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Spilitization, carbonation and hematite enrichment of a metabasaltic sill at Port Ellen, Islay, Scotland

Erik Hallin

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Department of Geological Sciences
Stockholm University
SE-106 91 Stockholm

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- *Erik Hallin*

Abstract

The spatial distribution of hematite enrichment, plagioclase phenocrysts, carbonation and spilitization of one metabasaltic sill at Port Ellen, Islay, Scotland was investigated. Profiles were constructed across the metabasaltic sill along which plagioclase and hematite concentrations were counted using a mechanical grid for point counting. A potential connection between hematite enrichment and vein systems was also explored by analysing the locations of hematite crystals on two large cliff faces. Furthermore, the extent of carbonation that has affected the metabasaltic sill was measured. The field-sampling allowed for the relationship between hematite, sill-margin carbonation, veining and mineralogical zoning to be analysed.

The study found that plagioclase phenocrysts are concentrated in the sill interior as a consequence of magmatic differentiation. Hematite, however, does not show a pattern of enrichment in the margins as initially expected. It is instead strongly correlated with veins, fractures and epidote segregations, which accounted for 98 % of the hematite found on the two large cliff faces. The carbonation of the sill was found to decrease from the margins inwards. A correlation analysis found no clear correlation between hematite and the carbonated margins, supporting the finding that hematite is vein related rather than related to margin enrichment as a consequence of magmatic differentiation processes.

The result of this study indicates that hematite enrichment was likely controlled by permeability-enhanced fluid pathways rather than magmatic differentiation. The finding that hematite is closely associated with veins, fractures and epidote segregations favours a later-stage fluid intrusion event as the cause of enrichments, since the epidote segregations did not undergo porosity-enhancements before greenschist facies metamorphism. These findings are highlighting the structural controls on ore-mineral enrichment processes in a metabasaltic sill.

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1 Introduction

1.1 Geological context

The metasedimentary rocks of the south-western Scottish Highlands are metapsammites, metapelites and metacarbonates. These rocks reflect different depositional environments in a transgressive regime, with a general increase in water depth over time (Roberts and Treagus, 1977). On Islay (shown in figure 1), the metasedimentary rocks are part of the Argyll Group of the Dalradian Supergroup (Harris and Pitcher, 1975). and at around 595 ± 4 Ma, this group of sedimentary rocks was intruded by mafic sills and dykes (Halliday et al, 1989).

Major metamorphic events of the south-western Scottish Highlands, including Islay, took place during the Caledonian Orogeny (Graham et al., 1983). There were four main deformation events, D_1-D_4, and peak metamorphism occurred during D_3, reaching greenschist to epidote-amphibolite facies (Roberts and Treagus, 1977). During the first two deformation events, D_1 and D_2, folding of the stratigraphy took place. It led to the formation of the two anticlines, Islay Anticline and Ardrishaig Anticline, as well as the Loch Awe Syncline (Roberts and Treagus, 1977). Despite reaching peak metamorphic temperatures during the D_3 deformation event (Harte et al., 1984), no textural evidence of this is preserved in the region (Roberts and Treagus, 1977). A consequence of peak metamorphism occurring during D_3 is that the metamorphic zones are overprinted on the D_1 and D_2 structures (Skelton et al., 1995). The final deformation event, D_4, caused renewed folding of the existing folds, leading to both a tightening and a collapse of these structures (Roberts and Treagus, 1977). It also folded the metamorphic isograds, which are dividing lines marking the appearance of new index minerals (Skelton et al., 1995).

The metabasaltic sills at the Ard of Port Ellen, Islay, have been metamorphosed under greenschist facies conditions. Two specific characteristics of these sills stand out, namely an abundance of large plagioclase phenocrysts within the sill interiors, and large iron-oxide crystals that have spatially been linked to the sill margins. The interior assemblage contains the following minerals: Epidote (Ep) + Albite (Ab) + Chlorite (Chl) + Amphibole (Amp) ± Quartz (Qz) ± Calcite (Cc). The margin assemblage contains the following minerals: Qz + Chl + Cc + Ab + iron-oxides ± Ep (Kleine et al., 2016). A distinguishing feature of these sill margins is the absence of amphibole, which similarly has been observed in other extensively carbonated sill margins found within the region (Harte and Graham, 1975; Graham et al., 1983). The difference between the other carbonated sills and these particular sills on Islay is the presence of large iron-oxide crystals in the Islay sills (Kleine et al., 2016).

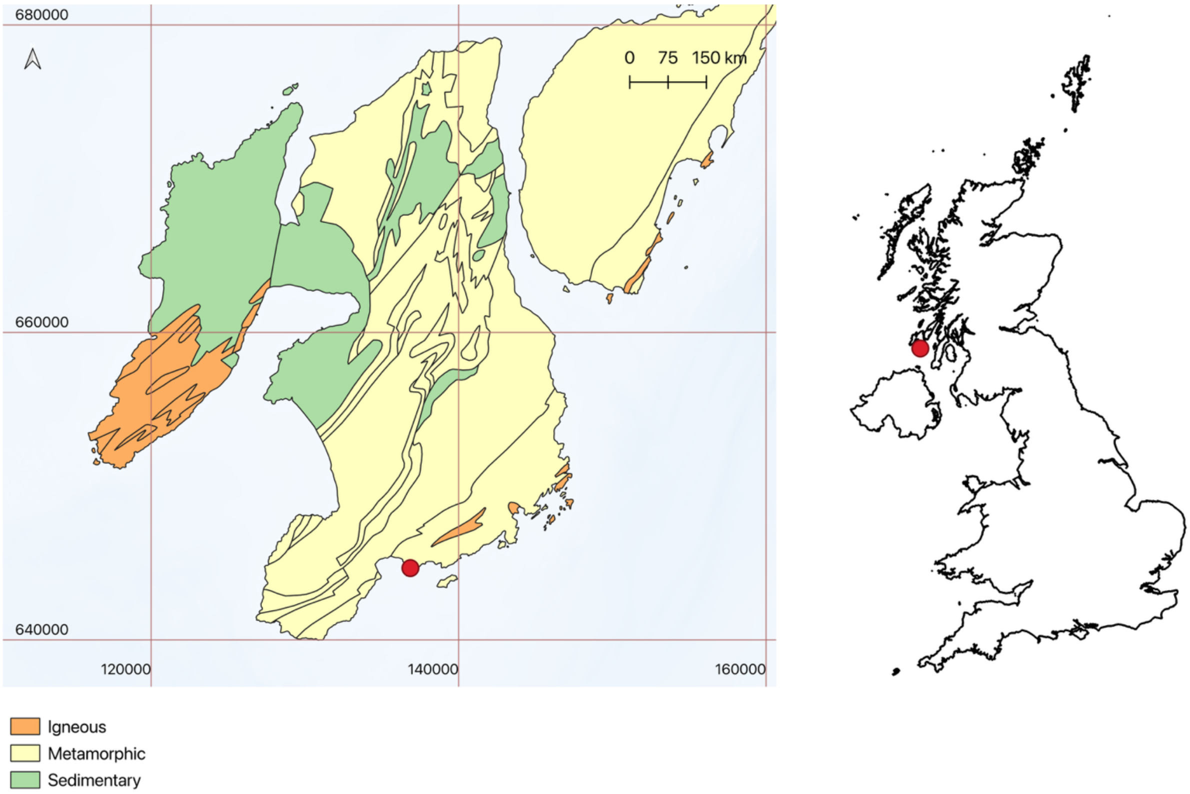


Fig 1. A map showing the location of the field study. Port Ellen is located at the red dot of the leftmost map of the island Islay. The rightmost map show the location of Islay in relation to the rest of the United Kingdom. Data for the construction of this map was taken from DIVA-GIS.org and from the British Geological Survey. Further information can be found as the last entry in the reference list at the end of this document.

1.2 Fluid flow

Three main fluid flow events have been recognised. These are events where fluids have infiltrated and reacted with the host rock. The events, in order of occurrence, are: spilitization (Graham, 1976; Skelton et al., 2010; Arghe et al., 2011), carbonation (Skelton et al., 1995; Arghe et al., 2011; Kleine et al., 2015) and quartz-carbonate-sulphide veining (Anderson et al., 2004; Baron and Parnell, 2005; Parnell et al., 2000). These are described in detail below. Syn-metamorphic fluid flow was structurally channelled upwards towards axial zones of anticlines, conforming to folded phyllite layers within structures such as the Ardrishaig and Islay anticlines. Metabasites were also functioning as fluid pathways, as fluid fluxes preferentially flowed via deformed metabasaltic sill margins and phyllites, rather than less permeable metapsammites (Skelton et al., 1995). Modal calculation of metabasaltic sills in the region suggests that both the spilitization and prograde metamorphism occurred before the fluid flow causing the carbonation of the sills (Arghe et al., 2011). This is corroborated by Kleine et al. (2015), via textural evidence of chloritized garnets, which suggests that the carbonation indeed happened after peak metamorphism.

1.2.1 Spilitization

The spilitization, caused by infiltration of saline fluids of oceanic origin, occurred before greenschist facies metamorphism. Firstly, the spilitization caused albitisation of calcic plagioclase. This is a process where calcium (Ca) is exchanged for sodium (Na). It occurred without apparent volume changes (Graham, 1976). The absence of volume changes in the plagioclase is suggested to relate to deposition of Ca in existing fractures, as either epidote or calcite. Secondly, the spilitization caused the formation of epidote in segregations, areas of particularly high epidote concentration within the sills in the shape of veins and lenses (Graham, 1976). This is of particular interest, as epidote is affected by volume changing reactions during metamorphism, creating areas of higher porosity. The textural and mineralogical changes in the epidote layers, i.e. lenses or veins, probably caused changes in flow pathways during later fluid intrusions, as the fluids preferentially flowed via these porous "pathways" rather than penetrating deeper into the less permeable sill interiors (Arghe et al., 2011; Kleine et al., 2016).

1.2.2 Carbonation

Prograde metamorphic reactions can cause the release of fluids in a process called devolatilization, which is when chemical reactions release volatiles as products of the reactants. Decarbonation and dehydration reactions are examples of such and often occur deeper down in the crust. The released fluids eventually rise upwards due to buoyancy forces and structural forcing (Skelton et al., 1995; Pitcairn et al., 2010).

This phenomenon is used to explain mineralogical changes within the metabasaltic sills, which require an intrusion of CO_2- and H_2O-rich fluids to have occurred. The devolatilization fluids were channelled upwards from below, via paths defined by the primary structures, such as the Ardrishaig Anticline (Skelton et al., 1995). This fluid infiltration caused the carbonation of the host rocks post peak metamorphism, and they were situated above the prograde rocks releasing the volatiles (Skelton et al., 1995; Kleine et al., 2015). Thus, the fluid source were rocks experiencing prograde metamorphism deeper down in the crust and the presence of these fluid fluxes enabled carbonation and hydration reactions to occur in overlying rocks, during retrograde metamorphism (Kleine et al., 2015; Kleine et al., 2016).

Evidence for the fact that carbonation of the sills occurred after peak metamorphic conditions can be found in textural characteristics of chloritized garnets at Loch Stornoway (Kleine et al, 2015). The margins of the metabasaltic sills were highly carbonated by the CO_2-rich fluid flow, whilst the interiors generally remained unaltered. This clear distinction between primarily unaltered sill interiors and highly carbonated margins is explained by textural and mineralogical differences. In the margins intergranular fluid flow was enhanced. In contrast, the massive and homogenous interiors experienced much less effective intergranular fluid infiltration (Kleine et al., 2016), resulting in the stark differences in carbonation levels between the margins and the interior.

The types of assemblages in greenschist metabasalts in the region, relevant to the study area, can be categorized into the following: Type I, II, III (Graham et al., 1983).

- **Type I**: Am + Ep + Chl + Qz + Ab and trace amounts of CC = Typical of greenschist facies metabasalts. Found in the sill interiors, massive in appearance, randomly oriented crystals. Relic igneous phenocrysts are preserved.
- **Type II**: Am + Ep + Chl + CC + Qz + Ab. Defines the reaction zone between type I and III (between interior and margin). + Lacks foliation.
- **Type III**: Chl + CC + Qz + Ab with only minor Ep and no Am. Sill margins. Produced from the type I assemblage by the fluid-driven carbonation reaction:
    - Am + Ep + H_2O + CO_2 = Chl + CC + Qz. Notice the lack of iron-oxides, e.g. hematite, magnetite etc., as major components.
      (Graham et al., 1983).

  In type III the foliation is defined by chlorite alignment. No igneous phenocrysts are preserved (Arghe et al., 2011).

1.2.3 Quartz-carbonate-sulphide veining

The third fluid event led to quartz-, sulphide- and carbonate-veins forming in a regionally extensive network. The veining formed either perpendicular to, or parallel to the lithological features, such as bedding or intrusions. In directional terms, the veining either followed a NE-SW or a NW-SE propagation (Anderson et al., 2004). The veining happened during exhumation of the now exposed rocks and can be categorised into at least two main subevents. The first one had high temperature conditions (> 300°C) and was of moderate salinity. The second one had low temperature conditions (< 200°C) and was also of moderate salinity (Anderson et al., 2004; Kleine et al., 2016).

The high temperature subevent mainly lead to the precipitation of quartz, and to a lesser extent sulphides. Analysis of sulphur isotope content suggests that the origin of sulphide minerals were likely from local metasedimentary lithologies (Andersson et al., 2004). The same study states that the veins share common features with globally widespread base metal veins, as well as gold veins of mesothermal type, both of which are found in the greenschist metamorphic facies rocks. Furthermore, the low temperature subevent led to the precipitation of the carbonates found in these veins. This time, the fluid source is thought to have been sedimentary basins situated above and the fluid infiltration was likely a consequence of structural overpressure. This second subevent led to the precipitation of dolomite and ankerite within already existing fractures and veins formed by the earlier subevent (Andersson et al., 2004).

1.3 Iron-oxide enrichment

A unique feature of some of the metabasaltic sills at the Ard of Port Ellen is the prevalence of large iron-oxide crystals, mainly consisting of hematite. Kleine et al. (2016) found that there is an overlap between the carbonated margins and the occurrence of these large hematite crystals. The origin of these iron-oxide crystals remains unclear, but it has been suggested that a likely source of this pattern of concentration is due to consequences of magmatic differentiation. Thus suggesting it happened before any of the fluid events described above (Kleine et al., 2016).

Galerne et al. (2010) proposes two different processes that can lead to iron enrichment in sill margins. Firstly, during early-stage crystallisation, there is the concept of inward cooling from the margins. The process is called Soret fractionation and it involves the partitioning of heavier from lighter elements, as a result of thermal differences within a magma body. In the process, heavier elements like iron preferentially moves away from warmer regions towards areas that have cooled relatively more, such as margin regions. The transportation is facilitated by thermal diffusion. Secondly, is a process that may occur during the later stages of crystallisation. The concept is very similar to the Soret fractionation, with thermal gradients being the cause of the differentiation. The temperature differences between regions creates a pressure gradient causing the remaining melt to, via porous flow, flow towards the margins. The margins are regions of lower pressure compared to central regions of the magma body. These processes can cause margin enrichment of iron in a sill. (Galerne et al., 2010)

2 Aims and objectives

The aim of this study is to investigate the spatial distribution of the enrichment of hematite in a metabasaltic sill at Port Ellen, Islay, Scotland. Furthermore, the aim is to investigate the relationship of the spatial distribution and the spilitization, carbonation and veining that has affected the metabasaltic sill. The objective is to map and count the distribution of these features. By doing so this thesis will answer the following research question:

- What is the spatial relationship between the carbonation, spilitization and veining with the iron-oxide enrichment of the metabasaltic sill?

3 Methods

A profile across the sill was constructed and data was sampled along the profile using a mechanical grid of 498 intersecting points. The mechanical grid had a length of 7.5 cm and a width of 12.5 cm. To sample the profile the grid was repeatedly placed perpendicular to the sill margin, in a sequence of connecting grids, from the margin towards the interior of the sill. See figure 2 for a visual overview of the concept. For example, four connected grids represent 4 x 7.5 cm, i.e. 30 cm, of a profile through the sill, perpendicular to the sill margin. For each grid placement, the number of points for macroscopic hematite crystals and plagioclase phenocrysts were counted. The area covered by epidote segregations were also estimated this way, as well as obstructed surfaces. Obstructed surfaces were for example surfaces covered by lichens and other types of vegetation. The grid points of obstructed surfaces were deducted from the total number of grid points when calculating percentages of hematite and plagioclase crystals within each grid.

The physical reality of the sill necessitated geometrical solutions to construct a continuous profile through the sill. There were three main obstacles:

i) The dip of the opposing margins of the sill were not parallel to each other. This led to the issue of constructing a continuous profile through the sill that is perpendicular to both margins. This meant that a constructed profile that is perpendicular to both margins will meet at an angle within the sill interior. A profile that does not change direction within the sill can at most, in this case of two margins with different dips, be perpendicular to one of the margins.
ii) Lack of exposed rock from the sill interior. The sill was in many places covered by vegetation, limiting the unobstructed propagation of a continuous profile through the sill.
iii) The dip of the sill margins combined with the availability of exposed rock prohibited the construction of one single profile from margin to margin. From upper margin, i.e. the footwall margin, the perpendicular profile quickly propagated into the ground. On the other end, the headwall side, the profile eventually propagated into the air above the sill.

To solve issue i) of different dips, the best solution was to construct two opposing profiles. Starting from each margin, with each profile propagating into the sill perpendicular to respective starting margin. This meant that the "meeting point" of the two profiles would be at an angle (see figure 2). The justification for this choice was firstly, that a profile perpendicular to the margin is of the utmost importance in order to determine the depth of each data point. Secondly, according to previous studies of sills in the areas, the interior of the sills are very homogenous both mineralogically and texturally (Graham et al., 1983; Arghe et al., 2011; Kleine et al., 2016). The homogeneity of the interior of the sills strengthens the viability of the chosen solution. Meeting at an angle in the interior is thus superior to propagating at an angle from the margins.

Solving issue ii) and iii) required the solution of constructing the continuous profile by patching together sequences. This solution was made simpler due to the previous choice of always propagating the sampling profile perpendicular to the margin. An example of profile construction using these solutions is illustrated by the following example: Profile A is initiated from the footwall margin with a dip angle of v. The first sequence is constructed out of 12 grids before it reaches the ground. To continue the profile the second sequence continues by "stepping sideways", by a 90° turn to reach another area that is exposed and simultaneously 12 grids deep into the sill. The second sequence then continues, still perpendicular to the margin angle v and at the same depth from the margin. Figure 2 illustrates how the combination of these solutions work together to construct a profile across the sill, where the key is continuous perpendicularity to the margin. The profile length, or depth, was calculated by the sum of number of grids times the length of the grid (7.5 cm). It was also approximated by tape measurement from the margin to the last grid, in segments, taking into consideration the dip angle as well as a secondary angle. The process can be described as follows: Segment 1 is measured with tape at an angle perpendicular to the dip of the sill. For this measurement to be perpendicular to the plane of the sill margin, another angle must be taken into consideration. This is the angle towards the line that is perpendicular to the dip and the strike, in other words the angle to a line entering straight into the sill. If the measured length is 3 meters, and this second angle is 30°, then the "real" length of the depth at that point is [3 x cos (30) = 2.6 m]. The concept is shown in figure 3. These values were afterwards compared. To reduce the error of the depth, care was taken to take grid measurements on areas being as close as possible to planar perpendicularity against the margin surface.

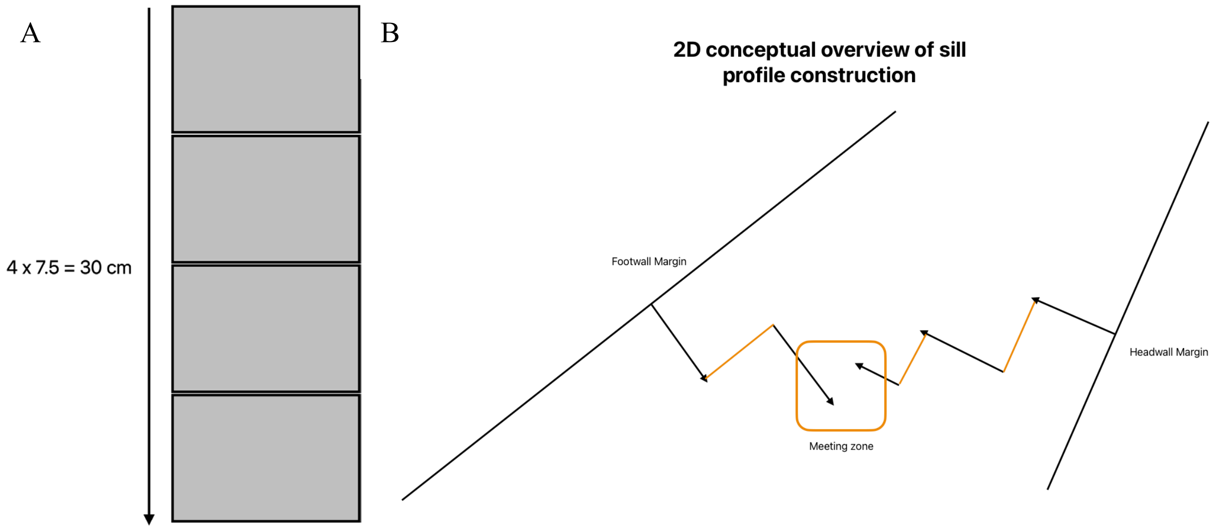


Fig 2. A) Concept of the connected grids to construct the profile. B) Concept of how the sill profile is constructed through the sill, to solve the practical issues encountered in the field.

The exact placement of the margin of the sill had to be assumed based on the apparent erosional differences between the metabasaltic sill and the adjacent and more easily eroded metasedimentary rocks (Kleine et al., 2016). To legitimise the assumed placements of the sill margin, the carbonate content of the assumed sill margin was compared to a known sill margin from previous studies. The estimation of carbonate content was calculated using a "fizz-o-meter", which is a device developed and described by Skelton (1995). The "fizz-o-meter" is illustrated and described in figure 4. It analyses a specific amount of crushed rock sample of a certain fineness by allowing it to react to 10% HCL of a specific quantity. The reaction takes place within a closed container that is equipped with a barometer to measure pressure buildup. The headwall side of the sill was analysed using this instrument, with a sample each 20 cm, totalling 8 samples, starting from the margin. The samples were collected using a hammer and chisel and later crushed using a hammer and a hollow steel cylinder with a "smashing pad" placed underneath. A smaller cylinder was hammered into the hollow cylinder containing the rock sample, thus crushing the samples therein to satisfactory fineness.

The most promising area of the sill to analyse was at certain times of the day inaccessible due to tidal changes. This necessitated planning of when it was possible to access the area for sampling and for how long.

The mineralogy was also analysed optically by looking at replacements textures of chlorite replacing plagioclase, as well as the presence of epidote segregations. The chlorite replacing plagioclase either fully pseudomorph the phenocrysts or preferentially replaced the interiors. The epidote segregations could be recognised from the parallel fractures that developed during volume changes caused by metamorphism. The segregations themselves are also subparallel, or even parallel, to the margins (Skelton et al., 2010; Kleine et al., 2016).

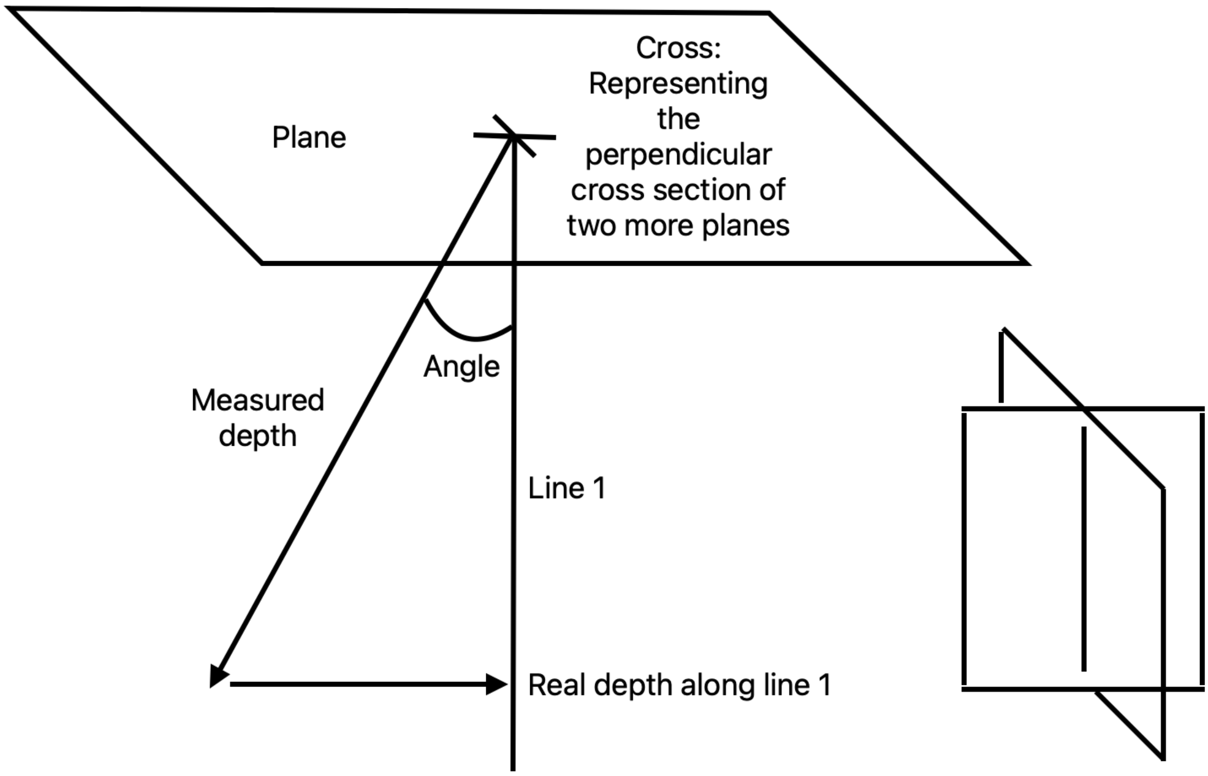


Fig 3. Showing the concept of approximating the real depth from the available angles. The margin is represented by the plane. Line 1 is a line that is perpendicular to this plane. Line 1 can also be represented by the line following the linear cross section of two planes that are both perpendicular to the main plane. The measured depth multiplied by cos(angle) gives the real depth.

It became apparent in the field that hematite might be concentrated within veining systems in the sill. This resulted in the decision to add one more data gathering method. Two large cliff faces with a close to planar perpendicularity to the margin were selected based on size, “freshness” i.e. lesser erosion and absence of vegetation, as well as accessibility. These two cliff faces were then analysed using 40 by 40 cm squares. Within the squares, the occurring hematite crystals were sorted into two categories: vein-related or not vein-related. Cliff face 1 required 19 squares whereas cliff face 2 required six squares.

The “fizz-o-meter”-device stopped working by the end of the week, causing problems to gather more carbonation data than the already collected headwall series. As a makeshift solution, samples were taken from the two cliff faces, both from within veins and elsewhere. These were then analysed using the 10% HCL. Fresh surfaces were arranged and HCL was applied and the presence of a reaction or not was noted.

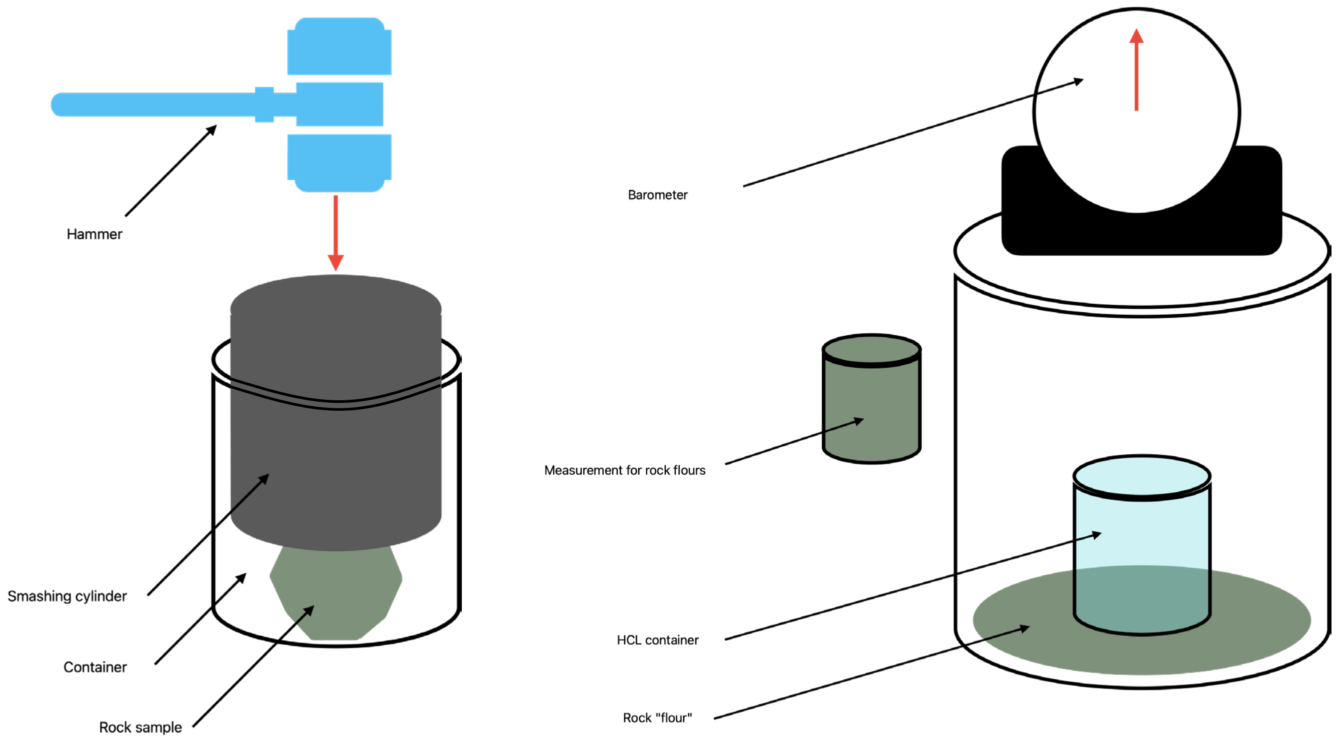


Fig 4. Showing the "fizz-o-meter" concept. The rock sample is hammered into finer fragments, using the hammer and the metal cylinders illustrated on the left. The fine-ground rock "flour" is then poured into the container to the right. A small vessel containing HCL is then placed inside the large container, on top of the rock flour. It is important that these two constituents do not react at this time. A lock with a mounted barometer is then attached to seal the container. The last step is to shake the container, i.e. the "fizz-o-meter", and read the pressure that builds up from the barometer.

4 Results

4.1 Grid data

The distribution of plagioclase phenocrysts increased towards the interior of the sill, remaining between 0-10% the first two meters and rising sharply thereafter. Figure 5 shows the difference between two grid measurements. Figure 5 A) was taken by the margin and figure 5 B) was taken within the sill interior. Figure 5 A) is lacking large plagioclase phenocrysts while figure 5 B) is showing a high concentration of plagioclase phenocrysts.

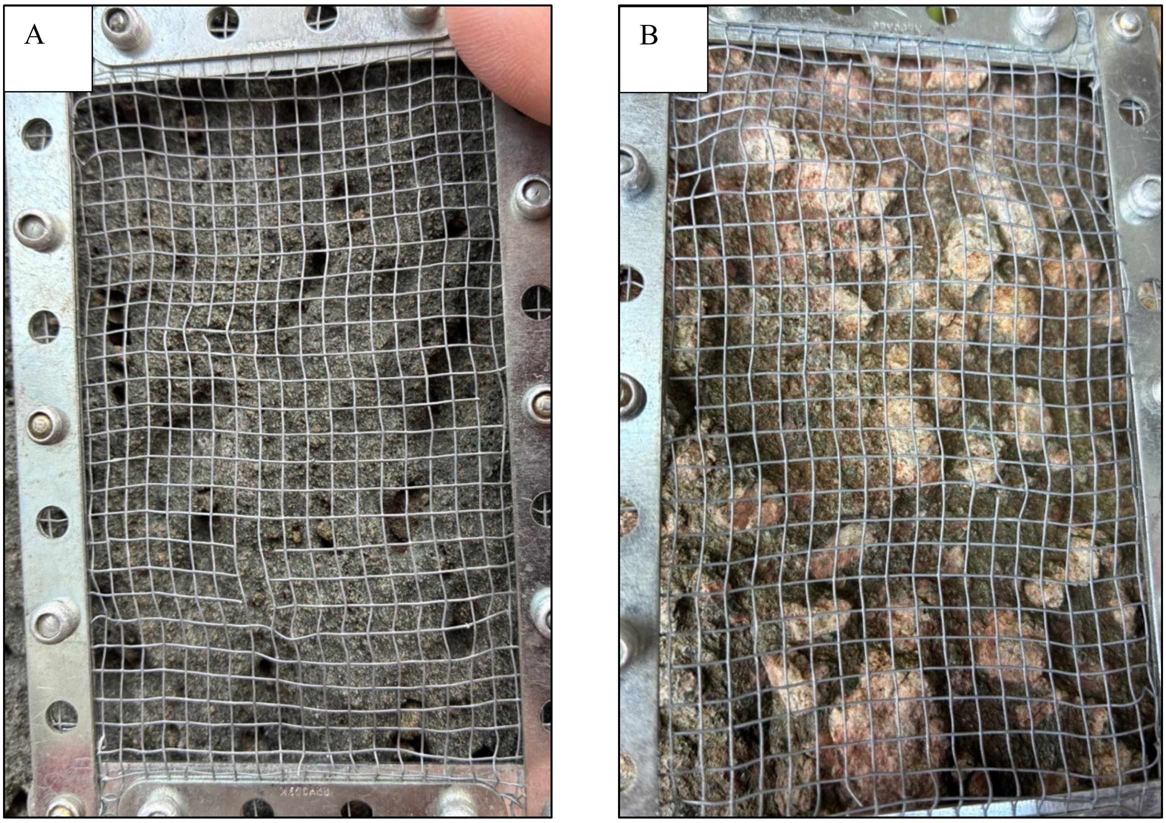


Fig 5. A) Grid photo of margin, lacking large plagioclase phenocrysts. B) Grid photo of interior, showing a great concentration of plagioclase phenocrysts.

The pattern of increasing prevalence of plagioclase phenocrysts towards the interior of the sill is obvious when plotted against distance, as done in figure 6. To plot a "complete" profile across the sill, two series were combined. One starting from the footwall side and one starting from the headwall side. From the footwall side two parallel series were sampled. The first one sampled (footwall series 1) had more obstructing elements, such as lichens and other types of vegetation, and was patched together from more individual segments than the second one (footwall series 2). For this reason, the second series was chosen when combining a footwall series with the headwall series to plot a "complete" profile through the sill. The process is described in more detail in the methods section of this paper.

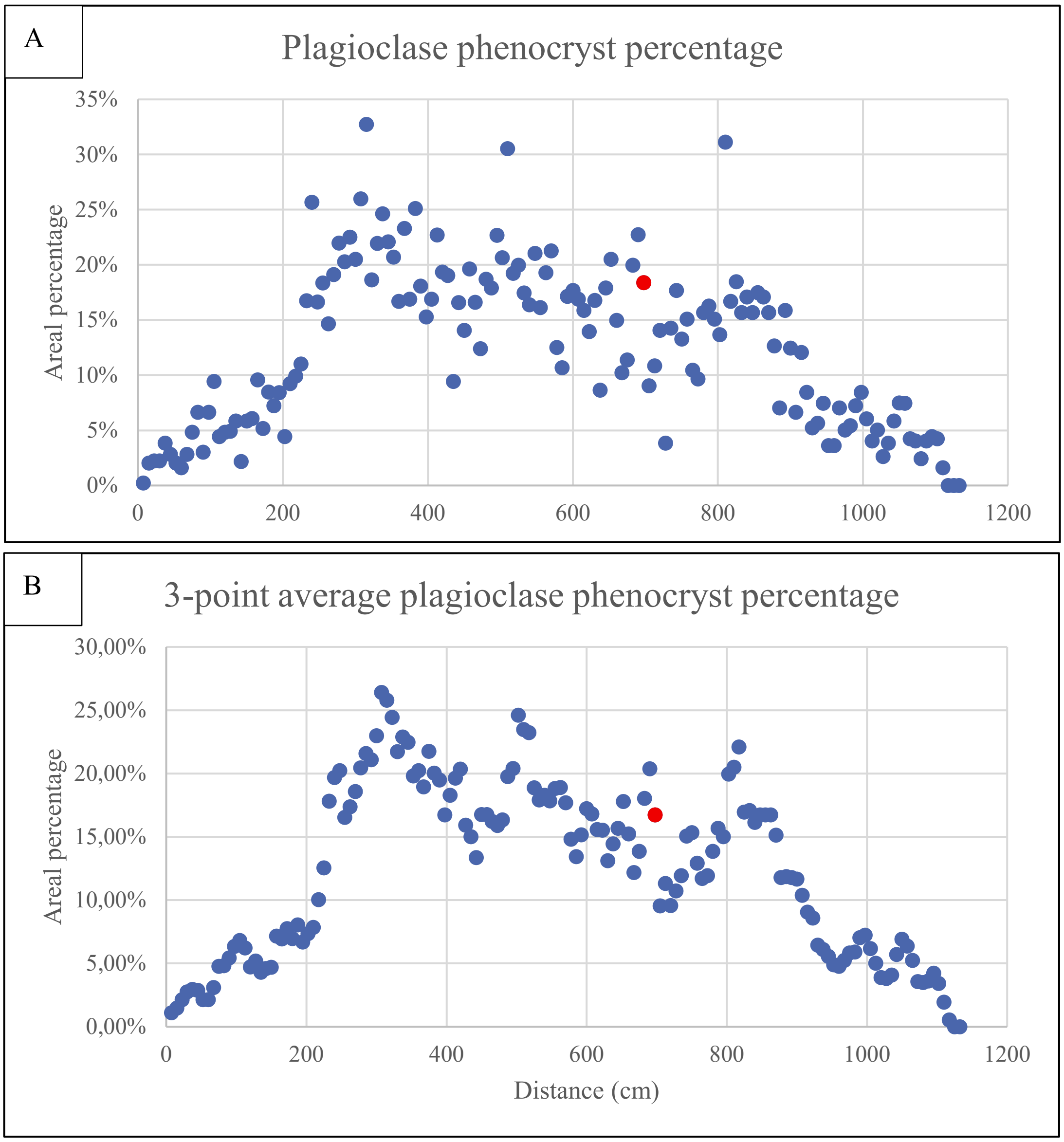


Fig 6. A) The distribution of plagioclase phenocrysts across the sill. The headwall margin starts at 0 and the footwall margin starts at ~1150 cm and they "meet" at the red circle B) The same profile of distribution of plagioclase phenocrysts but plotted using 3-point averaging of percentages to reduce the effect of extreme outliers from the narrow sampling band of grids. The headwall margin starts at 0 and the footwall margin starts at ~1150 cm and they "meet" at the red circle.

The occurrence of hematite crystals in the grid analysis is presented below in figure 7. The added trendline is showing an increasing trend from the headwall side of the sill towards the footwall side of the sill. This represents an increasing trend from the NW to the SE. R^2 is added to the trendline in figure 7 A, showing a value of 0.0249. The p-value for the trendline slope was calculated using Data > Data Analysis > Regression in excel for the hematite concentration as Y and distance as X. This gave a p-value of 0.0531. The statistical analysis shows a weak, but positive between correlation between the concentration of hematite by distance across the profile of the sill, starting at the headwall. However, the findings cannot be said to be statistically significant, as the *p-value* > 0.05 (Andrade, 2019).

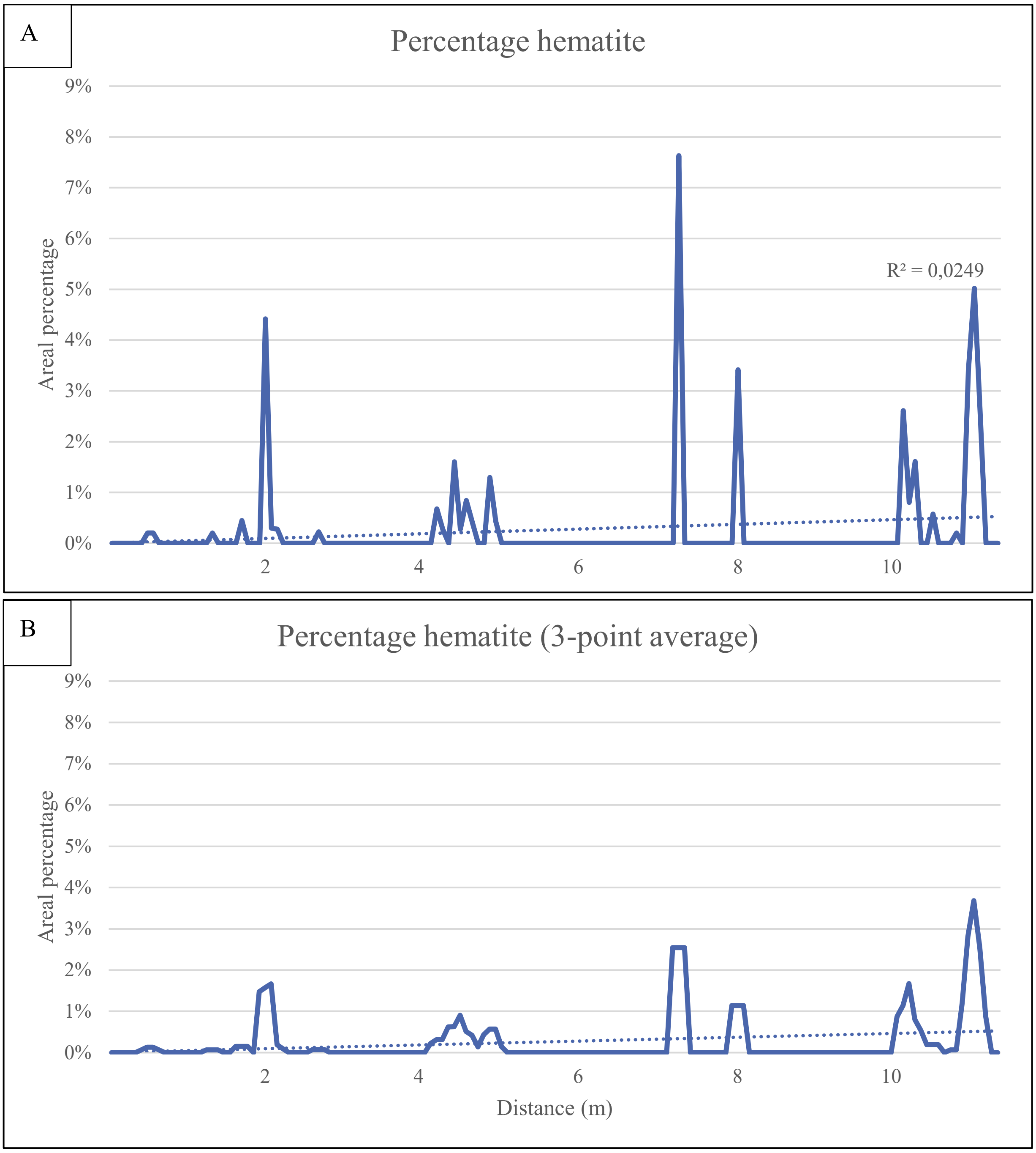


Fig 7. A) The distribution of hematite crystals through the sill, from the headwall margin to the footwall margin. The R^2 value is displayed on the right side of the graph. B) The distribution of hematite crystals through the sill, from the headwall margin to the footwall margin, using a 3-point average to reduce the effect of outliers from the small grid width of the sampling profile.

A spearman rank correlation was calculated on the profile, to analyse the relationship between plagioclase and hematite prevalence in the sill, yielding a -0.17305. This is a weak negative correlation, that could suggest an inverted relationship between hematite concentration and plagioclase concentration. However, given that it falls below 0.20 (in absolute values), the correlation is weak or small and not strong enough to draw any conclusions on this finding alone (Gignac and Szodorai, 2016). Figure 8 shows a plot representing the data series used for the spearman correlation calculation.

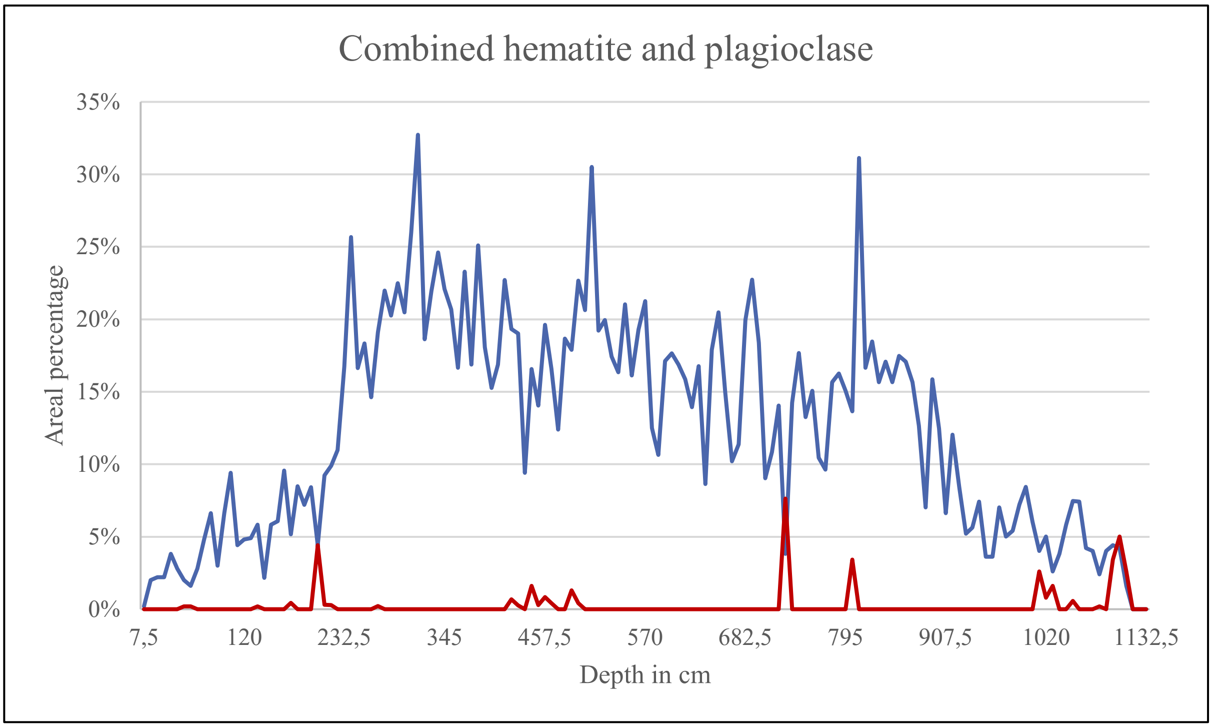


Fig 8. Showing the profile percentage of hematite and plagioclase per distance in cm, using the grid method. The two series were used to calculate a spearman ranked correlation coefficient.

The spearman rank correlation for the footwall series (series 1) that was not used in the combined sill profile series got a spearman coefficient of -0.589. This is a much stronger correlation. The values used for this computation is plotted in figure 9 below. The choice of using footwall series 2 in the combined profile was based on the fact that footwall series 2 was less covered in vegetation and constructed by fewer separate segments.

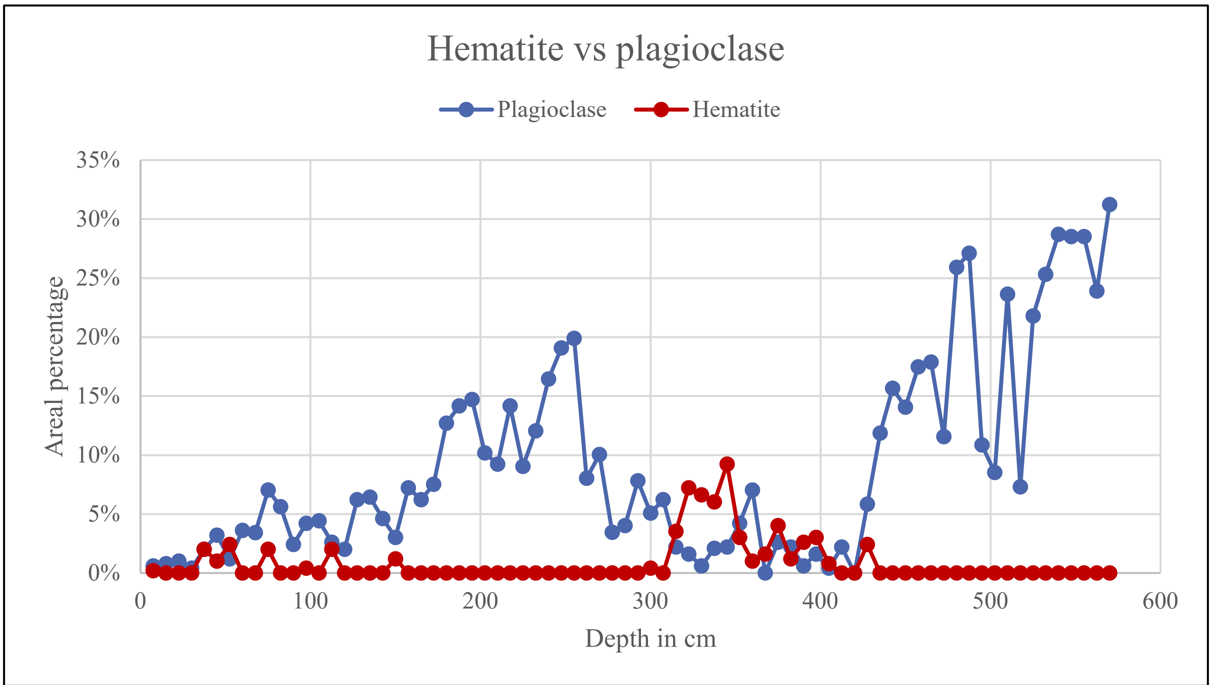
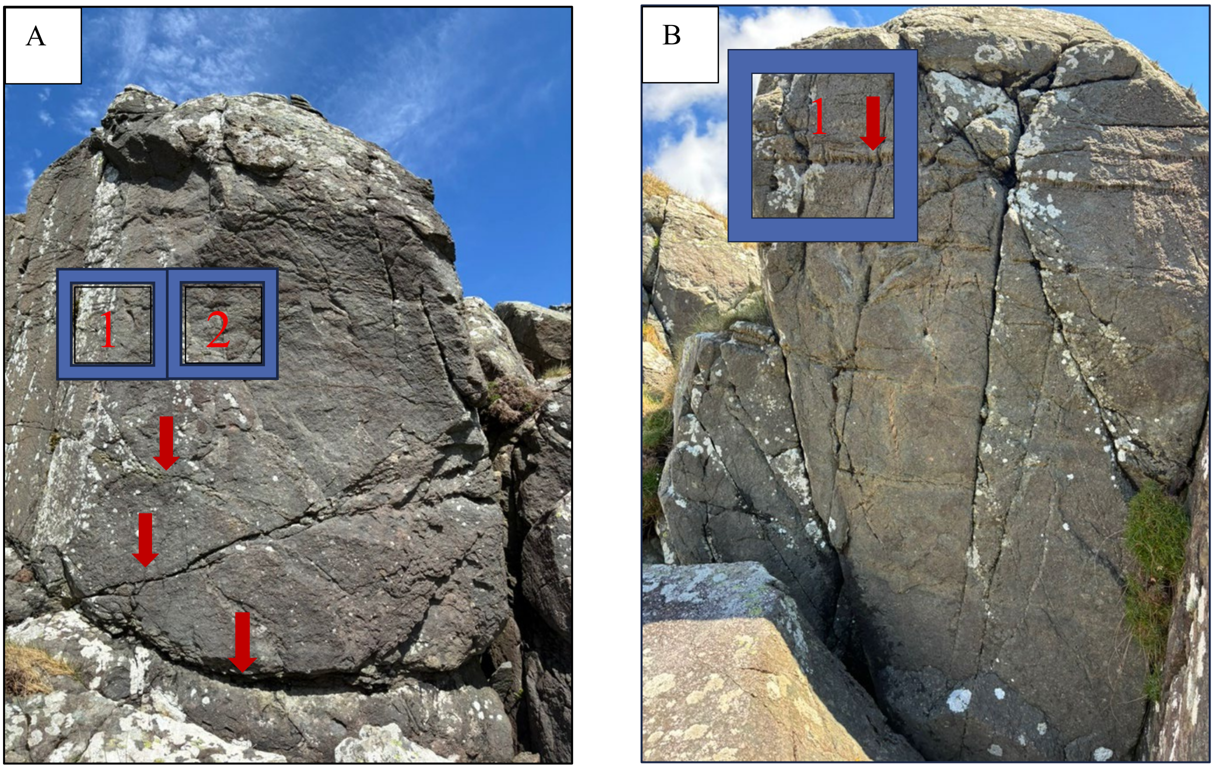


Fig 9. Showing a plot of the footwall series 1, that yielded a spearman ranked correlation coefficient of -0.589.

4.2 Hematite in veins

The two cliff faces, A and B, used for analysing the connection between hematite and veins are shown in figure 10. The height of cliff face A was 2.5 meters and cliff face B was 1.6 meters. However, due to impracticalities of both locations, only the lowermost 2 meters of cliff face A and the uppermost 1.2 meters of cliff face B were sampled.



1	2	3	4
5	6	7	8
9	10	11	12
13	14	15	16
	17	18	19

1	2	3
4	5	
	6	

Fig 10. A) Large cliff face requiring 19 40x40 cm squares, the squares are illustrated by the blue squares (approximate size) in order of placement. Below the photo is a schematic overview of the placement of each square, in numbered order. Red arrows pointing towards areas of high hematite concentration. B) Smaller cliff face of 6 40x40 cm squares, square 1 is illustrated by the blue square (approximate size). Below the photo is a schematic overview of the placement of each square. The red square is pointed at one of the veins, or epidote segregations, that are containing abundant hematite crystals.

The data for the two cliff faces sampled are presented as two tables in table 1. There is a striking difference between the occurrence of hematite crystals within veins compared to elsewhere on these two cliff faces. Combining both of the cliff faces, 98 % of hematite crystals were found within veins and only 2 % were found elsewhere. Only one hematite crystal was found outside of the veins on cliff face A, and four hematite crystals were found outside of the veins on cliff face B. In comparison, 135 and 100 hematite crystals were found within veins on cliff face A and B respectively. On cliff face B the veins are parallel to the sill surface and are in fact epidote segregations. On cliff face A some of the veins are parallel or sub-parallel, but one of them, the middle on in figure 10 A, is not.

Table 1. A) Table of hematite occurrence of cliff face A and where each hematite crystals were found. Elsewhere = not within veins. B) A similar table of hematite occurrence of cliff face B.

A)

Square number	Within vein	Elsewhere
1	5	0
2	1	0
3	0	0
4	0	0
5	8	0
6	4	1
7	12	0
8	0	0
9	5	0
10	4	0
11	0	0
12	8	0
13	5	0
14	5	0
15	11	0
16	15	0
17	28	0
18	19	0
19	5	0

B)

Square number	Within vein	Elsewhere
1	28	3
2	33	1
3	27	0
4	9	0
5	3	0
6	0	0

There is also visual evidence of hematite enrichment in epidote segregations, see figure 11 below. The “square” numbers in table 1 is connected to the overview tables in figure 10. For example, square number 1 in table 1 B is the same square as the cell numbered 1 in figure 10 B. In this case, the cell numbered 1 is the 40x40 cm square that was used in the upper left corner of the cliff face, encircling the area around the red arrow. The photo in figure 11 is a zoomed in photo of the area pointed at by the red arrow in figure 10 B. This illustrates the connection between epidote segregations and hematite rich zones. From the data in table 1 B one can see that in this square (1), 28 hematite crystals were found within the veins (Kleine et al., 2016) and only 3 outside of the veins, which in this square were epidote segregations.

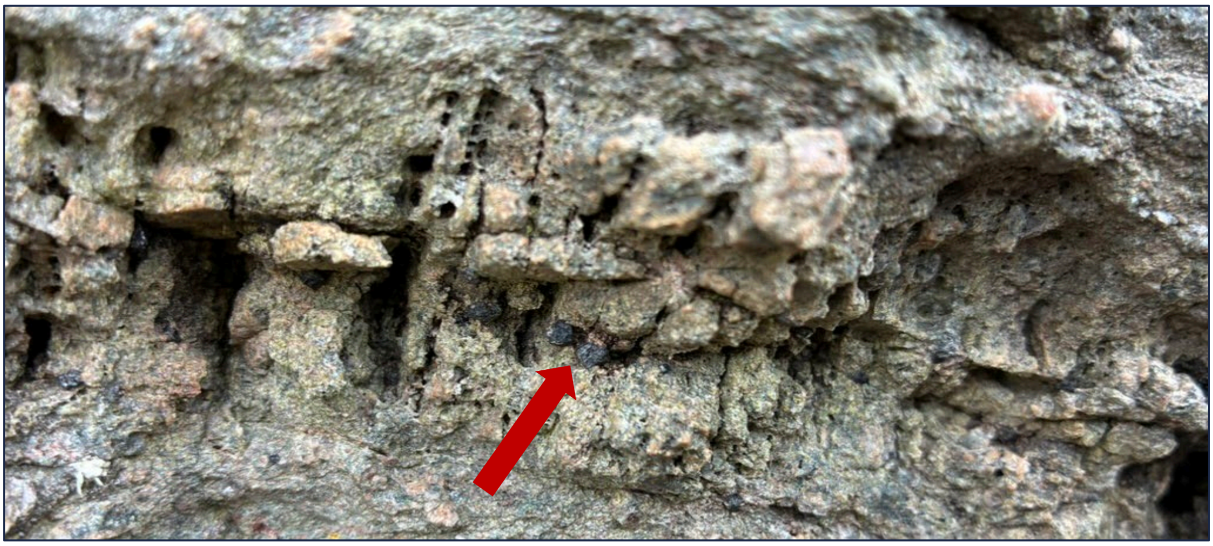


Fig 11. Showing an epidote segregation containing a high concentration of hematite crystals. This is a zoomed in photo of fig 10 B for a closer look at the epidote segregations seen there.

Further textural evidence of hematite precipitation within fracture systems and veins was found, and is best illustrated by the pictures in figure 12 B.

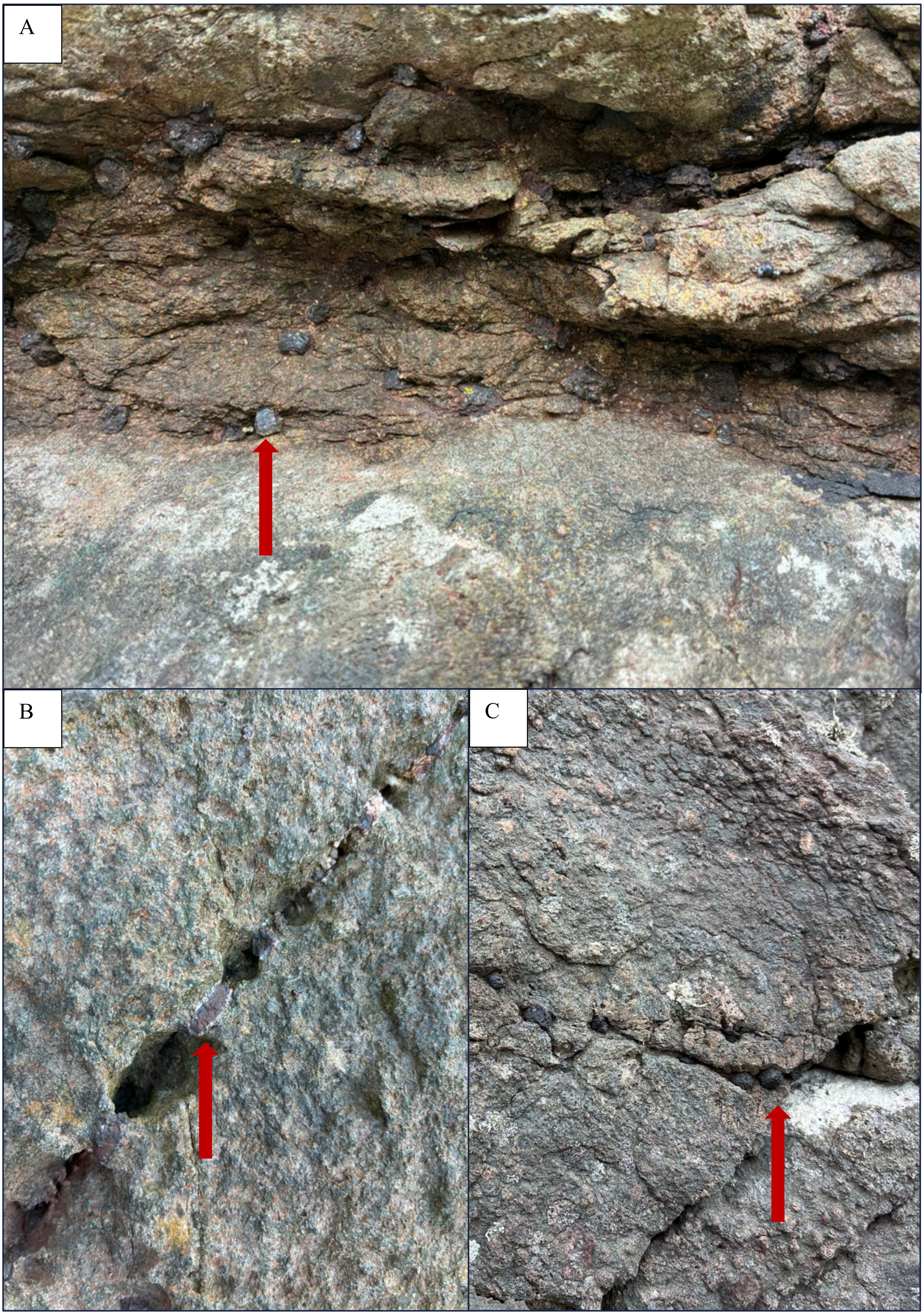


Fig 12. The red arrows points to hematite crystals, to illustrate their appearance in each picture. A) Extremely high concentration of hematite crystals found in vein at the bottom of cliff face A, seen in figure 10. B) Precipitated hematite within a fracture. C) Hematite within fracture system. Also, note the presence of the plagioclase phenocrysts surrounding the hematite "line".

4.3 Carbonation

Carbonation of the sill was measured by the "fizz-o-meter"-tool described earlier in the methods section. The carbonation of the headwall margin is described in table 2. The results show a decreasing pattern from the margin towards the interior of the sill. The first sample taken at the very start of the assumed margin gave the same output as the sample from the known margin, at 0.58 Bar. The spearman ranked correlation for hematite and calcite was calculated using the data from table 2, giving a value of -0.31. Combining the hematite data gathered in this study with the carbonation data from Port Ellen Sill 1, from the Kleine et al. (2016) study yielded a spearman correlation value of 0.12. The former is a low negative correlation, and the latter is a low positive correlation (Gignac and Szodorai, 2016).

Table 2. The table shows eight samples taken at 20 cm intervals from the margin towards the interior on the headwall side of the metabasaltic sill. The table also presents the rank used in calculating the spearman correlation for hematite and calcite at the margin.

Fizz-o-metry	Bar	Depth	Hematite	Rank Bar	Rank Hematite
1	0.58	0	0	8	3.5
2	0.4	20	0	7	3.5
3	0.21	40	0	6	3.5
4	0.11	60	0.2	5	8
5	0.1	80	0	3.5	3.5
6	0.1	100	0	3.5	3.5
7	0.05	120	0	2	3.5
8	0.04	140	0.1	1	7
Known sample (#10)	0.58				

Furthermore, samples were taken from cliff face A and adjacent cliff faces, comparing hematite rich areas to areas without visible presence of hematite crystals. The results are presented below in figure 6. Due to equipment failure, these samples were simply sampled by exposing fresh rock surfaces using a hammer and then wetted by 10% HCL.

Table 3. Showing a total of 10 samples, half from hematite rich areas and half from non-hematite areas. "Yes" indicates a clear reaction to wetting by 10% HCL on the fresh surface. "No" simply means that no reaction was observed at the area tested.

Hematite rich vein area	Non-hematite area
Yes	Yes
Yes	No
Yes	Yes
Yes	No
Yes	No

4.4 Chlorite replacement

Chlorite replacement was, at a systematic scale, only observed at the headwall side of the sill. It reached approximately 140 cm into the sill from the margin. Individual plagioclase phenocrysts could be observed further into the sill on both sides, but they were the exception rather than a systematic replacement of the majority of phenocrysts. Figure 13 shows the replacement texture of a plagioclase phenocryst by chlorite. It shows an internal replacement, leaving the rims unaltered.

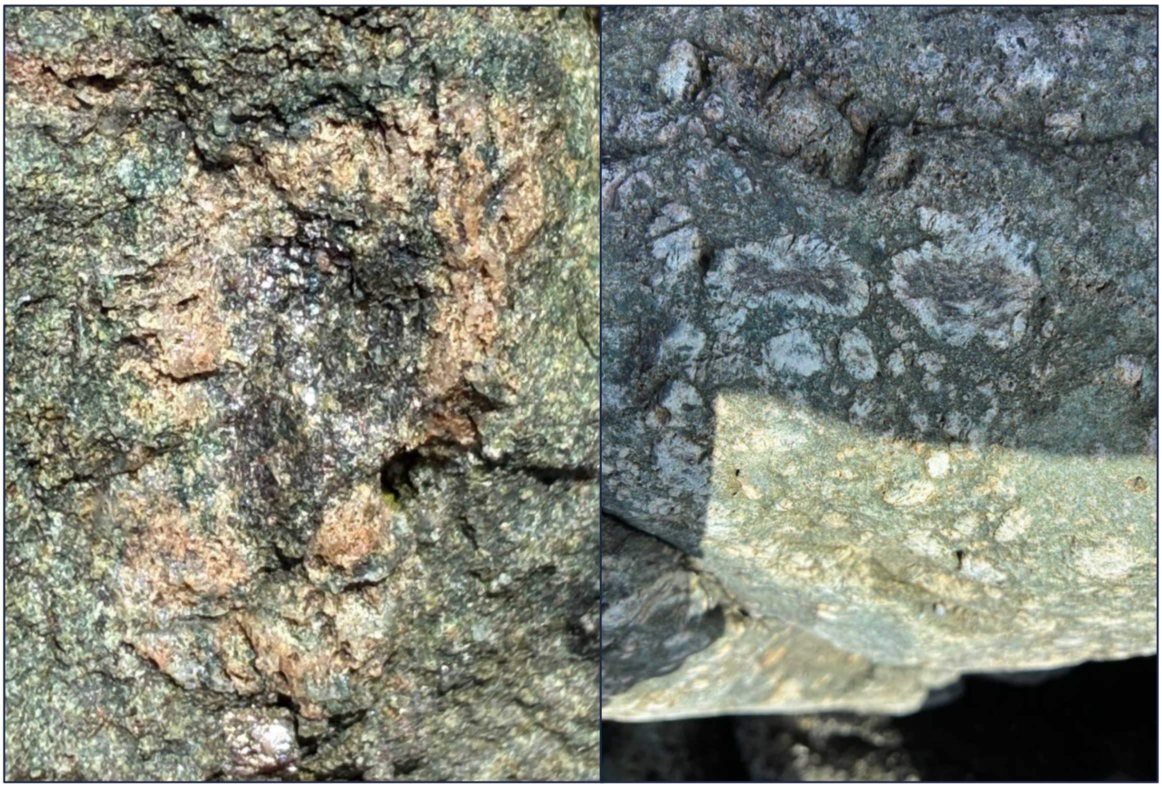


Fig 13. A) Chlorite replacement of plagioclase phenocryst.

4.5 Dip and Strike

The dip of the footwall was measured to ~ 29-32° towards the SSE. The headwall dip was measured to ~ 59-61° towards the SSE. The strike of the sill tended to follow a SWW-NEE trajectory. This was decided in field using a compass and confirmed by looking at satellite images on Google Earth. It is very similar to what is reported by Kleine et al. (2016) for other sills in the area.

4.6 Hematite within the interior

An area of particular interest is the one shown in figure 14. It shows an area of very high hematite concentration that is located below an area of high prevalence of the plagioclase crystals that are typical of the sill interior. The hematite rich area is located not within the foliated margin of the sill, but instead deeper down within the interior. This area is included in the footwall series 1, and is plotted in figure 9, showing that the hematite rich zone appears below 3 meters. The measured depth of the footwall series 1 from grids was 570 cm (7.5 x 76). The approximate depth from tape measurement and angles measured by clinometer was 555 cm. A possible interpretation is that this is a hematite rich vein seen from the side and found deeper within the sill. If it is a vein seen from the side, it is clearly cross cutting the stratigraphy, propagating perpendicular to the margin.

Footwall

Len of grid	7,5 cm
Num of grids	76
Measured length	570 cm
Sum grid length	555 cm
Error %	0,88%

Fig 14. A) Showing an area of very high concentration of large hematite crystals, as well as an area of very high concentration of large plagioclase phenocrysts. The plagioclase is concentrated and visible in the upper circle. The hematite are found with the lower circle. B) Zoomed out picture of a cross-section part of the sill. The arrow points to the place where the high hematite concentration is from A. The depth is between 3-4 meters approximately.

5 Discussion

5.1 Disconnecting the margins and hematite

The results of this field study are contradictory with the hypothesis that hematite was concentrated in the margins due to magmatic differentiation, presented by Kleine et al. (2016). As can be seen in figure 7, the hematite distribution across the sill appears not to be specifically correlated with the margin. Instead it appears as occasional spikes of very high hematite concentration in a groundmass normally absent of them. The spearman coefficients calculated for the hematite and carbonation concentrations yielded the values -0.31 and 0.12. These two values both indicate a weak, or low, correlation between the carbonation extent and hematite concentration (Gignac and Szodorai, 2016). The finding is relevant to this study due to the connection between the carbonation extent and the sill margins (Graham et al., 1983; Arghe et al., 2011; Kleine et al., 2016). The highly carbonated margins thus functions as a proxy for measuring the connection between hematite enrichment in the sill margins versus the sill interiors. The analysis shows that there is no clear connection between the carbonation of the margins and hematite enrichment. However, neither does is show a correlation between the absence of carbonation and hematite enrichment. Given the hematite concentration series illustrated in figure 7, with clear ‘spikes’ within the interior of the sill, the interiors of the sills cannot be said to be free of large hematite crystals. Combined with the finding that hematite is not correlated with the margins, the proposed cause of hematite enrichment within the sill margins as a result of magmatic differentiation seems improbable.

However, the general pattern of a metabasaltic sill where the interior is dominated by large plagioclase phenocrysts is confirmed by the data gathered in this field study, see figure 6. This pattern of distribution of plagioclase phenocrysts is probably due to magmatic differentiation processes, similar to the zoning examples in other basaltic sills, dykes and sheets (Mangan et al., 1993; Simura and Ozawa, 2006; Bédard et al., 2007)

5.2 The vein connection

As proposed above, the evidence from this field study suggests that the pattern of distribution of large hematite crystals, discernible in the field, is not due to the same processes of magmatic differentiation that caused the plagioclase phenocryst distribution across the sill. The areas with the highest concentration of hematite crystals are actually found quite deep within the sill (3-4 meters deep), away from the texture of the carbonated and foliated margins, and below areas that already contain large plagioclase phenocrysts. This means below areas belonging to the sill interior. For example, figure 14 shows the area of highest hematite concentration found within the sill, as well as an area of extremely high plagioclase concentration. The hematite rich region is here found *below* the region rich in plagioclase phenocrysts, at a depth of about three meters. This suggests that this area is well below the foliated and carbonated margin, that generally only reaches at most two meters (Kleine et al., 2016) and was found to only reach around 140 cm at the headwall side of the sill. One can also look at the distribution of large plagioclase phenocrysts across the sill. The pattern shows a steep increase at

1.4, 1.7 and 1.7 meters depth for the headwall, footwall series 1 and 2 respectively. This would suggest that the sill margins generally are less than 2 meter wide. This finding supports the claim that one of the most hematite rich regions is found within the interior of the sill, at a depth of 3-4 meters. In conclusion, the large plagioclase phenocrysts are spatially connected with the sill interiors, but hematite is not uniquely connected with the margins.

Looking at the data from the footwall, data series 1, shown in figure 9, there seems to be a negative correlation between the presence of hematite and plagioclase phenocrysts. A similar pattern can be seen in the complete profile presented in figure 8, as a combination of footwall series 2 and the headwall series. The complete plagioclase and hematite profiles can also be seen separately in figure 6 and 4 respectively. The pattern of correlation is not as clear in figure 8 as in figure 9. The spearman coefficient that was calculated for these two series show -0.17305 and -0.589, for the data in 5 and 6 respectively. The former is, although negative, not strong enough to determine a negative correlation. The latter is a much stronger negative correlation, considered relatively large, in agreement with the visual evidence of the plot (Gignac and Szodorai, 2016). By combining the spearman correlation with the visual clues of the plots it becomes apparent that there seems to exist a negative correlation between the concentration of plagioclase phenocrysts and of large hematite crystals.

Furthermore, there is evidence from the two large cliff faces of nearly planar perpendicularity that hematite crystals are heavily concentrated within specific areas, most aptly named veins (see figure 12). Out of all individual hematite crystals on both cliff faces, 97.92 % were found within veins and only 2.08 % were found elsewhere. It is worth noting that the hematite found elsewhere on the cliff faces were within close vicinity to the veins, at a distance of a couple of centimetres. The profile pattern of figures 4 and 5 also show a general complete absence of hematite within the sill, occasionally interrupted by spikes of very high concentrations of hematite, as one would suspect if the concentration was controlled by veins. These findings are contrary to what one would expect to find from a general zonation of high hematite concentration withing the margin, and low within the interior.

Simura and Ozawa (2006) describes veining systems related to magmatic differentiation processes, in the form of graded veins and rhythmic veins. The former can be likened to graded beds, and would show a size gradation of the phenocryst, which is not seen in these veins. The latter, the rhythmic veins, offers a more suitable potential explanation for the veining system in this sill. The rhythmic veins are regularly occurring veins, that appear parallelly to the contact zone. This is similar in appearance to the veins of cliff face B that have been identified as epidote segregations, which are a result of a different process (Skelton et al., 2010; Kleine et al., 2016). However, the veins in cliff face A are not parallel, and although they're generally not vertical, they're neither only horizontal/parallel to the sill margins. The two lowermost veins that are enriched in hematite in cliff face A meet each other at an angle of ca 40° in the lower left corner of the photo. This geometry, which is not aligning with the metamorphic textures, such as the foliation and margins, is likely formed during a later fracturing event. Therefore, these hematite rich veins in cliff face A, are more likely to be post-metamorphic.

Another interesting observation is that the appearance of the veins containing hematite is variable. The link between them is simply the high concentration of large hematite crystals. The differences can be seen when comparing the appearance of the veins in figure 12 (A, B and C) as well as the one in figure 11. An explanation for this could be that the process of hematite enrichment is not as much dependent on a specific mineralogy within a fracture or vein, but simply dependent on the existence of a reliable fluid pathway through the rock. If that pathway is a porous epidote segregation or a brittle fracture is of less importance than the shared characteristic of good permeability. Continuing this line of argumentation based on these findings suggests that the hematite enrichment occurred later rather than sooner, possibly entirely after metamorphism. It does, at least, undermine the idea of hematite enrichment as a consequence of magmatic differentiation.

The findings illustrated in figure 11, which is a zoomed in photograph of hematite enrichment in an epidote segregation, are similar to what is described by Plimer (1993). In the study epidote veins are described containing elevated levels of hematite and calcite, formed in rocks that have undergone retrograde metamorphism, as well as being hydrothermally altered in a submarine setting before metamorphism. Plimer continues to describe how fracture systems can act as pathways for fluid flows of oxidised hydrothermal fluids, that may precipitate veins of hematite + quartz + chalcopyrite ± gold ± epidote.

5.3 The fluid connection

The volume altering reactions that occur in epidote during metamorphism can lead to porosity enhancement in areas particularly rich in epidote. A great example are the epidote segregations found in the metabasaltic sills at Port Ellen (Skelton et al., 2010; Arghe et al., 2011; Kleine et al, 2016). The reactions are also connected to changing iron content within epidote grains themselves (Arghe, 2011). Spilitization is an open-system process, where elements can be displaced distances up to 10 meters. For example, iron can be mobile within epidote segregation systems, at least locally, as they are being linked to gradational differences between rim and interior of the segregations. The dark rims have higher iron concentrations than the lighter interiors (Skelton et al., 2010). This process could possibly be linked to the presence of large hematite crystals found in the veins, as evidenced by figure EPI. A study by Hung et al. (2020) describes how low temperature sea floor hydrothermal alteration is linked to the formation of fine-grained epidote veins with lesser levels of chlorite, quartz and *hematite*. This is a potential source of the hematite enrichment, as it is similar to the spilitization that is confirmed to have affected this sill and many of the sills in the area.

However, if the crystals precipitated within epidote segregations as a direct consequence of spilitization, why are large hematite crystals found in other fractures that are not epidote segregations? To explain that would necessitate two different genesis processes.

Another possible explanation is related to the fact that epidote segregations are thought to have functioned as fluid pathways for later fluid intrusions, e.g. carbonation fluids, intruding the sills after peak greenschist metamorphism (Kleine et al., 2016). These retrograde- or exhumation-related fluids would have been structurally directed by deformation textures (Skelton et al., 1995) and preferentially have permeated through features of increased porosity and permeability, such as the epidote segregations and the foliated margins (Skelton et al., 2010; Arghe et al., 2011; Kleine et al., 2016).

This justifies the idea that hematite enrichment might have been connected to carbonation fluids, or some other infiltration of fluids, after peak metamorphism rather than magmatic differentiation. Look at cliff face B in figure 10, with hematite heavily concentrated within the epidote segregations. If the carbonation fluids preferentially flowed within these epidote layers and other veins, as well as in the foliated margins, then precipitation of hematite crystals is possible. This is assuming that the carbonation fluids did contain iron. The common denominator between the hematite rich regions seems to be increased permeability creating fluid pathways, such as fractures and older veins.

A connection between the margins, based on carbonation data, and the concentration of the hematite crystals is not supported by the field evidence from this study. However, the hematite might still be linked to carbonation. There is data supporting the fact that some parts of the margin do contain higher concentrations of hematite crystals than much of the rest of the sill, which can be seen in figure 7. There was a plan to test this in the field, but it was not possible due to equipment failure and time constraints. An attempt to solve this problem was made by testing the reactivity of the veins containing hematite with 10% HCL. This test did show that the veins were more prone to reacting with 10% HCL than non-vein areas on average. However, the test is flawed in the sense that any exposed surface containing some calcite will react. This means that the relative amount of calcite, which is the main carbonation mineral in these sills (Arghe et al., 2011; Kleine et al., 2016), could not be determined. A fact that undermines any conclusion based on this data set, considering that varying levels of calcite was found throughout the sills (Kleine et al., 2016).

5.4 Drawbacks, setbacks and improvements

The constraint of time limited the possible extent of the survey. Given more time, more profiles could have been produced, and more time would also have allowed more carbonation data to be collected.

During the field work some complications occurred. One was the equipment failure of the "fizz-o-meter". This caused problems with the data gathering of the carbonation extent of the sill. For example, reliable data of carbonation of the cliff faces and veins could not be collected. As this was by the end of the field study, there was no time to seek replacement on Islay. Islay is generally lacking a wide range of options for tools, especially in my vicinity and being without a car. The solution was to test for reactivity with HCL on fresh surfaces, but the method is not very reliable and cannot reveal much about a potential connection to the carbonation of the sills.

The impact of tidal change also affected the work. In the field, the best areas for constructing the grid profile was at certain times of the day not safely accessible. However, as the quality of these areas were superior to the more accessible ones it was still worth adding this constraint.

The plan also changed towards the end of the week. The last profile series constructed, the footwall series 2, was also the series where it became apparent that there might be a connection between the veins and the hematite. This led to the decision to gather data from the two cliff faces. This decision also meant that less grid profiles were constructed, due to the time limitations.

5.5 Suggestions for further studies

Firstly, more sampled profiles would improve the reliability of the results. The results would also benefit from more data on the connection between veins and hematite. It would be especially interesting to further look into the mineralogy of the veins and the if they are carbonated or not. It would also be of interest to collect and prepare thin sections for analysis. Lastly, is the hematite enrichment a result of external fluid fluxes or local redistribution of an original iron enrichment?

6 Conclusions

- The negative correlation between the presence of hematite and plagioclase fit well with the finding that hematite is almost exclusively found in veins. This is due to the fact that the veins are either filled in fractures or epidote segregations and as such they would naturally be absent in large plagioclase phenocrysts, even within the interiors.
- The spatial distribution of hematite is found to be vein controlled rather than associated with the margins.
- The fact that hematite has precipitated in both epidote segregations as well as other brittle structures, suggests that the event was post greenschist facies metamorphism. This is based on the timing of the porosity increase of the epidote segregations, which occurred during greenschist facies metamorphism, as well as the occurrence of post-metamorphic fractures rich in hematite on cliff face A. The next known fluid infiltration event is the extensive carbonation event that occurred during retrograde metamorphism. The hematite precipitation could be related to this fluid event, or to later fluid events.
- The hematite enrichments is not related to the margins via the mechanisms of magmatic differentiation in the same way that the plagioclase is related to the interiors. The hematite found within the margins are more likely due to fluid infiltrations that preferentially flowed along the foliated texture of the margins.

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