Reply to comment on "Long or short silicic magma residence time beneath Hekla volcano, Iceland?" by Sigmarsson O, Bergþórsdóttir I A, Devidal J-L, Larsen G, Gannoun A.

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Abstract

We would like to thank Geist et al. (2023) for the opportunity to further discuss the arguments presented in our paper "Long or short silicic magma residence time beneath Hekla volcano, Iceland?" (Sigmarsson et al. 2022). The disagreement centres around the origin of the silicic magmas at Hekla, namely whether it is by (i) fractional crystallisation and a long crustal residence time before eruption or (ii) partial melting of altered basaltic crust and short transfer time to the surface. We disagree with the arguments presented by Geist et al. (2023) against the model for the origin of dacite at Hekla from dehydration melting of amphibolite, a model that still explains most if not all results obtained so far on the Hekla magma suite.

Introduction

The origin of silicic rocks at Hekla volcano, Iceland, has been discussed for more than a century. The German chemist Robert Wilhelm Bunsen (the one that improved the Bunsen-burner), the French mineralogist Des Cloizeaux and the Danish naturalist J. C. Schythe visited Mt. Hekla after its 1845-1846 eruption (i.e., Wentrup 2021). They proposed a classification of two rock types, a felsic and a mafic type originating from two different magma chambers (Bunsen 1851). A century later in 1947-1948, Hekla had one of its largest historical eruptions that prompted many detailed studies published by the Icelandic Science Society (the "Hekla Series"). For instance, based on Harker-diagrams, Einarsson (1950) proposed that fractional crystallization explained the compositional diversity of the tephra and lava produced. Also by combining accounts from written annals and experience drawn from the 1947-1948 eruption, Thorarinsson (1967) established the linear correlation between the length of the foregoing quiescent period and the initial SiO_2 concentrations in the first emitted tephra, however without proposing a specific mechanism for the observed relationship. The 1970 eruption (Thorarinsson and Sigvaldason 1972) drew the first attention of American researchers to Hekla with the study of Baldridge et al. (1973). They published electron-probe micro-analyses (EPMA) of Hekla products and concluded that its magma followed an evolution similar to the tholeiitic trend that had been established for the Thingmuli magma suite (Carmichael 1964). In marked contrast, Sigvaldason (1974), building upon the paper of Yoder (1973) on the concept of contemporaneous silicic and mafic magma, discussed the need for crustal origin of the more evolved rocks. By this time, all the hypotheses proposed for the origin of silicic magma at Hekla were based on major element criteria. However, major element variations cannot distinguish between a final silicic melt formed by extensive fractional crystallisation and a first melt generated by partial crustal melting. Both melts plot close to the eutectic in the "petrogenic residual system" (e.g., Johannes & Holtz 1996), where the final mineral assemblage controls the residual or the initial melt composition close to the solidus.

The utility of isotope ratios for discerning petrogenic processes and magma source compositions in Iceland was demonstrated by Muehlenbachs et al. (1974). They showed that silicic magma generally has lower $\delta^{18}O$ than basalt from the same volcano or volcanic system, later interpreted by Óskarsson et al. (1982) to reflect partial melting of the hydrated basaltic crust in amphibolite facies. However, Hekla dacite and rhyolite turned out to have similar $\delta^{18}O$ as its basaltic andesite (or icelandite), and not as low as silicic magma from the rift-zones. Soon thereafter, significantly lower ($^{230}Th/^{232}Th$) was measured in the silicic Hekla magma compared to the basalt and basaltic andesite, interpreted to reveal the crustal origin of Hekla dacite, consistent with higher Th/U in the silicic magma (Condomines et al. 1981; Sigmarsson et al. 1992). Geist et al. (2021) challenged that interpretation for Hekla silicic magma and preferred to return to the fractional crystallisation model that was first proposed by Einarsson (1950). They followed Chekol et al. (2011) by explaining higher Th/U in the silicic magma by apatite fractionation and magma dwelling timescales of tens of thousands of years beneath the volcano. How such a silicic magma chamber could have escaped all the Holocene magmatism and volcanic activity at Hekla is hard to understand. A few months later, Sigmarsson et al. (2022) published the partition coefficients of U and Th between several mineral phases and glass of basaltic andesite, dacite and rhyolite composition from Hekla. The D_U and D_{Th} between apatite and melt turned out to be within error (D_U/D_{Th}=1), a result that precludes apatite as a phase capable of fractionating the Th/U of the melt in the case of Hekla.

In the following, we will addresss all the comments by Geist et al. (2023) on our paper and demonstrate that they are logically inconsistent.

Discussion

Comment #1: Metaluminous vs peraluminous silicic magma:

Geist et al. (2023) argue that the alumnium saturation index (ASI), presumably calculated as the molar ratio $Al_2O_3/(CaO + Na_2O + K_2O)$, for glasses of amphibolite melting experiments are different from those of Hekla silicic rocks. They compare whole-rock analyses from their Hekla study (ASI < 1) with EPMA of experimental glasses (ASI > 1) obtained in diverse melting experiments of different amphibolites (see Geist et al. (2023) for references), metaluminous vs peraluminous silicic melt, respectively. Because of this difference they conclude that silicic magma of Hekla cannot be derived from partial melting of amphibolite. However, comparing whole-rock analyses to EPMA may lead to erroneous inferences. It is well known that spot analyses of Na_2O concentrations in hydrous Si-rich glasses by EPMA may underestimate the concentrations. For instance, Beard and Lofgren (1991), one of the study cited by Geist et al. (2023), estimated that the Na_2O loss during analyses of their experimental glass could have been as high as 32%. Moreover, small beam sizes may lead to overestimation of aluminium concentrations according to Acosta-Vigil et al (2003). Furthermore, the partial melting model of amphibolite discussed by Sigmarsson et al. (2022) for the generation of dacite beneath Hekla volcano is a fluid-absent, or dehydration, melting model where amphibole melts out and the residuum is free of amphibole.

Figure 1 shows the alumnium saturation index (ASI) of glass analyses of the Hekla 1104 CE pumice and melt inclusions from Geist et al. (2021) plotted against SiO_2 concentrations demonstrating the variability of ASI of the 1104 CE glass. The glass analyses of the 1104 CE pumice straddle the boundary between-per- and metaluminous devide. Similarily dehydration melting experiments of amphibolite (Beard and Lofgren 1991) with amphibole-free residuum produce dacitic melts that have both per- and metaluminous compositions. The experimental melts extensively overlap with the compositionof the silicic Hekla melt. A comparision of Hekla products with experimental results with amphibole still present should be considered irrelevant when discussing the proposed model of crustal origin of silicic melts beneath Hekla (Sigmarsson et al. 1992; 2022).

Comment #2: Torfajökull vs Hekla

Silicic magma with high $^{87}Sr/^{86}Sr$ (0.70334-0.70386) at Torfajökull (20 km east of Hekla) has been interpreted to reflect melting of compositionally evolved crustal material (with elevated Rb/Sr, e.g. Gunnarsson et al. 1998), whereas lower Sr isotope ratio at Hekla are consistent with melting of fairly young amphibolite with low Rb/Sr (Sigmarsson et al. 1992). In their comments, Geist et al. (2023) take the lower $^{87}Sr/^{86}Sr$ of Hekla rocks (0.70315), as an evidence against crustal anatexis. Such an argument would only be valid if the crust, in general, had higher Sr isotope ratio than basalt erupted around Hekla, which is unlikely for the following reasons. The $^{87}Sr/^{86}Sr$ of the basaltic crust will remain largely within the range of rift-zone basalt (where the crust is formed) because of its young age and the slow decay of ^{87}Rb generating ^{87}Sr. Therefore, partial crustal melt of amphibolite beneath Hekla will lead to silicic melts with similar or marginally higher $^{87}Sr/^{86}Sr$ compared to the basalt. The high $^{87}Sr/^{86}Sr$ at Torfajökull suggests partial crustal melts from different lithologies than amphibolite formed from rift-zone basalt, namely lithologies with elevated Rb/Sr as a magma source as discussed by Gunnarsson et al. (1998).

Comment #3: *Mobile vs immobile elements*

Geist et al. (2023) state that “mobile elements show similar variations as immobile elements …, and ratios of mobile to immobile elements in the dacites are precisely as predicted for crystallization differentiation of a basaltic andesite parent”. Such a strong statement is surprising given their earlier statement “that mineral/melt partition coefficients are uncertain”. Once again, Geist et al. (2023) prefer to compare Hekla silicic rocks with those of Torfajökull using a ratio that turns out to be indistinguishable between the two volcanos (Rb/Zr of 0.0463-0.0689 for Hekla compared to 0.028-0.232 for Torfajökull). Sigmarsson et al. (1992; 2022) concluded that dacite formation by either crustal anatexis or extreme fractional crystallisation could not be distinguished using conventional major- and trace element analyses in the case of Hekla. High-precision trace element analyses by isotope dilution mass spectrometry (with analytical errors less than 1%) are needed to unravel the natural variations such as that of Th/U for the different magma types. Fractional crystallisation alone cannot explain the increase from approximately 3.2 in basalt and basaltic andesite to 3.4 in dacite without crustal contribution. The D_U/D_{Th} indistinguishable from 1 between apatite and melt and the highly incompatible behaviour of U and Th request a magmatic process in addition to simple fractional crystallisation, namely an assimilation-fractional crystallisation (AFC) with crustal-derived dacite as an assimilant (Sigmarsson et al. 1992; 2022; Chekol et al. 2011).

Comment #4: *Absence or presence of amphibole in crustal melting residue*

Melting experiments should not be expected to mimic exactly natural compositions due to inherent experimental difficulties but rather to hint at likely magmatic processes or source rock compositions. In their effort to disprove the partial crustal melting model for the formation of Hekla dacite, Geist et al. (2023) pick results from run #1583 of Sisson et al. (2005) with 39% amphibole still in the residue, and calculate a trace element spectrum very different to those of Hekla dacite. Whether amphibole is exhausted or remains in the melting residue controls both the major- and trace element composition of the melt formed. Thy et al. (1990) showed that only dehydration-amphibolite melts with amphibole-free residuum have major-element composition comparable to silicic magmas in Iceland. Furthermore, Beard and Lofgren (1991) and Sisson et al. (2005) discussed the effect of the amphibolite source rock composition on the silicic melt produced during partial melting.

Systematic melting experiments at lower crustal conditions of amphibolite produced from the Icelandic rift-zone basalt could shed further light on Hekla dacite formation. Given the uncertainty regarding the exact source rock composition, hydrothermally altered basalt from a volcano (such as Krafla) in the middle of the rift-zone, where the crust of Iceland is being generated, must be considered a better source rock than diverse amphibolite from elsewhere in the world.

In a nutshell, calculations of trace element contents from the residue mode of melting experiments with abundant amphibole still present, have no bearing on the model for dehydration melting of amphibolite producing the silicic magma at Hekla.

Comment #5: *Hybrid origin for the andesites or not*

Geist et al. (2023) state that andesite at Hekla cannot be a hybrid between basaltic andesite and dacite melts. Figure 3 shows the Sr versus Th concentrations and a straight line representing binary mixing is drawn between basaltic andesite and dacite. Most points plot close to that line and the scatter is likely to reflect additional mineral-melt fractionation. It should be noted, however, that the compositions of the mingling endmembers can rapidly vary with time as has been observed during single eruptions, for example Eyjafjallajökull 2010 (Sigmarsson et al. 2011). Consequently, a single mixing line is not a proof for or against magma hybridisation, especially since the dacite crustal melt composition is expected to vary with time if the crustal source varies.

Comment #6: *Crystallisation differentiation or not*

Geist et al. (2023) discuss trace element modelling using their published results and come to the conclusion that fractional crystallisation (FC) can account for all trace elements. Sigmarsson et al. (1992 and 2022) stressed that, in the case of Hekla, conventional trace element analysis does not have the resolving power to distinguish between dacite melt origin by FC or partial amphibolite melting. High-precision analyses of U and Th demonstrate a significantly higher Th/U in the silicic magma of Hekla, which cannot be explained by the partion coefficients measured for basaltic andesite, dacite or rhyolite of Hekla. Once again, apatite-melt D^U/D^{Th} is indistinguishable from unity in the case of Hekla and other minerals in the basaltic andesite and the andesite have U and Th nearly perfectly incompatible.

Comment #7: *Krafla rhyolite?*

Geist et al. (2023) conclude their discussion by comparing rhyolite from Krafla volcano to silicic rocks of Hekla. In both cases, the silicic rocks have much lower ($^{230}Th/^{232}Th$) than the basaltic magma produced at both volcanoes, a fact that should not be ignored. In addition, $\delta^{18}O$ is much lower in the Krafla rhyolite than in the basalt while $^{87}Sr/^{86}Sr$ remains uniform (Nicholson et al. 1991). In both cases, the silicic magma with lower ($^{230}Th/^{232}Th$) is best explained by crustal anatexis. Ageing Th isotope ratio by tens to hundred of thousands of years in a Si-rich magma chamber beneath Hekla volcano would not explain linear decrease of ($^{230}Th/^{232}Th$) versus 1/Th shown in Fig. 4, but is fully explained by mixing of Si-rich crustal melt with incomming basaltic andesite. The extensive magmatic activity at Hekla during the Holocene would hardly escape a silicic melt waiting in a magma chamber beneath the volcano to be remobilished.

Future research needed to better understand Hekla magmatism

The general dehydration melting model with amphibolite protolith explains most silicic magma composition where the geothermal gradient of the Icelandic crust is elevated. The exact nature of the protolith composition is, however, challenging to assess although rift-zone tholeiite remains the best analogue being the most abundant rocks of the Icelandic crust. The crust is not only composed of basalt but also of an unknown proportion of silicic rocks, isostatically buried dacite-rhyolite and granite. These latter rock types may have formed by fractional crystallisation away from the rift-zones where the geothermal gradient is low, or by crustal anatexis where it is high (e.g. Martin & Sigmarsson 2007).

Sigurdsson (1977) suggested that plagiogranite melting could explain the abundance of silicic volcanic rocks in Iceland and Gunnarsson et al. (1998) used a variant of that model to account for the abundance of rhyolite at Torfajökull volcano. Hekla volcano frequently erupts a few light-coloured xenoliths of different composition than the eruptive products. Theses xenoliths have not been studied in much details yet but have been ascribed to products from Torfajökull that may underlie the young Hekla volcano (Sigvaldason 1974; Sigmarsson et al. 1992; Chekol et al. 2011; Geist et al. 2021). Similarily zircons from Hekla of heterogeneous composition (Carley et al. 2011) have been suggested to represent entrained crystal cargo of diverse origin (Bindeman et al. 2012). The zircons must be younger than 0.3 Ma, since they are in ^{238}U-^{230}Th radioactive disequilibrium, but much older than all known silicic tephra from Hekla (Carley et al. 2011). How relevant their model age is for the Hekla volcanism remains to be clarified but, in principle, they may be derived from Si-rich crustal melts of different age and origin, remobilized by the ascending Hekla magma. The presence of silicic crustal formations interbedded within the overall amphibolitic deeper crust, can also account for low $\delta^{37}Cl$ in Hekla pumice thought to represent crustal brine (Ranta et al. 2021). Indeed, almost complete melting of an old silicic protolith in U-Th radioactive equilibrium with zircon remaining in the residue withholding U relative to Th could be seen as a possible explanation for the low ($^{230}Th/^{232}Th$) in Hekla dacite-rhyolite magma. However, this possibility was rejected by Sigmarsson et al. (1992) and must be considered unlikely due to the rapid renewal of silicic magma beneath Hekla and the large Plinian eruptions forming the well-known prehistoric tephra layers.

Conclusion

In conclusion, we discuss the points criticised by Geist et al. (2023) and show that none of them provide compelling evidence against the dehydration melting of amphibolite, a model that still explains most if not all results obtained so far on the Hekla magma suite.

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Figure legends:

Figure 1. Alumnium Saturation Index (A.S.I.) versus **silica** oxide concentrations from electron microprobe glass analyses of the Hekla 1104 CE pumice (results from Geist et al. 2021). **Also plotted are experimental glass from amphibolite dehydration melting experiments with amphibole-free residuum (Beard and Lofrgren 1991). The overlap between the Hekla silicic glass and the experimental glass support the model of dehydration amphibolite melting for silicic magma at Hekla (see text for further discussion).**

Figure 2. Concentrations of U and Th measured with the isotope dilution technique demonstrating uniform Th/U from basalt to basaltic andesite due to fractional crystallisation, increase from Th/U of 3.2 to 3.4 from basaltic andesite through andesite to dacitic crustal melts and further increase in Th/U caused by zircon fractionation (further information is given in Sigmarsson et al. 1992 and 2022).

Figure 3. Strontium versus Th variation (Chekol et al. 2011; Geist et al. 2021; Sigmarsson et al. 2022).

Figure 4. Thorium isotope systematics of Hekla magma erupted last thousand years versus Th concentrations. Arrows for fractional crystallisation (FC) and assimilation fractional crystallisation (AFC; **De Paolo 1981**) with R being the ratio of crystalising mass over mass of assimilant (silicic crustal melt) with R of 0 representing a binary magma mixing. Here, the assimilant is silicic crustal melt (further details can be found in Sigmarsson et al. 1992 and 2022).

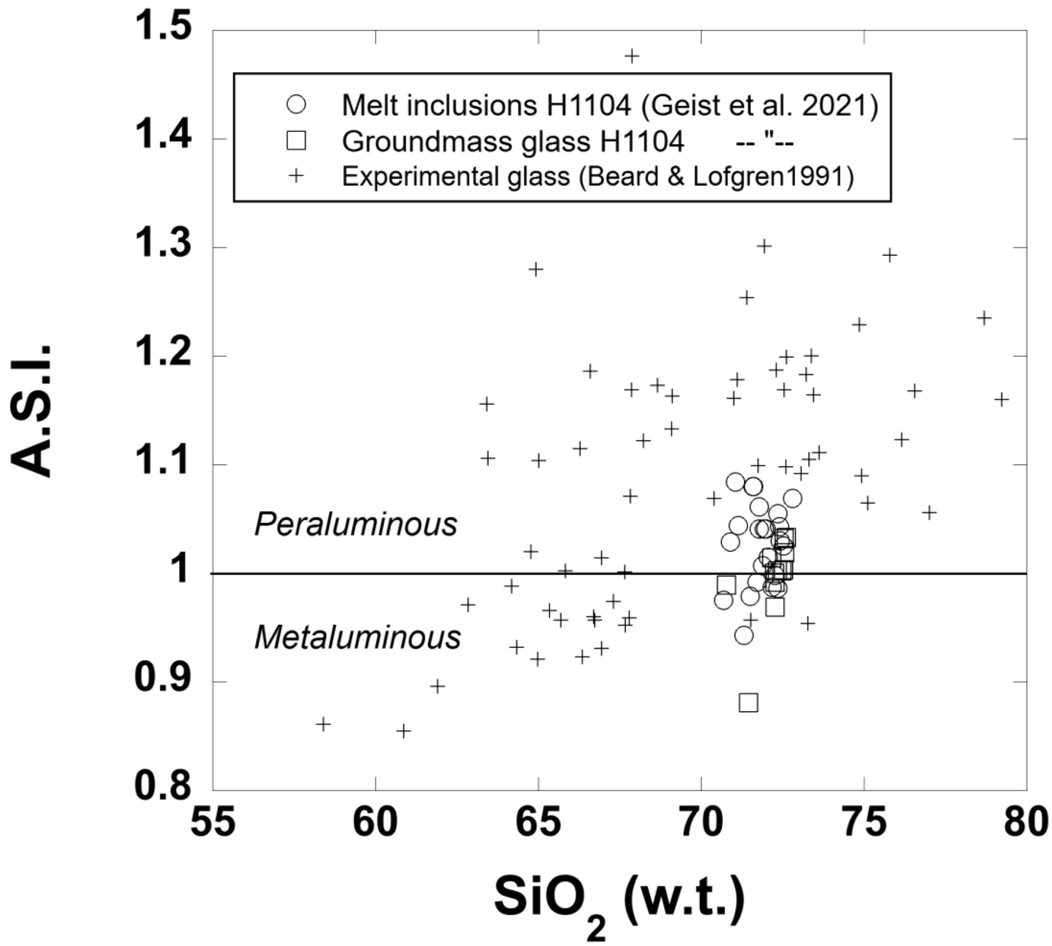

Melt inclusions H1104 (Geist et al. 2021)
Groundmass glass H1104 -- "--
Experimental glass (Beard & Lofgren1991)
Peraluminous
Metaluminous
A.S.I.
SiO2 (w.t.)
1.5
1.4
1.3
1.2
1.1
1
0.9
0.8
55
60
65
70
75
80

Figure 1.

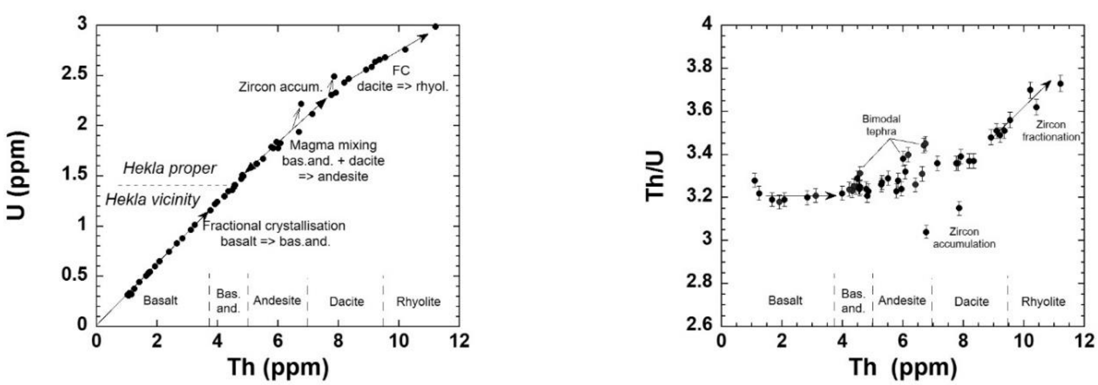
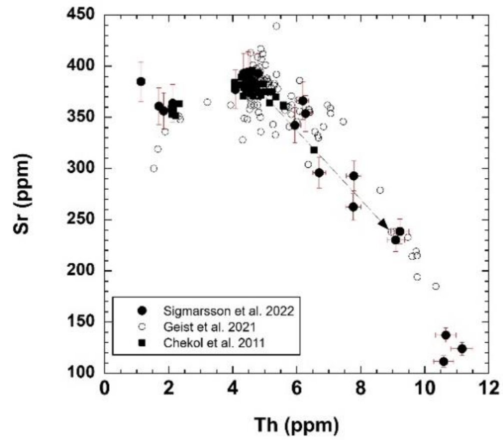
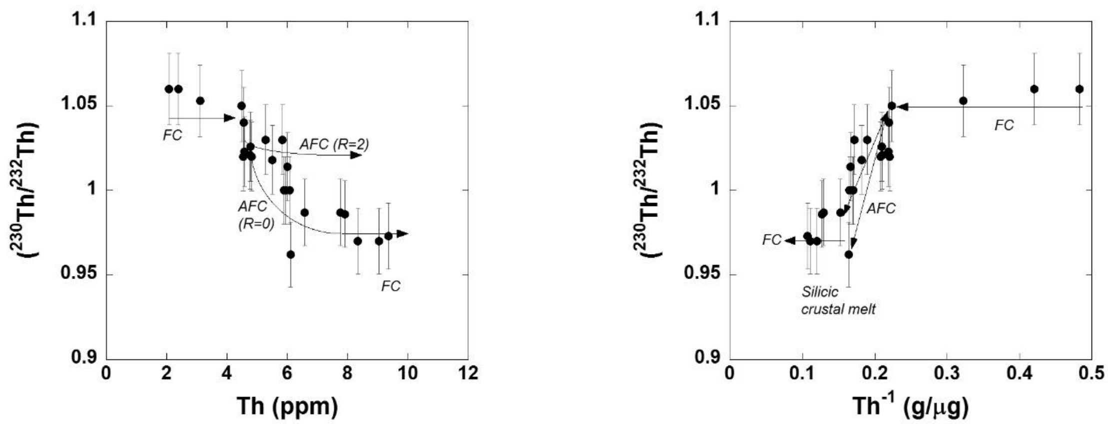

U (ppm)
Th (ppm)
Hekla proper
Hekla vicinity
Zircon accum.
FC
dacite => rhyol.
Magma mixing
bas.and. + dacite
=> andesite
Fractional crystallisation
basalt => bas.and.
Basalt
Bas. and.
Andesite
Dacite
Rhyolite
Th/U
Th (ppm)
Bimodal tephra
Zircon fractionation
Zircon accumulation
Basalt
Bas. and.
Andesite
Dacite
Rhyolite

Figure 2.



Sr (ppm)
Th (ppm)
Sigmarsson et al. 2022
Geist et al. 2021
Chekol et al. 2011
450
400
350
300
250
200
150
100
0
2
4
6
8
10
12

324 Figure 3.



325
326 Figure 4.