VARIATION OF SR AND PB ISOTOPES IN MEGACRYSTIC K-FELDSPAR FROM THE CATHEDRAL PEAK GRANODIORITE, CALIFORNIA

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A thesis submitted to the faculty at the University of North Carolina at Chapel Hill in partial fulfillment of the requirements for the degree of Masters of Science in the Department of Geological Sciences.

Chapel Hill 2016

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ABSTRACT

Jonathan Keith Munnikhuis: Variation of Sr and Pb isotopes in megacrystic K-Feldspar from the Cathedral Peak Granodiorite, California (Under the direction of Allen F. Glazner)

In-situ micro-drilling of K-feldspar megacrysts from the Late Cretaceous Cathedral Peak Granodiorite of the Tuolumne Intrusive Suite, California, provided material to study Sr and Pb isotopic compositions and trace element variation recorded by these crystals. Both cores and rims of megacrysts have isotopic signatures similar to previously published whole-rock values of the Cathedral Peak Granodiorite. Excursions in isotopic ratios demonstrate that crystal transects are not homogenous, indicating that the megacrysts crystallized in an open-system environment. However, variations in radiogenic isotopes of K-feldspars are not large as observed in other plutonic and volcanic settings, suggesting large scale magma mixing and contamination did not occur in the Cathedral Peak Granodiorite. Modeling of K-feldspar trace element concentration profiles in a closed system fails to mimic profiles observed in the megacrysts. This adds to the interpretation that the megacrysts coarsened late in the Tuolumne Intrusive Suite's history, rather than early nucleation and long-term growth.

ACKNOWLEDGEMENTS

I would like to thank Dr. Allen Glazner, Dr. Drew Coleman, and Dr. Ryan Mills for their academic support, teaching the importance of skepticism, and sharing their valuable knowledge and experience of the Sierra Nevadas. I would like to thank several graduate students; Ryan Frazer, Connor Lawrence, Jesse Hill, and Tyler Wickland. Ryan and Connor assisted and helped teach me methods of isotope geochemistry at UNC, Jesse helped with code development, and Tyler Wickland and I shared several cups of coffee with discussions about thermal cycling. I would also like to thank Christopher Doorn whose assistance and attitude during field-work was invaluable.

Funding for this thesis was provided by the Martin Research Fund (University of North Carolina), the Graduate Student Research Grant (Geological Society of America), and the Graduate Student Bursary (Mineralogical Society of Great Britain).

A final gratitude is for my family and Christine Lion, their support was essential to my spirits and studies.

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LIST OF ABBREVIATIONS

a	annum
А	Ampere
An	anorthite
amu	atomic mass unit
C_0	weight concentration element in unmelted solid
СА	California
C_l	weight concentration element in a liquid
Di	bulk distribution coefficient of element <i>i</i>
F	fraction of melt remaining in fractional crystallization
f	fraction of melt returned to magma chamber
EPMA	electron probe micro analysis
h	hour
g	gram
ka	thousands of years/ thousands of years before present
Kd	distribution coefficient
Кср	Cathedral Peak Granodiorite
Khde	equigranular Half Dome Granodiorite
Khdp	Half Dome Granodiorite porphyritic member
Kw	Whitney Granodiorite
KDE	Kernel density estimation
L	liter

М	molar
m	meter
mm	millimeter
mg	milligram
μm	micrometer
μL	microliter
Ma	millions of years/ millions of years before present
MWIS	Mount Whitney Intrusive Suite
Or	Orthoclase
ppm	parts per million
S	second
SNB	Sierra Nevada batholith
Sr _(i)	initial ⁸⁷ Sr/ ⁸⁶ Sr composition
TIMS	thermal ionization mass spectrometer / spectrometry
TIS	Tuolumne Intrusive Suite
wt%	weight percent
Xi	percentage proportion of a mineral in a rock

INTRODUCTION

The development of K-feldspar megacrysts in granitoids is important to understanding plutonic and tectonic histories, yet their origin is still debated. Classically, these crystals have been thought to nucleate early in a large magma chamber with a high melt fraction (Vernon, 1986; Vernon and Paterson, 2008; Moore and Sisson, 2008), developing observed megacrystic textures in granitoids through flow sorting (Paterson et al., 2005; Barboni and Schoene, 2014). An alternative hypothesis is that megacrystic textures develop during protracted, late-stage cooling with small magma increments providing thermal oscillations that drive coarsening (Higgins, 1999; Johnson and Glazner, 2010; Glazner and Johnson, 2013). Theory (Simakin and Bindeman, 2008) and experiments (Mills et al., 2011; Mills and Glazner, 2013) demonstrate that thermal cycling can lead to dissolution-reprecipitation which coarsens crystals via cannibalism of smaller crystals by larger crystals. These thermal variations have been suggested as the mechanism leading to crystal coarsening and megacrystic textures in granitoids (Johnson and Glazner, 2010).

Single crystal isotope and trace element profiles provide information on the chemical environment during texture development in igneous rocks. Strontium isotopic profiling of crystals (via microsampling) has been used to investigate residence times (Christensen and DePaolo, 1993), magma mixing (Geist et al., 1988; Davidson et al., 1990; Cox et al., 1996; Davidson and Tepley, 1997; Gagnevin et al., 2005a), metasomatic and metamorphic processes (Christensen et al., 1989), and protracted magma chamber cooling (Siebel et al., 2005). Lead isotopic transects of K-feldspars have been used to interpret open-system processes in granites

(Gagnevin et al., 2005b), study terrane correlations (Connelly and Thrane, 2005), and provide data for provenance studies (Tyrrell et al., 2006).

I analyzed Rb, Sr, Pb, and U concentrations and isotopes by thermal ionization mass spectrometry (TIMS) from core-to-rim using micromill techniques. The K-feldspar megacrysts were collected from zoned intrusive suites of the Sierra Nevada batholith (SNB) of California. K-feldspar in the granites and granodiorites of the Tuolumne Intrusive Suite (TIS) has high concentrations of Sr and Pb. Whole-rock Sr and Pb isotopic variability in the TIS is well documented (Kistler et al., 1986; Gray et al., 2008; Burgess and Miller, 2008). Comparing and contrasting the whole-rock isotopic data with microsampled, K-feldspar isotopic data provides information on the state of the magma system during K-feldspar crystallization and ultimately aids in understanding the textural coarsening process.

Geologic Setting

Due to the excellent exposure after Pleistocene glaciation, the TIS (Bateman, 1992) has been the subject of many geochronological (Coleman and Glazner, 1997; Glazner et al., 2004; Memeti et al., 2010), geochemical (Bateman and Chappell, 1979; Kistler et al., 1986; Gray et al., 2008; Burgess and Miller, 2008; Mills et al., 2009; Coleman et al., 2012) and petrological studies (Kerrick, 1969; Higgins, 1999; Paterson et al., 2005; Johnson and Glazner, 2010; Glazner and Johnson, 2013; Bauer, 2015).

Rock compositions in the TIS vary from tonalitic and granodioritic at the exterior to granitic in the interior (Bateman and Chappell, 1979). Mapped plutons in the TIS are (from oldest to youngest): Sentinel Granodiorite, Kuna suite (Kuna Crest Granodiorite, tonalite of Glen Aulin, tonalite of Glacier Point), equigranular Half Dome Granodiorite, porphyritic Half Dome Granodiorite, Cathedral Peak Granodiorite, and Johnson Granite Porphyry (**Fig. 1**) (Bateman, 1992). Although sharp contacts exist locally, most contacts are gradational over tens of meters

(Bateman, 1992; Coleman et al., 2012). The TIS displays variation in U-Pb zircon ages of ~ 8 Ma; ages monotonically decrease from 93.5 ± 1 Ma in the Kuna Crest to $85.4\pm.01$ Ma in the Johnson Granite Porphyry (Coleman et al., 2004; Memeti et al., 2010).



Figure 1: Simplified geologic map of the TIS (after Huber et al., 1989). Inset shows detail of TIS and sample locations. Blue hexagons indicate sample localities for this study.

The Cathedral Peak Granodiorite (Kcp) is the largest mapped intrusive unit in the TIS and its modal composition plots in the granite to granodiorite fields (Bateman and Chappell, 1979; Bateman, 1992). The major identifying feature of Kcp is its abundance of K-feldspar megacrysts (megacrysts are defined in this study as crystals with maximum dimension >5 cm). The megacrysts are hosted in a medium to coarse-grained matrix of oligoclase, quartz, biotite, and hornblende. The Half Dome Granodiorite consists of two facies, the equigranular member (Khd) and the porphyritic member (Khdp). The major identifying feature of Khd is its large euhedral hornblende and titanite phenocrysts. Khd granular groundmass consists of plagioclase, biotite, quartz, magnetite and apatite. Khdp contains similar modal compositions of minerals and the porphyry is defined by its abundance of K-feldspar phenocrysts.

K-feldspar megacrysts in Kcp are pink to white and range from 5 - 20 cm in longest dimension, with the largest crystals occurring near the contacts of Kcp and Khdp (Johnson and Glazner, 2010). The megacrysts are potassic (Or₈₅₋₉₅) (Kerrick, 1969; Johnson and Glazner, 2010) and color cathodoluminescence (**Fig. 2**) and backscattered electron (BSE) images (**Fig. 3**) show minor perthite texture within the crystals. BSE images also highlight the variability in Ba concentrations (Moore and Sisson, 2008; Johnson and Glazner, 2010).

Mineral inclusions are ubiquitous in the megacrysts. Mineral inclusions within K-feldspar megacrysts contain in decreasing abundance plagioclase, hornblende, biotite, quartz, titanite, Fe-Ti oxide, apatite, and zircon. These inclusions tend to be oriented parallel to the $\{010\}$, $\{001\}$, and $\{110\}$ forms of the host megacrysts and are concentrated into mineral bands (**Fig. 2, 3, 5**). Grains with the mineral bands tend to be segregated by size; in some mineral bands inclusions of hornblende, plagioclase, and biotite reach ~1 mm, although average size for inclusions is 0.1 - 0.5 mm. Plagioclase, biotite, and titanite inclusions are subhedral to euhedral. Hornblende

inclusions are commonly anhedral to euhedral. In megacryst JKCM3, a euhedral titanite crystal contains inclusions of biotite, Fe-Ti oxide and K-feldspar.



Figure 2: Color cathodoluminescence image of K-feldspar megacryst JKCM3. Image highlights perthite texture in the megacrysts and abundant mineral inclusions: biotite (bio), quartz (qtz), titanite (ttn), hornblende (hbl), plagioclase (pl)



Figure 3: Backscattered electron image of megacrysts JKCM3. Gray-scale brightness is positively correlated with Ba concentration (Johnson and Glazner, 2010).

METHODS

Sampling Strategy

Seven K-feldspar megacrysts from Kcp were selected for this study. Megacryst samples were collected from two localities on the western side of the TIS and one locality near the eastern margin where Kcp is in contact with Jurassic metavolcanic rocks (**Fig. 1**). Sample JKW is the only sample collected outside of the TIS and data for it are presented in the appendix. Megacrysts were collected from outcrops where preferential weathering of biotite grains surrounding the megacrysts allowed for easy extraction from the outcrop with a chisel and hammer.

JKCM samples are from the Daff Dome area in the central portion of Kcp, where the contact with Khd is ~2 km to the north and Khdp ~4 km to the west (**Fig. 1**). JTF samples are from the Tuolumne Falls area; the closest contact is Khd rather than Khdp, which is not the typical gradation in the TIS. JSL samples were collected from a locality, near Steelhead Lake where Kcp is in contact with Jurassic metavolcanic rocks. These samples were collected in order to compare geochemistry between adjacent megacrysts and if proximity to the wall rocks indicated any evidence of hydrothermal fluid exchange.



Figure 4: Sections of K-feldspar megacrysts samples in this study. All samples come from the Kcp of the TIS. Red circles indicate micromill drill positions. Blue dotted lines outline megacrysts JSL1 and JSL2.

Micromilling

All samples, except the Steelhead Lake samples, were cut through their centers to (010), polished to 800 µm, and microdrilled on a Leica Merchantek GZ6 Micromill. Samples from the Steelhead Lake group come from a feldspar mosaic, and were cut so that grain boundaries of megacrysts were exposed. Afterwards a cut was made through the mosaic as close as possible to parallel (010) planes of the two adjacent feldspars.

Microsample drill locations were designed so traverses avoided inclusions and areas of alteration (**Fig. 4**). Drill tracks were completed from core-to-rim with drill spacing 1 - 2 mm, depending on the crystal size. Drill depths ranged from 750 µm – 1000 µm. Sample weights varied between 0.01 - 0.1 mg, depending amount of drilled material that was recovered. For Pb isotopic analysis separate aliquots of sample were collected from drilling deeper into the pit from which the Sr aliquot was collected. Samples were extracted using weigh paper as a scoop. This was preferred over collecting a slurry of water and milled material (e.g., Tepley et al., 1999; Charlier et al., 2006) because it allowed for inspection under a binocular microscope to ensure milled material did not contain pieces of biotite inclusions which may have come from below the crystal's surface. Milled material was placed directly into clean *Savillex Teflon*® beakers.



Figure 5: a) K-feldspar megacryst from Kcp. Feldspar displays concentric bands of plagioclase and other minerals including nearly all phases in the host rock. *b)* Red circles indicate microdrilled transect. Mineral inclusions are highlighted; biotite (bio), quartz (qtz), titanite (ttn), hornblende (hbl), and plagioclase (pl).

Isotope Geochemistry

Rb/Sr

Microsampled material was dissolved using a mixture of approximately 50 μ L of 29 M HF and 1000 μ L of 8M HNO₃. Samples were spiked with a mixed ⁸⁷Rb–⁸⁴Sr Spike (UNC B-3, 0.9606 ppm ⁸⁷Rb: 0.1445 ppm ⁸⁴Sr), spike weights varied near 0.03 ± 0.01 g. After 24 h of dissolution at 170 °C samples were evaporated to dryness and approximately 500 μ L of 7 M HNO₃ was added to ensure conversion of fluorides to nitrates. After an additional 24 h on a hotplate at 170 °C, the samples were evaporated to dryness again to be dissolved in 550 μ L of 3.5 M HNO₃ as a loading solution for cation chemistry.

The sample solution was passed through microcolumns containing *Eichrom SR-B100-S*® Sr Resin. Strontium was set on the column by passing 500 μ L of solution through the column. Afterward the 1.49 mL of Rb-effluent HNO₃ was collected in separate *Savillex Teflon*® beakers and set aside. Next 1 mL of DirectQ water was used to elute the Sr from the resin. 1 drop (~30 μ L) of H₃PO₄ was added to prevent complete evaporation before being dried down to a small bead on a hot plate at 110 °C for 4 – 6 h.

Rubidium fractions were dried down on a hotplate and converted to chlorides by adding ~500 μ L of 2 M HCL. The solution was loaded into the *Dowex AG50 WX8*® (50 – 100 mesh) cation columns with 5.75 mL of 2M HCl. Another 1.5 mL of 2M HCl of eluted material was collected and dried down in another *Savillex Teflon*® beaker.

U/Pb

Sampled material was dissolved in the same manner as Sr procedure (50 μ L of 29 M HF and 1000 μ L of 8M HNO₃.) Before dissolution approximately 0.04 g of the mixed ²⁰⁵Pb-²³³U-²³⁶U GS-1 spike was added to the samples. After 24 h of dissolution at 170 °C, samples were evaporated to dryness and converted to chlorides by adding approximately 700 μ L of 6 M HCl.

After an additional 24 h on a hot plate at 170 °C, samples were evaporated to dryness again and converted to bromides by the addition of 550 μ L of 0.5 M HBr.

Approximately 1 mL of 0.5 M HBr was used to set the samples onto the *Eichrom AG-IX8*® anion resin. Uranium was eluted with 240 μ L of 2 M HCl and stored in separate beakers. The addition of approximately 1.44 mL of 6 M HCl was required for the elution of Pb, one drop of H₃PO₄ was added before drying Pb fraction on a hot plate.

The U fraction was dried down and redissolved with ~150 μ L of 6 M HCl. The sample was loaded on to the same *AG-1X8* resin and after setting with 1.2 mL of 6 M HCl the U was eluted from the resin with an addition of 1 mL of water. One drop of H₃PO₄ was added before drying down to a small ~1 μ L bead.

Analysis

Strontium samples were mixed with 2 μ L of HCl and 2 μ L of Ta oxide emitter solution (TaF₅) before being loaded on a Re filament. Rubidium samples were hydrated with 2 μ L of water and loaded in 1 μ l loads onto a Re filament, taken to a current of ~1 A and allowed to dry flat. Boise State University-Ta Gel mixed with 1M H₃PO₄ in a 2:1 ratio was mixed beforehand and 3 μ L was the current was increased at approximately 0.1 A per 10 s increments until a current of 2.5 A was reached and the bead was flat. Uranium and Pb samples were loaded directly onto Re filaments with 3 μ L of silica-gel and heated until flat on the filament.

A VG Sector 54 thermal ionization mass spectrometer (TIMS) was used to determine the isotopic composition of each separate. Rubidium isotopic analysis were conducted in the static collection mode using Faraday detectors. Strontium analyses used a 3 multidynamic analysis.

Rubidium abundances in samples were calculated from the 85 Rb/ 87 Rb ratios, using the natural Rb isotopic composition of 85 Rb/ 87 Rb = 2.59376 (Nebel et al., 2005). Long-term

measurements of NBS 727 in the UNC TIMS laboratory yields a Rb fractionation correction of $\pm 0.30823\%$ per amu.

Uranium was measured on the Daly detector for ${}^{233}\text{U}/{}^{235}\text{U}$ and ${}^{238}\text{U}/{}^{233}\text{U}$. Lead analysis utilized the Daly detector (${}^{205}\text{Pb}$) and a Faraday detector (${}^{206}\text{Pb}$) to measure ${}^{205}\text{Pb}/{}^{206}\text{Pb}$ ratios. A second analysis using only Faraday detectors provided ${}^{206}\text{Pb}/{}^{204}\text{Pb}$ and ${}^{207}\text{Pb}/{}^{204}\text{Pb}$ ratios. Because a Th spike was not employed, ${}^{208}\text{Pb}$ was not measured because it could not be age-corrected. Long-term measurements of NBS 981 in the UNC TIMS laboratory were used to determine a fractionation correction of $0.12 \pm 0.06\%$ per amu for all Pb analyses.

Rubidium, Sr, U and Pb isotopic data were first reduced using the Tripoli application (Bowring et al., 2011). Rubidium and Sr isotopic measurements were reduced in a Monte Carlo simulation developed using R (described in appendix), whereas U/Pb data were reduced with the Pb MacDat-2, program of Coleman (unpublished) utilizing data reduction and error propagation of Ludwig (1980).

RESULTS

This study yielded the first isotopic and trace element profiling of K-feldspar megacrysts from the SNB. Sixty-six individual Sr isotopic analyses (**Table 2-1**) and 36 Pb isotopic analyses (**Table 2-2**) are presented from seven megacrysts collected from Kcp. Ten Sr isotopic analysis are presented in the appendix for one micromilled transect completed on a megacryst from the Whitney Granodiorite of the Mount Whitney Intrusive Series.

Field Relationships

Megacryst ranging from 5 - 15 cm long are common in Kcp (**Fig. 6a**). Locally they are scattered throughout the unit with sizes varying from megacrystic to the size of the surrounding groundmass (1 - 4 cm) (**Fig. 6b**). The largest megacrysts, up to ~20 cm in length, occur near the contact between Kcp and Khdp (Johnson and Glazner, 2010).

Commonly K-Feldspar megacrysts occur in dense concentrations with greater than 80% K-feldspar (**Fig. 6c, 6d, 6f, 6g**). These dense concentrations, hereafter referred to as mosaics (Glazner and Johnson, 2013), are scattered throughout Kcp in patches a meter or two across. Mosaics with megacrysts over 10 cm in length occur in the Tuolumne Falls area (**Fig 6d**). These mosaics also occur in tabular concentrations which can reach 1 m in width and 15 m in length (**Fig 6f**). In some instances, high concentrations of megacrysts are bounded by rinds of biotite and hornblende, which separate the mosaics from normal megacrystic host rock.

In the eastern portion of the TIS near Steelhead Lake a megacrysts mosaic are in contact with the Jurassic metavolcanic wall rock (**Fig. 6e**). At this locality K-feldspar mosaics form an

irregular contact with the wall rock (**Fig. 6g**), and multiple megacrysts indent the contact, up to 5 cm in the wall rock (**Fig. 6h**).



Figure 6: Photographs of megacrysts from Kcp in different textural settings. **a**) Megacrysts showing inclusions of biotite and hornblende, hand lens for scale (~5 cm). **b**) Typical megacrystic size and distribution within Kcp. **c**) K-feldspar mosaic in Kcp where megacrysts are estimated to make up 80% of the bulk rock. **d**) K-felspar mosaic with largest megacrysts seen in this study near Tuolumne Falls. Here average feldspar exceeds 10 cm in length.



Figure 6 continued: e) K-feldspar mosaic in contact with Jurassic metavolcanic wall rock. f) K-feldspar mosaic near Tuolumne Falls bounded by a rind rich in biotite and hornblende. g) Close-up of Fig 6e, where megacrysts are seen with irregular contacts with Jurassic wall rock. h) Megacrysts were observed indenting \sim 5 cm into the contact with Jurassic wall rock.

Rb & Sr Trace Element Geochemistry

Strontium concentrations in megacrysts range from $\sim 300 - 4000$ ppm and Rb concentrations in megacrysts range from $\sim 60 - 800$ ppm (**Table 2-1**) and Rb/Sr ratios of s range from 0.14 to 0.46. There is no overall core-to-rim trend in Rb/Sr values, which scatter about a mean value of 0.22.

Daff Dome Rb & Sr Trace Element Geochemistry

Samples from the Daff Dome group range in Rb/Sr from 0.15 to 0.38. All samples from this locality show at least one major excursion for which Rb/Sr values deviates at least \pm 0.04 from the mean; in JKCM3 and JKCM5 the excursions are positive, whereas in JKCM4 the excursion is negative. Each sample contains their largest Rb/Sr ratio at the rim (**Fig. 7**).



Figure 7: Daff Dome sample group's Rb, Sr concentrations and Rb/Sr ratios plotted against normalized distance from core.

Tuolumne Falls Rb & Sr Trace Element Geochemistry

Samples from the Tuolumne Falls area show a narrower range in Rb/Sr values than samples from Daff Dome, with Rb/Sr values ranging from 0.17 to 0.26. Sample JTF1 records the most regular variation in Rb/Sr values, with changes of ~0.02 or greater occurring between each analysis along the transect. Rb/Sr ratios at the rims of JTF2 and JTF3 are lower than at the cores (**Fig. 8**).



Figure 8: Tuolumne Falls sample group's Rb, Sr concentrations and Rb/Sr ratios plotted against normalized distance from core.

Steelhead Lake Rb & Sr Trace Element Geochemistry

Both JSL1 and JSL2 are nearly 5 cm in length and have much higher abundances of mineral inclusions than other samples from this study. To avoid contamination from the high abundance of mineral inclusions in these megacrysts, milled transects were not set in a continuous line (see Fig. 4); the same spacing between analysis was maintained until transects spanned the length from core-to-rim. Rb/Sr ranges for each of transect of JSL1 and JSL2 are the greatest range in data set with values from 0.29 to 0.47. JSL1 shows a coarse increase in Rb/Sr values towards the rim, whereas Rb/Sr values in JSL2 decrease dramatically away from the core (Fig. 9).



Figure 9: Steelhead Lake sample group's Rb, Sr concentrations and Rb/Sr ratios plotted against normalized distance from core.

Sr Isotopic Transects

 87 Sr/ 86 Sr_(i) was calculated using the youngest recorded zircon age of Kcp at 88.1 Ma (**Table 2-1**; Coleman et al., 2004). In order to quantify the impact of age correction on the isotopic compositions, Sr isotopes were corrected to three different ages ranging in total 8 Ma. Calculated Sr_(i) values change less than 0.000025 over an 8 Ma age range (**Fig. 10**), which is much less than the calculated analytical uncertainty in each of the analyses (see appendix for methods of uncertainty calculation). Thus, age assumptions do not impact the interpretation of initial Sr isotopic compositions.



Figure 10: Different Sr isotopic composition based on different assumed crystallization ages (84.1 Ma, 88.1 Ma, 92.1 Ma). Difference in Sr isotopic compositions are less than 0.000025 between samples corrected at 84.1 Ma and 92 Ma.

Initial Sr isotopic values for megacrysts range from 0.706062 to 0.707651. The majority of these samples fall within previously reported ranges of Kcp (Kistler et al., 1986; Gray, 2003; Burgess and Miller, 2008), whereas two analyses from the JSL samples extend the range higher than previously reported values.



Figure 11: Initial Sr isotopic compositions of samples along their normalized distances from core-to-rim. Error bars indicate calculated 2σ uncertainty and may be smaller than symbol.

Daff Dome megacryst initial Sr isotopic composition

Samples from the Daff Dome sample suite have little variation in $Sr_{(i)}$ from core-to-rim. More abundant and larger magnitude excursions occur in JKCM3 than other JKCM samples; the largest negative excursion occurring 8 mm away from the rim, or approximately 2/3 of the distance across the crystal. JKCM4 and JKCM5 isotopic values from the transect closely parallel each other, with small increases and decreases in $Sr_{(i)}$ occurring near the core before becoming less radiogenic near the rim. All analyses at the rims for the samples of the Daff Dome suite are within analytical uncertainty at a value of ~0.70636 (**Fig. 11**).

Tuolumne Falls megacryst initial Sr isotopic composition

Samples from the Tuolumne Falls area contain generally more restricted values for initial Sr isotopic compositions than megacrysts from other sample locations. Samples JTF1 and JTF2 have excursions in $Sr_{(i)}$ of 0.0005 or greater occurring in the middle of the transects. Otherwise, $Sr_{(i)}$ compositions across the crystals are relatively flat compared to other sample suites. JTF1 is the only sample in which the rim values are significantly different than its core (> 0.0002) (**Fig. 11**).

Steelhead Lake megacryst initial Sr isotopic composition

Samples from the Steelhead Lake area are more radiogenic than the rest of the sample set from the TIS. The transect across each crystal shows the greatest variability in $Sr_{(i)}$ in this study. The isotopic profile across each of these crystals show a scatter around the mean of values of the transects. Core (< 50% of transect) values of $Sr_{(i)}$ each average near 0.70644, and each crystal contains radiogenic ($Sr_{(i)} > 0.707$) values near the rim (**Fig. 11**).
U/Pb Trace Element Geochemistry and Pb Isotopic Compositions

Four megacrysts were analyzed for Pb isotopic compositions, leading to thirty-six analyses (**Table 2-2**). Uranium concentrations in megacrysts range from ~0.005 – 2.5 ppm, Pb concentrations range from ~0.75 – 64.5 ppm, and U/Pb ratios are ~0.0005 – 0.08. Initial Pb isotopic compositions range from 18.78 – 18.99 for 206 Pb/ 204 Pb_(i) and 0.825 – 0.835 for 207 Pb/ 206 Pb_(i). The Pb isotopic compositions of megacrysts are similar to previously published whole-rock values for Kcp (Gray et al., 2008).



Figure 12: U/Pb ratios, ${}^{206}Pb/{}^{204}Pb_{(i)}$, ${}^{207}Pb/{}^{206}Pb_{(i)}$ compositions of megacrysts. Transect distances of each crystal are normalized and referenced from core-to-rim. 2σ uncertainty in isotopic compositions is smaller than symbol size.

JKCM3 U/Pb ratios and initial Pb isotopic composition

The U/Pb ratios in sample JKCM3 vary from 0.0016 to 0.02. The largest U/Pb ratio occurs at the rim. Excluding the rim analysis, the U/Pb ratios are fairly homogenous and low compared with other megacrysts. ²⁰⁶Pb/²⁰⁴Pb_(i) values range from 18.41 to 18.99, ²⁰⁷Pb/²⁰⁶Pb_(i) values range between 0.825 and 0.831. The largest excursion from the mean ²⁰⁶Pb/²⁰⁴Pb_(i) and ²⁰⁷Pb/²⁰⁴Pb_(i) values occur near the center of the transect (**Fig. 12**). The rim analysis has a ²⁰⁶Pb/²⁰⁴Pb_(i) value of 18.86 and ²⁰⁷Pb/²⁰⁶Pb_(i) value of 0.831, which is significantly different than analyses at the core.

JKCM4 U/Pb ratios and initial Pb isotopic composition

The U/Pb ratios in sample JKCM4 range from 0.0004 and 0.07 and the minima and maxima occur in analyses that are adjacent to each other along the transect. JKCM4's $^{206}Pb/^{204}Pb_{(i)}$ values range from 18.82 to 18.93, with the highest value being 4 mm from the center of the core and the lowest value at 8 mm away from the core. $^{207}Pb/^{206}Pb_{(i)}$ values range between 0.827 and 0.832. In this sample the highest $^{206}Pb/^{204}Pb_{(i)}$ value corresponds to the largest U/Pb ratio in the sample (0.07). Analyses near the rim (16 mm – 23.5 mm) average $^{206}Pb/^{204}Pb_{(i)}$ composition is 18.89, which is slightly higher than average values at the interior of the crystal (**Fig. 12**).

JKCM5 U/Pb ratios and initial Pb isotopic composition

The U/Pb ratios in sample JKCM5 are variable between 0.002 and 0.08. $^{206}Pb/^{204}Pb_{(i)}$ isotopic compositions range from 18.79 to 18.95, $^{207}Pb/^{206}Pb_{(i)}$ values range between 0.830 and 0.833. Only one large excursion occurs in $^{206}Pb/^{204}Pb_{(i)}$ near the core of the crystal with the maximum value of 18.95. $^{207}Pb/^{206}Pb_{(i)}$ values are fairly homogenous compared with other megacrysts, and show only slight deviation from the mean of 0.832 (**Fig. 12**).

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JTF2 U/Pb ratios and initial Pb isotopic composition

The U/Pb values range from 0.0015 to 0.032, and show a slight increase in the first 4 mm of the transect; the remainder of U/Pb ratios along the transect show little variability from the mean value of the transect. ²⁰⁶Pb/²⁰⁴Pb_(i) values range from 18.79 to 18.95, ²⁰⁷Pb/²⁰⁶Pb_(i) values range from 0.827 to 0.835. ²⁰⁶Pb/²⁰⁴Pb_(i) increases and ²⁰⁷Pb/²⁰⁶Pb_(i) decreases near the core of the megacryst, with the largest excursions of each ratio occurring near the center of the transect (**Fig. 12**). Rim values of ²⁰⁶Pb/²⁰⁴Pb_(i) show little variation from the core, whereas ²⁰⁷Pb/²⁰⁶Pb_(i) rim compositions are slightly larger than the core values.

DISCUSSION

Microsamples of megacrysts were analyzed for trace element and isotopic compositions in order to assess their origin in the TIS. If crystals nucleated and grew in a closed environment with a high-melt fraction, then microsamples should be isotopically homogeneous from core-torim. Alternatively, if magma mixing or mingling occurred the megacrysts could have recorded isotopic and compositional variations. If the K-feldspars formed from late stage coarsening, in a low-melt fraction, then their trace element and isotopic signatures should record the composition of the local, cannibalized material. In order to evaluate these hypotheses, it is necessary to describe and account for the isotopic and trace element profiles. Moreover, it is important to establish whether the analyses are primary feldspar values and do not represent contamination by inclusions or some other artifact that occludes a primary feldspar signature.

Isotopic Variation in the Megacrysts

Crystal transects from this study show little variation relative to the values recorded in the cores (**Fig. 13**). The variation in each transect in $Sr_{(i)}$ relative to the core analysis typically falls below 0.0002 (**Fig. 13a**). Differences in Pb isotopic compositions are similarly small with $^{207}Pb/^{206}Pb_{(i)}$, on average, recording less than 0.01 variation (**Fig. 13b**).



Figure 13: Absolute differences between isotopic compositions along transects relative to isotopic compositions in the grain core. Values are plotted along normalized distances of the crystals length from core (0)-to-rim (1). **a**) Differences between initial Sr isotopic compositions. Most differences between analyses from core value fall under 0.0002. Rim analyses of JSL1 (0.7) and JSL2 (1.0) have the greatest differences relative to their cores. **b**) Differences of 207 Pb/ 206 Pb_(i) relative to core values. Most differences between analyses and core value are less than 0.006. The most significant outlier is the rim analysis (1.0) of JTF2 that shows a difference of 0.022.

Isotopic compositions of K-feldspar megacrysts are more restricted than whole-rock compositions from the TIS. Kernel density estimations of $Sr_{(i)}$ values were plotted against compiled whole-rock values (Kistler et al., 1986; Gray, 2003; Burgess and Miller, 2008) to compare the isotopic variabilities (**Fig. 14**). K-feldspar megacryst mean $Sr_{(i)}$ is 0.7064, similar to mean $Sr_{(i)}$ of Kcp and Khdp. Initial Sr isotopic compositions of Khd have much higher variance and a distinctly different mean $Sr_{(i)}$ at 0.7062. Whole-rock data from the TIS span a large range in Sr and Pb isotopic compositions (~0.705 – 0.707 in $Sr_{(i)}$, ~18.3 – 19.2 in ²⁰⁶Pb/²⁰⁴Pb_(i), ~0.81 – 0.83 in ²⁰⁷Pb/²⁰⁶Pb_(i)), but Kcp and Khd have a much smaller range in $Sr_{(i)}$, 0.7058 to 0.7066. Although there are only two samples of whole-rock Kcp with both initial Sr and Pb reported in the literature (Gray, 2003), these data fall into the narrow range in initial Sr-Pb space defined by the megacrysts (**Fig. 15**).



Figure 14: Kernel density estimations (KDE) of $Sr_{(i)}$ of K-feldspar megacrysts and granodiorites of the TIS. The mean of the units of these compiled data are 0.70643 for megacrysts (n=63), 0.70641 for Kcp (n=32), 0.70640 for Khdp (n=17), and 0.70622 for Khd (n=33). Although the megacryst data fall into a sharp peak, there is some scatter which leads to a larger base than Kcp. Whole-rock data compiled from Kistler et al. (1986), Gray, (2003) and Burgess and Miller (2008). Data for units' age corrected for youngest recorded zircon ages. The uncertainty in the age of the whole-rock samples produces uncertainty in the calculated ${}^{87}Sr/{}^{86}Sr_{(i)}$ values. However, for interpretations here the uncertainty in the isotopic composition related to age uncertainty is negligible. For example, an age uncertainty of 4 Ma yields ${}^{87}Sr/{}^{86}Sr_{(i)}$ variations less than 1 x 10⁻⁴ for the whole-rock data.

The low deviations in isotopic compositions suggest either crystals did not record any hybridization within the magmatic system, or isotopic signatures were homogenized through interaction with metasomatic fluids. Fluid alteration resulting in loss of Sr, and shifts in ⁸⁷Sr/⁸⁶Sr toward altered material has been documented in other settings (Waight et al., 2001; Mills, 2007;

Gross, 2015; Doorn, 2016). Although this process seems unlikely in K-feldspar since Sr concentrations and isotopic retention would occur in all but the most extreme thermal events (Cherniak and Watson, 1992). Moreover, the comparable low variation in Pb isotopes reinforces the idea that Sr_(i) is a primary signal, and geochemical compositions were not reset through alteration or diffusion of Sr. The limited variability in Sr_(i) among individual megacrysts and the similarity of these values to the isotopic compositions of Kcp and Khdp suggest that megacryst Sr_(i) isotopic compositions were acquired through crystallizing from a magma of Kcp or Khdp, and likely not Khd or any of the other Tuolumne plutons.



Figure 15: Initial Sr plotted against initial Pb isotopic ratios of the feldspars. Whole-rock data plotted as grey squares from Gray (2003). Kcp samples plotted as blue squares. Daff dome samples plotted as colored circles. JTF2 plotted as purple triangles. **a**) ${}^{87}Sr/{}^{86}Sr_{(i)}$ vs ${}^{206}Pb/{}^{204}Pb_{(i)}$ **b**) ${}^{87}Sr/{}^{86}Sr_{(i)}$ vs ${}^{207}Pb/{}^{206}Pb_{(i)}$. Kernel density estimations (KDE) are plotted along their respective axes. Red lines indicate KDE for megacrysts, grey lines indicate KDE for whole-rock values for units excluding the Kcp, for which there are only two published measurements (blue lines).

Trace Element Variation in the Megacrysts

The trace element stratigraphy of megacrysts show scattered variability around a mean from core-to-rim. How this variability developed is important in understanding the crystallization settings in which the megacrysts grew. Although the trace element variations are small, interpretations from their radiogenic isotopes are crucial to this study. Therefore, it is important to address weather these heterogeneities are in fact primary signals recorded by the megacrysts, or an artifact of methodology.

Contamination During Microsampling

An unavoidable uncertainty in microsampling is that mineral inclusions below the surface of K-feldspar megacrysts were sampled. The most common inclusions in the megacrysts are biotite and plagioclase. Due to the low modal abundance of these inclusions in the megacrysts, the fact that the inclusions tend to be concentrated in narrow bands within the megacrysts (**Fig. 4**, **5**, **6**), careful placement of drill holes, it is unlikely significant phases other than K-feldspar were sampled. The isotopic compositions of these phases were not independently measured. However, the limited isotopic variability of the data suggests that, if biotite and plagioclase contaminated the analyses, they were isotopically similar to the host megacrysts. Otherwise, it would be fortuitous that random contamination of isotopically variable phases would yield such homogeneous results. It is, however, possible to evaluate potential contamination by examining trace element ratio (Rb/Sr) variations along the transects.

Two-component mixing plots were generated for Rb and Sr using the data from this study and concentration data from Kcp biotite and plagioclase (Kistler et al., 1986). These models indicate that micro-samples could have up to ~20 wt% biotite or ~50 wt% plagioclase and Rb/Sr ratios could fall within the range of measured values (**Fig. 16**). Although, for the reasons outlined

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above, it seems unlikely that such substantial fractions of inclusions were sampled, their potential impact on the results of trace element transects cannot be ignored.



Figure 16: Two-component mixing plots between K-feldspar and biotite (a) and plagioclase (b). Red lines indicate ranges for data of K-feldspar megacrysts, green line indicates mean value. Maximum Rb/Sr range in b) far exceeds mixing line, and is not plotted. Modeling indicates greater than 20 wt% biotite or 50 wt% plagioclase in sample would be required to offset Rb/Sr ratios dramatically. Trace element data for biotite and plagioclase from Kistler et al. (1986).

Trace Element Modeling

Several crystallization models were tested to evaluate their predicted impact on trace element profiles. Three variations of crystallization were modeled: equilibrium crystallization (**Eq. 3**), Rayleigh fractionation (**Eq. 4**), and *in situ* crystallization (**Eq. 5**). Trace element concentration profiles were calculated for K-feldspar crystallizing in each of these regimes.

The Nernst distribution coefficient (Kd) describes the equilibrium distribution of a trace element between a mineral and melt and is defined by:

$$Kd_{i} = \frac{C_{element \, i}^{mineral}}{C_{element \, i}^{melt}} \tag{1}$$

where C is the concentration of the trace element *i* in the mineral and melt. A bulk distribution coefficient (D_i) can be calculated for any element according to mineral proportions by weight (x_i) in a melt through:

$$D_i = \sum x_i K d_i \tag{2}$$

Equilibrium crystallization is the processes where there is complete equilibrium between all solids and the melt during crystallization. Trace element concentrations for this model can be estimated by:

$$\frac{C_R}{C_0} = \frac{D}{[D + F(1 - D)]}$$
(3)

where C_R is the concentration of a trace element in the solid and C_0 is the concentration of a given trace element in the original melt.

Rayleigh fractionation is a disequilibrium crystallization mechanism in which crystals are removed from being in equilibrium with melt, yielding constant changes in the melt's trace element concentrations. Rayleigh fractionation can be modeled as:

$$\frac{C_R}{C_0} = DF^{(D-1)} \tag{4}$$

In situ crystallization developed by Langmuir (1989) describes crystallization taking place on the cool exteriors of magma chambers and residual liquids reacting *in situ* with the crystal mush at the walls of a magma chamber. This is similar to Rayleigh fractionation but trace element distributions are dependent on the fraction of residual melt interacting with the solidification zone. This model can be modeled as:

$$\frac{C_R}{C_0} = DF^{f(D-1)/[D(1-f)+f]}$$
(5)

where *f* is the fraction of melt interacting with the solids near the edges of the magma chamber; the limiting case is when f = 1.0, which is Rayleigh fractionation. Modeling used values of *f* of 0.2, 0.5, and 0.8.

To model trace element concentrations in K-feldspar certain assumptions need to be made about the melt from which they crystallized. (1) Because K-feldspar is thought to be a late crystallizing phase in Kcp (Glazner and Johnson, 2013), modeling was only conducted on melt fractions (*F*) less than 50%. (2) Exact mineral proportions of the starting material are unknown; therefore, only modal fractions of early crystallizing phases (plagioclase, biotite, and hornblende) from Kcp are used to calculate concentration of trace elements in the melt remaining at F = 0.5. (3) Bulk partition coefficients were calculated based on *Kd* values (**Table 1**) from a rhyolitic melt (Arth and Barker, 1976; Nash and Crecraft, 1985), because the Kcp melts would have essentially been rhyolitic after 50% crystallization (Bachmann and Bergantz, 2004). (4) The bulk *D* was calculated using the modal proportions (Bateman, 1992) of the late crystallizing phases of Kcp: K-feldspar, quartz and plagioclase. Using these assumptions, the concentrations of Sr, Rb, Pb, and Ba are modeled according to the different crystallization scenarios outlined above. To model the concentrations of each trace element across a transect of a feldspar the concentrations are represented with respect to the cubic root of the total mass of the feldspar crystal formed in each interval.

	Plagioclase	K-feldspar	Hornblende	Biotite	Quartz
Weight Fraction	0.17	0.69	0	0	0.13
<i>Kd</i> _{Sr}	19.90	5.90	0.22	0.41	0.00
Kd_{Rb}	0.13	1.80	0.01	3.20	0.00
Kd_{Pb}	1.31	2.55	0.26	0.85	0.00
Kd_{Ba}	1.52	11.45	0.04	23.53	0.00

Table 1: Kds and modal percentages of minerals used in trace element modeling

Data sources: Arth and Barker, 1976; Nash and Crecraft, 1985.

Model results (**Fig. 14**) show simple equilibrium crystallization and fractional crystallization cannot produce the concentrations profiles of trace elements observed in megacrysts (**Fig. 7, 8, 9, 12**). At low melt fractions, and high *f* values, the *in situ* model results are the closest to matching the actual ranges of concentrations observed in the megacrysts, but not their profile.

All of the modeled crystallization scenarios fail to mimic trace element patterns observed in the K-feldspar megacrysts. Although the possibility of contamination by other phases during micromilling cannot be dismissed for the results from this study, the modeling also fails to account for the zonation of trace elements (notably Ba) documented in earlier work on the Kcp megacrysts (Johnson and Glazner, 2010). To overcome the limitations of simple crystallization from a static magma chamber, a new model that incorporates melt recharge was explored (see appendix for R code). The recharge model was initialized with same parameters listed for F = 1 to 0.5. For melt fractions less than F = 0.5, a new recharge melt with different trace element concentrations is added to the system, and new concentrations in the mixed liquid are calculated before the next crystallization cycle. The recharged magma is modeled with aplite trace element compositions. Two types of aplitic liquids are modeled in the recharge scenario. The first contains traceelement concentrations of aplites from Kcp. Aplite data from the TIS are used because their mineral compositions and textures suggest late-stage melt extraction from a granitoid plutons (Glazner et al., 2008). The second modeled aplite composition is a hypothetical enriched endmember with high concentrations of Sr, Rb, Pb and Ba. Trace element concentrations in the residue, the remaining liquid, and the feldspar are numerically estimated by using bulk partition coefficients of a rhyolite (**Table 1**).

Model results demonstrate saw-tooth zoning, or scattered, trace element patterns can be obtained through the recharge scenario (**Fig. 15**). Modeled mixing of Kcp aplites produced saw-tooth profiles of Rb and Pb in K-feldspar, but failed to produce this pattern in Sr and Ba (**Fig. 18a**). The concentrations of Sr and Ba are lower in that aplite than the original melt with which they are mixing yielding stair-step patterns in Sr and Ba, because the materials added are essentially diluting the total mixed liquid. When the hypothetical enriched aplite recharge was modeled, saw-tooth patterns in all elements (**Fig. 18b**) were reproduced. This suggests one mechanism to produce similar trace element profiles seen in K-feldspar megacrysts might be melt enriched in trace elements relative to the host being injected into a crystallizing magma chamber. This idea of a late-stage melt addition to a granitoid system is also consonant with a melt providing thermal oscillations to promote crystal coarsening.



Figure 17: K-feldspar concentrations for Sr, Rb, and Pb were modeled under for three crystallization models **a**) equilibrium crystallization **b**) Rayleigh fractionation **c**) in situ crystallization. f is the amount of magma chamber from melt fraction returned to a solidification zone. None of these models adequately accounts for the variations measured in this study (e.g., compare with Fig. 7, 8, 9, 12).



Figure 18: Recharge model trace element profiles of Sr, Rb, Pb, and Ba in K-feldspar modeled with (a) Kcp aplites (Glazner et al., 2008) and (b) hypothetical enriched endmember aplite. The overall trend of increase or decrease occurs relative to an elements compatibility in K-feldspar versus other crystallizing phases. Recharge of with Kcp aplitic material produces saw-tooth zoning in [Rb] and [Pb] during each incremental addition of the injected liquid as these elements are enriched in these elements compared to Kcp, but stair-step profiles in [Sr] and [Ba]. The hypothetical enriched aplitic material produces saw-tooth zoning patterns in all trace element profiles, similar to those observed in K-feldspar megacrysts.

Comparison of Data from Cathedral Peak Granodiorite Megacrysts with Megacrysts from Other Studies

Isotopic and trace element heterogeneity recorded by single mineral grains have been attributed to magma mixing and mingling in other studies (Cox et al., 1996; Davidson and Tepley, 1997; Tepley et al., 1999; Słaby et al., 2007). Gagnevin et al. (2005a) analyzed initial Sr isotopic compositions of K-feldspar megacrysts from the Monte Capanne monzogranite (Elba, Italy). The Sr isotopic profiles of megacrysts recorded large differences between cores and rims, often up to ~0.005, in contrast to the total variation of 0.0015 and the more typical variation of only 0.0002 measured in this study. The Pb isotopic profiles of the same Elba crystals (Gagnevin et al., 2005b) also showed larger variations in ²⁰⁷Pb/²⁰⁶Pb_(i) isotopic compositions than megacrysts from Kcp. The heterogeneity of isotopic transects in megacrysts from Elba, and other locations (Cox et al., 1996; Waight et al., 2001) has led authors to invoke scenarios in which K-feldspars initially grew in a relatively silicic melt, acquiring radiogenic cores, and then through mafic recharge and magma mixing recorded less radiogenic rims.

Similar scenarios of K-feldspar growth in a magma chamber with a high melt fraction have been used in the Sierra Nevadan granodiorites (Vernon, 1986; Vernon and Paterson, 2008). However, because the isotopic transects of megacrysts from Kcp show little variability, a recharge model seems unlikely and a different model for their origin is required.

The Origin of K-Feldspar Megacrysts in the Cathedral Peak Granodiorite

Since the isotopic and trace element data of the megacrysts show little variability between cores and rim, it is evident that mafic recharge did not occur during K-feldspar growth in Kcp. When isotopic data of the megacryst are compared to whole-rock values of the TIS, data only permit the possibility that the megacrysts were in equilibrium with Kcp or Khdp, and did not record growth within any other older units. Therefore, crystals could not have nucleated early

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in a magma chamber with a high melt fraction, or coarsen through the injection of additional material into the system.

Predicted profiles from trace element modeling suggests that recharge from an isotopically similar, yet trace element enriched aplite could have created the slight fluctuations in Rb, Sr, and Pb concentrations, and saw-tooth zoning patterns observed in Ba (Kerrick, 1969; Słaby et al., 2007; Moore and Sisson, 2008). This is consistent with a model that calls on minor, late melt retention in a diachronous and inward-moving melt zone during protracted accumulation of the TIS (Johnson and Glazner, 2010; Glazner and Johnson, 2013).

The existence of both small interstitial K-feldspar and megacrystic K-feldspar can be achieved through crystal cannibalization if temperatures are buffered near the liquidus of Kfeldspar (Higgins, 1999; Mills et al., 2011; Mills and Glazner, 2013). The trace element stratigraphy in this study add to this interpretation, in that the megacrystic textures which define some units of the TIS (**Fig. 6**) are developed through open-system processes rather than monotonous cooling of a pluton with a high-melt fraction.

CONCLUSIONS

This is the first study which conducted microsampling of K-feldspar megacrysts from the Cathedral Peak Granodiorite for Sr and Pb trace element and initial isotopic compositions. Several important observations and conclusions can be derived from the data:

- Isotopic signatures (Sr and Pb) show minimal variation, indicating that, if the feldspars crystallized in an open-system environment, recharge magmas were isotopically very similar.
- The variation from core-to-rim of radiogenic isotopes is not comparable to variations documented in other volcanic and plutonic systems. This suggests that mafic recharge was not as significant in the development of the Kcp as in the other systems.
- Initial Sr and Pb compositions form a tight cluster similar to Kcp whole-rock data, indicating feldspar growth occurred in Kcp.
- Trace element modeling of closed system crystallization (equilibrium, Rayleigh and *in situ*) fail to reproduce profiles seen in K-feldspar megacrysts from the TIS. Ranges of concentrations similar to those in megacrysts are only reproduced at low melt fractions. This could explain variability in trace element profiles only if the melt was remobilized or crystal cannibalization occurred.

APPENDIX 1: TABLES OF GEOCHEMCIAL DATA

Table 2-1: Rb	and Sr Tra	ce Element and	Isotopic Data

Sample	Distance from Core	Measured	Rb	Sr	Calculated	Initial	Uncertainty
. =	(mm)	⁸⁷ Sr/ ⁸⁶ Sr	ppm	ppm	⁸⁷ Rb/ ⁸⁶ Sr	⁸⁷ Sr/ ⁸⁶ Sr	(±)
JKCM3-1	2.00	0.707047	67	302	0.6424	0.706298	0.0000339
JKCM3-2	3.25	0.707066	59	265	0.7072	0.706328	0.0000506
JKCM3-3	4.50	0.707095	136	538	0.7337	0.706234	0.0000446
JKCM3-4	5.75	0.707123	379	1689	0.6489	0.706361	0.0000298
JKCM3-5	7.00	0.707043	203	905	0.6485	0.706265	0.0000283
JKCM3-6	8.25	0.707065	208	709	0.8495	0.706060	0.0000356
JKCM3-7	9.50	0.707035	155	640	0.7005	0.706209	0.0000309
JKCM3-8	10.75	0.707322	169	656	0.7434	0.706430	0.0000296
JKCM3-9	12.00	0.707229	176	582	0.8749	0.706194	0.0000338
JKCM3-10	13.25	0.707386	75	245	0.8847	0.706386	0.0000852
JKCM4-1	1.00	0.707216	207	899	0.6677	0.706422	0.0000199
JKCM4-2	3.50	0.707135	115	563	0.5947	0.706428	0.0000192
JKCM4-3	6.00	0.706964	165	1072	0.4469	0.706446	0.0000182
JKCM4-4	8.50	0.707022	127	751	0.4919	0.706442	0.0000187
JKCM4-5	11.00	0.707046	244	1310	0.5412	0.706408	0.0000187
JKCM4-6	13.50	0.707046	163	842	0.5603	0.706383	0.0000184
JKCM4-7	16.00	0.707099	73	343	0.6141	0.706388	0.0000217
JKCM4-8	18.50	0.707081	127	626	0.5898	0.706389	0.0000193
JKCM4-10	23.50	0.707651	190	505	0.3763	0.706345	0.0000258
JKCM5-1	0.00	0.707115	86	423	0.5846	0.706412	0.0000188
JKCM5-2	2.25	0.707408	768	2718	0.8174	0.706419	0.0000214
JKCM5-3	4.50	0.707116	211	1119	0.5453	0.706478	0.0000199
JKCM5-5	9.00	0.707078	137	661	0.5979	0.706373	0.0000212
JKCM5-6	11.25	0.707088	239	1162	0.5955	0.706376	0.0000192

JKCM5-7	13.5	0.707057	79	385	0.5947	0.706355	0.0000202
JKCM5-8	15.75	0.707056	252	1281	0.5698	0.706379	0.0000188
JKCM5-10	20.25	0.707359	105	359	0.8458	0.706359	0.0000238
JTF1-1	2.00	0.707112	101	492	0.5923	0.706398	0.0000191
JTF1-2	3.00	0.707086	155	839	0.5359	0.706437	0.0000183
JTF1-3	4.00	0.707136	346	1585	0.6320	0.706372	0.0000195
JTF1-4	5.00	0.707123	789	3981	0.5736	0.706430	0.0000187
JTF1-5	6.00	0.707238	178	788	0.6540	0.706504	0.0000201
JTF1-6	7.00	0.707091	172	900	0.5525	0.706429	0.0000185
JTF1-7	8.00	0.707051	180	928	0.5613	0.706371	0.0000189
JTF1-8	9.00	0.707113	226	1094	0.3461	0.706404	0.0000165
JTF1-9	10.00	0.707082	136	687	0.7786	0.706394	0.0000223
JTF1-10	11.00	0.707161	195	805	0.9542	0.706313	0.0000263
JTF2-1	1.00	0.707326	488	1852	0.7321	0.706406	0.0000210
JTF2-2	2.00	0.707243	280	1138	0.6607	0.706391	0.0000208
JTF2-4	4.00	0.707330	136	660	0.8309	0.706620	0.0000215
JTF2-5	5.00	0.707307	175	682	0.8946	0.706412	0.0000216
JTF2-6	6.00	0.707130	460	2213	0.6550	0.706399	0.0000191
JTF2-7	7.00	0.707018	464	2469	0.3664	0.706367	0.0000192
JTF2-8	8.00	0.707070	208	1117	0.5381	0.706430	0.0000189
JTF2-9	9.00	0.707064	292	1588	1.3140	0.706428	0.0000201
JTF3-1	1.00	0.707125	329	1536	0.6198	0.706379	0.0000192
JTF3-2	2.00	0.707186	245	1105	0.6420	0.706421	0.0000201
JTF3-3	3.00	0.707183	156	710	0.6361	0.706416	0.0000193
JTF3-4	4.00	0.707091	75	351	0.6169	0.706368	0.0000200
JTF3-5	5.00	0.707044	138	704	0.5672	0.706366	0.0000193
JTF3-6	6.00	0.707104	76	379	0.5792	0.706411	0.0000194
JTF3-7	7.00	0.707136	291	1365	0.6171	0.706407	0.0000205
JTF3-8	8.00	0.706992	106	623	0.4937	0.706411	0.0000193
JMSL1-1	1.00	0.707788	179	559	0.9255	0.706644	0.0000282
JMSL1-2	2.00	0.708003	211	508	1.2000	0.706538	0.0000328

JMSL1-3	3.00	0.707882	165	384	1.2445	0.706355	0.0000314
JMSL1-4	4.00	0.707855	173	453	1.1047	0.706502	0.0000270
JMSL1-5	5.00	0.709199	379	862	1.2742	0.707651	0.0000299
JMSL2-1	1.00	0.707978	191	544	1.2511	0.706454	0.0000289
JMSL2-2	2.00	0.707861	117	400	1.1286	0.706487	0.0000273
JMSL2-3	3.00	0.708199	365	786	1.3442	0.706558	0.0000458
JMSL2-4	4.00	0.708272	188	482	0.8494	0.707235	0.0000223
JMSL2-5	5.00	0.707735	156	360	1.0147	0.706486	0.0000279
JKW-1	1.25	0.708112	53	344	0.4467	0.707604	0.0000171
JKW-2	2.50	0.708261	117	627	0.5412	0.707635	0.0000180
JKW-3	3.75	0.708218	290	1638	0.5129	0.707636	0.0000187
JKW-4	5.00	0.708260	113	621	0.5275	0.707646	0.0000180
JKW-5	6.25	0.708228	100	611	0.4744	0.707677	0.0000174
JKW-6	7.50	0.708160	74	513	0.4157	0.707688	0.0000170
JKW-7	8.75	0.708199	56	344	0.4741	0.707654	0.0000173
JKW-8	10.00	0.708095	87	604	0.4158	0.707625	0.0000171
JKW-9	11.25	0.708109	75	512	0.4246	0.707620	0.0000174
JKW-10	12.50	0.708134	69	436	0.4592	0.707600	0.0000171

All samples from Kcp corrected to 88.1 Ma. Sample JKW corrected to 84.9 Ma. Data are normalized to naturally occurring 86 Sr/ 88 Sr = 0.1194. Ratios reported relative to 87 Sr/ 86 Sr = 0.71025 for NBS 987. Decay constant; 87 Rb = 1.393 x 10^{-11/}yr (Nebel et al., 2011).

	Measured								Initial	
	²⁰⁶ Pb	²⁰⁷ Pb	U	Pb	Age	²³⁸ U	²³⁵ U	²⁰⁶ Pb	²⁰⁷ Pb	²⁰⁷ Pb
Sample	²⁰⁴ Pb	²⁰⁴ Pb	ppm	ppm	(Ma)	²⁰⁴ Pb	²⁰⁴ Pb	²⁰⁴ Pb	²⁰⁴ Pb	²⁰⁶ Pb
JKCM3-1	18.9521	15.7118	0.009	2.161	88.1	0.130877	0.000949	18.9503	15.7118	0.8291
JKCM3-2	18.8919	15.6528	0.010	2.326	88.1	0.133481	0.000968	18.8713	15.6528	0.8295
JKCM3-3	18.8730	15.6454	0.005	1.416	88.1	0.11654	0.000845	18.8466	15.6454	0.8301
JKCM3-4	18.8520	15.6564	0.005	0.753	88.1	0.203524	0.001476	18.8871	15.6564	0.8289
JKCM3-5	19.0045	15.6750	0.006	3.818	88.1	0.047965	0.000348	18.9894	15.6750	0.8255
JKCM3-6	18.8821	15.6577	0.006	1.800	88.1	0.109688	0.000796	18.8670	15.6577	0.8299
JKCM3-7	18.8590	15.6516	0.005	1.683	88.1	0.083938	0.000609	18.8420	15.6516	0.8307
JKCM3-8	18.8652	15.6555	0.005	1.689	88.1	0.090942	0.00066	18.8596	15.6555	0.8301
JKCM3-9	18.8630	15.6448	0.004	0.833	88.1	0.148884	0.00108	18.8796	15.6448	0.8287
JKCM3-10	18.8834	15.6679	0.024	1.227	88.1	0.603437	0.004376	18.8662	15.6679	0.8305
JKCM4-1	18.8593	15.6598	0.095	53.685	88.1	0.053795	0.00039	18.8254	15.6598	0.8318
JKCM4-2	18.9259	15.6715	2.332	33.475	88.1	2.13482	0.015483	18.9323	15.6715	0.8278
JKCM4-3	18.8719	15.6582	0.139	14.599	88.1	0.291219	0.002112	18.8385	15.6582	0.8312
JKCM4-4	18.8496	15.6401	0.022	16.291	88.1	0.041307	0.0003	18.8197	15.6401	0.8310
JKCM4-5	18.8445	15.6389	0.026	20.440	88.1	0.038552	0.00028	18.8262	15.6389	0.8307
JKCM4-6	18.8629	15.6570	0.019	14.217	88.1	0.041534	0.000301	18.8389	15.6570	0.8311
JKCM4-7	18.9371	15.6777	0.030	64.216	88.1	0.014522	0.000105	18.9044	15.6777	0.8293
JKCM4-8	18.9201	15.6655	0.464	17.055	88.1	0.833471	0.006045	18.8955	15.6655	0.8291
JKCM4-10	18.8806	15.6912	0.064	9.781	88.1	0.199955	0.00145	18.8771	15.6912	0.8312
JKCM5-1	18.8281	15.6633	0.120	57.242	88.1	0.064113	0.000465	18.7981	15.6633	0.8332
JKCM5-2	18.8336	15.6597	0.404	64.661	88.1	0.190796	0.001384	18.8048	15.6597	0.8327
JKCM5-3	18.9726	15.7490	1.444	18.312	88.1	2.422716	0.017571	18.9531	15.7490	0.8309
JKCM5-4	18.8087	15.6314	0.078	15.448	88.1	0.153983	0.001117	18.7888	15.6314	0.8320
JKCM5-6	18.8591	15.6578	1.295	33.096	88.1	1.195257	0.008669	18.8311	15.6578	0.8315
JKCM5-7	18.8320	15.6443	0.121	46.295	88.1	0.079641	0.000578	18.7994	15.6443	0.8322
JKCM5-8	18.8436	15.6536	0.313	30.473	88.1	0.313567	0.002274	18.8191	15.6536	0.8318

 Table 2-2: U and Pb Trace Element and Isotopic Data

JKCM5-9	18.8602	15.6891	0.061	15.072	88.1	0.124057	0.0009	18.8345	15.6891	0.8330
JTF2-1	18.8240	15.6509	0.073	16.649	88.1	0.134434	0.000975	18.8105	15.6509	0.8320
JTF2-2	18.8176	15.6290	0.143	11.895	88.1	0.365699	0.002652	18.7982	15.6290	0.8314
JTF2-3	18.8611	15.6611	0.273	8.951	88.1	0.932488	0.006763	18.8734	15.6611	0.8298
JTF2-4	18.9075	15.6694	0.336	10.480	88.1	0.982775	0.007128	18.9541	15.6694	0.8267
JTF2-5	18.8314	15.6565	0.027	15.077	88.1	0.054163	0.000393	18.8116	15.6565	0.8323
JTF2-6	18.8298	15.6849	0.062	8.020	88.1	0.23838	0.001729	18.8779	15.6849	0.8309
JTF2-7	18.8898	15.6758	0.062	40.739	88.1	0.046903	0.00034	18.8571	15.6849	0.8318
JTF2-8	18.8482	15.6681	0.053	8.439	88.1	0.192186	0.001394	18.8513	15.6681	0.8311
JTF2-9	18.8069	15.6868	0.048	15.931	88.1	0.092738	0.000673	18.7889	15.6868	0.8349

Uncertainty in ${}^{206}Pb/{}^{204}Pb = 0.12\%$, ${}^{207}Pb/{}^{204}Pb = 0.18\%$. Decay constants ${}^{238}U = 1.55125 \times 10^{-10}/yr$, ${}^{235}U = 9.84858 \times 10^{-10}/yr$, (Steiger and Jäger, 1977).

APPENDIX 2: SCAN OF K-FELDSPAR SAMPLES AND CORRESPONDING DATA PLOTTED AGAINST TRAVERSE LENGTH







APPENDIX 3: ESTIMATION OF ERRORS IN MEASURED AND INITIAL ISOTOPIC RATIOS

Measured and Initial Pb Isotopic Ratios

Initial isotopic compositions were calculated through their respective age equations:

$$\left(\frac{{}^{206}Pb}{{}^{204}Pb}\right)_{i} = \left(\frac{{}^{206}Pb}{{}^{204}Pb}\right)_{m} - \left(\frac{{}^{238}U}{{}^{204}Pb}\right) \quad \left(e^{\lambda t} - 1\right) \tag{A1}$$

$$\left(\frac{{}^{207}Pb}{{}^{204}Pb}\right)_{i} = \left(\frac{{}^{207}Pb}{{}^{204}Pb}\right)_{m} - \left(\frac{{}^{235}U}{{}^{204}Pb}\right) \quad (e^{\lambda t} - 1)$$
(A2)

Where the subscript *i* indicates initial ratios and *m* the measured ratios. Decay constants $\lambda_{238U} = 1.55125 \times 10^{-10}/\text{yr}$, and $\lambda_{235U} = 9.84585 \times 10^{-10}/\text{yr}$ after Steiger and Jäger (1977).

Measured Pb ratios were corrected by reference to a standard material, NBS 981. Longterm measurements of NBS 981 in the UNC TIMS laboratory yielded a fractionation correction of $0.12 \pm 0.06\%$ per amu, yielding uncertainties during mass fractionation in 207 Pb/ 204 Pb, 206 Pb/ 204 Pb, 205 Pb/ 204 Pb of $\pm 0.18\%$, $\pm 0.12\%$, and $\pm 0.06\%$, respectively. Total uncertainty in the measurements is found by combining the fractionation uncertainty with the analytical uncertainty for each sample. Because these uncertainties are from two independent measurements, they are combined in quadrature (Taylor, 1997):

$$\sigma = \sqrt{\text{error in fractionation correction}^2 + TIMS \text{ measurment error}^2}$$
(A3)
$$\sigma \left(\frac{^{206}Pb}{^{204}Pb}\right)_m = \sqrt{(0.0012)^2 + TIMS \text{ measurment error}^2}$$
(A4)

$$\sigma \left(\frac{{}^{207}Pb}{{}^{204}Pb}\right)_m = \sqrt{(0.0018)^2 + TIMS \ measurment \ error^2} \tag{A5}$$

Sr Isotopic Data Reduction and Estimation of Uncertainties

Initial Sr isotopic values and combined uncertainties were corrected using an R program (code below description). Since complexity arises from the separate measurements of measurements of Rb and Sr, estimation of the total analytical uncertainty as well as $Sr_{(i)}$ value were calculated through a Monte Carlo simulation. The Monte Carlo simulation starts by creating normal distributions (n = 10,000) with means corresponding to the measurements of 85 Rb/ 87 Rb, 87 Sr/ 86 Sr and 84 Sr/ 86 Sr, and 2σ values based on the internal reproducibility of these measurements (0.01 for 85 Rb/ 87 Rb, 0.000015 for 87 Sr/ 86 Sr, 0.0003 for 84 Sr/ 86 Sr). With these distributions the data were then reduced to remove the tracer contribution to the measurement by normalizing to 86 Sr/ 88 Sr to 0.1194. This can be solved for a spiked sample by writing the following equations:

$$N_s \times h + N_t \times (1 - h) = N \times \left(1 + \frac{b}{m}\right)^{m \times \alpha}$$
(A6)

$$T_s \times h + T_t \times (1-h) = T \times \left(1 + \frac{a}{m}\right)^{m \times a}$$
(A7)

Where N is the normalizing isotopic ratio (84 Sr/ 86 Sr), T is the known tracer isotopic ratio, subscript *s* is the sample, *t* is the tracer, unscripted is the measured isotopic ratio, *h* is ratio of the number of atoms of the reference isotope in the sample to the total number of atoms of the reference isotope, m is the reference isotope (86 Sr), *a* is the mass difference for T (2), and *b* is the mass difference for N (2). These two equations can be solved and equated yielding:

$$X\left(1+\frac{b}{m}\right)^{m\times\alpha} - Y\left(1+\frac{a}{m}\right)^{m\times\alpha} + Z = 0$$
(A8)

Where:

$$X = \frac{N}{N_s - N_t} \tag{A9}$$

$$Y = \frac{T}{T_s - T_t} \tag{A10}$$

$$Z = \frac{T_{t}}{(T_{s} - T_{t})} - \frac{N_{t}}{(N_{s} - N_{t})}$$
(A11)

This equation (**Eq. A11**) can be solved for α , by defining:

$$F(a) = X \left(1 + \frac{b}{m}\right)^{m \times \alpha} - Y \left(1 + \frac{a}{m}\right)^{m \times \alpha} + Z$$
(A12)

and

$$F'(a) = X\left(1 + \frac{b}{m}\right)^{m \times a} \times \ln(m+b) - Y\left(1 + \frac{a}{m}\right)^{m \times a} \times \ln(m+a)$$
(A13)

Using Newton's method, alpha can be determined by iteration on the equation:

$$a = a_0 - F(a)/F'(a)$$
 (A14)

Through this, the first estimate of a_0 can be determined by solving the N and T equation (Eq. A6 & Eq. A7) using a linear fractionation law to get:

$$\alpha_{linear} = \frac{(Y - X - Z)}{(X \times b) - (Y \times a)}$$
(A15)

This is iterated until the 84 Sr/ 86 Sr value is 0.1194.

Once the spike and blank contributions have been removed from the measurement data, the corrected 87 Sr/ 86 Sr and 87 Rb/ 86 Sr ratios can be substituted into the decay equation to obtain 87 Sr/ 86 Sr_(i):

$$\binom{{}^{87}Sr}{{}^{86}Sr}_i = \binom{{}^{87}Sr}{{}^{86}Sr}_m - \binom{{}^{87}Rb}{{}^{86}Sr} \quad (e^{\lambda t} - 1)$$
(A16)

Where *i* is the initial isotopic ratio, *m* is the corrected measured ratio, *t* is time. A decay constant, ${}^{87}\text{Rb} = 1.393 \times 10^{-11}$ was used after Nebel et al., (2011). Once Sr_(i) has been obtained, the standard deviation of the entire "data cloud" of values which have gone through data reduction are obtained to estimate the 2σ uncertainty in the initial Sr isotopic composition.

APPENDIX 4: DEVELOPED SOFTWARE

Monte Carlo-ReduceR

####

```
#TIMS Rb / Sr Reducer with Monte Carlo simulation
#Jonathan Munnikhuis
#############Spike-b3
sp_86sr84sr=0.000589149
sp_87sr84sr=9.28084E-05
sp_88Sr84Sr=0.000387453
sp_ppm84Sr=0.144529
sp_85Rb87Rb=0.008030983
sp_ppm87Rb=0.968349
#Abundances
sp_AbSr84=0.998832
sp_AbSr86=0.000588461
sp_AbSr87=0.0000927
sp_AbSr88=0.000387
sp_AbRb87=0.992033
sp_AbRb85=0.007967
#Create delimited text file with sample info per column. If there are
multiple samples you can change sample by modifying the upcoming "co"
variable.
# Important = make sure Rb data is fractionated corrected before
inputting in 85Rb/87Data.
#Latest alpha value is = +.30823 3/16/2016
co=1 #change columns per sample here
# Reading in sample information
SampleName=input[1,co]
Sa_weight=input[2,co]
RbAliquotFrac=input[3,co]
sp_RbWeight=input[5,co]
RbBlank=input[6,co]
sa_85Rb87Rb=input[7,co]
SrAliquotFrac=input[8,co]
sp_SrWeight=input[10,co]
SrBlank=input[11,co]
sa_84Sr86Sr=input[12,co]
sa_87Sr86Sr_linNoRb=input[13,co]
sa_86Sr88Sr=input[14,co]
int_87sr86sr_guess=input[15,co]
guess_Age=input[16,co]
Bias=input[17,co]
blank87Sr86Sr=input[18.co]
normval_86Sr88Sr=input[19,co]
#######Monte Carlo portion########## roll the dice.
# Creates "data clouds" from UNC-TIMS internal reproducibility
#twosigabs85Rb87Rb
stdRb=.01/2
#twosigSr8786
stdSr_1=.000015/2
#twosigSr8486
```

```
stdSr_2=.000030/2
```

#Here's the number of iterations you want to send through the output $n\!=\!10e3$

#create random normal distribution of data

```
spreadRb=rnorm(n, sa_85Rb87Rb, stdRb)
spread8786=rnorm(n, sa_87Sr86Sr_linNoRb, stdSr_1)
spread8486=rnorm(n,sa_84Sr86Sr,stdSr_2)
#spread8688=rnorm(n.sa_86Sr88Sr.stdSr_3)
#Some histograms of the data clouds, make sure mean is where it should
be.
hist(spreadRb)
hist(spread8786)
hist(spread8486)
hist(spread8688)
###Monte Carlo is now about to roll several dice, before putting
through reduction.
for(i in 1:n)
{
  a=sample(n,i)
  b=sample(n,i)
  c=sample(n,i)
  rand_Rb8587=spreadRb[a]
  rand_Sr8786=spread8786[b]
  rand_Sr8486=spread8486[c]
}
sa_85Rb87Rb=rand_Rb8587
sa_84Sr86Sr=rand_Sr8486########
m8486=sa_84Sr86Sr
m8486=signif(m8486,6)
m8786=sa_87Sr86Sr_linNoRb
m8786=signif(m8786, 6)
m8886=1/sa_86Sr88Sr
m8886=signif(m8886, 6)
m8686=1
linnorm_amu=(((1/m8886)/normval_86Sr88Sr)-1)/2
linUNnorm_8486=m8486*(1+2*linnorm_amu)
linUNnorm_8786=m8786/(1+linnorm_amu)
linUNnorm_8886=m8886
linUNnorm_8686=1
sampleCont8686=((1/sp_86sr84sr)-linUNnorm_8486)/((1/sp_86sr84sr)-.0565)
SampleCont8486=.0565*SampleCont8686
TraceCon8486=m8486-SampleCont8486
TraceCon8786=sp_87Sr84Sr*TraceCon8486
TraceCon8886=sp_88Sr84Sr*TraceCon8486
TraceCon8686=TraceCon8486*sp_86Sr84Sr
```

SCont8686=linUNnorm_8686-TraceCon8686
SCont8486=.0565*SCont8686 SCont8786=linUNnorm_8786-TraceCon8786 SCont8886=linUNnorm_8886-TraceCon8886 Normd_8688=SCont8686/SCont8886 NormFac8486=((Normd_8688/ normval_86Sr88Sr)-1)/2 N8486=linUNnorm_8486/(1+2*NormFac8486) N8786=linUNnorm 8786*(1+NormFac8486) N8886=linUNnorm_8886*(1+2*NormFac8486) N8686=1 S_Cont8686=((1/sp_86sr84sr)-N8486)/((1/sp_86sr84sr)-.0565) S_Cont8486=.0565*S_Cont8686 TCont8486=N8486-S_Cont8486 TCont8786=sp_87Sr84Sr*TCont8486 TCont8886=sp_88Sr84Sr*TCont8486 TCont8686=sp_86Sr84Sr*TCont8486 SCont8686=N8686-TCont8686 SCont8486=.0565*SCont8686 SCont8786=N8786-TCont8786 Normd8688=SCont8686/SCont8886 Norm_Fac8486=((Normd8688/ normval_86Sr88Sr)-1)/2 N8486=N8486/(1+2*Norm_Fac8486) N8786=N8786*(1+Norm_Fac8486) N8886=N8886*(1+2*Norm_Fac8486) N8686=1 S_Cont8686=((1/sp_86Sr84Sr)-N8486)/((1/sp_86Sr84Sr)-.0565) S_Cont8486=.0565*S_Cont8686 TCont8486=N8486-S_Cont8486 TCont8786=sp 87Sr84Sr*TCont8486 TCont8886=sp_88Sr84Sr*TCont8486 TCont8686=sp_86Sr84Sr*TCont8486 SCont8686=N8686-TCont8686 SCont8486=.0565*SCont8686 SCont8786=N8786-TCont8786 SCont8886=N8886-TCont8886 #############################iteration Normd8688=SCont8686/SCont8886 Norm Fac8486=((Normd8688/ normval 86Sr88Sr)-1)/2 N8486=N8486/(1+2*Norm_Fac8486) N8786=N8786*(1+Norm_Fac8486) N8886=N8886*(1+2*Norm_Fac8486) N8686=1 S_Cont8686=((1/sp_86Sr84Sr)-N8486)/((1/sp_86Sr84Sr)-.0565) S_Cont8486=.0565*S_Cont8686 TCont8486=N8486-S_Cont8486 TCont8786=sp_87Sr84Sr*TCont8486 TCont8886=sp_88Sr84Sr*TCont8486 TCont8686=sp_86Sr84Sr*TCont8486 SCont8686=N8686-TCont8686

SCont8486=.0565*SCont8686

```
SCont8786=N8786-TCont8786
SCont8886=N8886-TCont8886
SampleComp8486=SCont8486/SCont8686
SampleComp8786=SCont8786/SCont8686
SampleComp8886=SCont8886/SCont8686
Normalized8688=SCont8686/SCont8886
#######
#######
a=Normd_8688/normval_86Sr88Sr
b=86/88
c=86*86
ExpCorr8786=SampleComp8786*sqrt((1-(log(a)/log(b)/c)))
ExpCorr8786=signif(ExpCorr8786. 6)
BiasCorr8786=ExpCorr8786+(ExpCorr8786*(Bias/1e6))
BiasCorr8786=signif(BiasCorr8786, 6)
###############
##Rb Ratios
Rb8587m=sa_85Rb87Rb #measured
Rb8587t=sp_85Rb87Rb #tracer
Rb8587s=Rb8587m-Rb8587t #sample
#########Isotopic concentrations
umo186Sr=(SCont8686/TCont8486)*(sp_ppm84Sr/83.913429)*sp_SrWeight
umol87Sr=BiasCorr8786*umol86Sr
umol84Sr=umol86Sr*SampleComp8486
umol88Sr=umol86Sr*SampleComp8886
##
umo187Rb=(Rb8587s/(2.59265-
Rb8587m))*((sp_ppm87Rb/86.909186)*(sp_Rbweight/RbAliquotFrac))#replace
1 here for Rb Aliquot fraction
umo185Rb=umo187Rb*2.59265
#####
Blcor_umol86sr=umol86sr-((.0986*srBlank)/(85.909256*1000))
BlCor_umol87Sr=umol87Sr-
(blank87Sr86Sr*((.0986*SrBlank)/(85.909256*1000)))
BlCor_umol84sr=umol84sr-(.056584*((.0986*srBlank)/(83.913429*1000)))
BlCor_umol88sr=umol88sr-
(blank87Sr86Sr*((.0986*SrBlank)/(87.905618*1000)))
##
BlCor_umol87Rb=umol87Rb-((.2785*RbBlank)/(86.909186*1000))
BlCor umol85Rb=BlCor umol87Rb*2.59265
####
BlCor87Sr86Sr=BlCor_umol87Sr/BlCor_umol86Sr
BlCor87Rb86Sr=BlCor_umol87Rb/BlCor_umol86Sr
Sr8786=BlCor87Sr86Sr
```

Sr8786=signif(Sr8786,6)

ppm87Rb=(BlCor_umol87Rb*86.9092)/(Sa_weight*RbAliquotFrac)
ppm87Rb=signif(ppm87Rb,5)

ppm86Sr=(BlCor_umol86Sr*85.9093)/(Sa_weight*SrAliquotFrac)

```
ppm86Sr=signif(ppm86Sr,5)
```

```
TotRb=ppm87Rb/((BlCor_umol87Rb*86.9092)/((BlCor_umol87Rb*86.9092)+(BlCo
r_umol85Rb*84.9118)))
TotRb=signif(TotRb,6)
```

ugRb=TotRb*Sa_weight
ugRb=round(ugRb, 3)

```
TotSr=ppm86Sr/((BlCor_umol86Sr*85.9093)/((BlCor_umol86Sr*85.9093)+(BlCor_umol87Sr*86.9089)+
```

(Blcor_umol88sr*87.9056)+(Blcor_umol84sr*83.9134)))
Totsr=signif(Totsr,6)

```
ugSr=TotSr*Sa_weight
ugSr=round(ugSr, 3)
```

RbSr_weight=TotRb/TotSr RbSr_weight=round(RbSr_weight,4)

Rb87_Sr86=BlCor87Rb86Sr Rb87_Sr86=round(Rb87_Sr86,digits= 4)

```
Sr_i=mean(Cal87Sr86Sr_i)
```

sr_i

#Here are some more things that might matter to you. #just uncomment the variables below if you care about it

```
# Sr8786=mean(Sr8786)
# TotSr=mean(TotSr)
# ppm86Sr=mean(ppm86Sr)
# TotRb=mean(TotRb)
# ppm87Rb=mean(ppm87Rb)
# ugRb=mean(ugRb)
# Rb87_Sr86=mean(Rb87_Sr86)
# Rbsr_weight=mean(Rbsr_weight)
```

```
# if you wanted a csv file of outputs uncomment this####
#outputs=rbind(Sr8786,TotSr,ppm86sr,ugSr, TotRb,
ppm87Rb,ugRb,Rb87_Sr86,RbSr_weight,Sr_i, twosigCal87sr86sr)
#print(outputs)
#write.csv(outputs,"")
print(Sr_i)
print(twosigCal87sr86sr)
```

RechargR – A Dynamic Crystallization Model

```
#RechargR#
#Developed by Jonathan Munnikhuis
#Simulate Recharge into a Crystallizing Magma Chamber
#Variables - Plag, Kspar, Hornblende, Biotite, Quartz
wt_frac = c(0.17301138, 0.699127742, 0, 0, 0.127860878
frac of hypothetical residual 25% of plg remaining, Kfs, and Qtz
                                                                      0.127860878) #wt
#####
              50%
                                %25
                                                                     %25
##Rhyolite Kds from Nash & Crecraft 1988
kd_sr=c(19.9, 5.9, 0.022, 0.4)
kd_rb=c(0.125, 1.8, 0.014, 3.2)
kd_pb=c(1.31, 2.545, 0.255, 0.85, 0)
kd_ba=c(1.515, 11.45, 0.044, 23.533, 0)
                               0.022,
                                               0.41, 0)
                                               3.2,
                                                       (0)
kd_feldspar = c(kd_sr[2], kd_rb[2], kd_pb[2], kd_ba[2])
### Bulk rock concentrations
##
Bulk_D_sr = sum(wt_frac*kd_sr)
Bulk_D_rb = sum(wt_frac*kd_rb)
Bulk_D_pb = sum(wt_frac*kd_pb)
Bulk_D_ba = sum(wt_frac*kd_ba)
bulkDs = c(Bulk_D_sr, Bulk_D_rb, Bulk_D_pb, Bulk_D_ba)
#######
ml=100; # units of initial mass of melt
mx=100; # initial mass of crystals
ppm_o=c(914, 176, 19, 587) #Original Concentration from Frac Xtlz
#ppm_add = c(604.25,128.5576923,17.38461538, 699.0730769 # Set recharge
liquid here
\#ppm_add = c(27, 297, 38, 20) \# Ex. Concentration of Elements in Kcp
aplites
### Set variables 1 = Sr, 2 = Rb, 3 = Pb, 4 = Ba
BulkD = bulkDs
cl = ppm_o
ca= ppm_add
D = kd_feldspar
############
dl = 6 #mass crystallized between melt additions
da = 3#mass liquid added
dx = 0.05 # increment of interval between cycles
n = 20 # number of cycles
############
mxc = seq(0,dl,dx) # range of crystal growth
ml=100; # units of initial mass of melt
mx=100; # initial mass of crystals
### initialize variabiles as emtpy vectors to be filled
C_sr <- vector()</pre>
Z_sr<- vector()</pre>
C_rb <- vector()
Z_rb<- vector()</pre>
C_pb <- vector()</pre>
Z_pb<- vector()</pre>
C_ba <- vector()
z_ba <- vector()
Fmelt <-vector()</pre>
R <-vector()
mxn = 0 # initial mass of feldspar</pre>
```

```
#initialize index values
ind = 1
###Loop####
for (i in 1:n){
  mel_sr = cl[1]*ml
  mel_rb = cl[2]*ml
mel_pb = cl[3]*ml
  mel_ba = cl[4]*ml
     for (k in mxc){
       ml = ml-dx #mass of liquid after xtls removed
       mxn = mxn+dx #mass of new x1 in this step
       mx = mx + dx
       Fmelt[ind] = ml/(ml+mx)
       R[ind] = (mxn^{1/3})
       #Sr
       Z_sr[ind] = c][1] * D[1]
CB_sr = BulkD[1] * c][1]
       C_sr[ind] = ((m]+dx) * cl[1] - CB_sr*dx) / ml
cl[1] = C_sr[ind]
       me\bar{1}_{sr} = c\bar{1}[\bar{1}]*m\bar{1}
       #Rb
       Z_rb[ind] = c1[2] * D[2]
CB_rb = BulkD[2] * c1[2]
       C_rb[ind] = ((ml+dx) * cl[2] - CB_rb*dx) / ml
cl[2] = C_rb[ind]
       mel_rb = cl[2]*ml
       #Pb
       Z_pb[ind] = c1[3] * D[3]
CB_pb = BulkD[3] * c1[3]
       C_pb[ind] = ((m]+dx) * c][3] - CB_pb*dx) / m]
       c\overline{1}[3] = \overline{c}pb[ind]
       me\bar{1}_{pb} = c\bar{1}[\bar{3}]*m\bar{1}
       #Ba
       Z_ba[ind] = c1[4] * D[4]
       _____CB_ba = BulkD[4] * cl[4]
C__ba[ind] = _((m]+dx) * cl[4] - CB_ba*dx) / ml
       cl[4] = c_ba[ind]
       mel_ba = cl[4]*ml
       ind=ind+1
     }
  ml = ml+da
  mel_sr = mel_sr+da*ca[1]
  cl[1]=mel_sr/ml
  mel_rb = mel_rb+da*ca[2]
  cl[2]=mel_rb/ml
  mel_pb = mel_pb+da*ca[3]
```

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