

## ***Interactive comment on “Quartz dissolution associated with magnesium silicate hydrate cement precipitation” by Lisa de Ruiter et al.***

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Response to reviewer 2

We thank the reviewer for constructive and helpful reviews and are pleased to read that the reviewer finds the manuscript interesting.

Below, we have repeated the questions/concerns of the reviewer followed by our response.

Quite a few observations presented in the Results section are interpreted in terms of their significance straight away (i.e., within the Results). The Discussion, then starts with the statement that ‘quartz grains with a diameter of 50  $\mu\text{m}$  within the cemented rocks are completely dissolved and (partly) replaced by magnesium silicate hydrate

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cement’ (L. 203-204). The latter interpretation may well be correct, but should follow on from discussing the observations. The paper would be stronger if the Results contained only minimal interpretations, and the Results started with a section that pulls together the key observations and comes to the conclusion as quoted above.

Response: We have rewritten part of the results and transferred interpretation to the discussion. We have removed the interpretation of the texture being the result of dissolution and replacement of quartz to the discussion and elaborated on the discussion of the observations.

Protolith: A bit more detail on the nature of the protolith would be useful: the different components are provided, but little detail on their relative abundance and size (other than ‘large’). As a result, it is difficult for the reader to form a mental image of what the till and cemented rock look like.

Response: We have added general information in the Geological setting to be clearer and more precise about the nature of the till and the cemented rock. Since both reviewers have questions about the protolith, we elaborated on that in the revised manuscript and added details on the components and abundance etc. We also present whole rock geochemistry of the till that locally covers the ground and which we interpreted as the protolith of the cemented rock. The analysis, (see supplement to this comment), demonstrates that the till is silica rich and the nearby cemented rock is similar in composition to the till, with a higher MgO and LOI content. This supports our interpretation that the M-S-H cemented rock is formed by adding MgO and H<sub>2</sub>O to the till and that the SiO<sub>2</sub> source is the till itself and has not been added. We have added a paragraph 4.3 on geochemistry and discusses this in 5.1 (this paragraph is copied below).

Deformation history: L. 125-126: ‘Many quartz fragments are characterized by small equigranular polygonal new grains with straight grain boundaries together with large old grains with undulose extinction and sometimes subgrains’. Neoblasts (for new) and unrecrystallised (for old) may be more appropriate terms?

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Response: We have changed the text in accordance with the suggestion.

L. 128-130: 'In addition, essentially all quartz grains, including the dynamically recrystallized grains, show undulose extinction, which indicates that the quartz was plastically deformed by dislocation processes again in a later stage'. It's not clear to me why undulose extinction requires a separate, later phase of deformation; could this not simply relate to the initial recrystallisation process itself?

Response: Recrystallization (=deformation phase 1) should lead to non-deformed crystals, which must go through a second deformation stage to become deformed and have undulose extinction. However, we agree that in practice this might not necessary be the later deformation phase and have changed the wording of the sentence.

Fig. 2c: this is a histogram of 1730 quartz grains in the cemented rock, implying an upper grain size limit of 45 micron. It is not clear to me how this relates to the photomicrograph shown in panel a, where there are clearly visible grains of up to 300 micron. Panel A is from a non-cemented rock, but what would have happened to the large grain during cementation? Presumably they would be more resistant to reaction than the small neoblasts?

Response: The histogram is about the newly recrystallized quartz grains and subgrains and does not include the 'old' grains which are present in panel A. Occasionally, we find these non-recrystallized grains within the cemented rock. However, the recrystallized grains appear to have a significant role in the cementation process as we can often find them partly or completely being dissolved within the cemented rock. Therefore, the histogram focusses on these grains only. The figure caption has been changed to avoid further confusion.

Microtextures: L. 148-151: 'When the cement has replaced the outer few  $\mu\text{m}$  of the grains, the dissolution is commonly no longer accompanied by cement precipitation, as indicated by the presence of honeycomb-like pore spaces, after the shape of quartz grains, in which sometimes relicts of quartz can be observed (Fig. 3c)'. Is there a

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possibility that these pore spaces did in fact contain quartz but that these grains have been plucked out of the sample during sample preparation?

Response: We are aware that sample preparation may create this texture that may lead to erroneous interpretations and we cannot be very sure that quartz has not been plucked. However, the delicate honeycomb texture is perfectly preserved, and it is difficult to see how the quartz can be plucked without damaging the honeycomb texture. Besides, the texture is also visible in the SE images of figure 4, where we used whole rock pieces that have not been through the process of cutting or polishing.

L. 182-184: 'the etch pits density is  $10^{10} \text{ cm}^{-2}$  with the reacted surface being larger than the non-reacted surface'. Two questions: 1) the text provides the etch pit density, but in the caption to figure 6 the same number is quoted as the dislocation density. Please clarify. 2) Is the sentence trying to say that the etch pit density is larger on the being larger on the reacted surface than on the non-reacted surface, or is it referring to the actual physical size of the reacted vs non-reacted surfaces? The caption is indeed not correct, this should be the etch pit density and has been corrected. The sentence was supposed to mean that the majority of the surface has been reacted, so that there is more reacted (physical) surface than there is non reacted surface. This is indeed not clearly written and has been improved.

Time line: 'these cemented rocks occur in the mine tailings of mines that were active until about 100 years ago, indicating that the grains dissolved in less than 100 years' (L. 205-206). This is only demonstrably true if the cement occurs between different rocks of the tailings pile, rather than within the individual rocks on the pile. Please link back to the description of the occurrence of the cement (e.g., on the wall of the tunnels) and provide a robust timeline.

Response: The cement does indeed form between the different rocks of the tailing pile, we added this to the description. MSH cement forms also outside the mining areas, typically where the soil develop openings due to mass movements downhill and on the

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downhill side of boulders where the groundwater can evaporate. For such cases, the cement can have formed in the period between the glaciation and present. Where the evaporation sites are created by the miners, we argue that the process started at the time the miners dug their trenches. Since the cement is developed in the evaporation zone and not behind this zone, we feel confident that this cement is formed post mining activity.

Reaction history: 'the fluid can still access the quartz surface since the cement is porous' (L. 363-364). Isn't the amorphous silica in the way? It has already precipitated in step 3 of the process as described. One of the aspects that is not discussed is a possible volume change during reaction. Do you have any constraints on this? It is possible that the honeycomb texture of MSH between polygonal grains is in part due to a positive volume change creating pathways for the reactive fluids to penetrate the quartz? This may provide an additional way in which the reaction proceeds, and could also contribute to the high reaction rate.

Response: We thank the reviewer for this very constructive question. We have added this possibility to the revised manuscript. It should be considered that, as is explained in the discussion, amorphous layer precipitation is usually thought of as slowing down or ceasing dissolution, since it covers the reactive surface. However, in multiple studies (e.g. Ruiz-Agudo et al., 2012) it is suggested that if the amorphous silica reacts to form a secondary phase (in this case cement), the surface is exposed again which leads to more dissolution. This would mean the amorphous silica is not in the way, as it will react to form the cement.

Figures: Fig. 5: In the caption, please describe the type of image this represents (SE/BSE/TEM), and whether or not this is a polished surface. Response: This is now added.

Fig. 7: In the text, it is written that 'these fibres of the cement are attached to and partly intergrown with the amorphous layer' (L. 191). Please highlight those areas in Fig. 7

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where the reader can make this observation. Response: This has been added.

Writing: L. 14: as a result L. 45-47: 'Also, the recent findings of De Ruiter and Austrheim 45 (2018) indicate dissolution of quartz in natural high pH conditions that is much faster than experimental studies and rate equations predict for the relevant conditions.' Suggested phrasing: 'Also, De Ruiter and Austrheim (2018) recently found that the dissolution of quartz in natural high-pH conditions is much faster than experimental studies and rate equations predict for the relevant conditions.' Response: This has been changed

L. 51, 90, 104, 206, 209, 217, 228, 241, 353, 359, 388: please use subscripts and superscripts where appropriate Response: We have changed this

L. 94: where=were Response: changed

L. 93-95: 'It is furthermore unlikely that the cementation started before the mines where abandoned in the 1920's, as this would influence the mine tailing in which the trench is present and would have made it unlikely that the cement is only present on the outer few cm of the trench.' This sentence is unclear to me; please rephrase. Response: We have reworded this sentence.

5.1. Geochemistry and M-S-H formation Till is produced by mechanical weathering and is assumed to produce a robust average composition of the upper continental crust (Goldschmidt 1933, Gaschnig et al. 2016). The composition of the till (Table 1) show no or little geochemical signature from the underlying ultramafite and suggest that the glacier must have collected an area much wider than the Feragen ultramafite. We can therefore not relate the till to a nearby lithology. The similarity in composition between the till and the M-S-H cemented tillite for most oxides, suggests that the till is the protolith to the M-S-H cemented tillite. The composition (reduced SiO<sub>2</sub> and increased MgO and LOI in the tillite compared to the till) gives support to the textural observation that quartz is replaced by M-S-H cement through a dissolution precipitation mechanism (Putnis 1992). We relate the cement formation to the weathering of the

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peridotite. Brucite ( $\text{Mg}(\text{OH})_2$ ) is leached during the weathering and leaves a  $\text{SiO}_2$  enriched weathering rim (Ulven et al 2018). The silicates remain inert during this process. The geochemical data is in accordance with this as there is no indication of Si addition. The  $\text{SiO}_2$  present in the cement must come from the till.

Please also note the supplement to this comment:

<https://se.copernicus.org/preprints/se-2020-34/se-2020-34-AC2-supplement.pdf>

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Interactive comment on Solid Earth Discuss., <https://doi.org/10.5194/se-2020-34>, 2020.