Hekla Revisited: Fractionation of a Magma Body at Historical Timescales

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Hekla is an elongate volcano that lies at the intersection of the South Iceland Seismic Zone and the Eastern Volcanic Zone. We report major and trace element, oxygen isotopic, and H$_2$O analyses on rocks, glass, melt inclusions, and minerals from almost all of the historical lavas and tephra deposits. This new data set confirms the remarkable observation that not only are many eruptions compositionally zoned from felsic to mafic, but the extent of zoning relates directly to the length of repose since the previous eruption. Compositional data are consistent with the origin of the basaltic andesites and andesites by fractional crystallization, with no measurable crustal interaction once basaltic andesite has been produced. Although the 1104 C.E. Plinian rhyolite and 1158 C.E. dacite are also created by fractional crystallization, uranium-thorium isotopic disequilibria measured by others require that it evolved in a separate body, where it is stored in a molten state for >10$^4$ years. Consistent trace element trends and ratios, as well as oxygen isotopic data preclude significant crustal input into the evolving magma. The phenocryst assemblages are dominated by crystals that formed from their host melt; an exception is the 1158 C.E. dacite, which contains abundant crystals that formed from the 1104 C.E. rhyolite melt. A suite of thermobarometers indicates that most crystals formed in the lower crust at temperatures ranging from ~1010 to 850°C. Hekla’s unique and systematic petrologic time series and geophysical activity are attributed to the unusual geometry of the magma body, which we propose to be a tabular, vertically elongate macrodike, extending from the lower- to the upper-crust. The vertical body is recharged with basaltic andesite magma at the end of each eruption, which then undergoes cooling and crystallization until the subsequent eruption. The entire system is supplied by a lower crustal body of basaltic andesite, which is produced by fractional crystallization of basaltic magma in a reservoir that is thermochemically buffered to ~1010°C. Cooling and crystallization of recharged basaltic andesite magma in a background geothermal gradient from the lower to the shallow crust accounts for the systematic relationship between repose and composition.

**Key words:** Iceland, Hekla, Fractional crystallization, igneous geochemistry, magma chamber
INTRODUCTION

Hekla volcano in Iceland is one of Earth’s best examples of the link between petrologic and volcanic processes (Figure 1). Not only do Hekla eruptions initiate with an explosive phase and culminate in effusion of lava, but most of the 18 historically documented eruptions are normally zoned, with more evolved magmas erupting first, followed by increasingly mafic products (Thorarinsson, 1950; 1967). Moreover, and most importantly, the extent of magmatic differentiation is a function of the repose time between eruptions (Figure 2; Thorarinsson, 1967), providing a unique opportunity to explore rates of magmatic differentiation over human timescales.

Owing to its classic volcanologic and petrologic characteristics, Hekla volcano has been subjected to nearly every modern geophysical and analytical method, including seismic and geodetic studies, analyses of actinide-series isotopes (Sigmarsson et al., 1992; Chekol et al., 2011), melt inclusion measurements (Moune et al., 2007; Portnyagin et al., 2012; Lucic et al., 2016), stable isotopes (Schuessler et al., 2009; Savage et al., 2011; Prytulak et al., 2017a,b; Deng et al., 2019; Inglis et al., 2019), compositional analyses of individual zircon (Carley et al., 2011; Bindemann et al., 2012; Carley et al., 2020), and radiogenic isotopes (Sigmarsson et al., 1991; 1992; Chekol et al., 2011; Carley et al., 2020). What have been missing are modern, state-of-the-art major and trace element measurements and crystal-scale studies of all the historical eruptive products, which are the focus of this study. Although much of the previous work on Hekla cited above includes elemental analyses, most studies include only a few analyses per eruption or only a few eruptive units, which misses two of the most important features of this volcano and its historical eruptive record: the zoned eruptions and the correlation between differentiation and repose (Figure 2). Moreover, the data sets are from different laboratories, using a variety of methods and standards. In addition to analyzing several samples from most of the historical eruptions in the same laboratories using consistent methods and standards, we also have performed detailed petrography on the rocks and measured the compositions of many of the phenocryst phases. We also include data on melt inclusions, including volatile contents, and oxygen isotopic ratios of olivine crystals.

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Geologic Setting

Hekla volcano is located near the intersection of the South Iceland Seismic Zone and the western margin of the Eastern Volcanic Zone (Figure 1). In southern Iceland, spreading partitions into the Eastern and Western Volcanic Zones, which are connected by the South Iceland Seismic Zone, a series of N-S trending right-lateral “bookshelf faults” that accommodate left-lateral transfer of plate spreading motion between the Reykjanes Ridge and the Eastern Volcanic Zone (Einarsson, 1991). Hekla diagonally cuts the inside-corner of the ridge-transform intersection (Figure 1). Northeast of Hekla, the Eastern Volcanic Zone is an active rift zone characterized by NE-trending normal faults and Holocene eruptive fissures that have been the source of large volume (2-15 km$^3$) eruptions of tholeiitic basalt (Jonsson et al., 1997; Einarsson, 2008; Pedersen et al., 2019), as well as numerous NE-trending hyaloclastite ridges that formed as subglacial fissure eruptions during the Pleistocene. Torfajökull volcano, which has a large caldera and is the largest silicic volcano in Iceland, lies ~25 km east of Hekla, on the western flank of the Eastern Volcanic Zone. Pyroclastic rocks erupted from Torfajökull crop out around the base of Hekla.

The Hekla edifice is a ~10-km-long ENE-trending volcanic ridge bisected by a 5 to 6-km-long fissure extending the length of the ridge (Figure 1). The ridge formed within the last 3000 years through the eruption and accumulation of lavas erupted from the summit fissure (Thorarinsson, 1950; Sverisdottir, 2007). The modern edifice is constructed on top of a basaltic hyaloclastite ridge, which is exposed along the north, west, and east sides of the volcano. Basalt has erupted from fissure systems a few kilometers east and northeast of Hekla as recently as 1878 and 1913, and to the southwest of Hekla in 1554 (Thorarinsson, 1967). The basalts erupted on the peripheral fissures are distinct from the intermediate and silicic rocks produced from the Hekla edifice itself, but because of their transitional alkalic character and geochemical similarities to Hekla products, the compositions of the peripheral deposits are considered by most investigators as parental to the Hekla magmatic system (Jakobsson, 1979; Sigmarsson et al., 1992). Rhyolite lavas with glassy, brecciated margins related to Torfajökull volcanic center are exposed at the surface 10 km east of Hekla. Xenoliths of rhyolite ignimbrite ejected during Hekla’s 1970 eruption (Thorarinsson and Sigvaldason, 1972; Sigmarsson et al., 1992) and clasts of rhyolitic hyaloclastite ejected from fissures on the NE shoulder of Hekla (our observations) indicate
that rocks related to the Torfajökull system, or an older equivalent, are also present at depth beneath Hekla.

In addition to its common intermediate effusive eruptions, Hekla has experienced 5 silicic Plinian eruptions, the oldest of which occurred ~7000 years ago. The tephra from these eruptions have volumes of 0.5 to 2.2 km$^3$ dense-rock equivalent (DRE) and have been deposited at intervals of 1,000 to 2,500 years (Larsen and Thorarinsson, 1977). Several of the prehistoric Plinian tephra deposits exhibit compositional zoning from rhyolite at the base and middle of the deposits to dacite and andesite in the upper parts of the deposits (Larsen and Thorarinsson, 1977; Sverrisdottir, 2007; Larsen et al., 2019). The most recent of the explosive silicic eruptions occurred in 1104 C.E., with the eruption of 0.9-1.0 km$^3$ of tephra (Thorarinsson, 1967; Larsen and Thorarinsson, 1977; Janebo et al., 2016a, b). In addition to the silicic Plinian events, more than 10 explosive eruptions, which are zoned from andesite or dacite to basaltic andesite, erupted between 1050 B.C.E. and 1104 C.E. (Larsen et al., 2020). In this study, we restrict all discussion to the historical eruptions, and therefore only consider the 1104 C.E. rhyolite eruption among the rhyolitic Plinian events in Hekla’s history.

In a seminal work combining observations from the tephra record, the geology of the volcano, and historical records, Thorarinsson (1967) documents 13 eruptions of Hekla in the period following the 1104 C.E. rhyolite eruption through the 1947-48 eruption. These eruptions occurred after repose periods ranging from 15 to 120 years, with an average repose of ~64 years. Thorarinsson (1967) notes that most eruptions a consistent pattern: 1) they begin with a highly explosive, presumably volatile-rich, initial phase of activity, which transitions to effusive activity that may last weeks to months; and 2) eruptions consistently exhibit compositional zoning in which the earliest erupted tephra is the most evolved and subsequent tephra and lavas are progressively less evolved. Thorarinsson (1967) also observes that the maximum SiO$_2$ content of the initial tephra is positively correlated to the length of the preceding repose period, and most eruptions terminated with a baseline composition of basaltic andesite (Figure 2).
Geophysical Constraints on Hekla’s Magmatic Plumbing

Hekla has been monitored by modern geophysical methods since the mid-20th century, providing important constraints on the transport and storage of magma. Geirsson et al. (2012) used continuous and campaign GPS data between July, 2000 (after the most recent eruption, which had a volume of 0.19 km$^3$) and January, 2010 to model the reinflation of the magma chamber. The geodetic data are best modeled by a sill-like chamber at 24 ± 4 km depth. Subsidence on the flanks of the volcano further suggests a vertical finger-like chamber extending between 17-28 km deep at its base to 9-13 km at its top. Pre-eruptive inflation measured by InSAR is modeled as a 14-20 km deep magma chamber, inflating at 0.003 to 0.02 km$^3$/y (Ofeigsson et al., 2011). Post-eruptive deflation in 2000 had a source centered at 14-18 km depth, and there is evidence of excess magma stored in a dike between 5.8 km and the surface. One study that took place around the 2000 eruption combines observations from InSAR, GPS, tilt, and a borehole strainmeter suggests that the depth of the pressure source is only about 10 km (Sturkell et al., 2013). These results also indicate that the dike supplying the eruptive fissure extends to <500 m depth, but that it does not connect along its entire strike to the deep magma reservoir.

Seismic studies reveal that Hekla magma does not slowly fracture its way to the surface preceding eruptions. Instead, Hekla displays a striking absence of precursory seismicity and strain. For example, seismicity began only 79 minutes before the initial breakout of magma in 2000, and a nearby strainmeter registered contraction, thought to be caused by ascent of the eruption-feeding dike 34 minutes prior (Soosalu et al., 2005; Sturkell et al., 2013). Interestingly, the earthquakes’ hypocenters appear to have propagated downward from 1-3 km to 12-15 km in the hour preceding eruption, which is not the signal expected from a dike rising from a deep magma reservoir. A similar phenomenon is described during the 2010 C.E. eruption of Eyjafjallajökull, where downward-propagating seismicity is attributed to a decompression wave caused by the initial tapping of a shallow magma body (Tarasewicz et al., 2012). In 1991, seismic activity began only 32 minutes prior to the eruption, with tremor detected only after the eruption commenced (Soosalu and Einarsson, 2002; 2003). Foci clustered in two zones, at 13-8 km and <4 km. The total seismic energy released over the course of the eruption only
amounted to the equivalent of a magnitude 3.4 earthquake. Soosalu and Einarsson (2004) used S-wave attenuation to conclude that no sizable body of magma exists between 4 and 14 km, indicating that any major reservoir must lie in the lower crust.

METHODS

In this study, we sampled all the historic tephras and lavas that we could confidently identify to construct a complete and internally consistent data set with modern analytical tools. We sampled rocks from all the exposed historical eruptive units produced by Hekla, guided by geologic maps (Thorarinsson, 1967; Hoskuldsson et al., 2007; Pedersen et al., 2019) and Thorarinsson’s (1950; 1967) extraordinarily detailed stratigraphic columns from dozens of excavated pits and natural outcrops (Electronic Appendix 1). To sample the widest compositional range for each historic eruption, care was taken to identify and collect tephra deposits along dispersal axes. Eruptive order was constrained by centimeter-scale stratigraphic documentation, extraction and analysis of single pumice fragments, and eruption narratives. We also report analyses of historically erupted basalts that surround the Hekla edifice, but we do not discuss their role in the petrogenesis (already thoroughly discussed by Sigmarsson et al., 1992).

All major elements are reported and plotted in units of percent oxide by weight; they are normalized to 100% but the analytical totals are reported. Trace elements are reported as parts per million by weight. Tephra clasts for major element analysis by X-ray fluorescence (XRF) were hand-crushed in an alumina mortar, and lava samples were chipped in a tungsten-carbide jaw crusher before pulverization in a tungsten-carbide shatterbox. Single pumice clasts were analyzed where possible (>2.5 grams). Major element analyses by X-ray fluorescence (XRF) were undertaken at the Peter Hooper GeoAnalytical Laboratory at Washington State University (Johnson et al., 1999; Electronic Appendix 2). Unground glass handpicked from the exterior rinds of individual tephra clasts and ground lava samples were dissolved for trace element analysis on a Varian 820MS quadrupole inductively coupled plasma-mass spectrometer (ICP-MS) at Colgate University, using methods described in Harpp et al. (2003) (Electronic Appendix 2). Precisions for XRF and ICP-MS are estimated from separate analyses of multiple tephra clasts from the same
horizon in tephra fall and are reported in Electronic Appendix 2 and shown on the variation diagrams.

Major element analyses of glass, including glassy melt inclusions, were determined on Cameca Camebax and JOEL 8500F microprobes at Washington State University (Electronic Appendix 3). Glass analyses were performed with a beam voltage of 15 kV, a 5–10 µm spot size, and a beam current of 10 nA. Sodium loss was minimized by measuring its abundance immediately after exposing the sample to the beam and correcting for time-dependent volatilization. Precisions based on replications of analyses are reported in Electronic Appendix 3.

Mineral modes were determined by counting ~1000 points on an automated stage mounted on a petrographic microscope. Modes are reported as the proportion of crystals > 0.2 mm in major dimension without interpretation of genetic origin. Most Hekla rocks have <6% crystals, thus mineral concentrates were mounted in epoxy disks for electron microprobe analysis. Crushed rocks were sieved to 0.35 to 1.0 mm and after separation of the magnetic fraction, treated with concentrated HBF₄ to remove adhering groundmass; optical and back-scatter electron imaging of grain boundaries and zoning indicates that the phenocrysts were not corroded by this treatment, only glass and fine-grained material. The grains were then mounted in epoxy and analyzed on 3 microprobes: a Cameca Camebax (Washington State University) and 2 JOEL 8500 instruments (WSU and University of Hawaii). Beam voltage was set to 15 to 20 kV, current to 15 nA, and the beam focused to ~1 µm for mineral analyses.

Oxygen isotopic analyses were carried out at the University of Oregon by laser fluorination using BrF₅ as a reagent and MAT253 mass spectrometer according to the methods of Bindeman et al. (2008a; Electronic Appendix 4). Owing to their small grain size, many analyses were performed on 2 to 14 olivine grains. In one basalt sample and the 1104 C.E. rhyolite tephra, olivine is large enough to permit analysis of single grains. Many olivine grains contain magnetite inclusions, which were culled with a magnet for selecting samples for oxygen isotopic analysis. Data were normalized to SCO= 5.25‰ and UWG garnet =5.80‰. Precision is estimated to be ±0.1 per mil (one standard deviation) on the basis of concurrently run standards.

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Naturally quenched melt inclusions (MI) in olivine were doubly intersected for analysis by Fourier-transform infrared spectroscopy (FTIR) at the University of Oregon (Electronic Appendix 5). Only olivine from tephra samples was used, with the exception of sample H-07-1 which is a glassy spatter bomb, to ensure rapid quenching and to minimize the effect of post-eruptive hydrogen loss that occurs in slowly cooled lava. Infrared spectra were collected between 6000 cm\(^{-1}\) and 1000 cm\(^{-1}\), and the main peaks of interest for this study were the total OH stretching vibration at 3570 cm\(^{-1}\) and the carbonate doublet at 1435 cm\(^{-1}\) and 1515 cm\(^{-1}\), although carbonate was detected in only 2 melt inclusions. Two to four replicate spectra were acquired for each MI. Thickness was measured using both a digital micrometer (± 2μm) and the reflectance interference fringe method described by Wysockanski and Tani (2006). Absorbances are converted to H\(_2\)O concentrations using the Beer-Lambert Law. We used an absorption value of 63 ± 3 L/mol·cm for H\(_2\)O (Dixon et al., 1988). An absorption coefficient of ~350 L/mol·cm was used for the CO\(_2\) doublet, calculated using the glass composition (Dixon and Pan, 1995). Peak heights were calculated using a straight-line background correction (Dixon et al., 1995), and glass densities, including the effect of H\(_2\)O, were calculated from the major element compositions as described in Luhr (2001). Combined uncertainties in density, absorption coefficient, and thickness cause precision to be approximately 10% (Dixon et al., 1988), where most of this error is due to the thickness measurement. Consequently, the average standard deviations (1σ) for H\(_2\)O are ± 0.25 wt.% (one standard deviation).

Apatite grains in a basaltic andesite and two andesites from the 1947 eruption were analyzed by laser ablation ICPMS in the Geoanalytical Laboratory at Washington State University, using a New Wave 213 nm laser and Element2 mass spectrometer (Electronic Appendix 10). Ablation troughs were ~10 micrometers wide and 10s of micrometer long with an irregular geometry. Counts per second are internally normalized to stoichiometric CaO contents and measured \(^{43}\)Ca counts per second, with BCR-2g and NIST610 glasses as external standards. Relative precision based on replicate analyses is estimated to be 10% for Th and 11% for U.
RESULTS

Major Elements

Our bulk-rock major element data from historical Hekla tephra and lavas (Electronic Appendix 2) are consistent with those reported in other studies (Baldridge et al., 1973; Sigmarsson et al., 1992; Checkol et al., 2011). The volcano is constructed of a transitional subalkaline suite, with whole rock compositions ranging from basaltic andesite through rhyolite (54 to 70% SiO$_2$; all oxides are reported as weight percent). Rocks with <54% SiO$_2$ have not erupted from the Hekla edifice in historic time, but basaltic material has been produced by nearby fissures as recently as 1878 C.E. and 1913 C.E. (Thorarinsson, 1967). In our study, any rock referred to as a Hekla rock erupted from vents at the summit or along the flanks of Hekla volcano; we exclude rocks erupted from surrounding fissures and monogenetic vents that are not directly on the edifice.

Hekla lava and tephra exhibit remarkably uniform major element trends at both the inter-eruption and intra-eruption level (Figure 3). Variations in MgO, TiO$_2$, FeO, CaO, and P$_2$O$_5$ all define negative correlations with SiO$_2$, whereas K$_2$O and Na$_2$O correlate positively with SiO$_2$. There is a strong inflection in Al$_2$O$_3$ at ~60% SiO$_2$ (Figure 3). Many of the elements, especially TiO$_2$, Al$_2$O$_3$, and P$_2$O$_5$, do not define linear trends. Silica contents of glasses tend to be slightly more evolved than the bulk rocks (Figure 4), especially the 1104 C.E. rhyolite. Otherwise, in comparison to whole rock compositions, glasses tend to be poorer in elements that are concentrated in the major phenocryst phases or microlites (Figure 4).

The 1104 C.E. and 1158 C.E. rocks are exceptional as the sole historic eruptions of Hekla to produce only silicic material, with no known late phase of basaltic andesite lava (Thorarinsson, 1967; Sigmarsson et al., 1992; Larsen et al., 1999; Figure 3). The 1104 tephra is relatively homogeneous at the two localities we sampled; bulk-rock analyses of individual pumice clasts vary from 70.0% SiO$_2$ in the bottom third of the deposit to 68.9% SiO$_2$ in the top third of the deposits (on a normalized, volatile-free basis). Glasses from the 1104 deposits range from 71.0 to 72.5% SiO$_2$ (Figure 4).
Pumice clasts from the 1158 C.E. eruption have uniform dacitic (~65% SiO$_2$) compositions despite a range of color and are similar to analyses reported by Larsen et al. (1999). The Haahraun lava flow on the south side of Hekla, now exposed as a kipuka in the 1991 C.E. lava flow field (Figure 1), is the only known silicic lava on Hekla and is thought to have also erupted in 1158 (Sigmarsson et al., 1992).

Eruptions after 1158 C.E. produced intermediate composition lavas and tephra, with whole rock compositions ranging from ~54 to 63% SiO$_2$ (Figure 3). Most of the higher SiO$_2$ rocks (~58 to 63% SiO2) are tephra and the lower SiO$_2$ rocks (~54-57% SiO2) are lava. The maximum whole rock SiO$_2$ is ~62 to 63% following repose periods >70 years (Figure 2), and glass compositions for the zoned historic eruptions reach 64% SiO$_2$, just slightly less evolved than the 1158 C.E. dacite (Figure 5). These results confirm the observation first made by Thorarinsson (1967) that the length of repose correlates to the SiO$_2$ content of the first-erupted material (Figure 2).

The most recent eruptions with substantial compositional zoning are the 1845 C.E. and the 1947-48 C.E. events, which occurred after repose periods of 77 and 102 years. Well-preserved deposits of the 1845 and 1947 tephra layers are characterized by a brown-to-black color transition from the bottom to the top of the tephra deposits, corresponding to a compositional change from ~63 to 59% SiO$_2$ (Figure 5). Lavas emitted during the waning phase of the eruptions have a minimum of ~54% SiO$_2$. The 1947-48 eruption produced materials with a compositional range nearly as large as that of all compositions erupted since the 1158 C,E, dacite (Figure 3).

Following the 1947-1948 C.E. eruption, Hekla erupted again after a repose of only 22 years. The products from the 1970 C.E. eruption have a much narrower compositional range, ~1% SiO$_2$ difference between tephra phase and last-erupted lava. Chekol et al. (2011) and Thorarinson and Sigvaldson (1971) report analyses of silicic pumice from the 1970 deposits, and Sigmarsson et al. (1992) report a silicic ignimbrite xenolith. We observed white frothy inclusions in 1970 tephra, but as discussed below, we interpret these to be fused rocks that were incorporated into the 1970 magma very near the surface and are volumetrically unimportant. Since 1970, Hekla has shifted to more regular eruptions, with repose periods of only 9-10 years, although the volcano has been quiet for more than 20 years since the 2000 C.E. eruption. The products of these most recent
eruptions exhibit minimal compositional variability compared to the previous historical eruptions (Figure 3). The most mafic lavas analyzed in this study are from the last phases of the 1947, 1970, 1980, 1991, and 2000 eruptions, which have SiO$_2$ contents approaching 54%.

**Phenocrysts**

Hekla lava and tephra are notably crystal-poor and have an anhydrous assemblage consisting of plagioclase + olivine + clinopyroxene + magnetite + apatite (irrespective of bulk-rock composition) ± orthopyroxene ± ilmenite ± zircon. The basaltic andesites have a maximum crystallinity of 9% (by volume), and crystallinity decreases with increasing SiO$_2$ content to the andesites (Electronic Appendix 3; Figure 6). In contrast, the silicic andesites, dacites, and rhyolites are richer in crystals. The relationship between crystallinity and composition is also reflected in zoned eruptions. For the 1947-48 event, the first-erupted, most evolved andesite contains ~3 modal % crystals, which decreases to nearly 0% in the upper part of the tephra deposit (Sigvaldasson, 1974). This finding is also consistent with crystallinity data from the 1104 C.E. rhyolite tephra and prehistoric H3 and H4 tephra, which also exhibit a correlation between crystal content and composition (Sigvaldasson, 1974; Sverisdottir, 2007). The size of mafic phenocrysts also correlates with rock composition. Olivine phenocrysts in basaltic andesites vary from 0.3 to 0.5 mm; pyroxene phenocrysts are from 0.5 to 1 mm. Olivine and pyroxene crystals in the andesite, dacite, and rhyolite samples are twice these sizes. Basaltic andesites and early-erupted andesites have euhedral phenocrysts characterized by sharp, well-defined grain boundaries, whereas phenocrysts in later-erupted, crystal-poor andesite tephra and lava have subhedral to anhedral habits. Most pyroxene and plagioclase in the 1104 rhyolite is subhedral, although most faces are faceted (Electronic Appendices 7 and 8). Many grains have concentric growth zones. Some grains enclose inclusions of apatite, zircon, magnetite, and pyrrhotite. The higher-Mg pyroxene in the 1158 C.E. dacite tephra is mostly euhedral, although many grains have inclusion-riddled and corroded lower-Mg cores. Some of these grains have zircon, magnetite, and pyrrhotite inclusions. Plagioclase is also faceted in the dacite and much is intergrown with anhedral olivine and pyroxene in glomerocrysts.
Plagioclase is the dominant phenocryst phase in all Hekla rocks, typically constituting between 50 and 80% of the phenocryst assemblage. Most rocks contain clinopyroxene, and orthopyroxene is present in some andesite and basaltic andesite. Orthopyroxene is relatively abundant in the upper parts of the 1845 C.E. and 1947 C.E. andesite tephra deposits, where it is more abundant than clinopyroxene. Sparse orthopyroxene also occurs in the 1389 C.E. and 1970 C.E. tephra, as well as late-stage spatter from the 1947-48 eruption.

Magnetite phenocrysts are ubiquitous in Hekla rocks and occur as solitary phenocrysts, in glomerocrysts, and abundant inclusions in olivine and pyroxene phenocrysts. Olivine-magnetite intergrowths are especially abundant in basaltic andesites. Ilmenite occurs as sparse ~100 µm euhedral inclusions enclosed within, or closely associated with, magnetite, clinopyroxene, and olivine phenocrysts. All rocks contain apatite, typically as 50-100 µm euhedral microphenocrysts along the margins of and as inclusions in magnetite, olivine, and clinopyroxene phenocrysts and, to a lesser degree, plagioclase. Apatite inclusions are larger and constitute a greater proportion of the assemblage in the basaltic andesites than in more evolved compositions. Zircon is a trace phase in the 1104 C.E. rhyolite tephra and 1158 C.E. dacite tephra but is otherwise absent. It occurs as 20-100 µm prismatic microphenocrysts on the edges of and as inclusions within clinopyroxene, magnetite, and olivine. Pyrrhotite is present as a trace phase throughout the Hekla suite. It occurs as 5-20 µm anhedral inclusions in magnetite, olivine, and pyroxene phenocrysts, as well as some plagioclase crystals from the 1947 C.E. andesite.

Olivine compositions range in composition from Fo_{11} to Fo_{59} and correlate with rock compositions and form a trend that is sub-parallel to the equilibrium olivine field predicted by standard olivine-liquid Fe-Mg partitioning models, but at lower Fo contents in the more silicic rocks (Rhodes et al., 1979; Putirka, 2008; Figure 7A). Experimental data on iron-rich compositions indicate that the $K_{\text{D}}^{\text{Fe/Mg}}$ increases to 0.41 for olivine of composition Fo_{27} (Toplis and Carroll, 1995; c.f. Fo_{11} in the 1104 rhyolite), which fit the Hekla data better than the standard value of 0.30.

One 1766 C.E. andesite contains sparse pl-px-ol-mt glomerocrysts with anomalously low-Fo olivine (Fo_{29-31}), compositions similar to some olivine from the 1158 C.E. dacite.
Euhedral olivine phenocrysts within the host lava have Fo_{37-39} (Figure 7A). The 1104 C.E. rhyolite and 1158 C.E. dacite tephra have a restricted range of olivine compositions, and the crystals are unzoned (Figure 7B). Most olivine in the first-erupted andesite tephra from the 1845 and 1947 zoned eruptions are compositionally uniform, whereas olivine from the later-erupted tephra are reversely zoned, with spatially broad zoning (Figure 7C). Basaltic andesite lava and spatter from the final phase of the 1947-48 eruption is unzoned or has olivine with narrow, normally zoned rims (Figure 7B). Basaltic andesite tephras from the last four Hekla eruptions contain unzoned olivine similar to the olivine from late in the 1947-48 eruption (Figure 7B).

Pyroxene compositions also correlate with rock composition but are more complexly zoned than olivine (Figure 8). Pyroxenes in the basaltic andesites exhibit no regular zoning pattern and have sectors that vary by <2% in Mg# (Figure 8C). Pyroxene from low-SiO_{2} andesites from the upper parts of the 1947 and 1845 tephra have ~50 μm reversely zoned rims that trend to compositions of augite from the basaltic andesites. One 1766 C.E. andesite lava sample contains sparse clinopyroxene in pl-px-ol-mt glomerocrysts, which have orthopyroxene reaction rims and are similar in composition to pyroxenes in dacites (Figure 7; H05-08). Pyroxene in the 1158 C.E. dacite is both normally and reversely zoned, but all measured grains have similar rim compositions (Mg# ~43; Figure 8B). The most Fe-rich cores in dacite pyroxene have the same composition as the rims and cores of pyroxene in the 1104 C.E. rhyolite. These evolved cores contain abundant glassy inclusions. The 1104 rhyolite contains pyroxene that has either oscillatory zoning (~ 5% variation in Mg#) or normal zoning across the entire grain, but rim compositions converge to Mg# ~27 (Figure 8B).

The compositions of plagioclase phenocrysts also correlate strongly with the composition of their host rocks (Figure 9A). Most of the plagioclase in the basaltic andesites are normally zoned, with cores of An_{54-60} and rims extending to An_{40}. Plagioclase in the 1158 C.E. dacitic tephra has oscillatory zoning, ranging in molar anorthite by ~4% (Figure 9B). Plagioclase in the 1104 C.E. rhyolite is broadly normally zoned, with rims up to 300 μm thick (Figure 9B). Unlike the reversely zoned pyroxene, plagioclase in the early erupted
andesitic 1947 tephra lacks zoned rims, but the cores are oscillatory zoned from An$_{51}$ to An$_{56}$.

**Melt Inclusions**

We analyzed glassy olivine-hosted melt inclusions in basaltic andesite tephra from the 1947, 1970, and 2000 C.E. eruptions, the zoned eruptions from 1510, 1845, and 1947 C.E., and the 1104 C.E. rhyolite. Major element compositions of the olivine-hosted melt inclusions are similar to those of the whole rock compositions, the same finding as earlier studies (Moune et al., 2007; Portnyagin et al., 2012; Lucic et al., 2016), although melt inclusions in olivine of similar composition have a small range in Mg#, suggesting post-entrapment crystallization. For most rocks, the range in glass Mg# is <5 and can be accounted for by <10% crystallization of olivine. Because of uncertainty in the Fe-Mg exchange coefficient for fayalitic olivine and the small amount of post-entrapment crystallization, we do not correct the melt inclusion compositions (Figures 4 and 5). In contrast to the Hekla suite, melt inclusions in olivine from primitive Icelandic basalts tend to be compositionally heterogeneous, differing from the carrier melt, which has been interpreted as entrainment of a cumulate mush that was precipitated from compositionally distinct melts (e.g., Maclennan, 2008). In general, melt inclusions in olivine from the basaltic andesites tend to be slightly poorer in SiO$_2$ than the carrier melts (Figure 4A), but the absence of any basaltic melt inclusions is notable. The melt inclusions in the 1104 rhyolite olivine are more silicic than the whole rock but are similar in composition to the carrier melt (Figure 4A-D).

H$_2$O concentrations in melt inclusions increase with SiO$_2$ and incompatible element concentrations, especially the maximum concentrations from any given deposit (Figure 10). This follows the pattern described in Moune et al. (2007), Lucic et al. (2016), and Portnyagin et al. (2012), but our analyses generally extend to slightly higher H$_2$O concentrations, 6.2 ± 0.25%. The average of the ten highest H$_2$O/K$_2$O measured in our melt inclusion set is 2.2, consistent with the 2.0 reported by Lucic et al. (2016) and 2.3 by Pornyagin et al. (2012) (Figure 10). H$_2$O concentration is zoned in the 1947 C.E. deposits, with the highest values in the more evolved tephra that erupted explosively at the start of the eruption and lower concentrations in the effusive near-vent spatter (Figure 5).
noted by others (Portnyagin et al., 2012; Lucic et al., 2016), H$_2$O concentrations are especially high (to 6.9%) in the 1104 C.E. rhyolite. Carbon dioxide concentrations in melt inclusions are lower than detection limits (typically about 50-100 ppm in melt inclusions of this size) in all of our samples. The low CO$_2$ is consistent with low concentrations measured by Lucic et al. (2016).

Sulfur concentrations of olivine-hosted melt inclusions correlate with the FeO$^*$ concentrations of the inclusions (Electronic Appendix 3). The sulfur concentrations in melt inclusions from the basaltic andesites agree well with those measured by Moune et al. (2007) and match the sulfur concentrations of pyrrhotite-saturated Hekla melts determined experimentally (Moune et al., 2008). The sulfur concentrations also agree well with the predicted sulfide solubility for models with $a_{FeS}=1$ (Smythe et al., 2017). Although the sulfur concentrations in the melt inclusions in the andesites are 100-150 ppm richer in sulfur than the thermodynamic model predicts, they are ~100 ppm poorer in sulfur than experimental glasses with 9% FeO$^*$ that are pyrrhotite-saturated (Moune et al., 2008).

Trace Elements

Rb is a highly incompatible element in the Hekla system; other elements that are typically incompatible in mafic systems are affected by the observed phenocryst phases, including apatite, ilmenite, and ferroaugite, as well as zircon in the most evolved rocks. Barium, U, Pb, and Th correlate almost perfectly with Rb (Figure 11); the slope and intercept on these plots indicate that Ba, U, Pb, and Th are slightly less incompatible than Rb, if they are related by crystallization. Yttrium (and the heavy REE elements) forms 3 distinct linear trends (Figure 11): tephra from the 4 late 20$^{th}$ century eruptions have distinctly higher Y at a given Rb concentration in comparison to the pre-1970 C.E. eruptions. Also, the 1158 C.E. dacite and 1104 C.E. rhyolite are offset to low Y concentrations. Strontium, much like Al$_2$O$_3$, correlates with Rb then inflects to define a negative relationship in the andesites. The tephra are systematically richer in Sr than the lavas for a given Rb concentration (Figure 11). Zirconium and Nb correlate positively with Rb, except for the 1104 C.E. rhyodacitic tephra, which have anomalously low Nb and Zr concentrations; in contrast, the 1158 C.E. dacite is unusually rich in these elements. The
trend of the 1947 zoned eruption is offset to slightly low Zr and Nb concentrations, especially at intermediate compositions. Uranium and Th have a strongly coherent linear trend, with an \( r^2 \) of 0.99 (Figure 12). Because the linear array has a non-zero intercept, the U/Th changes with the extent of evolution.

Owing to the large number of analyses, we apply ratios of REE elements rather than the standard REE pattern (Figure 13). The ratio Dy/Yb is a measure of the fractionation across the middle-to-heavy REEs, and Dy* (Davidson et al., 2013) quantifies the concavity of the REE pattern; Dy* values less than unity indicate patterns that are concave-up. In Hekla rocks, Dy/Yb and Dy* are strongly correlated. The patterns are increasingly concave-up with evolution, whereas Dy/Yb are greater than one and decrease with evolution, indicating decreasing middle/heavy REE fractionation. Hekla rocks also exhibit a correlation between La/Sm and La (Figure 13B), and La/Sm increases steadily with evolution of the Hekla suite. Curiously, the strongly zoned 1947 C.E. and 1845 C.E. tephra and lava are systematically shifted to higher La/Sm at a given La concentration.

Compatible trace elements have complex patterns, many of which define two trends; samples from 1970-2000 C.E. exhibit a different slope from the rest of the historical suite (Figure 11). For example, Sc variations have an overall negative slope, consistent with a system crystallizing ferroaugite. In contrast, rocks from 1970-2000 have a positive slope, indicating incompatible behavior by Sc. In fact, this correlation is largely due to tephra samples systematically having greater Sc concentrations for a given Rb concentration. Likewise, V decreases exponentially with respect to Rb, but the tephra samples are systematically offset to higher Rb and V than the lava samples.

**Oxygen Isotopes**

As is observed in many igneous rocks from Iceland (e.g., Bindeman et al., 2008a), olivine from Hekla rocks have \(^{18}\text{O}/^{16}\text{O} \) below typical mantle values (Figure 14). Moreover, individual olivine grains display within-sample heterogeneity. These olivine separates compare to whole rock values at Hekla of 4.7 to 5.1‰ (Figure 18; Sigmarsson et al., 1992). Single crystals of olivine from the 1104 C.E. rhyolite ranges from 3.4 to 4.2 ± 0.1‰, but there is no systematic change over the course of the eruption. The 1158 C.E. dacite has similar \( \delta^{18}\text{O} \) values to the andesites. Olivine from the strongly zoned 1947 C.E. eruption
ranges from 3.5 to 4.6 ± 0.1‰ as the eruption proceeded from andesite to basaltic andesite, but again, there is no systematic change over the course of the eruption (Figure 5), unlike the major and trace element compositions and H₂O concentration. The 2000 C.E. basaltic andesite, which is typical of Hekla’s mafic rocks, has olivine with 4.1‰, 1‰ lower than the mantle.

We also analyzed olivine from several historic and Holocene basalts erupted in the vicinity of Hekla, which range from 3.7 to 4.7 ± 0.1‰ (Electronic Appendix 4; Figure 14). The most notable observation is that olivine in the most primitive basalts has the highest δ¹⁸O, and δ¹⁸O decreases strongly with differentiation while still in the basaltic realm, a trend similar to Laki and other large volume basaltic systems in Iceland (Bindeman et al., 2008a).

Zoned Eruptions

As noted by Thorarinsson (1967; 1970), many Hekla eruptions are zoned, and the extent of the zoning depends on the repose between eruptions (Figure 2). We focus on the 1947 C.E. eruption as a type-example (Figure 5), although the 1845 eruption exhibits similar compositional diversity. The first erupted tephra is highest in SiO₂, with 62.4% SiO₂, corresponding to a glass composition of 63.6% SiO₂. Thirteen melt inclusions trapped in ten different olivine crystals have between 61 and 64% SiO₂, with an average composition of 62.5% SiO₂, virtually the same as the rock. Over the course of the eruption, whole rocks and glasses steadily decrease to 54.3% SiO₂; the glassy melt inclusions have practically the same composition as the whole rocks and glasses. Melt inclusions from the upper tephra, which is also poor in crystals, are more compositionally variable.

DISCUSSION

A wealth of geochemical data have been published on Hekla, which has been interpreted primarily in two ways. One line of hypotheses is that crystal fractionation of basaltic parent magma creates a range of intermediate to silicic magma compositions (Baldridge et al., 1973). The second class of models contends that the silicic magmas contain a large component of crustal rocks. In the latter models, intermediate compositions
are attributed to magma mixing of end-member compositions, a combination of crystal fractionation and magma mixing, or a combination of assimilation and fractional crystallization (Sigvaldason, 1974; Sigmarsson et al., 1991, 1992; Sverrisdotir, 2007; Chekol et al., 2011; Lucic et al., 2016). The comprehensive dataset presented here permits evaluation of these hypotheses, particularly given the constraints of timing and eruptive order.

**Conditions of Magma Storage**

**Magma Storage Temperatures**

Equilibration temperatures and oxygen fugacities from Fe-Ti exchange in magnetite-ilmenite pairs using the calibration of Ghiorso and Evans (2008) are calculated after verifying equilibrium using the Mg-Mn partitioning test of Bacon and Hirschmann (1988). Temperatures range from $1011 \pm 5^\circ C$ for basaltic andesite (single oxide pair from 1970 tephra) and $953 \pm 14^\circ C$ for andesite (three oxide pairs from 1947 tephra). Analyses of an oxide pair included within a clinopyroxene phenocryst from the 1510 C.E. andesite tephra yield a temperature of $897^\circ C$. This phenocryst also contains zircon inclusions, which otherwise are only present in dacite and rhyolite at Hekla. Calculated oxygen fugacities for all magnetite-ilmenite pairs fall within 0.25 log units of the FMQ buffer, except for the slightly more oxidized ($\log[O_2] = FMQ+0.5$) results for the 1970 basaltic andesite oxide pair. These temperatures and oxygen fugacities compare to an average of $870 \pm 26^\circ C$ ($\log[O_2] = 0.4$ log units more reduced than FMQ) for the prehistoric H3 tephra (Portnyagin et al., 2012).

We apply the 2-pyroxene geothermometer of Putirka (2008; equation 36) to all pyroxene pairs from each rock for which we have data. Pairs that are not in Fe-Mg equilibrium are discarded and the remaining pairs averaged. The only pyroxenes found to pass this equilibrium test are in the basaltic andesites and andesites, and they yield temperatures of $1003 \pm 8^\circ C$ and $978 \pm 14^\circ C$, in close agreement with the oxide thermometer (Figure 15). Forty-three clinopyroxene analyses from basaltic andesites are calculated to be in Fe-Mg equilibrium with their host glass or whole-rock ($K_p = 0.27 \pm 0.3$; Figure 8), and application of the clinopyroxene-melt geothermometer (Putirka, 2008, equation 33) yields $T = 1029 \pm 15^\circ C$. Forty-seven pyroxene analyses from andesite have
FeO and MgO concentrations that meet the equilibrium test and yield $T = 952 \pm 20^\circ C$.

Application of a more stringent filter involving 4 separate exchange equilibria removes 23 pyroxene analyses from the basaltic andesites and all but 2 analyses from the andesites. Calculated temperatures of this filtered subset are $1030 \pm 14^\circ C$ and $952$ to $984^\circ C$ for the basaltic andesites and andesites.

The apatite saturation thermometer (Harrison and Watson, 1984) applies to all Hekla rocks, owing to the ubiquitous presence of apatite phenocrysts. Calculated temperatures for basaltic andesite, andesite, dacite, and rhyolite are 1043, 997, 933, and 877°C (Figure 15). Only the 1104 C.E. rhyolites contain zircon in contact with glass, and it yields a saturation temperature of $878 \pm 6^\circ C$ (Boehnke et al., 2013), almost precisely the same as the other geothermometers (Figure 15). Carley et al. (2011) apply the zircon saturation geothermometer to 1104 C.E. rhyolite tephra, which yields $920^\circ C$ (we note that their interpretation was that their sample is an 1158 C.E. rock, on the basis of the glass composition, whereas we believe it is a 1104 C.E. rhyolite, which is confirmed by Calvin Miller, personal communication, 2020). The Ti-in-zircon geothermometer in the same rocks results in temperatures ranging from 750 to 930°C.

In summary, a series of geothermometers yield consistent results, with the most mafic basaltic andesites erupting between 1040 and 1000°C, the andesites (~60% SiO$_2$) between 1000 and 950°C, the 1158 C.E. dacite from 950 to 900°C, and the 1104 C.E. rhyolite from 890 to 870°C (Figure 15).

**Magma Storage Pressures**

Our interpretation of the melt inclusion H$_2$O concentrations is that the maximum values in any given sample represent the magmatic concentrations at the site where the crystals grew prior to eruption, owing to the excellent correlation of the maximum H$_2$O concentrations with incompatible element contents (Figure 10). The same interpretation has been made by Moune et al. (2007) and Portnyagin et al., (2012). Lower values are interpreted to be either the result of entrapment during ascent and degassing of the magma or diffusive hydrogen loss from the melt inclusions as the host melt dehydrates. If the magmas were undersaturated in H$_2$O-rich vapor as the crystals were forming, calculated vapor saturation pressure is a minimum estimate of the trapping pressure for the
inclusions. The calculated vapor saturation pressure for the highest H$_2$O concentration in the 1104 C.E. rhyolite is 220 MPa. Calculated saturation pressures are 106 MPa for the early erupted 1947 C.E. andesite tephra and 56 MPa for the 2000 C.E. basaltic andesite (Figures 10 and 15; assuming 80 ppm CO$_2$; Papale et al., 2006). Another solubility model (Ghiorso and Gualda, 2015) does not converge for the mafic and felsic melts, but the calculated saturation pressure for the andesite melt is 212 MPa. The H$_2$O solubility model of Moore et al. (1998) yields pressures of 215, 140, and 65 MPa for the rhyolite, andesite, and basaltic andesite (Figure 15). Given these very shallow depths relative to the geophysically determined depth to the magma reservoir and the mineral-melt geobarometric results reported below, it is clear that the Hekla magma cools and evolves in the absence of a water-rich fluid. Major vesiculation likely does not occur until the magma is at very shallow depths, < 2 km for the basaltic andesite.

We screened 143 clinopyroxene-melt pairs in the basaltic andesites and andesites for Fe-Mg, CaTs, EnFs, and DiHd equilibrium (bound set at ±20%) and found 90 analyses to be in Fe-Mg equilibrium with their host glass (or whole rock for samples lacking glass), and 22 analyses to be in equilibrium for all 4 of these exchange components. H$_2$O concentrations of melts are estimated on the basis of constant K$_2$O/H$_2$O, and we apply Putirka’s (2008) equation 31. Kernel density estimation (with a Gaussian filter) yields a maximum density of 570 ± 130 MPa for the basaltic andesites and 520 and 540 MPa for the andesites, when the more stringent filter is applied (interquartile range; total range is 490 to 710 MPa). When only the Fe-Mg filter is applied, estimated pressures are 510 ± 110 MPa for the basaltic andesites and 400 ± 130 MPa for the andesites, consistent with the depth to the main magma reservoir as determined by surface deformation since 2000 C.E, (Geirsson et al., 2012). Putirka’s (2008) equation 30 yields pressures approximately 100 MPa lower. Thus, the clinopyroxene compositions are consistent with the basaltic andesite magmas partially crystallizing at depths of at least 12 km and likely > 17 km, in the lower Icelandic crust. The large number of crystals that are not in equilibrium according to these 4 exchange reactions might be due to rapid crystal growth relative to diffusion rates, or alternatively the crystals grew from melts that no longer host them and they were picked up by the carrier melts as magma ascended over the course of ascent and eruption.
Moreover, the exchange reactions are calibrated mostly on basalts and may be imperfectly calibrated for the evolved Hekla compositions.

$H_2O$ concentrations of the melts can also be estimated with the plagioclase hygrometer (Waters and Lange, 2015). Average plagioclase and glass (where available) or whole-rock compositions are applied, along with temperatures from geothermometers and a pressure of 0.5 GPa. The hygrometer is only weakly dependent on temperature and pressure (Waters and Lange, 2015). The plagioclase compositions agree with the melt-inclusion data and indicate a steady increase in $H_2O$ with magmatic evolution (Figure 10) and an excellent correlation with $K_2O$, but at values that are 0.5 to 1.0% $H_2O$ lower than the maximum melt inclusion concentrations. The cause of this systematic difference is unclear. Waters and Lange (2015) report an uncertainty in their calibration of ± 0.35% $H_2O$. Uncertainty in temperature and pressure estimates results in increasing the uncertainty by an additional 0.2%. The congruence of the plagioclase-melt equilibrium calculations and the melt inclusion measurements confirms that melt $H_2O$ correlates strongly with incompatible elements in the Hekla suite, however.

Melting experiments on H3 tephra rhyodacite, which is a prehistoric Plinian deposit (about 1000 B.C.E.) similar in composition to the 1104 C.E. tephra studied here, show the melt to be saturated with olivine, plagioclase, clinopyroxene, and vapor at approximately $850 \pm 15°C$ and $P_{H2O} = 130$ to 175 MPa (Weber and Castro, 2017). This implies storage in the shallow crust.

**Magmatic Evolution at Hekla**

We approach the problem of the petrogenesis of Hekla’s diverse suite by breaking the problem into three parts: the relationship between the basaltic andesites and andesites (including the zoned eruptions), the significance of low-$\delta^{18}O$ values, and the origin of the dacite and rhyolite.

**Basaltic Andesites to Andesites (54 to 62% SiO$_2$)**

The strikingly coherent relationships among the major elements across Hekla eruptions suggests that crystallization is likely the dominant process relating the basaltic
andesite to the andesites (Figure 3). Basaltic andesites at Hekla contain the phenocryst assemblage pl + aug + ol + mt + ilm + ap. We apply a least squares mass balance method to determine whether these phases can account for the chemical variation of Hekla magmas with 54 to 62% SiO$_2$. Our results indicate that the eruptive products of the 1947 eruption, which range from 54.7 (H07-01) to 62.5% (H05-18) SiO$_2$ can be explained by fractionation of 4.2% olivine, 12.8% plagioclase, 6.1% augite, 4.0% magnetite, 2.0% apatite, and 1.1% ilmenite, with a sum of squares of residuals of 0.02; mineral compositions are averages from the parental basaltic andesite (Electronic Appendices 6-9). This amounts to a total of 30% crystallization, and according to the geothermometers, it is driven by cooling of the melt 50 ± 20°C.

As noted by others (Checkol et al., 2011), the basaltic andesite to andesite compositional changes can be modeled by the MELTS algorithm. The best-fit models are those at lower-crustal pressure conditions (400 to 500 MPa) and H$_2$O concentrations of 0.4% in the parental basalt. We also modeled the liquid line of descent of the 1947 basaltic andesite, 1947 andesite, and 1158 dacite using H$_2$O concentrations defined by the melt inclusion K$_2$O-maximum H$_2$O correlation. Models run at pressures of 200 to 600 MPa using rhyolite-MELTS (Gualda et al., 2012) closely match the major-element trends, with the exception of Al$_2$O$_3$ and P$_2$O$_5$. The highest Al$_2$O$_3$ concentrations at Hekla are 15.4%, in andesites with ~62% SiO$_2$, and these rocks contain plagioclase phenocrysts. Rhyolite-MELTS predicts that the inflection in Al$_2$O$_3$ is at 16.3% at 200 MPa or 16.8% at 600 MPa, alumina concentrations that exceed those in any Hekla rocks, most of which contain plagioclase phenocrysts. Rhyolite-MELTS also does not predict saturation with apatite in Hekla andesite and dacite, despite that the rocks contain euhedral apatite phenocrysts and have the compositional fingerprint of having crystallized apatite. Villiger et al. (2004) and Balta and McSween (2013) note that MELTS is successful at predicting experimental liquid lines of descent to only within 1 weight % for most oxide components, and with that criterion, the Hekla suite is well modeled by rhyolite-MELTS with the caveat that the model underestimates the temperature of plagioclase saturation at these higher pressures and H$_2$O concentrations.

The coherent linear trends between incompatible elements are also consistent with crystallization processes to explain the relationship between basaltic andesite to andesite
The array of data between compatible and incompatible elements (e.g. Rb vs. V, Sc, and Sr) are curved, also consistent with crystallization. To model all of the trace elements quantitatively, we adopt the partition coefficients in Table 4 of Nielsen (1992), supplemented by more recent data from the Geochemical Earth Reference Model (https://earthref.org/GERM/) (Electronic Appendix 7). Mineral and melt fractions determined by the least-squares major element model are applied to calculate the bulk partition coefficient, and the Rayleigh equation is used. The concentrations predicted by this method match the incompatible element contents in Hekla lavas closely (Figure 16), certainly within the uncertainty in the partition coefficients. The poorest fits are for U and Th (see below) and the middle-to-heavy REEs, both of which we attribute to poorly determined partition coefficients for apatite, which are likely higher in the Hekla suite than those adopted here (c.f. Prowatke and Klemme, 2006). Also, the distribution coefficients for heavy REEs are known to be substantially higher for iron-rich pyroxene (Olin and Wolff, 2010) than for the compositions we used.

Mineral compositions in Hekla lava and tephra support the fractional crystallization model. With few exceptions, olivine, pyroxene, and plagioclase solid-solution compositions correlate strongly with their host rock compositions (Figures 7-9). This would not be the case if the intermediate rocks resulted from magma mixing (assuming some crystals survived hybrization), or if the andesites were formed by crustal melting or wall rock assimilation (assuming that some crystals would have derived from the protolith or wall rock). With few exceptions, the cores of most crystals have only minor oscillatory zoning, fluctuating by only small variations in Mg# or An (Figures 8, 10, and 12), and the most common style of zoning is narrow, normally zoned rims.

Melt inclusions in olivine in the basaltic andesites tend have the same composition as the matrix glass or to be slightly more mafic (<2% SiO₂), which is consistent with the olivine crystallizing from a slightly hotter ancestor of the host melt, whose composition shifted due to crystallization of the phenocrysts themselves. If the evolution from basaltic andesite to andesite is caused by 60°C cooling (Figure 15), then the <2% change in SiO₂ between the melt inclusions and host glass could be driven by <30° of cooling. A few of the melt inclusions in basaltic andesites have anomalously low MgO concentrations, no doubt due to small amounts of post-entrapment crystallization (Figure 4), but the vast majority of
them have compositions matching those of the host glass, indicating a near absence of exotic melts that would be indicative of hybridization.

The sulfur concentrations in olivine-hosted melt inclusions in both the basaltic andesites and the andesites are close to those predicted at sulfide saturation (Smythe et al., 2017) and produced experimentally (Moune et al., 2008). Moreover, most Hekla rocks contain pyrrhotite, generally as inclusions in phenocrysts. The measured sulfur concentrations in melt inclusions in the andesites are consistent with fractionation of 0.10 to 0.15% by mass pyrrhotite from a basaltic andesite melt.

Origin of Low $\delta^{18}O/^{16}O$

Because the oxygen isotope ratios of nearby basalts overlap with those of the evolved rocks erupted from Hekla (Figure 14), our interpretation is that basalt with typical upper mantle $\delta^{18}O$ assimilates low $\delta^{18}O$ Icelandic crust in the lower crust, creating an evolved basaltic melt with low $\delta^{18}O$, consistent with the interpretations of Hemond et al., 1988, Sigmarsson et al., 1992b, and Bindeman et al., 2008a. Alternatively, low $\delta^{18}O$ may be a characteristic of the Icelandic mantle (e.g. Winpenny and Maclellan, 2014). Irrespective of the origin of low $\delta^{18}O$ in the basalts, the low $\delta^{18}O$ in Hekla basaltic andesites compared to estimates of the upper mantle composition is likely inherited from the parental basalts (Sigmarsson et al., 1991; 1992).

Models of crystallization that include changing isotopic fractionation factors with temperature and mineral and melt composition predict a ~0.2‰ increase in melt $\delta^{18}O$ as the $SiO_2$ concentration increases from 55 to 70% (Bindeman et al., 2004). The model predictions are in close agreement with empirical data from suites of magmas that have evolved to dacite and rhyolite by fractional crystallization at the Galápagos Spreading Center (~0.4‰ difference between basaltic andesite and rhyolite; Muehlenbachs and Byerly, 1982) and the Galápagos Islands (Geist et al., 1998). Superimposed on the melt trend is an increase in $\Delta^{18}O$(melt-olivine) by 1‰, because of compositional and temperature effects on olivine-melt fractionation: olivine is more Fe-rich and the melt more Si-rich as the magma cools. Therefore, $\delta^{18}O$ in olivine is expected to decrease by about 0.6‰ from basalt to rhyolite, or 0.2‰ as basaltic andesite magma cools from 1010°C to
875°C and the melt evolves from 55 to 70% SiO$_2$, precisely what is observed in the Hekla basaltic andesite to rhyolite suite (Figure 14; Bindeman et al., 2008b). The moderate scatter in δ$^{18}$O in olivine from the same eruption likely involves inheritance from isotopically heterogeneous basaltic melts, comparable to those in large-volume fissure system of Iceland (e.g. Bindeman et al. 2008a). In both cases recycled olivine will preserve heterogeneous isotopic compositions of melts even if they have equilibrated with respect to Fe-Mg, owing to slow diffusion of oxygen relative to Fe-Mg diffusion (Ryerson et al., 1989).

**Origin of Dacite and Rhyolite**

The origin of the dacites is particularly important, because they have been proposed to be the result of crustal melting or assimilation coupled with crystallization, on the basis of evidence of U/Th isotopic disequilibrium (Sigmarsson et al., 1992; Chekol et al., 2011). Dacite magma is posited to be the parental magma to Hekla’s Plinian rhyolites, which formed by fractional crystallization of the dacite. Our findings, in contrast, indicate that the dacites and rhyolites adhere nearly perfectly to a fractional crystallization model, whereby the dacites derive from crystallization of silicic andesite. The composition of sample H05-63, from the 1158 C.E. dacite (whose composition is the same as that of 1158 tephra, H09-01) can be created by removal of 2.3% olivine, 6.8% plagioclase, 1.1% augite, 1.0% magnetite, 0.4% apatite, and 0.4% ilmenite from an andesite melt (H05-18) with 62.5% SiO$_2$ (mineral compositions are averages of compositions from the parental andesite, Electronic Appendix 6-9). This leaves 88% dacitic melt, with a sum of the squares of the residuals of 0.86. We note, however, that there is strong petrographic evidence that the petrogenesis of the dacite is more complicated, because it contains xenocrystic crystals, including zircon, that we interpret as having formed from the 1104 C.E. rhyolite and being incorporated in a dacitic melt. Trace element systematics also support the crystallization model, although the partition coefficients for Sc, V, and Sr appear to be underestimated (Figure 16B), and high U, Th, and Hf concentrations no doubt reflect the xenocrystic zircon observed in these rocks. As with the basaltic andesite-to-andesite model, U and Th are
predicted to be richer in the evolved rocks than is observed, which we attribute to underestimation of the partition coefficients of U and Th in apatite.

The major element mass balance model relating the 1158 C.E. dacite to the 1104 C.E. rhyolite (H07-25) is an almost perfect fit, with an \( r^2 \) < 0.01, and previous work by Sigmarsson et al. (1992) has also proposed that Hekla’s rhyolites are generated by fractional crystallization of dacite melt. The model predicts crystallization of 3% olivine, 10% plagioclase, 1.4% ferroaugite, 1.3% magnetite, and 0.3% apatite (mineral compositions are averages of analyses of crystals in the parental dacite, Electronic Appendix 6). The low concentrations of sulfur in the melt inclusions in olivine from the rhyolite (96 ± 50 ppm) also require that pyrrhotite, which is observed in the rocks, also fractionates, but we do not include it in the trace-element model. We adopt the partition coefficients of Bacon and Druitt (1989) for dacite and interpolate for elements not included in their data set (Electronic Appendix 10). The amount of zircon fractionation was calculated by mass-balance of Zr, and partition coefficients calculated from average Hekla zircon from Carley et al. (2011) were applied. Again, the fit is excellent except for the compatible elements (which are particularly sensitive to the uncertain values of the partition coefficients) as well as U and Th. Thus, our proposal for the formation of the 1158 C.E. dacite requires a combination of fractional crystallization and incorporation of the cumulate complement to the 1104 C.E. melt-rich magma that erupted 154 years previously.

Pyroxene from the 1158 C.E. dacite contains reversely, normally, and un-zoned grains, indicating that many of the grains did not simply crystallize from their host melts. Some plagioclase and pyroxene in the 1104 C.E. rhyolite is broadly normally zoned, consistent with its growth from a cooling melt, whereas olivine and some pyroxene and plagioclase is unzoned (Figures 7-9). Melt inclusions in olivine in the 1104 rhyolite are slightly more evolved (by about 4% SiO\(_2\)) than the host rock, but the compositions are the same as the host glass (Figure 4), indicating that these crystals formed from their carrier liquids.

**Sparse Evidence for an Open System**

In our experience, the Hekla rocks form one of the most compositionally coherent igneous suites identified to date. With few exceptions, major and trace elements form
strikingly consistent trends, and most phenocryst compositions correlate strongly with those of the carrier melts. There is a complete absence of exotic melt inclusions (Figure 4), and crystals that might have been inherited from mafic and silicic endmembers are absent. If the intermediate rocks are hybrids between basaltic andesite and a silicic magma, nearly all the crystals would have had to have formed after hybridization, and we know of no locality where hybrids do not contain inherited crystals. In the thousands of rocks we examined in the field, we observed only one xenolith locality (silicic hyaloclastites in a prehistoric fissure on the NE shoulder of the volcano) and only a single silicic rock with mafic enclaves.

That said, there are several specific examples of crystals that are distinctly out of equilibrium with their carrier melts:

1. The uppermost tephra of the 1947 eruption, which is andesite, contains reversely zoned olivine, with rims that are more magnesium rich than the cores, by about 5% Fo. These reversely-zoned olivine have the same composition as the olivine in the first-erupted andesitic tephra (Figures 5D and 7), and they contain evolved melt inclusions that have the composition of the first-erupted rocks (Figures 4 and 5B). The late-tephra also contain reversely zoned pyroxene whose cores are the composition of pyroxene in the early erupted andesite, but rims that are ~7 mole % richer in diopside component than the cores (Figure 8). Our interpretation is that the crystals initially grew from the evolved andesitic melt, and then were immersed subsequently in a more Mg-rich andesite melt shortly before eruption; there must have been convective stirring of the magma to some degree. Application of Fe-Mg diffusion coefficients of Chakaborty (2010) at 1000°C yields residence timescales of 70 days for the olivine to account for the thickness of the boundary layer (assuming a well-mixed open boundary). The late-erupted, less-evolved tephra also has anomalous trace element compositions, with Nb and Zr concentrations that are ~6 and 15 ppm lower than expected from their Rb concentrations (Figure 11). This offset can be explained by mixing with a small fraction (<10%) of cooler melt that had fractionated ilmenite and zircon, a composition similar to the 1104 C.E. rhyolite, or assimilation of a rock of similar composition.
2. The 1158 C.E. dacitic lava contains both normally and reversely zoned ferroaugite, whose cores are about 6 mol % richer and poorer in diopside component than the rims. This suggests that some pyroxene cores grew from slightly less evolved dacite and rhyolite melts, followed by growth from the carrier dacitic melt. The boundary layers separating these zones are 20-30 µm thick. As noted above, some of the phenocrysts in both the dacite lava and tephra contain zircon, even though the melt was not zircon-saturated (the zircon saturation temperature for this rock is calculated to be 619°C, >200°C below its eruption temperature). Our interpretation is that the 1158 C.E. dacite contains phenocrysts that originally crystallized from the 1104 C.E. rhyolite, and the 1158 magma is a hybrid between dacitic melt and crystal mush residual from the 1104 rhyolite. This makes sense given the close temporal relationship between the two eruptions.

3. A single plagioclase from the 2000 tephra has a core that is An_{47}, then is reversely zoned with a wide rim close to equilibrium with the carrier melt (An_{57-60}). This plagioclase was likely inherited from a dacite (Chekol et al., 2011), but it resided in the basaltic andesite magma long enough for a 500 µm rim to grow. This plagioclase crystal is only one of dozens we analyzed from this eruption, the others having close to equilibrium compositions.

4. The only mafic enclaves we observed and sampled in the historical rocks of Hekla is in the 1766 C.E. andesitic lava, which contains the most forsteritic olivines of any sample, up to Fo_{86}. In all likelihood, these olivines crystallized from a basaltic melt that has not been sampled otherwise at Hekla. As other authors have pointed out (Sigmarsson et al., 1992, and others), the likely parent to Hekla basaltic andesite is a composition similar to that of the nearby 1913 Lambafitarhraun basaltic fissure eruption.

5. A pyroxene in the 1510 C.E. andesite contains magnetite, ilmenite, and zircon inclusions and clearly formed from a dacitic magma. This is the only example of such a crystal in the rocks we inspected. Its origin may be from mixing with silicic magma that had survived for 352 years or partial assimilation of solidified 1158 C.E. dacite.
Subtle differences in the trace element concentrations of the 1947 C.E. eruptive products suggest that its magma might have assimilated a small amount of silicic rocks or mixed with silicic magma, comparable in composition to the 1104 C.E. rhyolite. Specifically, the intermediate 1947 rocks are characterized by anomalously low Nb/Rb and Zr/Rb, which could be accounted for by addition of <5% rhyolitic melt (or granite) with the composition of the 1104 C.E. tephra. On the basis of U-Th isotopic disequilibrium and a correlation with SiO$_2$, Chekol et al. (2011) also find that the 1947 intermediate magmas also are affected by assimilation of young rocks, although they invoke interaction with rocks that we attribute to Torfajökull.

In summary, phenocrystic and melt-inclusion evidence for magma mixing, assimilation, or re-entrainment of cumulate crystals in Hekla's silicic rocks is sparse. Most crystals in Hekla magmas formed from the melt that hosts them at the time of eruption. No cumulate xenoliths or “antecrysts” have been sampled from the volcano, and no material that might be attributed to a cumulate mush have been observed. Consequently, crystal-liquid segregation must be very efficient at all stages of magmatic evolution at Hekla.

The Case Against Crustal Melting

Previous studies have invoked several pieces of evidence to argue that Hekla dacites and rhyolites result from crustal melting or extensive crustal assimilation and are only related to the basaltic andesites and andesites via the transfer of heat.

1. Hekla's silicic rocks have low $^{18}$O/$^{16}$O compared to the Earth's mantle (Sigmarsson et al., 1991; Bindeman, 2008). The origin of the low $\delta^{18}$O in olivine is discussed above: all evidence points to it being inherited from low $\delta^{18}$O basalts, followed by isotopic fractionation during crystallization.

2. Dacites erupted in 2000 have distinct Sr, Nd, and Th isotopic ratios (Chekol et al., 2011);

3. Rocks from the large-volume silicic eruptions have fractionated Th/U relative to the mafic rocks (Sigmarsson et al., 1992). U-Th isotopic disequilibrium indicates that this fractionation must have occurred tens of thousands of years ago,
In addition to hypotheses that propose that Hekla’s silicic rocks are produced by anatexis, Lucic et al. (2016) propose that the basaltic andesites are due to melting of gabbro. They base their hypothesis on the very low abundance of CO$_2$ and variable H$_2$O concentrations in melt inclusions in Hekla rocks.

The exotic Sr and Nd isotopic compositions of dacites from the 2000 C.E. eruption noted by Chekol et al. (2011) prove that these rocks could not have originated by fractional crystallization of the co-eruptive basaltic andesites. We question, however, the ancestry of these dacites and suggest that they might be very local and low-volume, re-melted pieces of silicic rocks from around the vent in the shallow subsurface, as also found in the 1970 deposits (Sigmarsson et al., 1992). Thus, their origin bears little on the petrogenesis of the voluminous 1158 C.E. dacite or the Plinian rhyolites. None of the 5 lava and tephra samples from the 2000 eruption that we analyzed and no other published analyses (Moune et al., 2007; Höskuldsson et al., 2007; Portnyagin et al., 2012) show any sign of a silicic component. The dacites reported by Checkol et al. (2011) have the isotopic composition of rhyolites from Torfajökull (Martin and Sigmarsson, 2010), and young Torfajökull tephra are exposed all around Hekla. All other analyses of 2000 rocks indicate that the eruption was strongly homogeneous basaltic andesite (Figure 3), as is be predicted by the short repose between 1991 and 2000. Although this contamination is local and not of great petrogenetic significance, the compositions of the rare contaminated rocks in the 2000 C.E. deposits indicate that crustal contributions are readily detectable at Hekla.

Sigmarsson et al. (1992) make the strong case that recent fractionation of U/Th between the andesite and dacite requires that the dacite be derived by anatexis of amphibolite from the Icelandic crust. The covariation of U and Th of the basaltic andesite-to-andesite sequence is extraordinarily linear and nearly the same as the andesite-to-dacite-to-rhyolite sequence, especially considering that the rhyolites have crystallized a small amount of zircon, and the dacites have accumulated it. Previous investigators have agreed that the transition from basaltic andesite to andesites is caused by fractional crystallization (e.g. Baldridge et al., 1973; Lucic et al., 2016) or in combination with lesser extents of assimilation (AFC; Sigmarsson et al., 1992; Chekol et al., 2011): the interpretation that is disagreed upon is the origin of the dacites and rhyolites. The linear trend of U and Th in our suite could be explained by extraction of a cumulate with low but
constant U and Th concentrations. Note that linear regression of U versus Th results in a line that intersects the U axis (Figure 12), meaning that if the rocks are related by fractional crystallization, U is slightly less incompatible than Th. Because of the non-zero intercept, the U/Th changes as both U and Th abundances increase with differentiation, a phenomenon observed and emphasized by Sigmarsson et al. (1992). Our laser ablation ICPMS data on apatite are consistent with this hypothesis, although we recognize that the uncertainty is large (Figure 12). Checkol et al. (2011) note that literature data indicate that $D_{\text{U,apatite}} > D_{\text{Th,apatite}}$, which well explains the Hekla U-Th trend, although experimental data indicate that the relative partitioning is not well known (Prowatke and Klemme, 2006 find values of $D_{\text{U,apatite}}$ ranging from 0.2 to 1.4 and $D_{\text{Th,apatite}}$ ranging from 0.7 to 1.3 between apatite and basaltic andesite and andesite melt).

The hypothesis that the dacites are the result of anatexis (Sigmarsson et al., 1992) is untenable for other reasons. The compositions of Hekla dacites are vastly different than melts produced by melting of natural amphibolite at pressures of between 0.3 and 1.0 GPa, which are typically tonalitic and peraluminous. The experimental melts have $\text{Al}_2\text{O}_3$ ranging from 14 to 19% in dacitic compositions and $\text{K}_2\text{O}$ concentrations ranging from 0.9 to 2.8% (e.g. Beard and Lofgren, 1991; Rushmer, 1991; Zhang et al., 2013).

The experiments also indicate that amphibole is in the residue up to 20 to 40% melting, at temperatures $>1000^\circ\text{C}$, and trace element concentrations also disprove an origin of the dacites by melting of amphibolite. For example, Brophy (2008) has shown that partial melting of amphibolite yields distinctive REE patterns. Specifically, partial melting of amphibolite yields La concentrations that are very slightly enriched over the concentrations in the source and Yb concentrations that are depleted. In contrast, Hekla dacites and rhyolites have La concentrations that are up to 150% enriched relative to the most mafic rocks at Hekla and even more so compared to regional basalts (Figures 3 and 16). Yb is also higher in the silicic rocks than the mafic ones. Thus, the REE are inconsistent with melting an amphibolite source to generate the silicic magmas at Hekla.

The basement beneath Hekla is unlikely to have the same trace element and radiogenic isotopic composition as Hekla rocks. The oldest crust around Hekla formed at the Western Volcanic Zone, ~60 km west of the volcano. At a half-spreading rate of 18 km/My (Einarsson, 2008), the oldest rocks beneath Hekla should be $>3$ Ma. Many cycles of
volcanism over that time span will result in an isotopically heterogeneous crust (e.g., Hemond et al., 1993 find a range in $^{87}$Sr/$^{86}$Sr from 0.7029 to 0.7032 in the Western Volcanic Zone). In contrast, the Sr and Nd isotopes of Hekla’s dacites and rhyolites are the same as the mafic rocks (Sigmarsson et al., 1992), and it would be fortuitous if the rocks that melted to form the dacitic magma had the precise Sr-isotopic composition of Hekla basaltic andesite. Moreover, the near-perfect correlation between incompatible elements across the Hekla suite (Figure 3) further argues against older crustal sources for the silicic Hekla material. One possibility is that crustal sources of the silicic magmas are older Hekla magmas, thus having isotopic and elemental compositions similar to the material erupted historically (Sigmarsson et al., 1992). Most potential protoliths in the buried crustal section at Hekla will have undergone some combination of weathering, hydrothermal alteration, zeolite-, greenschist-, and amphibolite-facies metamorphism, each of which would have been accompanied first by the introduction of fluids at temperatures up to 400°C, followed by production and loss of fluids by dehydration reactions beginning at the greenschist facies (e.g. Oskaarson et al., 1982). For example, a series of Icelandic rocks that have undergone simple burial metamorphism (to 90°C) in the upper few kilometers of the crust (Neuhoff et al., 1999) have undergone large amounts of mineralogical and major-element transformation, and mobile trace elements have been altered by more than 10-fold: their composition is unlike that of the unmetamorphosed basalt. Amphibolite-facies metamorphism would then alter the compositions of the original basalts and gabbros even more. If amphibolites melted, they would have left a residue comprising amphibole, pyroxene, and plagioclase up to 20-40% melting and 1000°C (Rushmer, 1991; Zhang et al., 2013; Jagoutz and Klein, 2018). It is highly unlikely that such metamorphic and anatectic processes could have occurred without strongly fractionating Pb/Rb, Zr/Rb, K/Rb, Th/Rb, U/Rb, Dy*, and Ba/Rb (Figures 14 and 15). It is challenging to speculate on what the sulfur concentration of a basaltic andesite melt produced by melting of gabbro would be (Lucic et al., 2016), and likewise for melting of amphibolite to produce the dacite (Sigmarsson et al., 1992). If the crustal rocks contained residual sulfide, then the observed concentrations would be produced, but we know of no way of testing that hypothesis.

Hekla has been investigated for the stable isotopic composition of Li, Si, Ti, V, Fe, Mo, Zr, and Tl, owing to the large compositional range of the eruptive rocks (Schuessler et al.,
The results fall into two broad categories: nuclides that show isotopic fractionation (Si, Ti, V, Zr, and Fe) and those that do not (Li, Mo, and Tl). A crustal melting model for the origin of the dacites would require that hydration and metamorphism of the mafic crust be without any stable isotope fractionation, then partial melting of this hydrated metamorphic rock without isotopic fractionation of lithium, molybdenum, or thallium. Moreover, thallium and lithium concentrations correlate with other incompatible elements in the Hekla suite (Schuessler et al., 2009; Prytulak et al., 2017b), which would be exceedingly unlikely if hydrated crustal rocks partially melted. In contrast, iron, titanium, zirconium, and silicon isotopes change in the basalt to rhyolite sequence by an amount predicted by fractional crystallization models (Schleussler et al., 2009; Savage et al., 2011; Deng et al., 2019; Inglis et al., 2019). Vanadium isotopes also correspond to crystallization models for the basaltic andesite to andesite sequence (Prytulak et al., 2017a), but unfortunately the silicic rocks are too poor in V to measure the isotopic composition. A crustal melting model for the origin of the dacites would require that hydration and metamorphism of the mafic crust, followed by partial melting of this hydrated metamorphic rock, fractionate the stable isotopes of Fe, Ti, Zr, and Si in exactly the same magnitude as predicted for crystallization.

The case against crustal melting is further supported through examination of a nearby center whose silicic rocks have been shown convincingly to be due to anatexis. In contrast to the tight, coherent trend of incompatible elements at Hekla, compositions of Torfajökull rocks, especially the silicic ones, are strongly scattered (Figure 17; Gunnarsson et al., 1998; Zellmer et al., 2008), particularly when variations in relatively immobile incompatible elements such as Zr are compared with those of mobile ones, like Rb. Moreover, amphibole-bearing xenoliths have been described at Torfajökull (Gunnarsson et al., 1998) but have not been discovered at Hekla. Although the two volcanoes are close, they lie in different tectonic settings, and Torfajökull has a more prolonged hydrothermal and volcanic history. Nevertheless, it provides an excellent example of how crustal melting and extensive assimilation can be identified via petrologic and geochemical tools.

Constraints from Volatiles
The correlation of the maximum H$_2$O concentrations in melt inclusions and the plagioclase hygrometer with K$_2$O (Figure 10) confirms that H$_2$O is an incompatible element in the Hekla system (Moune et al., 2007; Portnyagin et al., 2012; Lucic et al., 2016). The calculated saturation pressures indicate the minimum pressure at which the inclusion was trapped by the host grain (Figures 10 and 15). The inclusions with lower H$_2$O contents are likely to have lost hydrogen via diffusion through the host as the surrounding magma degasses during ascent. Although the diffusivity of H$^+$ is poorly known for fayalitic olivine, existing data (Bucholz et al., 2013; Barth et al., 2019) suggest that the H$_2$O contents in Hekla melt inclusions could be reduced by ~50% during ascent in ~2 days (for 1020°C basaltic andesite) to ~12 days (for 870°C rhyolite; assuming a 40 µm inclusion that is 460 µm from the grain boundary along the a-axis of olivine; A. Barth, written communication, 2020). In fact, most of the melt inclusions in this study are <100 µm from the grain boundaries, thus the diffusive time scales could be far shorter (Electronic Appendix 5). The very narrow range of plagioclase compositions in rocks that have a wide range of melt inclusion H$_2$O concentrations are evidence against the measured range of the melt inclusions being due to different magmatic H$_2$O concentrations (Figure 10; Waters and Lange, 2015). The melt inclusions measured by Lucic et al. (2016) appear to be more strongly affected by degassing than other melt inclusion studies and our sample set, although this is not their interpretation.

The concentration of H$_2$O in melt inclusions in the silicic rocks and that calculated with the plagioclase hygrometer agree with H$_2$O concentrations predicted by the crystallization model but could not have been generated by the melting of crustal materials (Portnyagin et al., 2012). A typical amphibolite with a basaltic or gabbroic protolith has ~50% amphibole and thus contains ~1.0% H$_2$O. It is theoretically possible to generate a dacite with 5% H$_2$O by melting the protolith by 20% if H$_2$O is perfectly incompatible, but because amphibole is in the residue, melting would more likely require melting ~5% to create a melt with 5% H$_2$O (Jagoutz and Klein, 2018). Melting of either a Hekla-related rock or one from the Western Volcanic Zone <5% would create a melt that is enriched in incompatible trace elements many times that of Hekla dacite, and trace element ratios indicative of residual amphibole would be strongly fractionated. Similar arguments can be made against the hypothesis of melting of gabbro to create basaltic andesite magma (Lucic et al., 2016).
et al., 2016): gabbro contains too little H$_2$O to produce basaltic andesite magma with 2-3% H$_2$O, especially if it only contains the anhydrous minerals known to have crystallized from Hekla magmas and regional basalts.

The very low CO$_2$ concentrations in the melt inclusion glasses is perplexing: if one assumes a constant CO$_2$/Nb of 400 in Icelandic magmas (within the range estimated by Maclennan, 2017), then one would predict CO$_2$ concentrations of 2.6% in a typical Hekla basaltic andesite, far greater than the solubility of CO$_2$ at the conditions of magma storage and olivine growth at Hekla (calculated to be ~2500 ppm at 0.45 GPa; Shishkina et al., 2010). In other words, at any depth in the crust and well into the upper mantle, Hekla’s magmas are likely saturated with a CO$_2$-rich fluid. One way to explain the low CO$_2$ concentrations is that the basalts that are parental to Hekla basaltic andesite are created by remelting of strongly hydrated rocks in the lower crust, as hypothesized by Bindemann et al. (2008a). This hypothesis is supported by the oxygen isotope data in both the basalts and the Hekla rocks (Figure 14), which demonstrate that individual olivine grains crystallized from melts with variable oxygen isotope compositions. For the Fa-rich olivine in the rhyolite, melt inclusions could have trapped melt that had evolved by large extents of vapor-saturated fractional crystallization from less differentiated magma at > 200 MPa (Wallace, 2005), and this would have produced rhyolitic magma with little dissolved CO$_2$. The differentiation to form rhyolitic melt could also have occurred at higher pressures than this, as long as the melts ascended to ~200 MPa and crystallized their phenocrysts at that depth, consistent with experimental phase equilibrium evidence. The lack of CO$_2$ in the more mafic melt inclusions could potentially be the result of post-entrapment bubble formation (e.g., Moore et al., 2015; Wallace et al., 2015), as bubbles are common in the melt inclusions we analyzed, but we note that Lucic et al. (2016) only analyzed melt inclusions without vapor bubbles and found similarly low to below detection values. A final possibility is decrepitation of the melt inclusions during ascent (e.g. Maclennnan, 2017). For example, it is estimated that fewer than 10% of the melt inclusions in olivine at Laki record magmatic CO$_2$ concentrations. Further study of Hekla melt inclusions by microanalysis and Raman measurements of vapor compositions in included bubbles are called for in order to solve this CO$_2$ paradox.
The low CO$_2$ concentrations might also be explained by circulation of magma to shallower depths, degassing, and return to the lower crust, as has been proposed at Kilauea (Wallace and Anderson, 1998; Barsanti et al., 2009), although this is unlikely to occur in the more evolved rocks, which are buoyant relative to the basaltic andesite irrespective of volatile content.

Thorium Isotopic Disequilibrium and Origin of the 1104 Rhyolite

A feature of the Hekla magmatic suite is yet to be explained and is inconsistent with a crystallization origin for the silicic rocks: model ages from Th isotope analyses of the dacites and rhyolites, and their constituent zircon, are much greater than the repose times between eruptions (Thorarinsson, 1950; 1967; our Figure 2). Model ages based on whole rock Th isotopic analyses of the dacites and rhyolites are as high as 76,000 ± 14,000 years (Sigmarsson et al., 1992; Chekol et al., 2011). Model ages of zircon that crystallized from a rhyolite magma range from modern to 43,000 ± 7,000 years, and they have heterogeneous oxygen isotope compositions (Carley et al., 2011; Bindemann et al., 2012). These model ages are in stark contrast to the observation that the rhyolite erupted no more than 480-720 years after the previous explosive eruption from Hekla, which is zoned from dacite to basaltic andesite (Larsen et al., 2020). In addition to the incontrovertible evidence of zircon recycling, the relatively low ($^{238}$U/$^{232}$Th) in the rhyolites almost certainly involves removal of zircon, but the old model ages of both whole rocks and individual zircon crystals require that this occurred 10s of thousands of years before the eruptions. The effect of the incorporation of zircon into the 1158 C.E. dacite and the control on the model ages are unknown and would require detailed single-crystal study of those rocks. Simple mass balance (Figure 12) suggests that only a small fraction of the U and Th in the dacite derives from xenocrystic zircon.

As noted by Sigmarsson et al. (1992), there is little difference between thorium isotopic compositions of the basaltic andesites ($^{230}$Th/$^{232}$Th = 1.014 ± 0.020) and the andesites ($^{230}$Th/$^{232}$Th = 1.003 ± 0.020), meaning that the isotopes are consistent with the andesites originating by fractional crystallization of basaltic andesite over the course of a century. The U-Th isotopic disequilibria of the dacites and rhyolites and the model ages derived from them preclude that they could be generated by recent (<10,000 years)

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fractionation of andesite melts. Given that the strong correlations of trace elements, including U and Th (Figure 12), preclude an origin of the dacites and rhyolites by extensive assimilation or anatexis, we propose that the 1104 C.E. rhyolite was produced in a separate, crustal magma body by extensive crystal fractionation of the more commonly erupted mafic magmas from Hekla. One observation that may support the hypothesis of a separate magma body is that there is no known late-erupted basaltic andesite lava associated with the 1104 C.E. and 1158 C.E. eruptions, although we recognize that it is possible that they could be buried by younger rocks.

We propose that once produced by extensive fractionational crystallization, either the silicic magma or mostly solidified granodiorite to granite is stored over the course of >10⁴ years, conditions that are respectively referred to as “hot” and “cold” storage. The cold storage hypothesis is motivated by the commonly observed discrepancy between ²³⁸U-²³⁰Th model ages and other geochronometers (Cooper and Kent, 2014; Cooper, 2019), precisely the observation at Hekla, although in this case the discrepancy is between U-Th isotopic model ages of rocks (and zircon) and eruption interval. Although we recognize that the U-Th isotopic disequilibrium can be explained in other ways, we contend that the Hekla rhyolite must be stored in a largely molten state for at least tens of thousands of years, under hot, largely molten conditions. Beyond zircon with old model ages, there is no evidence for remobilized mush (or rock) in the 1104 C.E. rhyolite. Most systems that we are aware of that have been attributed to cold storage (e.g. Mt. Hood, Okataina, Kneeling Nun) are characterized by rocks with an abundance of phenocrysts that bear evidence of being reheated and mobilized, including reverse zoning and multiple crystal populations. Moreover, the relatively H₂O-rich compositions of Hekla’s silicic rocks are nearly impossible to have been derived by remelting of solid rock. Storage of molten magma for long periods is facilitated by the high temperatures (600-800°C) of the Iceland lower crust, as well as recharge by the steady-state basaltic andesite magma (Figure 19).

**Difference between Tephra and Lava**

The systematic difference in compositions between tephra and lava in all eruptions (Figure 11) remains enigmatic, and we do not have a satisfactory explanation for it. We believe that it is not an analytical artifact, because we analyzed the tephra and lava samples
in random order and by 3 different methods (XRF, ICP-MS, and electron microprobe). Contamination due to different grinding protocols is unlikely, as the tephra is systematically richer in Sr, Y, V, and Sc, but it never came in contact with metal during sample preparation. Analyses were completed on scoria and pumice clasts 0.5 to >2 cm in diameter, so this is not a manifestation of crystal-glass fractionation in the eruption plume, although it is possible that the rims of clasts, which we preferentially sampled, have fewer crystals than the interiors. The primary compositional differences are that the tephra is richer in both compatible (Sc, V, and Sr) and incompatible (Rb, Nb, La) elements than the lavas that erupted shortly after the tephra. In terms of major elements, the major differences are that tephra has less TiO$_2$, MgO, and P$_2$O$_5$. One possibility is that part of the magma body assimilated a rock with these characteristics, but we are not familiar with rocks that are rich in TiO$_2$, MgO, P$_2$O$_5$, Sr, Sc, V, and Rb. Alternatively, the earlier-erupted rocks may have crystallized and fractionated olivine, magnetite, and apatite (but not feldspar or clinopyroxene), or the later-erupted (lava) magma accumulated these crystals. Least-squares mass-balance on the most extreme lava-tephra pair from the 1991 eruption indicates that the major element differences are explained by 2.8% fractionation of olivine and apatite (about 2:1 proportions), thus whatever process responsible for the systematic differences between the lava and tephra had only a minor effect. The greater abundance of Sc and Rb in the tephra, however, requires up to 27% accumulation of olivine and apatite in the lava. The sample used for the mass-balance calculations (H05-02) only has 4% crystals dominated by plagioclase and clinopyroxene. The most credible explanation is that the tephra derived from a magma body that crystallized the assemblage plagioclase + clinopyroxene + olivine + magnetite + apatite ± ilmenite, and the olivine and apatite (± magnetite) was removed from this magma, perhaps by density sorting.

**Hekla’s Magmatic Plumbing**

A number of distinctive aspects of Hekla require consideration in any hypothesis that addresses its magmatic evolution. First, a multitude of geophysical and petrologic evidence indicates that inter-eruptive cooling and crystallization over the past 50 years occurs in the middle and lower crust, centered at about 20 km depth (pressures ~ 0.6 GPa). Second, deformation and seismicity related to an eruption progresses extraordinarily
rapidly, in the matter of 30 to 60 minutes (e.g., Soosalu et al., 2002, 2003, 2004; Geirsson et al., 2012). Third, chemical evidence is most consistent with fractional crystallization, and the eruptive rocks are crystal-poor (Figure 6), indicating that crystal-liquid differentiation is efficient. Fourth, the chemical processing of the magma occurs on a time scale of decades to centuries.

Because most eruptions return to a basaltic andesite composition during the waning stages of the eruptions, and basaltic andesite is the most voluminous eruptive product, there must be a large supply of this ~1020 ±20°C magma in the lower crust at Hekla. The basaltic andesites are almost certainly formed by cooling and crystallization of basaltic magma (Sigmarsson et al., 1992). The constancy of the baseline composition and deformation data suggests that basalt intrudes into the base of the crustal magmatic system. The heat input from the basaltic recharge magma must balance (at least over the course of recorded history) that lost to the surroundings to keep the magma body in a thermochemical steady-state, buffered at about 1020 ± 20°C, and the melt composition evolving to basaltic andesite. We emphasize that the Hekla plumbing system is special, in that it leads to extremely efficient crystal-liquid segregation: there is no evidence in the erupted basaltic andesites for the existence of the basaltic parents or the cumulate complement, including crystals with primitive compositions, mafic melt inclusions, and mafic enclaves (one exception in an andesite lava).

Previous studies (e.g., Thorarinsson and Sigvalason, 1971; Sigmarsson et al., 1992; Gudmundsson et al., 1992) have proposed a distinctive geometry for the Hekla magma reservoir, a tall inverted-funnel-shaped cupola that extends ~7 km vertically and ~5 km wide, located above a lower-crustal sill. At least one geodetic study (Geirsson et al., 2012) suggests a different geometry: an extended vertical reservoir, located from 21 to 10 km depth. We hypothesize that a vertically elongate tabular shape is responsible for the unique characteristics of the volcano: time-dependent, rapid (~1 century), zonation of the magma between eruption (Figure 19). In turn, the tall, thin geometry of the magma body is a manifestation of Hekla’s tectonic location at the inside corner of the intersection of the Eastern Volcanic Zone and the South Iceland Fault Zone. An elongate, upright tabular magma body is also consistent with the seismic observations (Soosalu et al, 2002; 2003; 2004). If molten magma persists between eruptions from a depth of 20 km to the shallow
crust, pre-eruptive seismicity would be minimized, because the magma is stored and ascends through a long-established, trans-crustal molten conduit.

Heat flow measurements indicate that the background geothermal gradient in the shallow crust near Hekla is approximately 100°C/km (Arnorsson et al., 2008). The gradient must be significantly lower in the deep crust, as it approaches the adiabat of the shallow mantle beneath Iceland. For example, the steady-state geotherm in the lower crust of 5 Ma oceanic lithosphere is ~45°C/km (e.g., Turcotte and Schubert, 2002). Thus, the temperature difference between basaltic andesite melt and the far-field wall-rock is likely to be between 300 and 700°C greater at the top of Hekla’s magma reservoir than it is at the bottom (recognizing that the geothermal gradient around a volcano is exceedingly complex owing to transfer of heat by precursory magma and active hydrothermal circulation).

We propose that more cooling and crystallization takes place in the upper part of the tall magma storage reservoir than in the lower section, owing to different far-field temperatures with depth, resulting in variable extents of differentiation. To test this hypothesis, we apply a simple thermal model (Figure 18), based on equation 4-113 of Turcotte and Schubert (2002), which describes conductive cooling of a semi-infinite half space. To account for latent heat of crystallization, we add an energy term: the latent heat divided by the amount of crystallization (0.3) over a specified temperature range (60°C). This simplified model assumes the magma is stagnant and develops a thermal boundary layer. If convection in the magma body was vigorous, the Nusselt number would be 2 (Marsh, 1989), and the resulting heat loss would be at twice the rate calculated below.

Our thermal model parameters are based on the 1947 C.E. eruption. Geothermometry indicates that the most silicic andesites of the 1947 eruption, which erupted explosively and first, were ~50°C cooler than the later-erupted and parental, effusive basaltic andesites. Preceding this eruption was the well documented 1845 C.E. eruption, which was similarly zoned (up to 62% SiO₂) after a 79-year hiatus. Because the 1845 eruption ended with the eruption of basaltic andesite, we assume that the conduit extending from the surface to the lower-crustal magma reservoir was completely filled with ~1010°C basaltic andesite magma at the end of that event. Geochemical mass-balance indicates that 50° of cooling corresponds to ~30% crystallization. For the thermal model
(Figure 18), we apply standard values of thermal conductivity of 1.4 J/msK, heat capacity of 1 J/K, a density of 2440 kg/m³, and heat of crystallization of 400 J/gm.

This thermal model suggests that the interior of a vertical tabular body slightly wider than 100 m (50 m half-width) would cool to the temperature of the erupted andesite with 600°C wall rocks after approximately 100 years, whereas the magma in contact with 800°C wall rock would have only cooled about 25°C during that interval (Figure 18). If the igneous body flared at depth to a half-width >100 m, as envisioned by Sigmarsson et al. (1992), cooling over the same period would be insignificant (Figure 18). The volume of the 1947 eruption has recently been recalculated by Pederson et al. (2018) to be 711 ± 122 million m³ dense rock equivalent. A tabular magma body that is 100 m wide and 7 km tall would only have to be 1 km long along strike to accommodate this volume, which is realistic given that the vents of the 1947 eruption are >10 km apart along strike (Figure 1). This thickness calculation does not account for the volume of crystals that forms as the result of cooling, which is ~30% for the differentiation of basaltic andesite to silicic andesite. The strike length therefore would have to be 1300 m, again a small proportion of the distance between the vents. If the tall magma body were more cylindrical, as proposed by Geirsson et al. (2012), the cooling times would be slightly longer.

The 1947 C.E. eruption is the best-documented eruption that followed a long hiatus and consequently is strongly zoned. In contrast, the more recent eruptions in the 20th century followed much shorter repose, between 9 and 23 years. Application of the dimensions of a tall and thin magma body cited above (1000 x 100 x 7000 m) to the cooling model indicates that heat loss over 10 years would be about 1°C (Figure 18), which would lead to inconsequential extents of fractionation, as has been observed. Observations of the repetitive eruption sequences, each eruption returning to a baseline composition of basaltic andesite, suggest that recharge after each eruption resets the magma in this vertical body to ~1010°C.

The increasing buoyancy of the melt due to crystallization is a critical factor that controls the zonation of the tall and thin Hekla magma reservoir, because the more evolved melts are progressively less dense, even though they are cooler, owing to crystallization of iron-titanium oxides and removal of iron from the melt. Basaltic andesite melt at 1010°C has a density of 2450 kg/m³, whereas the evolved andesite that erupted early in 1947 C.E.
has a density of 2350 kg/m³ at its eruption temperature of 950°C (Lange and Carmichael, 1987). Thus, if the evolved melts are made by greater extents of cooling in the upper, cooler part of the tall magma body, the reservoir will be stably stratified (Figure 19. Consequently, there will be no driving force for compositional convection, preserving the heterogeneity from top to bottom of the elongate reservoir.

Differentiation of the magma by cooling in a narrow body that extends for >10 km vertically also might explain the variable H₂O concentrations and very low CO₂ concentrations. Replenishment into this body is always of the steady-state basaltic andesite composition, and if some of this magma were stored at depths of only a few kilometers, the melt would be poor in both CO₂ and H₂O (Figure 19). Basaltic andesite magma that is progressively richer in volatiles would extend down to the steady-state reservoir in the lower crust. The magma stored in the vertical magma body supplies the magma that is variably degassed and erupts in the early explosive phase of Hekla eruptions. We note that all of our melt inclusion analyses are from the early explosive phase, a bias that is introduced because it is well known that tephra preserves melt inclusion H₂O concentrations better than lava. One way to test this hypothesis is that it predicts that melt inclusions in the more voluminous later-erupted lavas should have higher CO₂ concentrations. Evidence against this hypothesis are the mineral compositions: early erupted materials have pyroxene and plagioclase that is no different in composition than the late-erupted minerals, and crystallization at systematically different pressures and in the presence of different volatile concentrations would produce heterogenous populations.

The greatest conundrum in the evolution of Hekla’s magmas is the extraordinary efficiency of the crystal-melt segregation and the absence of evidence for a cumulate mush. Crystal-rich clots, phenocrysts out of equilibrium with the carrier melt, exotic melt inclusion compositions, and cumulate xenoliths are exceedingly rare. We speculate that Hekla’s suite of compositions might be due to the unusual magma chamber geometry. As pointed out by a number of authors (e.g., Marsh, 1989; Maclennan et al., 2001; Gudmundsson, 2012; Geist et al., 2014), most magma reservoirs emplaced in the oceanic crust are sill-like, especially those at shallow levels, and that geometry may be more conducive to remobilization of crystal-rich mush. Some argue that the advance of a solidification front is more rapid than the gravitational separation of residual liquids in a
sill (Marsh, 2006; 2013), although this principle is controversial (e.g., Latypovov et al., 2015; Bachmann and Huber, 2018). Owing to the dominance of horizontally extensive bodies (i.e., sill-like reservoirs), most modern work on crystal-liquid segregation considers mushy and crystal-settling systems dominated by gravitational forces (e.g., Bachmann and Huber, 2018). In contrast, there has been little research into crystallization on the sides of tall magma bodies since McBirney et al. (1985). In fact, we envision the differentiation in the Hekla magma body to be much like that invoked by McBirney et al. (1985), although we attribute the accumulation of silicic melt at the top of the system to result from crystallization in the presence of a geothermal gradient as opposed to buoyant boundary layers of melt rising along the sidewall.

CONCLUSIONS

On the basis of our detailed analysis of Hekla materials, the preponderance of evidence is that the sequence of magmas ranging from 54 to 70% SiO$_2$ at Hekla volcano results from fractional crystallization, with at most minor contributions from crustal melting or assimilation of crustal materials. On the basis of evidence from zoned eruptions, most of the range of magmatic compositions must exist contemporaneously in the plumbing system at any given time. Intra-eruption mixing is rare, suggesting that the eruptive sequence taps a stably stratified plumbing system. The systematic relationship between repose time and the extent of evolution is remarkable and explained by the marginal cooling of a tall, thin magma body that is surrounded by wall rock that is hundreds of degrees cooler at its top than its base.

The strong correlations between trace elements of different mobilities during hydrothermal and metamorphic processes in the Hekla suite, along with the incompatible behavior of H$_2$O, argue strongly against significant crustal interaction in the basaltic andesite to rhyolite compositional spectrum. The basaltic andesite-to-andesite sequence develops over the course of a century in the tabular conduit that extends from the lower to middle crust, perhaps more than 10 km high. Actinide-series disequilibria suggest that the rhyolite developed in a separate magma body, one that also evolved by crystallization, but for a protracted time (>50,000 years). The depth at which the silicic magma is stored is
poorly constrained, beyond that H$_2$O-rich vapor is saturated in the melt at 7 km depth, providing a minimum estimate. The low CO$_2$ concentrations in melt inclusions suggest, however, that the silicic magma has undergone substantial degassing in the middle to upper crust. The dacites that followed the eruption of the rhyolites by 54 years contain abundant crystals inherited from the crystal mush complement to the rhyolites and one of the very few candidates for hybridization within the Hekla system.

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**FIGURE CAPTIONS**

**Figure 1:** Geologic map of Hekla showing the distribution of historical lavas, after Höskuldsson et al. (2007). Inset shows the regional tectonic setting of Hekla, at the intersection of the left-lateral South Iceland Seismic Zone and the Eastern Rift Zone.

**Figure 2:** Data from this paper overlying diagram constructed by Thorarinsson (1967; dashed line), showing that the extent of differentiation and compositional zoning within individual eruptions is dictated by the repose between eruptions.

**Figure 3:** Major element variation diagrams show the strongly coherent variation of the historical Hekla suite. Uncertainty is reported in Electronic Appendix 2 and is smaller than the symbols used.

**Figure 4:** The compositions of matrix glasses and glassy melt inclusions compared to those of bulk-rock analyses. A. Most tephra glass is slightly richer in SiO$_2$ than whole rock compositions. Melt inclusions in the basaltic andesites are both more mafic and more evolved, whereas those in the andesites and rhyolites are similar to the compositions of the matrix glass. B-D. Tephra glass and melt inclusions parallel the trend of the bulk rocks, except some tephra glasses are poor in Al$_2$O$_3$ owing to feldspar microlites, and some melt inclusions are poor in MgO owing to olivine crystallization.
**Figure 5:** Compositions of rocks, matrix glasses, melt inclusions, and mineral phases change consistently over the course of the 1947 eruption. Horizontal lines indicate analytical uncertainty. Eruptive order is lowest tephra, middle tephra, upper tephra, main lava phase, waning lava phase. Analytical precision is indicated by horizontal bars on each of the plots where it is greater than the size of the symbols.

**Figure 6:** The crystallinity of Hekla lavas is strongly related to rock composition, with a crystal content minimum observed in the silicic andesite. Crystal content of the 1104 C.E. rhyolite is from Sigvaldason (1974). Dacites are the most crystal-rich rocks and contain a large proportion of xenocrysts.

**Figure 7:** A. Olivine core (solid symbols) and rim (open symbols compositions correlate strongly with whole rock and glass Mg#. Dotted lines show $K_{\text{Fe/Mg}}$ between olivine and melt = 0.41 ± 0.3. Average matrix-glass and phenocryst-rim data also shown for available samples. B. Zoning profiles in olivine phenocrysts from andesites, dacites, and rhyolites. C. Olivine zoning profiles from all phases of the zoned 1947 eruption.

**Figure 8:** A. Pyroxene compositions plotted against whole rock Mg#. Dotted lines show $K_{\text{Fe/Mg}}$ between pyroxene and melt = 0.20 to 0.36, indicating approximate equilibrium compositions. The set of pyroxenes from the 1766 andesite are thought to be inherited from a dacitic magma (Mg# ~14). B. Representative zoning profiles of clinopyroxene phenocrysts from the silicic rocks. The 1158 C.E. dacite contains phenocrysts with both normally and reversely zoned cores. C. Pyroxene from the zoned 1947 C.E. eruption shows mostly equilibrium compositions of augite in the basaltic andesites and equilibrium cores with reversely zoned rims in the andesite from the late explosive phase of the 1947 eruption.

**Figure 9:** A. Plagioclase compositions compared to whole rock compositions, indicating strong correlation. B. Representative zoning profiles through plagioclase phenocrysts in a basaltic andesite (3 profiles), a dacite, and a rhyolite.

**Figure 10:** H$_2$O concentrations in glassy melt inclusions in olivine determined by FTIR. Line is drawn through constant H$_2$O/K$_2$O of 2.2, the average of the 10 highest measured ratios. Pressures indicate saturation of the most H$_2$O-rich compositions according to the algorithm of Papale et al. (2006). Error bars are shown for representative samples. Also shown are data from Lucic et al., 2016 (LBS) and Moune et al., (2007). Open circles are the result of the plagioclase hygrometer of Waters and Lange (2015). Downward trend for each sample at constant K$_2$O indicates H$_2$O loss from melt inclusions or entrapment at lower pressures.

**Figure 11:** Trace element concentrations determined by ICP-MS plotted against Rb, the most incompatible element in the Hekla suite. Note that linear trends of Zr, Nb, La, and Ba intersect the y-axis, indicating that element is less incompatible than Rb. Lava and tephra samples are separated by a dashed line: for many of the elements, tephra clasts have
distinct trace element compositions compared to the lava. Zirconium variations in 1104 C.E. lavas indicate zircon saturation and removal from the system.

**Figure 12:** U and Th are almost perfectly correlated in the historical Hekla suite. Note the non-zero intercept along the y-axis (U), which indicates that although the samples are colinear, the U/Th ratio changes. Laser ablation data from apatite from a basaltic andesite and two andesites are indicated. The arrow extending from the regression line shows the effect of adding 0.2% zircon (average of Carley et al., 2011 analyses) to a melt that lies on the regression line. Xenocrystic zircon explains the anomalous composition of the dacite. Likewise, most data from the 1104 C.E. rhyolite have slightly lower U concentrations, consistent with zircon fractionation.

**Figure 13:** Rare Earth Element (REE) compositions of Hekla rocks, expressed as different ratios. (Top) Dy/Yb reflects fractionation of the middle/heavy REEs; the decrease of this ratio with evolution indicates progressively flatter heavy-REE patterns. Dy* (Davidson et al., 2013) compares the estimate of Dy concentration from interpolation between La and Yb to the actual measured Dy content; values >1 indicate concave-down patterns, whereas values <1 indicate concave-up patterns. Hekla REE patterns become increasingly concave-up with differentiation, owing to the crystallization of apatite. (Bottom) REE patterns also become progressively steeper with evolution.

**Figure 14:** Oxygen isotope ratios in olivine crystals separated from Hekla rocks and basalts erupted in the vicinity of Hekla compared to bulk-rock data of Sigmarsson et al. (1992). Large symbols indicate composites of 2-14 grains; smaller symbols are single-grain analyses. Illustrated uncertainty is 2 * sigma. Arrow shows predicted trend for olivine due to fractional crystallization: although melt δ18O increases due to Fe-Ti oxide fractionation, the fractionation factor between olivine and silicic melt is larger in magnitude.

**Figure 15:** Pressure-temperature phase diagram constructed for the Hekla system: BA = basaltic andesite, A = andesite, D = dacite, and R = rhyolite. Vertical solid lines show range of temperatures calculated from the 2-oxide (BA FeTi and R FeTi) and 2-pyroxene (BA PYX and A PYX) geothermometers. Vertical dashed lines show temperatures calculated from the apatite- (BA Ap, D Ap, and R Ap) and zircon-saturation (R Zrn) geothermometers. The liquidus curves from the experiments of Weber and Castro (2017) on a Hekla rhyolite are in the lower left. Minimum pressures are estimated from volatile saturation from the models of Papale et al. (2006) and Moore et al. (1998) (indicated by lines and boxes labeled VS P and VS M). Pressure is also estimated by application of the clinopyroxene-melt geobarometer of Putirka (2008) (gray box, top, labelled CPX-Melt for both andesite and basaltic andesite). The gray box at the top represents the depth to the magma reservoir since the 2000 C.E. eruption, from Geirsson et al., 2012). All of the thermobarometers are consistent with crystallization of the phenocrysts at this depth.

**Figure 16:** Trace element compositions and fractional crystallization models for the sequence basaltic andesite – andesite – dacite – rhyolite. (A) Basaltic andesite to andesite, applying Nielsen’s (1992) partition coefficients to the mineral assemblage and melt fraction calculated from least-squares mass-balance of the major elements. Compositions
are normalized to a primitive basaltic andesite. Note that the vertical axis is linear, not logarithmic. Propagated analytical uncertainty is ~3% RSD for most of these elements, thus smaller than the symbols, with the exception of V. B: same as above but showing fractionation model for dacite from andesite and normalized to andesite. Note the positive excursions of U, Th, and Hf, which are attributable to the xenocrystic zircon in the dacite. C: Fractionation model for dacite to rhyolite, applying the partition coefficients of Druitt and Bacon (1988). Zircon abundance is estimated by mass balance of zirconium.

**Figure 17:** Comparison of Hekla trace element variations to those of a volcano where extensive crustal contributions have been documented, Torfajökull, and Hekla. Torfajökull data are compiled from the GeoROC database ([http://georoc.mpch-mainz.gwdg.de/georoc/](http://georoc.mpch-mainz.gwdg.de/georoc/)). We attribute the scatter in the Torfajökull data to contributions of crustal rocks that have been altered by hydrothermal activity and metamorphism, then fractionated by partial melting in the presence of amphibole and other metamorphic phases.

**Figure 18:** Cooling model for vertical tabular magma reservoirs of various half-widths in the presence of wall rocks at different temperatures. Equation 4-113 of Turcotte and Schubert (2002) for conductive cooling of a semi-infinite half space is applied, with the modification that a term is added to the heat capacity to account for the latent heat of crystallization. Model curves are for different dike half-thicknesses and far-field wall-rock temperatures ranging from 400 to 800 °C. Horizontal lines indicate temperature estimates for basaltic andesite and andesite. Vertical line indicates the typical hiatus observed at Hekla preceding eruptions that first erupt andesite; the intersection of this line with the cooling curves indicates that andesite can be made from a cooling basaltic andesite dike that is 100 m wide in 100 years, if the background geothermal gradient is 500 to 600°C over the height of the magma body.

**Figure 19:** Conceptual cross section of the Hekla magmatic plumbing system, drawn perpendicular to the NE-SW strike of the volcano’s fissure system and using the same scale vertically and horizontally. Left panel shows the thermal and compositional evolution after 20 years of repose. Right panel shows the thermal and compositional evolution after a century of repose. The geothermal gradient is such that the far-field temperature is 400° near the top of the system and 800° near the base. The thin body extending from the lower crustal magma reservoir is only 100 m wide, and the inset shows it expanded 20x in the horizontal. This body barely cools in 20 years, but over a century its upper segment cools 65°, and the melt evolves from basaltic andesite (red) to andesite (purple shades). The rhyolite from the 1104 C.E. and dacite from the 1158 C.E. eruptions derived by fractional crystallization of basaltic andesite, but in an independent magma body that is molten for >10,000 years. Its location in the crust is poorly constrained but > 6 km depth.

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Figure 1: Geologic map of Hekla showing the distribution of historical lavas, after Höskuldsson et al. (2007). Inset shows the regional tectonic setting of Hekla, at the intersection of the left-lateral South Iceland Seismic Zone and the Eastern Rift Zone.

279x215mm (300 x 300 DPI)
Figure 2: Data from this paper overlying diagram constructed by Thorarinsson (1967; dashed line), showing that the extent of differentiation and compositional zoning within individual eruptions is dictated by the repose between eruptions.

215x279mm (600 x 600 DPI)
Figure 3: Major element variation diagrams show the strongly coherent variation of the historical Hekla suite. Uncertainty is reported in Electronic Appendix 2 and is smaller than the symbols used.

215x279mm (300 x 300 DPI)
Figure 4: The compositions of matrix glasses and glassy melt inclusions compared to those of bulk-rock analyses. A. Most tephra glass is slightly richer in SiO2 than whole rock compositions. Melt inclusions in the basaltic andesites are both more mafic and more evolved, whereas those in the andesites and rhyolites are similar to the compositions of the matrix glass. B-D. Tephra glass and melt inclusions parallel the trend of the bulk rocks, except some tephra glasses are poor in Al2O3 owing to feldspar microlites, and some melt inclusions are poor in MgO owing to olivine crystallization.
Figure 5: Compositions of rocks, matrix glasses, melt inclusions, and mineral phases change consistently over the course of the 1947 eruption. Horizontal lines indicate analytical uncertainty. Eruptive order is lowest tephra, middle tephra, upper tephra, main lava phase, waning lava phase. Analytical precision is indicated by horizontal bars on each of the plots where it is greater than the size of the symbols.
Figure 6: The crystallinity of Hekla lavas is strongly related to rock composition, with a crystal content minimum observed in the silicic andesite. Crystal content of the 1104 C.E. rhyolite is from Sigvaldason (1974). Dacites are the most crystal-rich rocks and contain a large proportion of xenocrysts.
Figure 7: A. Olivine core (solid symbols) and rim (open symbols) compositions correlate strongly with whole rock and glass Mg#. Dotted lines show KDFe/Mg between olivine and melt = 0.41 ± 0.3. Average matrix-glass and phenocryst-rim data also shown for available samples. B. Zoning profiles in olivine phenocrysts from andesites, dacites, and rhyolites. C. Olivine zoning profiles from all phases of the zoned 1947 eruption.
Figure 8: A. Pyroxene compositions plotted against whole rock Mg#. Dotted lines show KDFe/Mg between pyroxene and melt = 0.20 to 0.36, indicating approximate equilibrium compositions. The set of pyroxenes from the 1766 andesite are thought to be inherited from a dacitic magma (Mg# ~14). B. Representative zoning profiles of clinopyroxene phenocrysts from the silicic rocks. The 1158 C.E. dacite contains phenocrysts with both normally and reversely zoned cores. C. Pyroxene from the zoned 1947 C.E. eruption shows mostly equilibrium compositions of augite in the basaltic andesites and equilibrium cores with reversely zoned rims in the andesite from the late explosive phase of the 1947 eruption.
Figure 9: A. Plagioclase compositions compared to whole rock compositions, indicating strong correlation. B. Representative zoning profiles through plagioclase phenocrysts in a basaltic andesite (3 profiles), a dacite, and a rhyolite.

215x279mm (600 x 600 DPI)
Figure 10: H2O concentrations in glassy melt inclusions in olivine determined by FTIR. Line is drawn through constant H2O/K2O of 2.2, the average of the 10 highest measured ratios. Pressures indicate saturation of the most H2O-rich compositions according to the algorithm of Papale et al. (2006). Error bars are shown for representative samples. Also shown are data from Lucic et al., 2016 (LBS) and Moune et al., (2007). Open circles are the result of the plagioclase hygrometer of Waters and Lange (2015). Downward trend for each sample at constant K2O indicates H2O loss from melt inclusions or entrapment at lower pressures.
Figure 11: Trace element concentrations determined by ICP-MS plotted against Rb, the most incompatible element in the Hekla suite. Note that linear trends of Zr, Nb, La, and Ba intersect the y-axis, indicating that element is less incompatible than Rb. Lava and tephra samples are separated by a dashed line: for many of the elements, tephra clasts have distinct trace element compositions compared to the lava. Zirconium variations in 1104 C.E. lavas indicate zircon saturation and removal from the system.
Figure 12: U and Th are almost perfectly correlated in the historical Hekla suite. Note the non-zero intercept along the y-axis (U), which indicates that although the samples are colinear, the U/Th ratio changes. Laser ablation data from apatite from a basaltic andesite and two andesites are indicated. The arrow extending from the regression line shows the effect of adding 0.2% zircon (average of Carley et al., 2011 analyses) to a melt that lies on the regression line. Xenocrystic zircon explains the anomalous composition of the dacite. Likewise, most data from the 1104 C.E. rhyolite have slightly lower U concentrations, consistent with zircon fractionation.

215x279mm (300 x 300 DPI)
Figure 13: Rare Earth Element (REE) compositions of Hekla rocks, expressed as different ratios. (Top) Dy/Yb reflects fractionation of the middle/heavy REEs; the decrease of this ratio with evolution indicates progressively flatter heavy-REE patterns. Dy* (Davidson et al., 2013) compares the estimate of Dy concentration from interpolation between La and Yb to the actual measured Dy content; values >1 indicate concave-down patterns, whereas values <1 indicate concave-up patterns. Hekla REE patterns become increasingly concave-up with differentiation, owing to the crystallization of apatite. (Bottom) REE patterns also become progressively steeper with evolution.
Figure 14: Oxygen isotope ratios in olivine crystals separated from Hekla rocks and basalts erupted in the vicinity of Hekla compared to bulk-rock data of Sigmarsson et al. (1992). Large symbols indicate composites of 2-14 grains; smaller symbols are single-grain analyses. Illustrated uncertainty is 2 * sigma. Arrow shows predicted trend for olivine due to fractional crystallization: although melt $\delta^{18}$O increases due to Fe-Ti oxide fractionation, the fractionation factor between olivine and silicic melt is larger in magnitude.
Pressure-temperature phase diagram constructed for the Hekla system: BA = basaltic andesite, A = andesite, D = dacite, and R = rhyolite. Vertical solid lines show range of temperatures calculated from the 2-oxide (BA FeTi and R FeTi) and 2-pyroxene (BA PYX and A PYX) geothermometers. Vertical dashed lines show temperatures calculated from the apatite- (BA Ap, D Ap, and R Ap) and zircon-saturation (R Zrn) geothermometers. The liquidus curves from the experiments of Weber and Castro (2017) on a Hekla rhyolite are in the lower left. Minimum pressures are estimated from volatile saturation from the models of Papale et al. (2006) and Moore et al. (1998) (indicated by lines and boxes labeled VS P and VS M). Pressure is also estimated by application of the clinopyroxene-melt geobarometer of Putirka (2008) (gray box, top, labelled CPX-Melt for both andesite and basaltic andesite). The gray box at the top represents the depth to the magma reservoir since the 2000 C.E. eruption, from Geirsson et al., 2012). All of the thermobarometers are consistent with crystallization of the phenocrysts at this depth.
Figure 16: Trace element compositions and fractional crystallization models for the sequence basaltic andesite – andesite – dacite – rhyolite. (A) Basaltic andesite to andesite, applying Nielsen’s (1992) partition coefficients to the mineral assemblage and melt fraction calculated from least-squares mass-balance of the major elements. Compositions are normalized to a primitive basaltic andesite. Note that the vertical axis is linear, not logarithmic. Propagated analytical uncertainty is ~3% RSD for most of these elements, thus smaller than the symbols, with the exception of V. B: same as above but showing fractionation model for dacite from andesite and normalized to andesite. Note the positive excursions of U, Th, and Hf, which are attributable to the xenocrystic zircon in the dacite. C: Fractionation model for dacite to rhyolite, applying the partition coefficients of Druitt and Bacon (1988). Zircon abundance is estimated by mass balance of zirconium.
Figure 17: Comparison of Hekla trace element variations to those of a volcano where extensive crustal contributions have been documented, Torfajökull, and Hekla. Torfajökull data are compiled from the GeoROC database (http://georoc.mpch-mainz.gwdg.de/georoc/). We attribute the scatter in the Torfajökull data to contributions of crustal rocks that have been altered by hydrothermal activity and metamorphism, then fractionated by partial melting in the presence of amphibole and other metamorphic phases.
Cooling model for vertical tabular magma reservoirs of various half-widths in the presence of wall rocks at different temperatures. Equation 4-113 of Turcotte and Schubert (2002) for conductive cooling of a semi-infinite half space is applied, with the modification that a term is added to the heat capacity to account for the latent heat of crystallization. Model curves are for different dike half-thicknesses and far-field wall-rock temperatures ranging from 400 to 800 °C. Horizontal lines indicate temperature estimates for basaltic andesite and andesite. Vertical line indicates the typical hiatus observed at Hekla preceding eruptions that first erupt andesite; the intersection of this line with the cooling curves indicates that andesite can be made from a cooling basaltic andesite dike that is 100 m wide in 100 years, if the background geothermal gradient is 500 to 600°C over the height of the magma body.
Conceptual cross section of the Hekla magmatic plumbing system, drawn perpendicular to the NE-SW strike of the volcano’s fissure system and using the same scale vertically and horizontally. Left panel shows the thermal and compositional evolution after 20 years of repose. Right panel shows the thermal and compositional evolution after a century of repose. The geothermal gradient is such that the far-field temperature is 400° near the top of the system and 800° near the base. The thin body extending from the lower crustal magma reservoir is only 100 m wide, and the inset shows it expanded 20x in the horizontal. This body barely cools in 20 years, but over a century its upper segment cools 65°, and the melt evolves from basaltic andesite (red) to andesite (purple shades). The rhyolite from the 1104 C.E. and dacite from the 1158 C.E. eruptions derived by fractional crystallization of basaltic andesite, but in an independent magma body that is molten for >10,000 years. Its location in the crust is poorly constrained but >6 km depth.
