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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 1.

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

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

1) While I’m inclined to believe the mechanisms outlined in the ms from the evidence provided, have the authors considered that the source of Si may be from the parent rock rather than the dissolving quartz grains. Is there any possibility that some of the Si originates from there?

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Response: We cannot totally exclude that some of the Si comes from weathering of primary minerals, but we observe hydro-carbonates where quartz is absent in the mine tailings and in the mine shafts. The MSH cement forms in the till where quartz is abundant. This allow us to conclude that quartz must provide Si for the cement otherwise we cannot explain that the tailings do not develop cement (see discussion). The textural relationships e.g. the honeycomb texture with quartz relicts and abundant etch pitch on quartz also suggest that quartz provided Si to the cement. In the revised version, we say that some Si may come from the weathering of silicates in the ultramafic rocks, but that Quartz must be present to form M-S-H cement. This is also supported chemistry of the surface water in the area (which is rich in Mg but not in Si) and the composition of the till (see table 1 in supplements) compared to the composition of the cemented rock. This shows that the compositions are very similar, except that the cemented rock has a much higher Mg and LOI than the till. This corresponds to the idea that the cemented rock is indeed formed from adding Mg and H2O to the till and not by adding SiO2, which is already abundant in de till (this is now explained in the discussion part, which we copied at the end of this document).

2) The chronology behind deriving the weathering rate seems speculative (i.e., the quartz only started dissolving following the closure of the mine. It’s not clear how the rate was derived, was it calculated from a shrinking core model?

Response: MSH cement forms also outside the mining areas, typically where the soil develop openings due to mass movements downhill and on the 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. We further infer that the etch-pits and quartz dissolution relates to the cement formation. The mining activity at Feragen started in 1824 which leaves 200 years for cement formation and not 100 as argued in the earlier version. This has been changed throughout the manuscript and dissolution rates has been recalculated accordingly. The dissolution rate has been calculated using the dissolved radius (as observed from microstructures with SEM), the molar volume of quartz and time during which the dissolution has taken place, as explained above (see Lasage 1984 for the equation). It should be noted that using a timeframe of 100 or 200 years wouldn't change the order of magnitude.

3) The overall evolution of the ground is not well described. This can be improved in the 'geological setting' section. What is the source of the glacial till? When did the primary minerals weather (if they weathered post-glaciation, could they provide some of the Mg to the cements?). How does ground water flow into the till, and what is its chemical composition?

Response: Both reviewers ask for the source or the protolith of the glacial till. In order to answer this, we present geochemistry of the till and its nearby cemented rock/tillite and add a table in the appendix. The till has elevated SiO2 contents and no clear geochemical signature from the underlying peridotite. When comparing the chemistry of the till and cemented rock it can be observed that the MgO and LOI are higher in the cemented rock. This is in accordance with our model for cement formation. As pointed out in earlier papers from this area, brucite is easily weathered and is absent in the weathering rind, leaving a Mg depleted and Si enriched residue. We thus feel confident that brucite is the main source of Mg, but we cannot exclude that some Mg is derived from silicates. The chemistry of the groundwater has been reported by Beinlich & Austrheim (2012) and shows a relatively high concentration of Mg and a high pH. This corresponds to the idea that brucite weathering is an ongoing process, providing most of the Mg for the cementation process. Fig 1c is an attempt to explain the groundwater flow and we can add to that that the cement forms where the groundwater is able to evaportate. This occurs where soil movement creates cracks and in small puddles next to boulders as also explained in De Ruiter & Austrheim 2018.

4) In similar environments, when Mg-OH ground waters contact the atmosphere, the

formation of Mg-carbonatets are typically observed (see mine tailings e.g., Turvey et al., 2018 International Journal of Greenhouse Gas Control V79, or the classic O'Neil and Barnes 1971 GCA). Is it not strange that Mg-carbonates were not observed here? Especially if evaporation was your primary mechanism for driving precipitation.

Response: The tailings from the Cr-mines are carbonated with hydro-carbonates (nesquehonite, dypingite and landsfordite). The carbonation starts 5-10 cm below the surface. Hydro-carbonates also coats the walls of the mine shafts as described by Beinlich and Austrheim 2012. We have added on the occurrence of hydrocarbonates in the geological setting. In the till where quartz is the dominant mineral carbonates are missing and msh cement is formed. This must mean that the presence quartz prevents the formation of hydro carbonates. Although he reason for this is unclear, it has significant consequences. The Si in the MSH cannot be derived solely from the weathering of primary silicates (see question 1 above). The presence of quartz will also prevent CO2 sequestration as discussed under application (section 5.5).

5) Section 5.5., regarding the applicability of the results need substantial expansion. How could M-S-H cements be used and how does this information in the ms held constrain this use? What is the long-term fate of M-S-H (e.g., potential for carbonation or weathering?).

Response: We have added on the role of H-M-S in CO2 storage with reference to Ninomiya et al. 2007 and Turvey et al 2018 and discussed our findings on this background as outlined under point 4 above.

We also addressed the specific comments in the text and have modified the text according to the suggestions. However, we lack answers to some of the questions. The question about mechano-chemical activation through deformation below the glacier is an interesting question we are working on, but at present we cannot say for sure how this influences the process. Glaciers leave shiny surfaces referred to as Blankenberg. To our knowledge, there has been no microstructural study of such surfaces. It would

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be an exciting result if glaciation could precondition quartz to increase solubility and reaction. We have added a sentence saying that we speculate if this may be the answer to the enhanced dissolution.

References Ninomiya, J., Mizuochi, Y., Katoh, T., Okamoto, M. and Yajima, T., 2007. CO2 fixation in the serpentine-groundwater system. Nendo Kagaku (J. of the Clay science society of Japan), 46, 28-32 Turvey, C.C., Wilson, S.A., Hamilton, J.L., Tait, A.W., McCutcheon, J., Beinlich, A., Fallon, S.J., Dipple, G.M. and Southam, G. 2018. Hydrotalcites and hydrated Mg-carbonates as carbon sinks in serpentinite mineral wastes from the Woodsreef chrysotile mine, New South Wales, Australia: Controls on carbonate mineralogy and efficiency of CO2 air capture in mine tailings. International journal of greenhouse gas control, 79, 38-60

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 underlaying 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 SiO2 and increased MgO and LOI in the tillite compared to the till) gives support to the textural observation that quartz is replace by M-S-H cement through a dissolution precipitation mechanism (Putnis 1992). We relate the cement formation to the weathering of the peridotite. Brucite (Mg(OH)2) is leached during the weathering and leaves a SiO2 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 SiO2 present in the cement must come from the till.

Please also note the supplement to this comment:

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https://se.copernicus.org/preprints/se-2020-34/se-2020-34-AC1-supplement.pdf

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