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Interactive comment on "Constraints on Enhanced Weathering and related carbon sequestration – a cropland mesocosm approach" *by* Thorben Amann et al.

Thorben Amann et al.

science@thorbenamann.de

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Dear Søren, we are grateful for your extremely thorough review of our manuscript. In the following we document the changes that we made according to your suggestions, based on the text that you wrote. Annotations that you made in the original manuscript were considered and changed directly without additional documentation here, unless we found it to be necessary to be explicitly addressed.

Reviewers comment

Our reply

1. A set of CO2-sequestration rates makes use of a correction for preferential flow. I find this problematic. The underlying reasoning for factoring up the inferred CO2- sequestration is that lots of weathering take place in the statically held back solution, which is allegedly continuously bypassed by macropore flow. However, even in the case of a dominantly preferential flow system, the net weathering rate would still be a function of the effluent water flux multiplied by its weathering product concentration and a stoichiometrical coefficient. The deduction of a preferential flow factor therefore is, as far as I am convinced, purposeless for estimating the CO2 -sequestration.

This is correct for the net flux from the mesocosm in this experiment. However, we point out, that this is a relevant process to be considered in nature, possibly biasing results from field experiments if trials are compared to controls. We tweaked the wording in the direction that we don't want to publish "corrected" CO2 sequestration rates, but that we can learn from the experiment, that there is a process potentially influencing the results, which is specifically important if those are used to estimate method potentials or even CO2 sequestration values for CO2 capture rewards.

Furthermore, elsewhere in the manuscript the authors state that "extended periods of drying out" resulted in "slowed down or ceased chemical weathering processes", which is quite the opposite to the assumption made above of quantitatively important reactions in zones of static water. So, there is a contradiction here which also needs to be solved.

We think by modifying the text according to your remarks above, this point is implicitly addressed.

2a. The manuscript concludes that CO2-sequestration "was shown" (without the above correction). I disagree: DIC needs to percolate to the groundwater table and beyond, and/or form (stable) carbonate minerals and/or org. C stocks. The latter two are not measured, and effluent DIC concentrations are (according to

the manuscript) measured too infrequently to be applicable. The use of Mg as a proxy for DIC need to be carefully documented by data (not just by theoretical reaction stoichiometry) before it can be used as direct evidence.

Mg was compared to controls, so only Mg from applied material is released and used to estimate CO2 consumption. To improve the chemical background understanding, we added a weathering reaction formula for forsterite in section 3.3 and some points in the introduction, addressing your next remark at the same time.

I think the authors should include a discussion (in the Introduction) of the requirements for enhanced weathering to actually be achieved.

The dissolution of silicates stores CO2 in form of alkalinity in oceans on longer timescales. This process previously mentioned somewhat indirectly and is now elaborated a bit in the introduction.

2b. In my opinion, the 'fixes' for the manuscript to 1) and 2) above might include to acknowledge that the elements under consideration (e.g., Mg) are non-conservative, and that the retarding processes are not investigated mechanistically in this study (at least, the data were not shown). Therefore, the preferential flow-calculation (which uses Mg) must be skipped, and the tight conclusion regarding the sequestration most be softened.

Yes, there is a potential influence of plants and other processes (removal by cation exchange and precipitation). However, we try to give a first order estimate for the process of preferential flow to point out its potential to change the impact of CO2 sequestration or weathering estimates. Rebuttal Fig. 1 (reply to your remark 5c) shows that if we add the error bars (standard deviations) the differences between crop and no crop treatment are not significant. Nonetheless, we changed the discussion to point out that we do a rough estimate rather than a process based detailed analysis.

While the sequestration cannot be said to be "shown", in my opinion, I do

think the authors could safely say that their results 'indicate a potential' CO2-sequestration of X t C/ha/yr.

We concur and toned down the wording a little bit here and there.

3. A thorough analysis of the water balance for the mesocosms needs to be presented. The water balance needs to include an evaluation of the transport time for water through the mesocosms.

The water balance data we have is shown in Fig. 2 of the MS. The transport time of water through the mesocosm was not tracked. We don't have any other information.

4a. The overall purpose of the study appears to be slightly blurred. The focus on estimating CO2-sequestration rates infers that this was the main aim. (However, one must then ask why DIC in the effluent was not more carefully measured, ie., what were the mesocosms designed for?)

This is a very good point, which we didn't address originally. The experiment was designed to evaluate elemental cycles in typical crops affected by rock powder application. The idea to evaluate weathering fluxes came in later. Therefore, the experiment seems not to be designed towards the questions raised in the manuscript. Also, this explains why DIC data is patchy: We could only use what was left over. To justify the "purpose-deviating" setup, we added a sentence in the beginning of the methods section.

4b. Another purpose appears to be to demonstrate the use of dunite as a "model mineral" for enhanced weathering experiments. (But then, why the strong focus on the estimation of CO2-sequestration rates and trace elements, which implies a focus on field applicability?)

We use the mineral because of its relative simplicity in terms of geochemical composition and thermodynamic response. As the complexity of weathering effects, resulting from deployment of large amounts of small grain size powder, is not understood at all at the moment, we abstain for the moment from a more complex mixture of elements and minerals like in basalt or other more differentiated rock types. Yes, it is unlikely that dunite is used, if EW is deployed, but it can be a good starting point to identify the various pitfalls of field deployment. The overarching aim of the manuscript is to work out some general statements of what must be considered in future experiments, let alone open field deployment.

4c. The authors should state the objective(s) more clearly.

We sharpened the text, especially the last paragraphs of the introduction to be more precise.

5a. Presentation and structure: The manuscript needs to undergo a major revision in terms of the structure, conciseness of the text, and its figures. For example, the manuscript contains many repetitions and many imprecise statements. Also, many results are presented in the Discussion and some discussion take place in the Results section. Some results were not presented (or did I miss them during my reading?) but were still referred to/used in the Discussion.

We took your advice and reworked the text.

5b. The artwork needs to be polished; generally, the figures in the supplementary information seem to be better worked through than the figures in the manuscript, although the depths (e.g., cm below soil surface) needs to be added in the supplementary material, rather than using a 'depth number'.

We tried to streamline the artwork a bit. However, due to the complexity of the data, it is hard to find a good way of presenting it. We are open for further suggestions. The issue of depth indications was fixed in all figures and also in the text.

5c. Five replicates for each treatment combination were conducted, but this need to be visualized by statistics in the figures.

In general, we agree. However, due to the complexity of the data, we think the ab-

sence of error bars in the main text can be justified. It seems unreasonable to put the appendix figures into the main document, since it would expand the document significantly. Exemplary, we created the figure for Mg concentrations as in the manuscript this time including error bars (Rebuttal Fig. 1). As can be seen, the addition of error bars decreases readability, but does not influence the major conclusions from the mean values presented in the figure in the main text.

Rebuttal Fig. 1: Recreation of Fig. 5 of the main manuscript to show readability reduction through inclusion of error bars. Green: fine olivine, red: coarse olivine, blue no olivine.

Comments within the manuscript

Please use lower case: "enhanced weathering", to comply with the most frequent use in the literature.

We explicitly chose to write it capitalized to indicate that we don't talk about the process but a "method" of carbon sequestration. In previous studies this style of writing was already adapted, to show the difference between the geological process and the method.

The authors should comment on the differences between "unplanted+olivine" and "unplanted-no olivine". Could indicate significantly different storage properties. If so, it is not a total game changer, but still needs to be understood/explained.

It looks truly interesting if we leave away the indicators for standard deviations. Yet, the variance is very high and a statistical test (Mann Whitney U) reveals that differences are rarely significant at the 5

Rebuttal Fig. 2 Exemplary comparison of statistical differences between outflow volumes of mesocosms with fine, coarse and no olivine treatment. Mann-Whitney-U p-Values below the red line are <0.05 and therefore considered to indicate a significant *outflow volume difference in treatments.*

This [preferential flow effect; author's note] **is an important point in the manuscript. The underlying reasoning (for factoring up the inferred CO2-sequestration) is that lots of weathering take place in the still standing solute which is claimed to be continuously bypassed by the macropore flow. However, the net weathering would still be expressed by the effluent water flux multiplied by its weathering product concentration and a stoichiometric coefficient. The deduction of a preferential flow factor therefore is, as far as I am convinced, useless for the purpose of estimating the CO2-sequestration. [**...**]. Preferential flow may play a role, but will not change the final weathering rate. DIC needs to percolate to the groundwater table and beyond, and/or form (stable) carbonate minerals within the soil. The latter is not shown.**

This is an important point. The final weathering rate is of course determined by what leaves the system. This is why we provide the "observed" sequestration rate. Also, we don't specifically mention the higher weathering rate, factoring out the preferential flow effect, in the main text of the discussion, to avoid inconsiderate use of the number. However, we think that the experiment points out nicely, that there are probably processes in play, which were not considered so far, and we suggest that preferential flow is one of them. To point out the differences more clearly, we changed the wording, to be more precise and to avoid misunderstandings in the direction of calculated sequestration rates. Since we changed a lot of words all over the manuscript, it is not possible to point out specific sections.

What the authors state [in the hydrology section; author's note] is that there was no relation between irrigation scheme and weathering rate, CO2 uptake, etc. (all other monitored parameters). This is an important result! It should be emphasised more and data or statistics should be presented for support.

We created an exemplary figure to show the differences resulting from the two rain

regimes and added a remark in section 3.1 and supplied the comparison in the supplementary material (Suppl. Fig. S2-1). It can be deduced from the Mg data (selected as example), that there are some points where differences between rain treatments are significant but generally, they are not. Furthermore, there is no systematic deviation between the rain treatments. This is why we decided to lump them together in the study at hand.

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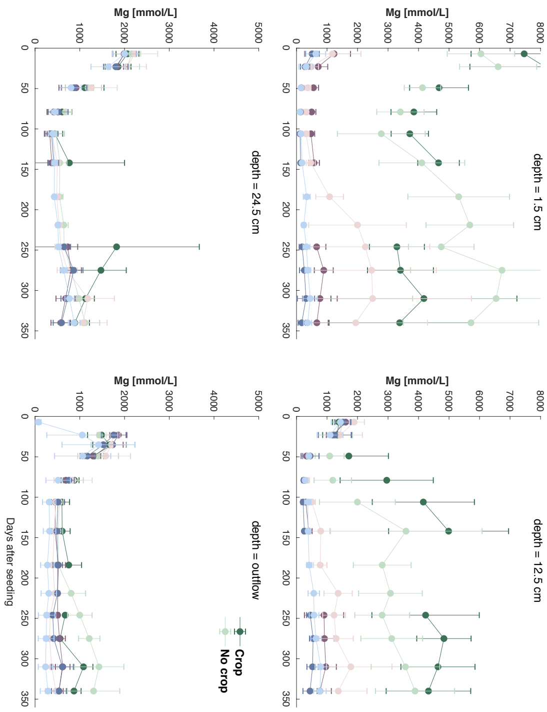
depth = 1.5 cm
depth = 12.5 cm
depth = 24.5 cm
depth = outflow
Mg [mmol/L]
Days after seeding
Crop
No crop

Fig. 1.

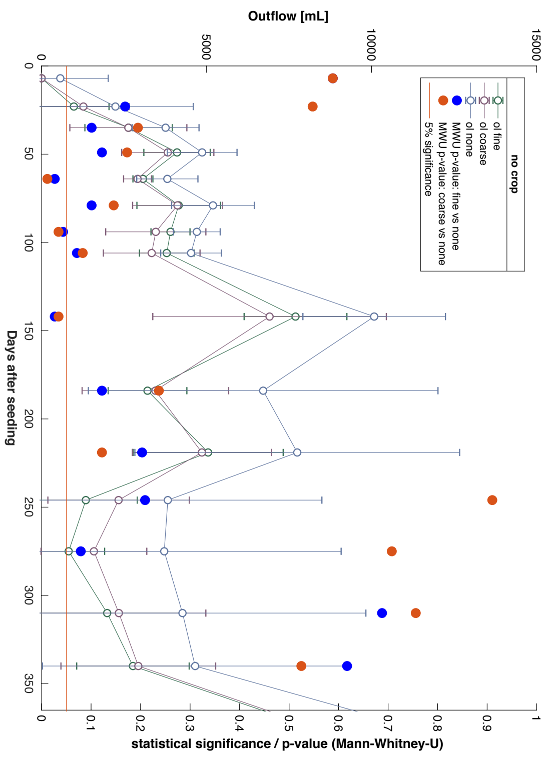
no crop
ol fine
ol coarse
ol none
MWU p-value: fine vs none
MWU p-value: coarse vs none
5% significance
Outflow [mL]
Days after seeding
statistical significance / p-value (Mann-Whitney-U)

Fig. 2.