyroxene inclusion. Orthopyroxene began crystallizing after magnetite and ilmenite.

There are no mutual inclusions between orthopyroxene and zircon, making their relationship texturally ambiguous.

LB1: magnetite \rightarrow ilmenite; oxides \rightarrow zircon

Magnetite contains no ilmenite inclusions but ilmenite contains magnetite inclusions, meaning magnetite began crystallizing earlier.

Magnetite and ilmenite contain no zircon inclusions but zircon contains 4% and 6% of magnetite and ilmenite inclusions, respectively, meaning zircon began crystallizing later than the oxides.

LB2: oxides \rightarrow zircon; oxides \rightarrow orthopyroxene

Magnetite contains a single inclusion of ilmenite, while ilmenite contains no magnetite inclusions. Their order is therefore texturally ambiguous although it is possible ilmenite began crystallizing first.

Magnetite and ilmenite contain no zircon inclusions, but zircon contains 4% and 6% magnetite and ilmenite inclusions, respectively.

Orthopyroxene contains 21% and 27% magnetite and ilmenite inclusions, respectively, but magnetite contains no orthopyroxene and ilmenite contains a single orthopyroxene inclusion. Orthopyroxene began crystallizing after the oxides.

Orthopyroxene and zircon have no mutual inclusions.

JV1: magnetite \rightarrow zircon

Magnetite and ilmenite each contain equal proportions of mutual inclusions, making their crystallization order texturally ambiguous.

Neither magnetite nor ilmenite contain zircon inclusions but zircon contains 3% magnetite inclusions, meaning zircon began crystallizing after magnetite.

JV2: hornblende \rightarrow ilmenite; hornblende \rightarrow zircon

Ilmenite contains 19% hornblende inclusions. Hornblende only contains 7% ilmenite inclusions, meaning it started crystallizing before ilmenite.

Zircon contains a single ilmenite inclusion and 7% hornblende inclusions (n=5). Ilmenite contains a single zircon inclusion and hornblende has no zircon inclusions. Zircon began crystallizing after hornblende but its relationship with ilmenite is texturally ambiguous.

JV6: magnetite → ilmenite

Magnetite contains no ilmenite inclusions but ilmenite contains 12 magnetite inclusions (21%), so magnetite began crystallizing first.

Magnetite and zircon contain no mutual inclusions. Ilmenite and zircon are mutually included to a statistically indistinguishable level (2% and 3%). Zircon's order relative to the oxides is texturally ambiguous.

Eastern Peninsular Ranges Batholith

LP3: ambiguous

Ilmenite and zircon are mutually included; there is more ilmenite in zircon but not to a statistically significant level.

Hornblende and ilmenite are mutually included with slightly more ilmenite in hornblende but not to a statistically distinguishable level. There are no hornblende inclusions in zircon but hornblende contains a single zircon inclusion.

LP1: ilmenite \rightarrow zircon

Zircon contains 11% ilmenite inclusions but ilmenite contains only 2% zircon inclusions, so ilmenite began crystallizing before zircon.

Ilmenite and hornblende, along with zircon and hornblende, are mutually included to a statistically indistinguishable level.

LP2: ambiguous

Ilmenite and zircon order cannot be distinguished by mutual inclusions.

LP5: ambiguous

Ilmenite and zircon order cannot be distinguished by mutual inclusions.

LP6: zircon \rightarrow ilmenite; zircon \rightarrow monazite

Zircon contains no ilmenite inclusions but ilmenite contains 5% zircon inclusions (n=2). Zircon began crystallizing before ilmenite.

Zircon contains no monazite inclusions but monazite contains 33% zircon inclusions (n=24). Zircon began crystallizing before ilmenite.

Ilmenite and monazite contain no mutual inclusions.

IG3: ambiguous

Zircon contains no hornblende but hornblende contains a single zircon inclusion; this is statistically nonsignificant but zircon possibly began crystallizing before hornblende.

Zircon and ilmenite are mutually included to a statistically indistinguishable level, as are hornblende and ilmenite.

IG4: ambiguous

Zircon and hornblende relative order cannot be distinguished based on mutual inclusions.

RG1: ilmenite \rightarrow zircon

Zircon contains 6% ilmenite inclusions, but ilmenite contains a single zircon (1%). Ilmenite began crystallizing before zircon.

Hornblende and zircon contain no mutual inclusions. Hornblende and ilmenite are mutually included to a statistically indistinguishable level.

MCG1: ilmenite \rightarrow zircon

Ilmenite contains no zircon but zircon contains 7% ilmenite inclusions (n=6). Ilmenite began crystallizing before zircon.

MCG3: ilmenite \rightarrow zircon

Ilmenite contains no zircon but zircon contains 5% ilmenite inclusions (n=7). Ilmenite began crystallizing before zircon.

ML1: oxides \rightarrow zircon

Zircon contains 4% each magnetite and ilmenite inclusions (n=4 each), but magnetite and ilmenite contain no zircon inclusions. The zircon began crystallizing after the oxides.

Magnetite and ilmenite are mutually included to a statistically indistinguishable degree.

GM1: ambiguous

Ilmenite contains no zircon but zircon contains a single ilmenite inclusion. This is statistically indistinguishable but it's possible ilmenite began crystallizing first.

Big Bear Intrusive Suite:

BB6: zircon → magnetite

Magnetite contains 3% zircon inclusions (n=2) but zircon contains no magnetite. Zircon began crystallizing before magnetite.

Magnetite and hornblende are mutually included to a statistically indistinguishable extent. Hornblende and zircon contain no mutual inclusions.

BB12: ambiguous

Zircon contains no mutual inclusions with the other phases. magnetite and hornblende are mutually included to a statistically indistinguishable level.

BB14: zircon \rightarrow oxides; magnetite \rightarrow monazite

Zircon contains no oxide or monazite inclusions. Magnetite contains 26%, ilmenite 31%, and monazite 9% zircon inclusions, respectively. Zircon began crystallizing before these other phases.

Monazite contains 9% (n=3) magnetite inclusions and no ilmenite inclusions. Magnetite contains a single monazite inclusion and ilmenite contains no monazite. Magnetite began to crystallize before monazite but there is no constraint on ilmenite vs monazite.

Magnetite and ilmenite are mutually included to a statistically indistinguishable level.

Colorado River Extensional Corridor:

SpLa: ilmenite \rightarrow magnetite (??); magnetite \rightarrow monazite

Zircon contains a single inclusion each of magnetite and ilmenite and no monazite, although magnetite contains 4% and ilmenite 5% zircon inclusions respectively. This is not statistically distinguishable, however it is likely zircon may have begun to crystallize before the oxides.

Based on mutual inclusions, magnetite's 3 ilmenite inclusions and ilmenite's lack of magnetite inclusions suggests ilmenite began crystallizing first. This is somewhat at odds with the petrographic thick section observations of euhedral magnetite rimmed by apparently later subhedral ilmenite, although euhedral free grains of ilmenite also exist in the matrix. It is possible there are several generations of ilmenite or that ilmenite despite beginning to crystallize earlier than magnetite continued to crystallize later. Late-stage crystallization is also consistent with its higher capture rate for Nb-Ta and LREE phases as well as fluorite. Although they did not resemble exsolution lamellae in electron imaging, it is possible that the ilmenite inclusions interpreted here were mistakenly misidentified and are exsolution.

However, we tentatively identify ilmenite as beginning to crystallize before magnetite, while probably continuing after magnetite crystallization had ceased.

Monazite contains 21% magnetite inclusions while magnetite contains a single monazite inclusion. Magnetite began to crystallize before monazite.

SLT1: ambiguous

There are either no mutual inclusions or there are only mutual inclusions that do not reach statistical significance for interpreting crystallization order among the studied accessory minerals.

SLT2: magnetite \rightarrow zircon; magnetite \rightarrow thorite

Magnetite contains no zircon but zircon has 3% magnetite inclusions. Magnetite began to crystallize before zircon.

Magnetite contains to thorite but thorite has 24% magnetite inclusions. Magnetite began to crystallize before thorite.

Magnetite and ilmenite are mutually included to a statistically indistinguishable level.

HR1: magnetite \rightarrow zircon

Magnetite contains no zircon inclusions, but zircon contains 12% magnetite inclusions. Magnetite began to crystallize before zircon.

SLP1: ambiguous

Magnetite contains no zircon inclusions, but zircon contains a single magnetite inclusion. Although it is not statistically distinguishable, it is possible magnetite began to crystallize before zircon.

Supplemental Figures



Fig. S1: Effects of adjustments to the primary inclusion totals for various inclusion species and categories. "Preadjustment" refers to inclusions isolated from cracks as determined by backscattered electron imaging, as for the procedure for other accessory minerals. "Post-adjustment" refers to this isolated inclusion assemblage after

further screening for subsurface cracks and embayments as well as inherited cores or altered regions revealed by cathodoluminescence imaging.



Figure S2: There is little coherent relationship between apatite content and Q/QAP in the inclusion assemblage as a whole. Within some individual plutons there is a rough positive or negative relationship. A) all peninsular

ranges samples, B) all Big Bear samples, C) all Spirit Mountain Leucogranite samples, D) San Jacinto pluton samples (PRB), E) La Posta pluton samples (PRB).



Figure S3: A) Q/QAP and B) K-feldspar/total feldspar variations for the studied accessory mineral inclusion assemblages. Zircon assemblages are shown as a probability density distribution, with other minerals shown as points offset on a dimensionless y-axis. As seen in Figure 5 of the main text, most accessory minerals have inclusion assemblages shifted towards higher Q/QAP and K-feldspar/total feldspar than the CIPW norms calculated from their associated whole-rock chemistries. Ilmenite and magnetite tend to show higher Q/QAP on average than zircon, with large overlap.


Figure S4: Whole rock chemistry for the various samples in which crystallization order of at least two of the studied minerals could be inferred. As for M vs SiO_2 (Fig. 9 of the main text), there is little relationship between whole rock chemistry and apatite behavior with respect to crystallization order.



Figure S5: An alternative view of feldspar chemistry for matrix grains, inclusions in zircon and mafic phases, and post-magmatic alteration phases in the matrix. For the K-feldspar panels, the "all in zrc" population includes a

handful of potential mixed albite/K-feldspar inclusions that were more albitic than the matrix grains and notably more albitic than the majority of the K-feldspar inclusion data for the same rock. "Zrc, no mixed" excludes these potentially mixed analyses.