

1 Long or short silicic magma residence time beneath Hekla 2 volcano, Iceland?

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10 11 12 **Abstract**

13 Timescales of magma transfer and differentiation processes can be estimated when the
14 magma differentiation mechanism is known. When conventional major- and trace-element
15 analyses fail to distinguish between various processes of magma differentiation, isotope
16 compositions can be useful. Lower Th isotope ratios in silicic relative to basaltic magmas at a
17 given volcano, could either result from magma storage over a period of several tens of
18 thousands of years of if the differentiation process was fractional crystallisation alone, or from
19 crustal anatexis on a much shorter timescale. Recently mapped bimodal tephra layers from
20 Mt. Hekla, Iceland, confirm lower (²³⁰Th/²³²Th) and higher Th/U in silicic versus mafic
21 magmas. Higher Th/U has been taken to indicate either apatite fractionation or partial crustal
22 melting. In-situ trace element analysis of apatite and the enveloping glass in basaltic andesite,
23 dacite and rhyolite was undertaken to examine its capacity to fractionate trace elements and
24 their ratios. Both Th and U are compatible in apatite with a partition coefficient ratio D^U/DTh
25 of 1. Hence, apatite crystallization and separation from the melt has a negligible effect on
26 Th/U in Hekla magmas. Partial melting of hydrothermally altered crust remains the preferred
27 mechanism for producing silicic melt beneath Hekla. Ten to twenty percent partial melting of
28 metabasaltic crust with 0.4-1.2 wt% H₂O produce dacite magma with 4-6% water. Absence of
29 low δ¹⁸O values in Hekla magmas compared to silicic magmas of the rift-zones suggests mild
30 hydration of the hydrothermally altered crust. Silicic magma formation, storage,
31 differentiation and eruption at Hekla occurred over a timescale of less than a few centuries.
32 Decreasing production of rhyolite and dacite during the Holocene lifetime of Hekla suggests
33 changes in the crustal magma source and readjustment of the magma system with time.

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38 **Keywords** Magma differentiation timescales, tephra, apatite, partition coefficients, Th/U

1 **Introduction**

2 An important research goal for better understanding volcanic behaviour is the evaluation of
3 how fast magma differentiates at depth and how quickly it ascends through the crust. The
4 timescales of magma transfer after a mixing event of two magma types, or thermal
5 perturbations, is often recorded in mineral zonation (e.g. Costa et al. 2020 and references
6 therein) whereas zircon-ages suggest several order of magnitude longer timescales (e.g.
7 Cooper & Reed 2008). Information on the timescale of magma differentiation derived from
8 U-series disequilibria on whole-rocks depends on a good understanding of the magma
9 fractionation process(es) and how different elements of the U-series respond (e.g.
10 Condomines et al. 2003). The occurrence of low Th isotope ratio in more silicic magmas from
11 several active volcanoes such as Teide (Tenerife, Canary Islands), Snæfellsjökull and Hekla
12 volcanoes (Iceland; Fig. 1a) have been interpreted as reflecting several tens to hundreds of
13 thousands of years of closed- or open-system fractional crystallisation of mafic magma before
14 the eruptions (Hawkesworth et al. 2000; Kokfelt et al. 2009; Chekol et al. 2011). Similarly,
15 ^{238}U - ^{230}Th radioactive equilibrium in continental rhyolites from North America were
16 explained by inferring a crustal residence time of $> 10^5$ years (Reagan et al. 2003). These long
17 timescales have also been suggested from zircon compositions in rhyolite at the Yellowstone
18 caldera (Stelten et al. 2015) and in a silicic tephra from Hekla (Carley et al. 2011). In marked
19 contrast, the complete magma suite from basalt to rhyolite of Hekla volcano would have been
20 produced over a period that was one or two orders of magnitude shorter (hundreds to
21 thousands of years) if the silicic magma result from crustal anatexis (Sigmarsson et al. 1992).
22 Therefore, estimations of time elapsed from magma generation to eruption depend on a
23 detailed knowledge of the magma generation, and thus of the differentiation mechanisms.

24 Magma differentiation processes generating silicic melt have been debated for a long time.
25 Fractional crystallisation, assimilation fractional crystallisation (AFC), melting of crustal
26 silicic segregations or fluid-absent melting of hydrated metabasalts (amphibolites) have been
27 proposed to explain the abundance of silicic magmas in Iceland (see review by Jónasson 2007
28 and references therein). All of these processes have been shown to be capable of producing
29 silicic melts with the appropriate major-element composition, though their efficiency most
30 likely depends on the crustal thermal regime (Martin and Sigmarsson, 2007; Schattel et al.
31 2014). Silicic magma at Askja, Krafla and Torfajökull volcanoes, all of which have developed
32 powerful geothermal systems, have different isotope ratios to contemporaneously erupted
33 basalts, and are thus ascribed to crustal melting associated with an AFC process (Nicholson et al.
34 1991; Sigmarsson et al. 1991; Gunnarsson et al. 1998; Martin and Sigmarsson 2007;
35 Zellmer et al. 2008; Chekol et al. 2011; Kuritani et al. 2011; Bindeman et al. 2012).
36 Elsewhere, such as at Hekla volcano, different magma types have less contrasting isotope
37 ratios with the exception of Th and a few heavy stable isotopes.

38 Identifying crustal melts and their proportions in mingled or mixed magma is straightforward
39 when the crust has a contrasting isotope composition compared to mantle derived basalts.
40 Such a discrimination is not straightforward for partial melts of young oceanic or oceanic
41 island crusts where classic radiogenic isotope ratios, such as those of Sr and Nd, remains
42 indistinguishable from the pristine mantle melts. This is because the low ratio of radioactive
43 versus radiogenic isotopes together with long half-life of the radioactive parent isotope lead to
44 slow production of the radiogenic daughter isotope in a crust of young age. Such is the case of
45 the young Icelandic crust. Therefore, isotopes with a short half-life (such as that of ^{230}Th with

1 a half-life of only 75 kyrs) are needed to detect contributions from young mafic crust. Lower
2 Th isotope ratio in a metabasaltic crust that is a few million years old relative to recently
3 erupted basalts is thus an excellent tracer to decipher oceanic crustal contamination (e.g.
4 Nicholson et al. 1991; Sigmarsson et al. 1991). Recently, it has been proposed that the Hekla
5 silicic magma with low Th isotope ratios resulted from fractional crystallisation of basaltic
6 parental magma and “storage in a molten stage over tens of thousands of years” before its
7 eruptions (Geist et al. 2021) rather than being the product of crustal anatexis and rapid
8 transfer to the surface (Sigmarsson et al. 1992).

9 The Hekla magma suite is frequently chosen for testing the effects of terrestrial magma
10 differentiation on heavy stable isotope composition (e.g. Tuller-Ross et al. 2019). However, a
11 lack of consensus concerning the exact differentiation mechanism producing the Hekla
12 magma suite, namely whether the suite is co-genetic or not, hampers its value for studying
13 magmatic processes. The disagreement appears to lie in the understanding of different tools
14 for identifying the differentiation processes in operation. The most important divergence in
15 view concerns the role of apatite during magma differentiation and the storage time of silicic
16 melts at depth. Can the observed increase in Th/U from basalt to rhyolite (Sigmarsson et al.
17 1992) simply be explained by extraction of apatite from the melt (Geist et al. 2021)? Or does
18 the higher Th/U in silicic Hekla magma reflect an origin from anatexis melting (Sigmarsson
19 et al. 1992)? Lower ($^{230}\text{Th}/^{232}\text{Th}$) in the silicic magma in the former case could be interpreted
20 as being the result of fractional crystallisation of basalt to produce the entire Hekla magma
21 suite, followed by a long storage time at depth (tens of thousands years). In the second case,
22 the silicic magma could have erupted shortly after anatexis with the rhyolite simply
23 originating from fractional crystallisation of the dacitic crustal melt.

24 In order to assess if apatite fractionation lowers Th/U in the derived melt, selected trace
25 element partition coefficients between minerals and melt were measured in four rock types
26 chosen to represent the Hekla magma series. Moreover, additional field and geochemical data
27 have been integrated to discuss the different hypotheses proposed for the origin of the silicic
28 magma. It is shown that apatite fractionation from Hekla magma has a negligible effect on the
29 Th/U in the fractionated melt. Furthermore, new Th isotope ratios in two-coloured tephra
30 layers confirm lower ratios in the more evolved magma. Rapid magma differentiation and
31 eruption is shown to be fully compatible with the lower Th isotope ratios in the silicic
32 magmas compared to the basalt and basaltic andesite magmas. Short timescales of magma
33 evolution concur with the well-known eruption history whereas long-term storage of silicic
34 melt somewhere beneath the volcano which is not involved in the numerous eruptions of the
35 whole magma suite, from basalt to rhyolite, is considered unlikely. Finally, the erupted
36 magma compositions are used to infer the nature of the crustal magma source and the partial
37 melting conditions during the formation of the silicic magma under Hekla.

38

39 **Hekla magmas and samples**

40 The Hekla volcanic system (Fig. 1a) is constructed on older crust at the junction between a
41 transform fault and a southward propagating rift into the metabasaltic crust (e.g. Sæmundsson
42 1979; Meyer et al. 1985, Óskarsson et al. 1982). Its basement is built up of Pleistocene
43 hyaloclastite ridges of basaltic composition and Mt. Hekla is only known to have produced
44 differentiated magma during the Holocene. Prehistoric Plinian eruptions of Hekla (Hekla-5, -4

1 and -3 eruptions) produced compositionally zoned tephra layers ranging from rhyolite at the
2 base to andesite and basaltic andesite tephra at the top, whereas basaltic andesite and basalt
3 are respectively erupted from Hekla proper and fissures around the volcano (e.g. Sigvaldason
4 1974; Larsen and Thorarinsson 1977; Jakobsson 1979; Sigmarsson et al. 1992; Sverrisdóttir
5 2007). Recent work on the prehistoric tephra stratigraphy has led to an increased
6 understanding of it (Larsen et al. 2019), yielding a better time series of the volcanic and
7 magmatic evolution of Mt. Hekla. After the Hekla-3 eruption (3000 years ago) a series of
8 two-coloured (or bimodal) tephra were erupted (Fig. 1b), followed by more mafic
9 compositions until historical times in Iceland (~ 870 CE). The first two historical eruptions
10 produced rhyodacite (Hekla-1 eruption; 1104 CE; Thorarinsson 1967) and dacite (1158 CE)
11 magmas. Since then, basaltic andesite to evolved iron-rich andesite (icelandite) have been the
12 dominant eruption products.

13 *The bimodal tephra suite*

14 The well-known silicic tephra layers from the Plinian Hekla-5, -4 and -3 eruptions (7100,
15 4250 and 3050 years ago, respectively) characterize the early history of Hekla activity (Fig.
16 2). Smaller silicic eruptions also produced compositionally zoned tephra layers such as the
17 HÖ eruption, approximately 6100 years ago (Gudmundsdóttir et al. 2011; Jónsson et al.
18 2020). Recently, Larsen et al. (2019) demonstrated a shift in the eruption mode of Hekla
19 which occurred approximately 3000 years ago, or soon after the large compositionally zoned
20 Hekla-3 Plinian eruption. From 3050 b2k (age before the year 2000 CE) the volcanic activity
21 changed from infrequent large explosive eruptions to frequent smaller eruptions until the first
22 historical eruption occurred, namely the Plinian 1104 CE, or Hekla-1, eruption. For a 600-
23 year period after the Hekla-3 eruption, nine two-coloured tephra layers were formed with a
24 volumetrically smaller light coloured lower part (evolved iron-rich andesite) and a dominant
25 dark upper unit of basaltic andesite composition (Fig. 1b). This was followed by a 500-years
26 period of andesitic production with an increase in eruption of basaltic tephra towards the end
27 of the prehistoric period prior to the Hekla-1 eruption. Eight units were selected from the four
28 largest two-coloured tephra layers, H-X (2260 b2k), H-Y (2680 b2k), H-B (2800 b2k) and H-
29 C (2840 b2k), from which cm-sized pumice fragments were collected and analysed together
30 with twenty lavas and tephra from Sigmarsson et al. (1992). The sample suite represents the
31 entire age range of products from the Hekla volcanic system.

32 *Tephra for mineral-melt partition analysis*

33 Four tephra of lapilli size were collected for in-situ measurements of selected trace element
34 partition coefficients, namely basalt scoria from the Lambafit 1913 CE crater row (Moune et
35 al. 2007), basaltic andesite from the 1970 CE craters (Baldrige et al. 1973), the 1158 CE
36 dacitic tephra (Larsen et al. 1999) and a rhyolite from the first phase of the Hekla-3 eruption
37 (Larsen and Thorarinsson 1977). The samples were chosen to represent the range in magma
38 composition of the Hekla volcanic system. They correspond to tephra of different eruption
39 types, from scoria derived from a low-energy fire fountaining basalt fissure eruption to silicic
40 pumice of a Plinian eruption, which produced cubic kilometre of tephra. All the analytical
41 methods and results are given in Supplementary Information 1 and 2, respectively.

42

43 **Results**

1 *Mineral-melt equilibrium assessment*

2 Partition coefficients, or the distribution of an element between two phases in thermodynamic
3 equilibrium, can be measured if equilibrium conditions are met. Textural equilibrium between
4 minerals and glass was examined with a scanning electron microscope (SEM) and assessed
5 based on the sharpness of the crystal faces and signs of resorption in contact with the
6 surrounding glass (see Fig.1 in Supplementary Information 1). Once textural equilibrium had
7 been established, chemical equilibrium was evaluated using existing minerals-melt
8 equilibrium relationships. For olivine-, plagioclase- and pyroxene-melt equilibrium, the
9 criteria discussed in Putirka (2008 and references therein) was employed together with that of
10 Bacon and Hirschmann (1988) for coexisting FeTi-oxides.

11 Ninety-six olivine crystals, 148 plagioclases and 117 clinopyroxenes were analysed for major
12 elements. No pyroxene was found in the 1970 basaltic andesite, but 30 FeTi-oxides could be
13 analysed. Limited chemical zonation was observed in most macrocrysts apart from the
14 plagioclase in H1970 which was normally zoned from An₅₇ to An₄₅. The mineral composition
15 is within the published range for the 1913 basalt, the 1970 basaltic andesite and the H3
16 rhyolite (Baldrige et al. 1972; Moune et al. 2007; Chekol et al. 2011; Portnyagin et al. 2012;
17 Lucic et al. 2016, Weber and Castro 2017; Geist et al. 2021). Olivine in the H1158 dacite was
18 homogeneous with a median composition of Fo_{23.6} (rim: Fo₂₃₋₂₅; core: Fo₂₃₋₂₄). In contrast,
19 both normal and reversed zoning was observed in H1158 plagioclase, ranging in composition
20 from An₅₆ to An₃₆. Similarly, clinopyroxene ranged in composition from Mg# 40-46 at the
21 rim to Mg# 39-48 in the cores (median Mg# of 44 and 40, respectively) and thus mostly
22 displayed reverse zoning.

23 *Major-element concentrations*

24 Whole-rock major element concentrations were measured in ten new tephra and lavas. The
25 results represent the melt composition because of the aphyric character of the Hekla products.
26 On the total alkali vs silica (TAS) diagram (Fig. 3) the analyses form a complete magma suite
27 when plotted together with those previously published by Sigmarsson et al. (1992), Chekov et
28 al. (2011) and Geist et al (2021). A linear correlation is observed from the more evolved
29 basalt to the rhyolite composition. In order to minimize inter-laboratory differences, only
30 whole-rock analyses from Clermont-Ferrand and Misasa laboratories are shown in Fig. 3,
31 together with the recent results of Geist et al. (2021) and the detailed basalt study of
32 Jakobsson (1979). Several Harker diagrams are displayed in Supplementary Information 3.
33 The most primitive basalts are pillow fragments from the Pleistocene hyaloclastite (Hek-4 and
34 -10; Supplementary Information 2) that forms the basement upon which Hekla volcano is
35 constructed ($\text{MgO} \leq 7.73\%$).

36 The variations in P₂O₅ concentrations with magma differentiation are shown against those of
37 MgO in figure 4 together with whole-rock lava values from the literature (Jakobsson 1979;
38 Chekol et al. 2011; Geist et al. 2021) and lapilli-sized tephra by Larsen et al. (2019) for
39 comparison. Historical basalt lavas fall on a slightly different curve compared to prehistoric
40 basalt, both of which reach the maximal P₂O₅ values of 1.4-1.6% in the relatively primitive
41 basaltic andesite of the Valagjá chasm (sample Hek-11 and Hek-11b; Table 1 in
42 Supplementary Information 2). From this peak two linear trends of falling P₂O₅ vs MgO are
43 observed. The main trend extends from the Valagjá basaltic andesite to the initial tephra of the
44 1947 eruption (sample H1947F in Supplementary Information 2 with MgO = 1.46%) and

1 olivine-hosted melt inclusions from dacite and rhyolite (Portnyagin et al. 2012; Lucic et al.
2 2016; not plotted for clarity). A secondary trend is formed by several basaltic andesites that
3 pre-date the H3 eruption (3000 a) originating from craters on the northern part of the Hekla
4 ridge and lavas close to the Older Valagjá crater.

5 *Trace-element concentrations*

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7 Increasing SiO₂ and decreasing MgO concentrations are often used as differentiation index for
8 magma suites but highly incompatible elements amplify the variability of elemental
9 concentration during magma differentiation. While SiO₂ concentrations increase from 45% to
10 75% on an anhydrous basis from basalt to rhyolite (Fig. 3), the Th concentrations increase by
11 a factor of 6 (Fig. 5a). Thorium is therefore used to measure the evolution of the magma
12 composition. Variations of TiO₂ with Th concentrations are shown in Fig. 5b. A sharp
13 increase in TiO₂ is observed with increasing Th in the basalts up to a TiO₂ concentration
14 around 5 %. From there, TiO₂ concentrations falls from basaltic andesite to dacite and
15 rhyolite.

16 Forty-eight trace elements, analysed for 20 samples representing the whole Hekla magma
17 suite, complement the trace element analyses of Sigmarsson et al. (1992) and the study of the
18 most recent Hekla lavas by Chekol et al. (2011) and Geist et al. (2021). The multi-element
19 ICP-MS analyses yield that are results coherent with the ID-MS analyses for U and Th
20 concentrations albeit less precise (see Supplementary Information 2 and 4). Uranium and
21 other large-ion lithophile element concentrations (such as those of Cs and Rb; see
22 Supplementary Information 4) form an excellent linear correlation with Th, as expressed by
23 squared correlation coefficient, R², higher than 0.96. Basalt to basaltic andesite lavas fall on a
24 line passing through the origin for light and middle Rare Earth Elements (LREE and MREE,
25 respectively) versus Th concentration diagrams. Smaller increase in LREE is observed from
26 basaltic andesite to silicic magma while MREE decrease from basaltic andesite to dacite and
27 then rise again in the most evolved rhyolite (Supplementary Information 4). Increasing Th/U
28 with differentiation (Sigmarsson et al. 1992) is confirmed by the new results (Fig. 6). The
29 change in REE occurs at a Th concentration equal to 4 ppm whereas a rise in Th/U occurs at
30 Th of 5-6 ppm. Zirconium concentration deviates from the line defined by the basalts at a
31 similar Th concentrations to those of the REE. It varies by a factor of 5 in the silicic magmas
32 while Th differs by less than a factor of 2 (Fig. 6). Evolved andesite from the bimodal tephra
33 frequently deviate from the main evolution trends on concentration plots.

34 Samarium concentration in basalts correlate linearly with that of Th with a line extending
35 from the origin towards basaltic andesite at approximately 15 ppm of Sm (Fig. 7). From there
36 a flat trend connects basaltic andesite (via andesite) with dacite/rhyolite until a final rise is
37 observed in the most evolved rhyolite of the zoned H4 eruption (H4-5). The evolved andesite
38 from the bimodal tephra layers falls well below the general trend. Basalt and basaltic andesite
39 magmas have uniform Gd/Yb of 2.45-2.70, falling to approximately 2 in the andesite (Fig. 7).
40 The lowest ratio of 1.7 is measured in HY-A, an evolved andesite, but remain constant at 1.8-
41 1.9 in silicic magma from Hekla. Silicic magma from Torfajökull, Tindfjallajökull and
42 Snæfellsjökull (Martin and Sigmarsson 2007; Zellmer et al. 2008; Kokfelt et al. 2009;
43 Tomlinson et al. 2010) have Gd/Yb ranging from 1.54 to 2.2, similar to that of Hekla.

44

1 *In-situ trace-element analyses and partition coefficients*

2 A total of 111 laser-ablation analyses were performed on mineral and glass phases in H1913,
3 H1970, H1158 and H3 tephra and 71 partition coefficients were calculated (Supplementary
4 Information 5 and 6; Fig.8). In plagioclase, Sr is the only element of those analysed that is
5 considered compatible ($D_{\text{min/melt}} > 1$), together with Eu ($D^{\text{Eu}} = 1.21$) in the silicic rocks. The
6 $D_{\text{plag-melt}}$ for Sr and Eu increase by up to an order of magnitude from the more primitive
7 H1913-Lambafit basalt ($D^{\text{Sr}} = 1.53$ and $D^{\text{Eu}} = 0.14$) to the H1158 dacite ($D^{\text{Sr}} = 3.73$ and D^{Eu}
8 $= 1.21$). In clinopyroxene, Sc, Cr, Co and Ni are compatible together with a few REE in the
9 more evolved H1158 dacite and H3 rhyolite, $D^{\text{Sm}} = 1.62-1.75$, $D^{\text{Gd}} = 1.93-1.96$ and $D^{\text{Yb}} =$
10 $1.59-1.70$. Yttrium is also compatible in clinopyroxene from H1158 and H3 with $D^{\text{Y}} = 1.57-$
11 1.66 . None of the elements measured are compatible in olivine, and the highest D calculated
12 is that of Sc (~ 0.5).

13 Primary emphasis was put on measuring the partition coefficients for apatite-melt pairs in
14 Hekla and the mean D is found to be consistent from one rock type to another (Supplementary
15 Information 5; Fig. 8a). Rubidium, Nb and Ba are incompatible in apatite, as is Zr to a lesser
16 degree. The highest partition coefficients are observed for the MREE (Nd, Gd, Sm, Pr) then
17 the LREE (La, Ce) and Y with the HREE (Yb and Lu) having the lowest D. Strontium has a
18 somewhat lower D but Th and U are equally compatible in the apatite with $D^{\text{ap-melt}}$ ranging
19 from approximately 1 in the basaltic andesite [$D(\text{Th}) = 0.80 \pm 0.16$ and $D(\text{U}) = 0.95 \pm 0.22$
20 (2σ)] to 2 in the rhyolite [$D(\text{Th}) = 1.9 \pm 0.4$ and $D(\text{U}) = 1.9 \pm 0.4$ (2σ)]. The $D^{\text{apt-melt}}$ of this
21 study agree with experimental results (Watson and Green 1981; Klemme and Dalpé 2003;
22 Prowatke and Klemme 2006; Fig. 8b). It is noteworthy that there is again a consistent increase
23 in $D^{\text{apt-melt}}$ from the basaltic andesite through the dacite to the rhyolite by a factor of
24 approximately 3 for each element. Apatite has D^{U} and D^{Th} approaching a value of 2 and the
25 ratio of D^{Th} over D^{U} is always within error of 1 (Table 1).

26 *Thorium isotope ratios, Th/U and ^{230}Th - ^{238}U disequilibria*

27 The Th/U of 3.2 in Recent Hekla basalts and basaltic andesite contrasts with increasing ratio
28 of 3.4 in the dacite and 3.6-3.7 in the rhyolite. Zircon fractionation and accumulation
29 observed in the zoned H4 eruption caused large variations in Th/U (Sigmarsson et al. 1992),
30 whereas relatively high ratios are measured in two of the bimodal andesites (Fig.6b).

31 Four pairs of evolved andesite or basaltic andesite from the sequence of bimodal tephra layers
32 yielded significantly lower ($^{230}\text{Th}/^{232}\text{Th}$) and higher Th/U in the silicic part (Fig. 9 and
33 Supplementary Information 2). As an example the light-coloured (Fig. 1b) silicic part of the
34 HY tephra layer has ($^{230}\text{Th}/^{232}\text{Th}$) and Th/U of 0.936 (± 0.008 ; 2SD) and 3.44 ($\pm 1\%$),
35 respectively, compared to 1.030 (± 0.014 ; 2SD) and 3.25 in the darker part. Two magma types
36 of a contrasting composition are thus erupted in a single eruption. The evolved andesites of
37 the bimodal tephra have relatively low ($^{230}\text{Th}/^{232}\text{Th}$) for a given Th concentration (Fig. 9).
38 Moreover, these evolved andesites have significantly lower ^{230}Th -excess over ^{238}U than
39 dacites or rhyolites from Hekla (Fig. 10). The new results confirm the already observed
40 difference between silicic and more mafic magmas from Hekla (Sigmarsson et al. 1992).

41

42

1 **Discussion**

2 *The Hekla magma suite revisited*

3 Recently, studies of “non-traditional” stable isotopes of a number of elements have used
4 Hekla volcano as a case study for evaluating the effect of magma differentiation on the
5 systematics of a given isotope. Clearly, understanding of terrestrial fractionation is needed
6 before the physical and chemical processes occurring on a planetary scale can be discussed.
7 While isotopes of several elements either show no isotope fractionation at Hekla (e.g. Zn, Mo
8 and K; Chen et al. 2013; Yang et al. 2015; Tuller-Ross et al. 2019), or non-systematic
9 variations (e.g. Li and Tl isotopes; Schuessler et al. 2009; Prytulak et al. 2017a), other isotope
10 systems (Fe, Si, V, Ti and Zr) display clear evolutionary trends (Schuessler et al. 2009;
11 Savage et al. 2011; Prytulak et al. 2017b; Deng et al. 2018; Inglis et al., 2019). The heavier
12 Fe, V, Ti and Si isotope ratios with magma differentiation have been suggested to reflect
13 either Rayleigh fractionation, redox variations (with FeTi-oxide fractionating Fe, V and Ti
14 isotopes) or thermogravitational differentiation (e.g. Zambardi et al. 2014 and references
15 therein), whereas the possibility of crustal contamination is rarely explored. Similarly,
16 conflicting interpretations have arisen with respect to Zr isotope results in Hekla rocks, with
17 either isotopically light zircon being extracted during magmatic differentiation (Inglis et al.
18 2019) or isotopically heavy Zr being inherited from crustal anatexis (Ibañez-Mejia and Tissot
19 2019). The latter proposition is in accordance with the interpretation of B isotopes (Rose-
20 Koga and Sigmarsson, 2008). Therefore, the lack of consensus concerning the exact
21 differentiation mechanism operating beneath Hekla hampers accurate interpretation of several
22 heavy stable isotope systematics when applied to a magma suite.

23 Tight correlations of major- and trace-elements have been used as an argument for magma
24 differentiation by fractional crystallisation alone (e.g. Kokfelt et al. 2009; Geist et al. 2021).
25 However, melt and rhyolite-melt models of fractional crystallisation processes have failed to
26 explain several major element variations, such as those of Fe₂O₃ and K₂O (Chekol et al. 2011)
27 and Al₂O₃ and P₂O₅, in the Hekla magma suite (Geist et al. 2021). Moreover, the analytical
28 precision of major- and trace-element measurements is not always good enough to distinguish
29 between magma differentiation from a basaltic parent magma by fractional crystallization
30 alone or by assimilation fractional crystallization (AFC) process with dominant magma
31 mixing between silicic crustal melt and more mafic melts (e.g. Sigmarsson et al. 1992). The
32 complication is illustrated by the TiO₂ vs Th concentration diagram (Fig. 5b), which could be
33 used to support either interpretation. The divergent view on silicic magma formation at Hekla
34 (e.g. Sigmarsson et al. 1992; Chekol et al. 2011; Portnyagin et al. 2012; Geist et al. 2021)
35 centres around the role of apatite fractionation and its effect on trace element systematics
36 during magma differentiation together with interpretations of low Th isotope ratios in the
37 silicic magma.

38 *Apatite fractionation*

39 Highest P₂O₅ concentrations are observed in the basaltic andesite of the Valagjá lava chasm
40 (Hek-11) and associated volcanic bomb (Hek-11bis), which tephrochronology shows to be
41 older than the H3 eruption (3050 years ago) but younger than H4 (4250 years ago). Apatite
42 fractionation would cause a decrease of P₂O₅ in the derived melt, which coincides with the
43 transition from evolved basalt to basaltic andesite. However, the straight line of decreasing
44 P₂O₅ and MgO could also represent magma mixing. The decrease in P₂O₅ is different before

1 and after the H3 eruption as reflected by different slopes on Fig. 4 which might suggest either
2 more apatite fractionation in the past or different silicic end-members during magma mixing
3 or AFC-

4 Fractional crystallisation of apatite has been called upon to explain the REE patterns in
5 several magma suites such as that of Snæfellsjökull and Hekla volcanoes (Kokfelt et al. 2009;
6 Chekol et al. 2011). Apatite fractionation is known to deplete the residual melt in REE,
7 especially the MREE. Experimental studies have shown that $D^{\text{ap/melt}}$ is higher for MREE than
8 light or heavy REE (Prowatke and Klemme 2006; Fig. 8). The deflection in REE
9 concentrations when examined against an index of magma differentiation for Hekla occurs at
10 a Th concentration equal to 4 ppm (Fig. 7). It appears to coincide with apatite fractionation,
11 starting at magma composition corresponding to the basaltic andesite of Valagjá with
12 approximately 1.6 % P_2O_5 and Th close to 4 ppm (Figs. 4). However, the rise in Th/U starts
13 later in the magma evolution at a Th concentration of 5 to 6 ppm (Fig. 6b).

14 *Apatite partition coefficients*

15 The measured $D^{\text{ap-melt}}$ confirm the correlations with SiO_2 concentrations, or the melt
16 polymerization, as experimentally observed (e.g. Prowatke and Klemme, 2006; Fig. 8b). The
17 partition coefficients are thus strongly composition dependent. For example D_{Sm} between
18 clinopyroxene and melt increases by a factor of 4 from 0.41 (± 0.04 ; 2σ) in the 1913 CE
19 basalt to 1.75 (± 0.22 ; 2σ) in the H3 rhyolite and that of apatite-melt reaches values as high as
20 40 for the MREE.

21 The new partition coefficients allow better quantification of the differentiation process as
22 illustrated by the Sm vs Th diagram (Fig. 7). The best-fit linear correlation between Sm and
23 Th concentrations in basalts passes through the origin with a slope corresponding to a bulk
24 D_{Sm} of 0.002, readily explained by small proportions of clinopyroxene in the crystallising
25 assemblage. Slightly decreasing Sm concentrations from basaltic andesites (Th = 4 ppm) to
26 Hek-8 dacite (Th = 7.75 ppm) and an abrupt decrease to evolved andesite HY-A (Th = 6.68
27 ppm) would correspond to bulk D_{Sm} of 1.15 and 1.7, respectively, with an increase in apatite
28 fraction in the crystallising assemblage from 5 to 8%. The final rise in Sm from dacite to
29 rhyolite could then indicate the end of apatite fractionation. Fractional crystallisation of
30 clinopyroxene and significant fraction of apatite could thus, in principle, account for the Sm -
31 Th systematics in Hekla products, although it can by no means be used as proof for that
32 process. Indeed, the flat trend around 15 ppm Sm from basaltic andesite through andesite and
33 dacite towards rhyolite (from 4 to 10.5 ppm Th) is equally well explained by binary mixing
34 and/or AFC with a small mass of crystal fractionation relative to melt assimilation.

35 Further assessment of the fractional crystallisation mechanism for magma composition
36 varying from basaltic andesite to dacite can be obtained from the measured $D^{\text{ap-melt}}$ for Gd, Yb
37 and Th. Five percent apatite in the fractionating mineral assemblage has been suggested from
38 major-element modelling (Chekol et al. 2011). Apatite is the only mineral, in the present case,
39 that incorporates Th with a $D^{\text{ap-melt}}$ of 0.8 in the basaltic andesite. From these parameters, 50%
40 fractional crystallisation of basaltic andesite with 4 ppm Th would produce dacitic magma
41 with 7.8 ppm Th. With $D^{\text{ap-melt}}$ in the basaltic andesite of 13 for Gd and 4.9 for Yb
42 (Supplementary Information 5), and assuming an average $D^{\text{cpx-melt}}$ for basaltic andesite from
43 those measured in the basalt and dacite and further assuming that clinopyroxene makes up
44 half the mass of the fractionating phases, the Gd concentrations would decrease from 16.1 in

1 basaltic andesite (with 4 ppm Th) to 8.87 ppm, whereas Yb would only slightly decrease from
2 6.29 ppm to 6.19 ppm. This would lead to a decrease in Gd/Yb from 2.6 to 1.43, which is
3 significantly lower than that found in Hekla silicic magma (Fig. 7). In addition, the calculated
4 Gd and Yb concentrations in the hypothetical dacite are too low compared to those measured
5 in Hekla dacites, which have values of 14.6 - 15.1 ppm for Gd and 7.33 – 8.66 ppm for Yb.
6 Lowering the proportions of clinopyroxene fractionation to only 10%, for the sake of
7 argument, yields Gd and Yb concentrations (12.4 and 8.51 ppm, respectively) closer to the
8 observed values but still results in a Gd/Yb of 1.43, which is significantly lower than the
9 actual value. A simple fractional crystallization model thus underestimates MREE
10 concentrations and their ratios in the derived melt compared to those measured in Hekla
11 dacite.

12 The strongest argument against major apatite fractionation is the indistinguishable D^U and D^{Th}
13 between apatite and melt, which clearly eliminates the theory that apatite fractionation could
14 explain the higher Th/U in the silica rich magma of Hekla. Therefore, fractional crystallisation
15 of parental basalt cannot explain all the trace element systematics of the Hekla magma suite.

16 *Source of silicic Hekla magmas*

17 Silicic magmas from the rift-zone in Iceland are characterized by low $\delta^{18}O$ whereas those far
18 from the rift-axis have higher $\delta^{18}O$ (Martin and Sigmarsson 2007; Schattel et al. 2014 and
19 references therein). Less intense basaltic magmatism away from the rift-zones leads to a lower
20 geothermal gradient and cooler crust that causes incoming basalts to lose heat and
21 differentiate mainly via fractional crystallisation to produce silicic derivatives (e.g.
22 Öraefajökull and Eyjafjallajökull volcanoes; Selbekk and Trønnes 2007; Sigmarsson et al.
23 2011). In contrast, hotter crust from intense basaltic magmatism is expected at the rift-axis,
24 which is consequently easier to partially melt. High permeability of the intensively fractured
25 crust in rift-zones leads to hydrothermal alteration of the dominant basaltic lavas and intrusive
26 rocks. The oxygen isotope composition of the altered bulk crust depends on several factors
27 such as alteration temperature, secondary mineral fractionations of O-isotopes, degree of
28 hydration and degree of metamorphism together with the composition of the thermal water,
29 which in Iceland ranges from $\delta^{18}O$ of -20.8 to +2.3 ‰ (SMOW; Stefánsson et al. 2017).
30 Consequently, hydrothermally altered basalts have large range of $\delta^{18}O$ or from -11 to +6 ‰
31 (Hattori and Muehlenbachs 1982). Mount Hekla, being located on a flank zone with no
32 extensional tectonics, produces dacite and rhyolite with relatively high $\delta^{18}O$ compared to rift-
33 zone rhyolites (e.g. Muehlenbachs et al. 1974; Sigmarsson et al. 1992; Bindeman et al. 2012;
34 Geist et al. 2021). The water content of the crustal source for Hekla silicic rocks can be
35 estimated from the olivine-hosted melt inclusions of Portnyagin et al. (2012) and Lucic et al.
36 (2016). Dacite melts within Fo₂₅ olivine were shown to contain approximately 4% H₂O. If the
37 dacite were formed by 10-20% dehydration melting of amphibolite under fluid-absent
38 conditions (Thy et al. 1990; Sigmarsson et al. 1992) then the water content of that amphibolite
39 source would only have to be 0.4-0.8% since water behaves incompatibly with respect to
40 “water-free” residual minerals after amphibole breakdown. Such a low water content would
41 be expected if the hydrothermal alteration occurred at a relatively low water-rock ratio
42 (Hattori and Muehlenbachs 1982), explaining why the silicic rocks of Hekla do not have very
43 low $\delta^{18}O$.

1 Zircons extracted from a single Hekla tephra layer (most likely that of the H3 eruption) by
2 Carley et al. (2011) exhibit complex compositional zonation, have an order of magnitude
3 variation in REE concentrations and $\delta^{18}\text{O}$ that varies from 1.8 to 4.8‰ (SMOW) suggesting
4 an entrained crystal cargo of diverse origin (Bindeman et al. 2012). These zircons also span a
5 surprising range in Th isotope ratio with ($^{230}\text{Th}/^{232}\text{Th}$) ranging from 0.18 to 2.22. None of
6 these zircons have reached radioactive equilibrium between ^{238}U and ^{230}Th and must therefore
7 be younger than ~400 ka. The zircon results demonstrate important crustal heterogeneity
8 beneath Hekla and they record crystal ages of several tens to hundreds of thousands of years
9 beneath Hekla volcano, possibly from silicic segregations present within the old metabasaltic
10 crust and defrosted by thermal pulses from the southward propagating rift (Sigurdsson 1977;
11 Gunnarsson et al. 1998). At first glance, this timescale is comparable to that discussed by
12 Chekol et al. (2011) for the production of andesite from a basaltic parent. However, such a
13 long timescale is inconsistent with the eruption history of the silicic Hekla magma. It calls for
14 isolated silicic magma pockets to have remained molten over tens to hundreds of thousands of
15 years, beneath the volcano but to have escaped ascending basalt, basaltic andesite or andesite
16 during the entire volcanic history of Hekla, which appears rather fortuitous. Elsewhere,
17 zircons have been suggested to be recycled between solidification and defrosting (Cooper and
18 Kent, 2014) in mature magma systems, which is an unlikely scenario for Hekla due to its
19 aphyric magma and young age. Therefore, these heterogeneous zircons are most likely
20 xenocrysts and do not convey much information about the formation of the silicic magma.
21 The rarity of crystals in Hekla magmas and the absence of complex zoning in those present
22 (e.g. Sigvaldason 1974; Geist et al. 2021) is best explained by crustal melting and dominant
23 melt mixing at the origin of Hekla evolved magmas (Fig. 9).

24 *Radioactive disequilibrium and time constraints*

25 Decreasing ($^{230}\text{Th}/^{232}\text{Th}$) and ($^{238}\text{U}/^{232}\text{Th}$) with magma differentiation for the entire Hekla
26 magma suite is shown in Fig. 10. If the whole suite were generated by fractional
27 crystallisation alone, a period of 130 thousand years would be needed to decrease the
28 ($^{230}\text{Th}/^{232}\text{Th}$) from 1.08 (the highest ratio in basalts) to 0.94 (lowest ratio in evolved andesite).
29 The dacite and rhyolite would have to remain for $10^4 - 10^5$ years at depth, which is clearly not
30 supported by the volcanic history of Mt. Hekla. Therefore, the lower ($^{230}\text{Th}/^{232}\text{Th}$) in the
31 silicic magma of Hekla is best explained by metabasaltic crustal anatexis (Sigmarsson et al.
32 1991).

33 The tectonic situation of Hekla volcano, namely at the intersection of a transform fault and a
34 propagating rift segment (Saemundsson 1979), suggests a source of relatively fertile
35 metabasaltic crust generated and hydrothermally altered at a rift-axis, and metamorphosed as
36 a consequence of isostasy from the loading of volcanic products and plate spreading (e.g.
37 Óskarsson et al., 1982). To produce ^{230}Th excesses, or ($^{230}\text{Th}/^{238}\text{U}$) higher than 1, with partial
38 melting of metabasaltic crust, or amphibolite, a residual mineral phase during melting is
39 needed that is capable of retaining U relative to Th. The most likely candidate is a Ti-bearing
40 phase (hemoilmenite-ulvospinel solid solutions or rutile) with higher D^{U} than D^{Th} . Klemme et
41 al. (2006) measured partition coefficients between FeTi-oxides and melt and found that D^{U}
42 was less than or equal to 0.01 and that of D^{Th} an order of magnitude lower. Such low partition
43 coefficients cannot explain ($^{230}\text{Th}/^{238}\text{U}$) of 1.10-1.20 in silicic magma generated by 10 to
44 20% dehydration melting of amphibolite. The melting degree would have to be an order of
45 magnitude smaller to yield a reasonable residue with 5-20% FeTi-oxides. In addition, a melt

1 fraction of only 1% would yield high-silica rhyolite rather than dacite. Dacitic melt with
2 ($^{230}\text{Th}/^{238}\text{U}$) of 1.10 to 1.20 can be generated by 10-20% amphibolite melting if the
3 proportions of rutile in the residue are between 0.5 and 2.5%, assuming rutile-melt D^{U} of 2
4 and D^{Th} of 0. Indeed, Klemme et al. (2005) found D^{Th} to be less than 0.004 for rutile whereas
5 that of D^{U} was frequently higher than 1 and even reached 3.8 in their melting experiments.
6 Residual rutile after 20% melting of a mafic crust with Nb/La of 1.3 yields dacite with Nb/La
7 of 0.93 (assuming 0.5% rutile in the residue and D^{Nb} and D^{La} between rutile and melt of 20
8 and 0.003, respectively) as observed at Hekla (range: 0.9-1.1). Thus residual rutile is the most
9 likely cause of the 10-20% ^{230}Th excess over ^{238}U in the silicic rocks of Hekla, and their
10 formation must occur in the deepest part of the crust due to the pressure dependent stability of
11 rutile (Xiong et al. 2005; Angiboust and Harlov 2017). Melt inclusions (Portnyagin et al.
12 2012; Lucic et al. 2016) reveal H_2O contents as high as 5-6 wt %, which suggest an elevated
13 crystallisation pressure for the olivine to explain the high water solubility in the entrapped
14 silicic melt. Furthermore, high volatile solubility results in low enough viscosity and density
15 to generate the buoyant force for the crustal melt to migrate to higher level where the final
16 phase equilibria of the silica-rich magma took place (5-6.6 km according to the melting
17 experiments of Weber and Castro 2017).

18 The crustal melt produced over the Holocene cannot have had a constant composition as
19 demonstrated by variable ($^{230}\text{Th}/^{232}\text{Th}$) in the evolved andesite, dacites and rhyolites (Fig. 9).
20 Lower SiO_2 and Th concentrations as well as lower ($^{230}\text{Th}/^{232}\text{Th}$) and ($^{230}\text{Th}/^{238}\text{U}$), compared
21 to earlier formed dacites, occur in the evolved andesite of the bimodal tephra that were
22 erupted soon after the H3 eruption. Between H3 (3050 BP) and H1 (1104 CE) basaltic
23 andesite became the dominant magma type produced by Hekla (Larsen et al. 2019; Fig. 2).
24 From 3050 BP to approximately 2100 BP, ten eruptions emitted both evolved andesite and
25 basaltic andesite forming the bimodal tephra layers, with no evidence for magma mingling
26 between the two magma types (Fig. 1b). The composition of these younger evolved andesites
27 suggests a separate crustal source, with lower ($^{230}\text{Th}/^{232}\text{Th}$) in the crustal melt. That source
28 would have produced higher melt fractions (to explain lower SiO_2 and Th concentrations as
29 well as lower ^{230}Th excess over ^{238}U ; Fig. 10). The increased eruption frequency of the
30 basaltic andesite after the H3 eruption implies a greater flux of hot mafic magma into the
31 magma system and consequently increased latent heat from basalt crystallisation forming the
32 basaltic andesite. Both these factors would facilitate a larger degree of crustal melting if other
33 parameters remained constant. Melting experiments studying the phase equilibria of these
34 evolved andesites are needed to better understand whether the crustal source was at a different
35 level than for the previous magmas.

36 *Field evidence on time constraints*

37
38 The elevated H_2O and F concentrations in Hekla melts at depth (Moune et al. 2007;
39 Portnyagin et al. 2012) and the nearly aphyric character of the erupted magma does not
40 indicate a gas-filter pressing mechanism to separate silicic melt from crystals. Rather the high
41 volatile solubility suggests a relatively low viscosity of buoyant melt and both rapid and
42 efficient separation from the solid residue after partial crustal melting. The lack of magma
43 mingling in the two-coloured tephra implies separate magma storage for the bimodal magmas
44 and a short contact time between the two magmas before eruptions. The time interval between
45 these “two-magma” eruptions range from only 30 to 200 yrs. (Larsen et al. 2019) showing

1 that silicic magma was generated by crustal melting beneath Hekla on a decadal timescale and
2 erupted without accumulation into a large magma body as suggested, for example, before
3 caldera-forming eruptions, such as the Minoan eruption on Santorini (Druitt et al. 2012).
4 Further field studies are needed to establish the length of the quiescent period before the large
5 zoned Plinian eruptions H3 and H4, when compositionally stratified magma chambers were
6 emptied. Judging from the repose time preceding the last rhyodacite eruption in 1104 CE (H1
7 tephra) at least 200-250 years are needed for the accumulation of silicic magma beneath
8 Hekla before a large explosive eruption.

9 10 **Conclusion**

11 The principal conclusions of this study are the following:

- 12 • The partition coefficients of U and Th between apatite and melt are identical.
13 Therefore, apatite fractionation cannot explain the higher Th/U in the silicic magma
14 from Hekla compared to the basalt and basaltic andesite.
- 15 • The higher Th/U and lower Th isotope ratios in the silicic magma are best explained
16 by partial melting of a metabasaltic crust and rapid magma ascent through the crust.
- 17 • The diverse composition of the Hekla evolved andesite and dacite, the bimodal tephra
18 versus those produced during the large Plinian eruption, suggest a heterogeneous
19 crustal source and variable crustal melt fractions.
- 20 • The short quiescent time between the prehistoric eruptions and the lack of magma
21 mingling after the H3 eruption suggest that the time elapsed between crustal melting
22 episodes and eruptions is in the range of only 10-100 years.
- 23 • The time needed for establishing of larger magma reservoirs before the zoned Plinian
24 eruptions is only a few hundred years. The lifetime of the magma reservoirs appears to
25 have shortened with time, becoming only a few tens to a hundred years during the
26 historical period.
- 27 • Although Hekla appears to produce more mafic magma with time, the magma system
28 has not yet developed a high-level magma chamber, suggesting that partial melting of
29 the lower to middle crust has given rise to the explosive silicic magma.

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1

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17 **Table legend.**

18 Table 1. Mean partition coefficients between minerals and melt for Th and U.

19

20 **Figure legends.**

21 Fig. 1. Left panel shows a digital elevation map of Hekla volcano and the Valagjá chasm. In
22 the right panel the peculiar sharp boundary between evolved andesite and basaltic andesite is
23 shown for the HY bimodal tephra layer.

24 Fig. 2. Simplified stratigraphic column for tephra layers from Hekla volcano. Note that basalt
25 units are not shown. Historical dates are given as CE (Common Era) and prehistoric dates as
26 before 2000 CE (b2k). Different colours are shown in the on-line version with yellow as
27 rhyolite, orange as dacite and blue for andesite-basaltic andesite tephra.

28 Fig. 3. The sum of sodium and potassium oxides plotted against silica oxide concentrations
29 (TAS diagram) for Hekla lavas and tephra. Results from Jakobsson (1979), Sigmarsson et al.
30 (1992), Chekol et al. (2011) and Geist et al. (2021) are plotted together with results of this
31 study. All results have been recalculated on an anhydrous basis.

32 Fig. 4. Concentrations of P_2O_5 in historical basalts are higher than in prehistoric basalts for a
33 given MgO . In contrast, P_2O_5 decreases faster in older basaltic andesite from the northern part
34 of the Hekla ridge and craters further north along the same strike (Fig. 1) relative to basaltic
35 andesite and andesite younger than the H3 eruption (ca. 3000 a). Two double-headed arrows
36 indicate either apatite fractionation or magma mixing of basaltic andesite with differentiated
37 magma components, with the older one (between Valagjá and Older Valagjá) being of less
38 evolved composition. Results from Jakobsson (1979), Sigmarsson et al. (1992), Chekol et al.

1 (2011) and Larsen et al. (2019) are plotted together with those of this study. See text for
2 further discussion.

3 Fig. 5 a) Thorium vs SiO₂ concentrations in Hekla volcanics. The over five-fold rise in Th
4 corresponds to only a 1.6 times increase in SiO₂ concentrations demonstrating that fine details
5 of magma differentiation are better represented by Th variations than by variations in silica.
6 Only Th measured by ID-MS are plotted together with results from Sigmarsson et al. (1992;
7 filled circles) and those of Chekol et al. (2011; empty circles). b) Titanium oxide vs Th
8 concentrations in Hekla volcanics. Onset of FeTi-oxide fractionation in the basalts is marked
9 by the spike in TiO₂ concentrations. The decreasing TiO₂ with differentiation (increasing Th)
10 can be explained by either progressive fractional crystallisation or by magma mixing of
11 different components. Such a mixing line is drawn from the basaltic andesite through andesite
12 towards the dacite.

13 Fig. 6. a) Zirconium versus Th concentrations. The observed differences between the new
14 results and those of Sigmarsson et al. (1992) reflects the different analytical methods (isotope
15 dilution mass spectrometry, ICP-MS multi-element analysis and XRF on pressed pellets) used
16 in three separate laboratories. Open headed arrows depict fractional crystallisation from basalt
17 to basaltic andesite and from dacite to rhyolite. Binary magma mixing between basaltic
18 andesite and dacite is indicated by the double-headed arrow. Zircon fractionation and
19 accumulation controls the zirconium concentration in products from the H4 eruption. Also
20 notable is the peculiarity of the evolved andesite from the bimodal tephra layers (HY-A and
21 HX-A). b) Only U and Th results obtained by isotope dilution mass spectrometry are plotted.
22 The Th/U in basalt and basaltic andesite is uniform at ~3.2 up to Th~5-6 ppm, from where the
23 ratio increases to ~3.5. Redistribution of zircon in the H4 zoned eruption causes Th/U to vary
24 between 3.04 and 3.73.

25 Fig.7. Rare Earth Element variations examined with increasing magma differentiation in the
26 Hekla magma suite. Analytical errors on the REE concentrations are shown as 3%. Range of
27 Gd/Yb in Icelandic silicic magmas is shown as well. See text for further discussion.

28 Fig. 8. a) Mean partition coefficients between apatite and melt for intermediate to silicic
29 magma from Hekla. Note the consistent increase in D from basaltic andesite through dacite to
30 rhyolite. b) Increasing $D^{\text{ap-melt}}$ with silica content of the melt. Filled symbols are averages
31 from this study; associated error is in most cases smaller than the size of symbols (see
32 Supplementary Information 5). The $D^{\text{ap-melt}}$ of U and Th are within analytical error and thus
33 overlap each other. Open symbols are experimental apatite/melt partition coefficient from
34 published studies (Watson and Green 1981; Klemme and Dalpé 2003; Prowatke and Klemme
35 2006; Hammouda et al. 2010) and the calculated exponential best fits are shown as dashed
36 lines for comparison with the new results.

37 Fig. 9. Thorium isotopes in Recent Hekla magmas plotted against Th concentration and
38 $1/(^{232}\text{Th})$ (in grams per disintegration per minute (dpm)). Identical ($^{230}\text{Th}/^{232}\text{Th}$) in basalt and
39 the most primitive basaltic andesite reflects fractional crystallisation (FC). Decreasing
40 ($^{230}\text{Th}/^{232}\text{Th}$) from basaltic andesite through andesite towards dacite strongly suggests magma
41 mixing with limited crystal-melt separation causing the observed scatter. Fractional
42 crystallisation of dacite produces rhyolite. Evolved andesite from the bimodal tephra sequence
43 are explained by crustal melt of different composition than for the other dacites. See text for
44 further discussion.

1 Fig. 10. Equiline diagram schematically displaying the mantle source and the metabasaltic
2 crust in U-Th radioactive equilibrium ($(^{230}\text{Th}) = (^{238}\text{U})$) together with the approximate fields
3 for rift-zone basalts Kokfelt et al. (2006 and references therein) and partial metabasalt melts.
4 Ageing and metamorphism of basalts, produced by mantle melting and erupted in the rift-
5 zone, together with isostasy and crustal spreading form amphibolite crust that upon
6 dehydration melting produces the Hekla silicic magmas (Sigmarsson et al. 1991). The spread
7 of results for silicic magma on the diagram suggest a heterogeneous crustal source. Excess
8 ^{230}Th over ^{238}U results from dehydration crustal melting and subsequent zircon fractionation
9 as observed for the rhyolites. The maximum time needed for ^{230}Th decay alone is also
10 indicated. See text for further discussion.

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14 **Supplementary Information**

15 **1- Methods and analytical technics**

16 **2- Analytical results (Major and trace element concentrations, U and Th
17 concentrations by isotope dilution and $(^{230}\text{Th}/^{232}\text{Th})$ measurement results)**

18 **3- Harker diagrams**

19 **4- Trace element plots**

20 **5- Measured partition coefficients**

21 **6- In-situ trace element results**