Review of Cipolla et al EGUsphere-2022-196

General comments

Enhanced weathering (EW) is a biogeochemical carbon sequestration strategy which is currently gaining interest in the light of climate change. This paper considers several case studies of EW in the USA and Italy, and considers important aspects of soil moisture and rainfall effects which have been largely neglected in previous studies. The authors have used an existing EW model (Cipollo et al. 2021, *Adv. Water Res.* **154**:103934) to determine the seasonality of leached bicarbonate and carbonate as the metric of carbon sequestration. Their weathering rates and carbon sequestration rates are within the range of other studies, but water fluxes (precipitation, evapotranspiration) reducing soil moisture greatly reduced leached carbon during the growing season.

Specific comments

Generally, this is a good study but there are a number of omissions in the manuscript that need to be rectified. For example, no details are provided about the olivine application rate or whether the application was repeated. More detail about the tuning of the model, e.g. the background weathering rates, is required; it is unclear whether cation exchange capacity or other parameters were also tuned. The chemistry of rainfall and irrigation affecting key aspects of the model, such as the effect of soil moisture on pH and weathering, needs to be clarified. Comparison of the grain-scale weathering rates to those of Amann et al (2020) should be revisited as it seems the modelled rates are actually higher rather than similar. Some of the figures do not include enough information or could be improved by better labeling. These weaknesses should be easy for the authors to address; places in the text where these specific aspects should be addressed in the manuscript are detailed below amongst the English language and other technical corrections.

Technical corrections

Figures

As many readers may look at the figures without reading the text beforehand, it would be helpful to make it easier to understand the main elements of the story at a glance, with an indication of where more information can be found in the text. As they stand, some of the figures (especially the heatmaps) currently require careful study and references to the text to understand them.

Figure 3. Rainfall frequency is noted as a reason why Iowa has higher weathering rates than California, and it would have been useful to see λ for the states superimposed. The top row could show λ for Padan, Sicily and IA, CA and the bottom row could show α for the same cases. This arrangement would still allow readers to compare α and λ seasonality for individual sites.   In any case, please make it clear what α and λ are. Y axis labels should read "Mean rainfall α [mm]" and "Rain frequency λ" or something similar. In the caption, refer readers to section 2.2.1 for more details.

Figure 4. Y axes labels for Kc should read "Crop coefficient Kc". In the caption, clarify that ADD is added organic carbon. Refer readers to Section 2.2.3 for more details.

Figure 5. It would be helpful if the labels "Corn Clay Loam" etc could appear on the left of each row of this figure, the individual panel colorbars were removed, and a single colorbar with legible label e.g. "soil moisture ratio S_{IA}/S_{CA}" appeared at the top of each row. Not all software makes it easy to do this but it would greatly improve the readability of the whole figure. The bottom row lefthand label would be "Average daily ratio" and the individual Y-axes labels would be e.g.

"S_{IA}/S_{CA}". The label for column 3 should make it clear what scale is being considered, e.g. "Grain scale weathering rate ratio".

Figure 6. Please add letter designations to the panels (a, b, c, d). The Kc panel Y axis label should say "Crop coefficient Kc" and that for the bottom left panel should say "Specific soil moisture s".

Figures 7, 8, 10: Similar remarks as for Figure 5. Please add column and row labels. Room for these labels will be available if the individual colorbars are removed and master colorbars for each column appear at the top with the column labels. Also, make it clear that the weathering rates are per mineral surface area rather than per land area, e.g., mol/(m^2 olivine s) in Figure 10.

Figure 11. Are the samples contributing to the boxplots monthly values from the ten years of a single run, or from several runs where some parameter(s) varied? Which crops and soils are involved in these simulations; which simulations from Table 1 are included? Please clarify. Consider adding the place names to the figures so that readers can see this information at a glance.

Introduction

line 20: allows

lines 20 and 21: Yes, there are many studies about olivine because it is widely distributed and has relatively fast dissolution rates. However, the sentence is a little awkward and I had to read it several times. Better to say that "many studies discuss using olivine (often modelled as the end-member forsterite Mg_2SiO_4) ..." Olivine is a solid solution series between forsterite (Mg_2SiO_4) and fayalite (Fe_2SiO_4), but the common ones tend to be more Mg-rich and rate laws for forsterite dissolution are freely available.

line 31: "...(i.e., fungi and bacteria) that, ..."

line 42: please make it clear that the weathering rate is per square meter of mineral, not per square meter of land.

lines 50–52: The sentence about the models summarized by Taylor et al is a little bit difficult to understand. The models do indeed vary in their degree of complexity and plant processes may well be absent or oversimplified. A better wording might be: "The reactive transport models summarized by Taylor et al. (2017) vary in their degree of complexity and plant processes may be absent or oversimplified."

2.1 Methodology

Please describe the olivine applications: one-time or annual, mass per unit area applied, specific surface area modelled, depth of soil into which the olivine is mixed. This information deserves either a subsection or, if journal guidelines and space permits, a table. The source of the weathering rate law for olivine used in the model should also be cited.

2.2.2 Soil type and composition

How was the Hartmann and Moosdorf (2012) lithological map used? Were minerals assigned to the different lithological classes and then weathered individually in the soil, or was a generic rock defined for which the rate constant (rather than the apparent surface area of the rock/mineral) was tuned to the reported pH for the soils? What stoichiometry (base cations, Al, C, Si) was assigned to the native minerals/rocks?

Soil properties which differ for the four sites should be tabulated, perhaps extending Table 3: CEC, mean initial pH, bedrock type from Hartmann and Moosdorf (2012).

2.2.3 Crop cycle

Please clarify what the "crop coefficient" represents as soon as it is introduced, i.e., it is a proportionality constant relating actual evapotranspiration to potential evapotranspiration and depends on the crop and stage of growth. Are you using the Kc and/or crop stage length values tabulated in Tables 11 and 12 of the FAO website (https://www.fao.org/3/X0490E/x0490e0b.htm), or following the procedures outlined on that website? In either case the FAO guidelines and tables deserve a proper citation.

Lines 204–215: This sentence is very long and the beginning of it is awkward. It is obvious that root exudation products are connected to the vegetation cycle so this does not need to be stated. Reword as follows: "Root exudation products consist of carbon-based compounds ... (Shen et al.,2020). Their contribution ... during the initial growing stage (... all four locations). During the development phase ..."

Results

3.1 The role of rainfall seasonality on EW dynamics

line 239: The comma after "... the figure" is unnecessary.

Most of the paragraph about irrigation does not belong here; it deserves its own subsection. A bit more information would help explain why irrigation lowers the pH, and whether rainfall does the same. Are any ions being included in the irrigation water and does this differ from rainfall? Are rainwater and/or irrigation water in equilibrium with atmospheric pCO_2? Is the saturation state of the olivine playing a role where soil moisture is low? Discussion of the heatmaps and the influence of the irrigation shown there can remain in Results.

lines 259–260: Awkward sentence. Reword: "These considerations about soil moisture and pH affect weathering rates, which are higher in California in summer due to irrigation."

Lines 265–266: Reword: "... Julian day 300). Combined with low transpiration during the initial growing stage, this leads to higher soil water content."

Line 268: Edit: "... resulting from slightly higher average soil moisture and slightly lower pH."

Lines 270–271: Reword the whole sentence: "Similar considerations apply to corn grown in Italy (Figure S1). In summer, corn requires irrigation in Sicily but not on the Padan plains. During the rest of the year, the Sicilian and Padan plains soil have similar soil moisture and the soil moisture ratio is near 1."

Line 276: Reword: " Therefore, the Pandan soil tends to be more acidic and weathering rates tend to be higher ...).

Line 277–279: The word "significantly" implies that a statistical test has been done but this does not seem to be the case. The sentence would also benefit from rewriting, e.g., "For the same soil and vegetation, higher rainfall leads to considerably faster olivine dissolution in Iowa than in California due to higher soil moisture driven by higher seasonal rainfall frequency (λ). For the Italian case studies ...".

Line 281: What is meant by "More relevant differences..."? Were your selected case studies not relevant in terms of MAP? Do you mean larger differences?

Line 282: Too many instances of "emphasizing"; reword as "2021b), emphasizing the effect of rainfall seaonality and climatic conditions on olivine dissolution and EW."

3.2 The role of soil type on EW dynamics

As the range of soil textures for the case studies was somewhat limited, it would have been interesting to see what the model predicts with more extreme textures, such as clay and sandy soils.

3.3 The role of vegetation on EW dynamics

Line 311–313: Too many commas and repeated words here. Please edit: "... when either of the two crops is in the rest phase, water losses due to bare soil evaporation are similar in magnitude to transpiration for the other crop. The fact that wheat and corn cycles are not in phase ..."

3.4 EW case studies

Line 322: Is this a one-time application of 10kg/m^2 or is it repeated annually as in some other studies? As stated above, information about the olivine treatments needs to be either tabulated or presented in Section 2.

Lines 330–332: Awkward sentence. Reword: "... spring months, but in summer soil moisture is low due to high transpiration losses associated with a peak of the corn crop coefficient."

Line 333: "... one can observe that Iowa ..."

Line 337: The first sentence ("A similar situation ... ") looks like it belongs at the bottom of the previous paragraph as it compares the two Italian sites with similar conclusions to the two American sites. This paragraph is about comparison of Sicily with California, and Padan with Iowa.

Line 338: Remove "thus"

Lines 340–341: Please reword as "... can be observed in soil pH and the order of magnitude of the olivine weathering rate."

Line 347: Amann et al. (2020) used a Belgian soil with pH~6.6 (their table S1) which is very similar to the average annual Iowa soil pH of 6.61 given in line 335. However, the weathering rates (mol Olivine m^{-2} s^{-1}) modelled for Iowa (2.13e-12 = $10^{-11.67}$), California (1.61e-12 = $10^{-11.79}$), Padan (3.17e-13 = $10^{-12.50}$), and Sicily (4.78e-13 = $10^{-12.32}$) are all at least an order of magnitude faster than Amann et al's rates ($10^{-13.12}$ and $10^{-13.75}$ for coarse and fine dunite respectively). This is not necessarily a problem but the model rates do not really "reflect" those of the Amann et al study. Their specific surface areas (their Table 2, m^2 g^{-1}) were measured with gas adsorption which likely overestimates the actual reactive surface area of the dunite, unfortunately they do not also present more conservative geometric surface areas. What specific surface area was used in the model? As stated above, basic information about the olivine treatments should be tabulated or described.

Line 348: Section 2.2.2 discusses the calibration of the background weathering, but says that CEC was set based on existing CEC data (Ballabio et al. 2019 and USDA) which does not necessarily imply that CEC was calibrated; it seemed reasonable to assume that the CEC values used were simply means of CEC measurements from the cropland areas in the four regions (pink areas of the maps in Figure 2). If CEC or any other parameter of the weathering model was calibrated, please give details in Section 2.2.2. Then explain how these parameters affect the weathering fluxes and link to the rest of this paragraph comparing the Italian and US case studies.

Line 357: This sentence is ambiguous; it is not clear which study the 22 kg m^{-2} applies to. Amann et al. (2020) seem to have applied 22 kg dunite m^{-2} to their mesocosms; they said the dunite was about 90% olivine of which 92% was forsterite, so they applied 19.8 kg olivine m^{-2} and 18.216 kg forsterite m^{-2}. What was the application rate here? it is never stated anywhere in this manuscript as far as I can see.

line 362: Replace "correspondence" with "corresponding".

Line 395: Here olivine-derived CO_2 leached and CO_2 in soil water are distinguished, with leached CO_2 being the preferred metric of carbon sequestration. If EW were rolled out on a large scale and CO_2 consumption then calculated based on river water samples, then the leached DIC (dissolved inorganic carbon), alkalinity or HCO_3 based on major cations, is indeed relevant rather than the chemistry of the soils. It could also be argued that DIC stored for centuries to millennia in groundwater is actually sequestered at least on those timescales. If those long flowpaths comprise closed systems, the total carbon will be constant and speciation of the carbonate system will not lead to degassing. when that water eventually enters streams or rivers the likelihood of degassing due to turbulence or seasonal mixing in estuaries should be similar to waters entering at similar points on shorter flowpaths. Following the carbon all the way to the ocean where it is believed to be sequestered on 10^5-year timescales (e.g., Renforth and Henderson 2017, *Rev. Geophys.* **55**:636–674) is not really straightforward! In any case, it is not clear how the Cipolla et al. (2021) model calculates loss to groundwater. Please clarify.

Lines 410–413: Nitric acid weathering may well weather both olivine and carbonate rocks, resulting in loss of N_2O (a potent greenhouse gas) either on site or downstream, as well as CO_2 degassing in the latter case . The beginning of this sentence suggests that nitric acid is beneficial for EW. Reading the whole sentence several times, it seems this is not the intention, but rather that nitric acid is not beneficial even though it may increase olivine weathering rates. Please reword, e.g. "Even though acidification may increase olivine weathering rates, nitric acid (i.e., NHO_3) from nitrogen fertilizer would react with carbonate rocks such as those comprising the bedrock in Sicily and the Padan plain, releasing CO_2 to the atmosphere (reference) and reducing carbon sequestration potential." Please find another reference because Hartmann and Moosdorf (2012) do not mention nitrogen, although several other Hartmann papers do.

Line 414: "Despite we here considered olivine application for EW" is a bit awkward; please consider rewording, e.g.: "Even though we considered olivine application for EW here"