## **Review of Cipolla et al EGUsphere-2022-196**

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#### **General comments**

Due to the climate crisis emergency, research into enhanced weathering (EW) as a potential method for carbon dioxide removal (CDR) has increased exponentially over the past decade. Land based application of EW is thereby tested from lab over mesocosm to field scale experiments, representing increasingly more realistic conditions which are however also increasingly more complex - and require increasingly more time (from weeks-months in lab experiments to up to 10 years in field experiments). As time is of the essence when it comes to climate change mitigation, model computations of EW scenarios can play an important role in assessing the potential for CO<sub>2</sub> sequestration under specific climate, soil and crop conditions. To achieve this, close collaboration between EW 'lab/field' and 'computer' based researchers is necessary to coordinate their research and continuously use insights gained from one field as new input for the research in the other field.

This manuscript thus represents a very relevant study on modelling the effects of rainfall seasonality, irrigation, crop growth cycle and soil type at 4 different cropland sites across the world. The complexity of the authors' EW model and the as realistic as possible input data for most of their model's variables make it stand out and represent an important contribution to research into CDR potential of terrestrial EW.

The main weakness of this manuscript is the lack of some relevant background knowledge regarding mineralogy, petrology and soil formation. This is reflected in a rather poor and unrealistic modelling of the mineralogy of the soil, and the absence of necessary information on the 'olivine' material used as soil amendment for EW. This can, however, certainly be addressed in a revised version of the manuscript.

Comparison of the dissolution and  $CO_2$  sequestration rates obtained from the current model with those from the (few) published lab and field experiments could also use further discussion. It would be more valuable when the conditions (crop, olivine amendment, soil type, water availability, ...), and the methods to calculate these rates, are also compared for the lab/field and model studies. Further exploration of the plausible reasons for any observed differences between lab/field and model results would also be interesting.

Future further improvements of this excellent EW model could be to introduce a combination of different minerals as EW source material, reflecting the reality of Ca-Mg silicate rock powders proposed for EW. A multi-mineral design of the soil's mineral composition would also greatly benefit the computed background weathering ratio prior to EW. Using model parameter data from ongoing field experiments could be a next step to overall improve the EW model, which then in turn will provide relevant insights into real life EW experiments.

# Specific comments

Below follows a list of all my comments, ordered according to the manuscript's structure. Besides language corrections (yellow) there are "requests and suggestions to rephrase" as well more explanatory paragraphs to clarify (geological-mineralogical) concepts relevant to EW and this manuscript.

The changes suggested above to improve the manuscript are presented in more detail within these comments.

# Title

As the title is now, it suggests that mainly rainfall seasonality, vegetation cycle and irrigation have been studied in detail to assess their effect on EW. The manuscript however also investigates the effect of having two (not too) different soil types. So perhaps include this as a fourth variable, and also point out that this is a model. For example

"Effects of precipitation seasonality, irrigation, vegetation cycle and soil type on enhanced weathering – Modelling of cropland case studies across four sites."

As pointed out above and further discussed below, the mineralogical composition of the soil used in these models is significantly less representative or realistic than the other four model parameters mentioned in the title.

If this is corrected in a revised version of the manuscript, 'soil type' in the above suggested title can be replaced by 'soil composition'.

# Abstract

- <u>lines 9-10</u>: ... strongly affected also by the pre-EW soil pH, which is one of the main factors controlling soil pH before olivine amendment. The same parameter is referred to her: pre-EW soil pH = soil pH before olivine amendment. After having read the rest of the manuscript, it seems that 'pre-EW soil pH' should be replaced by 'background weathering flux' or the 'mineral composition of the soil', which largely determines the background weathering flux.

- <u>lines 10-11</u>: Looking at the numbers presented here for sequestered CO<sub>2</sub>, and without further explanation on the modelled rainfall seasonality, crop cycle or soil type here in the abstract, this sentence does not make so much sense. How are 4.20 and 0.62 the largest compared to 2.21 and 0.39? Do you mean to compare the two US sites with one another, and the two Italian sites with one another? After reading the manuscript I understand what is meant here, but the abstract should make sense on its own. Please rephrase to make the 'take home' message more clear.

# **1** Introduction

- line 20: bicarbonates (as on line 21 there is also the plural carbonates)

- <u>lines 21-22</u>: please consider rephrasing "which are then leached out of the soil, transported by groundwater, and eventually reach the oceans or precipitate as carbonates" to clarify that

carbonate precipitation may happen at any stage from (bi)carbonate formation in the soil to transportation into groundwater and transfers via rivers into the ocean.

- <u>lines 22-25</u>: please rewrite/revise the sentence "Many studies.....(Hartmann et al.,2013)." to clarify/correct the following:

- Olivine is the general name for the solid solution series between the ideal end member minerals forsterite (Mg<sub>2</sub>SiO<sub>4</sub>) and fayalite (Fe<sub>2</sub>SiO<sub>4</sub>), where the Mg richer varieties are more common and also more reactive with CO<sub>2</sub> and H<sub>2</sub>O. (Generally, it is the Mg-Ca-silicates that have the most potential for CDR - forsterite and wollastonite CaSiO<sub>3</sub>.) So for ease of representation/calculation Mg<sub>2</sub>SiO<sub>4</sub> is often used to represent an olivine mineral with real formula (Mg<sub>1-x</sub>, Fe<sub>x</sub>)<sub>2</sub>SiO<sub>4</sub>.

- The mineral olivine is found in igneous rocks: whereas (1) volcanic rocks such as basalt and (2) plutonic rocks such as gabbro typically have up to ca 10-20vol% of olivine; (3) ultramafic rocks such as parts of the earth's mantle exposed on the surface in ophiolite assemblages can have much higher olivine contents up to 95%. So most 'olivine' mines across the world are quarrying ultramafic mantle rocks (for example dunite, peridotite) as they have higher olivine contents, but 'basalt' is also quarried and used for EW as despite its somewhat lower olivine contents it consists of other silicates that provide plant nutrition upon dissolution.

#### (Gabbro is NOT a volcanic rock)

- <u>lines 28-29</u>: There is indeed still a discrepancy between silicate dissolution rates observed in labs (where they are more easily measurable) and in the field (where the main challenge is to differentiate the EW signal from the other biogeochemical processes going on). However a lot of lab, mesocosm and field experiments have been carried out since the reference to this issue in White & Barley (2003). As the research field for EW as CDR method has exponentially grown in the last 1-2 decades, it seems better to provide a more recent reference on this issue.

- <u>line 34</u>: "any other Ca-Mg-silicate mineral" (see comment lines 22-25); basalt is NOT a mineral, it is a rock containing different minerals one of which can be olivine  $\rightarrow$  "such as basalt  $\rightarrow$  wollastonite"

- <u>line 35</u>: the single-mineral particle lab experiments of dissolution you refer to here are not on Ca-Mg-silicates most often considered for EW, but instead on other silicate minerals that are relevant to natural weathering and soil formation (albite feldspar in Hellman and Tisserand, 2006; illite clay mineral in Koehler et al., 2003). Perhaps you can replace these with references to olivine, wollastonite, ... dissolution rate experiment studies which are more relevant to this study? (for example: Pokrovsky & Schott, 2000 <u>https://doi.org/10.1016/S0016-7037(00)00434-8</u>; Oelkers et al 2018 <u>https://doi.org/10.1016/j.chemgeo.2018.10.008</u>...)

- <u>lines 35-36</u>: Please rewrite to clarify and correctly group the different types of EW experiments. Besides single mineral grain dissolution experiments (see above), terrestrial EW experiments can be classified in the following 3 categories: (1) Laboratory experiments involving soil cores/columns to which silicate rock powder (SRP) is added, under controlled T and irrigation conditions, without biological processes (Renforth et al. 2015, Dietzen et al., 2018). (2) Mesocosm or pot experiments where plants and/or soil organisms are added to larger containers of soil with SRP, representing more closely 'real life' conditions but still closed and controlled system (ten Berge et al. 2012, Amann et al. 2020, Kelland et al. 2020). (3) Field trials where SRP is added outdoors to a field, grassland, forest soil representing complex open system of real life conditions (published study results so far only with wollastonite: Haque et al. 2020, Taylor et al, 2021 <u>https://doi.org/10.5194/bg-18-169-2021</u>).

- line 41: "magnesium and silica concentrations"

- <u>lines 42-43</u>: Please clarify that weathering rates of  $10^{-13}$  mol/(m<sup>2</sup>.s) refers to the surface of the mineral grain in contrast to sequestration rates in kgCO<sub>2</sub>/(ha.year) which refers to land surface on which mineral dust is spread on.

- <u>line 46</u>: Although White and Brantley (2003) indeed compare field and laboratory observed dissolution rates, the subject of this study is natural weathering of plagioclase and other non Ca-Mg-silicates present in a granite. Could you perhaps find more recent references pointing out the discrepancy of lab, mesocosm and field derived dissolution rates of Ca-Mg silicate minerals relevant for enhanced weathering?

- <u>line 60</u>: "suggesting that the model estimates approach a condition that is more similar to what happens in the field" (mesocosm experiments still do not represent the full complexity of field trials)

- <u>lines 63-64</u>: "Many of the model components are characterized on the base of measurements (i.e. pH and cation exchange)" perhaps better formulated as "Many of the model parameters are obtained from measurements"?

- line 66: The acronym "MAPs" is used here without introducing/explaining it.

# 2.1 Methodology

- <u>line 88</u>: Long sentence which might be more easily readable by splitting as "... to which we refer for details. It links ecohydrological and ..."

- <u>line 90</u>: "The model is composed of four closely related components." After reading this paragraph a number of times it is not clear to me which one are these four. Could you please sum them up here, or number them in the following description?

- <u>line 93</u>: "... of soil water **ions** released by olivine dissolution..." as you refer both to silicates which are anions and magnesium which forms cations

- <u>line 94</u>: Mg<sup>2+</sup> can be removed here as base cation as it is already referred to as one of the main ions formed upon olivine dissolution

- line 95: "the ... (CEC) accounts for the process between": which process?

## 2.2 Study areas and data

- <u>line 116</u>: there seems to be a mistake with the web link: there is a ´c´ in subscript <u>https://ipad.fas.usda.gov/rssiws/al/globalcropprod.aspx</u> and I get an error message when trying this link - <u>line119</u>: "active root zone depth of the involved crop." Could you please already here write these specific depths chosen for the corn and the wheat crops in the models?

### 2.2.1 Rainfall seasonality

- line 123: The acronym MAP is used again without writing it out in full before

- <u>line 126</u>: Since acronyms SIAS and USGS in the previous and next line, respectively, are fully written out perhaps this might also be done here for the acronym ARPA.

- <u>lines 133-134</u>: "... are two months out of phase, ..." If 'out of phase' refers to different trends for  $\alpha$  and  $\lambda$ , one increasing and the other decreasing, it seems to me this happens in more than 2 months (from 2 through to 6, and from about 9 to 11).

### 2.2.2 Soil type and composition

- <u>line 151</u>: SOC estimations are derived from the GSOCmap which represents "organic carbon content of the first 30 cm soil layer" – are the retrieved  $C_0$  and  $C_b$  values in the model applied only to the top 30 cm, or also further down to the root depths of 40 cm and 60 cm for corn and wheat, respectively?

- <u>line 163</u>: ... consume H<sup>+</sup> ions...

- <u>line 165</u>: "existing bedrock. This last information was extracted from the lithological map presented in Hartmann and Moosdorf (2012)." *Although a very valuable publication, it is too general to derive soil mineralogical input data for these 4 respective regions in comparison to the rainfall input data that are carefully derived from real meteorological measurements at these locations. The here used mineralogical/background lithological input data would compare to using the most common meteorological pattern in south Europe, west and central USA. So either acknowledge that the input data for the soil mineralogy of the four sites is much less representative for the real locations than the rainfall data. Or try to find more accurate data for the local geology of these four areas.* 

In case of the latter, please take into account that soils in plains retrieved a lot of their minerals from the weathering of surrounding mountains and might hence not only reflect the local bedrock of the plain but also the mineralogical composition of surrounding mountains. Furthermore, weathering of bedrock and surrounding rocks creates new minerals that end up in the soil. Eventually, the most accurate model input for the mineralogical composition of a soil is obtained from XRD measurements of that soil.

- <u>lines 168-170</u>: Carbonate sedimentary rocks are NOT calcite which is a mineral - carbonate sedimentary rocks (e.g. limestone, dolostone, ...) are mainly composed of carbonate minerals (e.g. calcite, dolomite, ...). Siliciclastic sedimentary rocks are NOT quartz which is a mineral - siliciclastic sedimentary rocks (e.g. sandstone, conglomerate, siltstone, shales, breccia, ...) are mainly composed of silicate minerals (e.g. quartz, feldspars, micas, clay minerals, ...). Please correct this by rewriting the sentence.

-<u>line 170</u>: "Lasaga (1984) and <mark>44</mark> (1979)" Please correct the later reference and perhaps also check more recent references on dissolution rate constants for carbonate and siliciclastic rocks.

- <u>line 171</u>: …"calcite and quartz minerals…" It seem from the text that just these two minerals are used for the modelling of background soil weathering, calcite for the Italian sites and quartz for the US sites? If so, please do mention that this is a big simplification of the real soil's mineralogy which is highly unlikely to exist only of calcite or only of quartz. In case a more accurate estimation of the soil mineralogy is used, a combination of mineral dissolution rate constants of the main occurring minerals should be taken into account.

## 2.2.3 Crop cycle

- line 184: What does FAO stand for? Reference please?

- <u>lines 190-198</u>: When introducing these important computation calculations (1) and (2), please clarify all the different variables in them, as was done for the next computation calculation (3). For example Crop transpiration loss T(s) where s refers to varying soil moisture - refer to Table 2 – and Bare soil evaporation E(s) where s represents...

- <u>lines 208-218</u>: The details on the crop cycle's different stages and their length at each of the four sites and for each specific crop is better represented in a figure/graph than written out in detail here, introduced in the first paragraph (183-189). For example with an horizontal axis representing the year and a vertical axis which reflects different sites and crops, showing horizontal bars divided in blocks which represent the different stages, having number of length in days inside and a specific colour for each of the specific crop cycle stage.

# **3 Results**

- <u>lines 226-228</u>: Rainfall seasonality, soil type, crop phenology and soil composition are correctly mentioned as some of the factors mostly affecting EW dynamics. And parameter input data of these variables for the model calculations are carefully determined based on real life data from the 4 sites. Except when it comes to the soil's mineral composition, where general, non-site-specific and somewhat unrealistic mineral assemblages are used (quartz for the US sites and calcite for the Italian sites) to derive background weathering fluxes. Please either clearly state that these parameter input values are less site accurate than the other ones. Or better find more accurate mineral assemblages typical for each of the four sites and use these to calculate a background weathering flux based on each mineral's relative presence and dissolution rate constant.

- <u>line 229</u>: Another major control factor of EW dynamics is the silicate rock dust powder (SRP) applied for EW. Its mineral composition greatly determines CO<sub>2</sub> sequestration potential (for example whether it is mostly olivine in ultramafic matle rocks, or olivine along with feldspars and volcanic glass in basalt). CO<sub>2</sub> sequestration potential is also influenced by how much the SRP's mineralogy differs from the soil mineralogy (see Swoboda et al, 2022 - 10.1016/j.scitotenv.2021.150976).

In general, information seems to be missing on the 'olivine amendment' that is used in these EW models. It seems that the same imaginary 100% forsterite rock dust is used across the four sites, keeping this input parameter simple and the same everywhere to allow investigation of the effects of rainfall seasonality (with/without irrigation), soil type and composition, and crop cycle – which is the main aim of this study? Or is real olivine rich rock dust modelled, for example the

one used in the mesocosm experiments of Amann et al. (2020) to which results the outcome of these models are compared?

Besides the mineralogical composition of the applied silicate rock dust powder there is other information that is important to better compare the model results to insights from field scale experiments: what is the grainsize of the rock dust? How much of it is applied per m<sup>2</sup>? Is it left on top or worked into the soil? If the latter, to which depth is it mixed with the soil? Is this application repeated annually throughout the 10 years, or is it a one time application? These SRP parameters also have an important influence on EW dynamics (see Swoboda et al, 2022) and are therefore usually well defined in lab, pot/mesocosm or field experiments. So in order to allow better comparison of EW models and EW field trials, as well as better communication between the scientists carrying out these two kinds of studies, please also include this information as a separate subsection of 2.2 Study areas and data, for example "2.2.4 olivine amendment".

# 3.1 The role of rainfall seasonality and irrigation on EW dynamics

- line 235: "...between soil moisture (S), pH and weathering rate (Wr) achieved..."

- <u>line 236</u>: Before describing the top 4 rows with heat panels, it would be helpful for scientists not familiar with such diagrams to shortly describe how to interpret them. For example, blue colours indicate higher values for a parameter (soil moisture, pH, weathering rate) in California than in Iowa at a specific time and under specific crop and soil conditions. Red colours indicate that at a given circumstances of soil, crop type and rainfall seasonality the soil moisture, pH or weathering rate is higher in Iowa than in California.

- <u>line 240</u>: Is it necessary to use the computation term 'Julian day' here as the model output data are shown horizontally as a year from day 0 to day 365, so one could say "from day 150 through to about day 250" which is more easily understandable for non-modelling scientists? If 'Julian day' needs to be mentioned perhaps shortly explain what exactly this means?

- line 241: As before, Julian day needed or is "some days around day 300" also ok?

- <u>line 254</u>: Soil moisture time-series in the figure 6 caption is referred to as panel b), not c)
- line 255: Please rewrite as "the field capacity in the days from about day 100 up to day 250".

- <u>lines 242-257 until "... is provided.</u>": This paragraph introducing irrigation for the Mediterranean climates – the reason why it is necessary and how it is implemented in the model – should be moved to '2.2 Study area and data' as a new subsection right after 2.2.1 Rainfall seasonality. So 2.2.2 Irrigation, 2.2.3 Soil type and composition, 2.2.4 Crop cycle, 2.2.5 Olivine amendment. Figure 6 should then also be moved to this earlier section of the paper. The stress-avoidance irrigation procedure for corn planted in Sicily should also be shown in 2.2.2 Irrigation for one of the two soil types, either added to Figure 6 or as a new Figure.

- <u>line 240</u>: When the irrigation paragraph is moved to an earlier section, you can then refer back to it here "soil moisture is higher in California than in Iowa due to irrigation".

- <u>line 259</u>: What is the reason that the soil pH becomes lower, more acid, with increased soil moisture, irrigation? Please briefly clarify.

- <u>line 261</u>: Please be consistent, in line 255 'Julian' was omitted when describing the period from day 100 to day 250. So perhaps generally remove the word 'Julian' throughout this document.

- line 263: Please replace "the Julian day 300" with "the 300th day" or "around day 300".

- <u>lines 267-269</u>: In the concluding sentence "On average, …. weathering rates derived for Iowa are about seven times higher than those in California…" This refers to the cases where wheat is the crop so please clarify this by adding "with wheat". Likewise it might be beneficial to repeat once again in the conclusions of the previous paragraph, lines 259-261, that these are model observations with corn.

Also: Where is this 7X higher weathering rate for Iowa compared to California derived from? The average daily ratio of Wr in Figure 5? Please clarify where this number comes from.

- <u>lines 270-276</u> where the role of rainfall seasonality on EW dynamics is discussed for the <u>Italian</u> <u>sites</u>: It is unclear why the time-series heat map for the Italian sites is put as Supplementary material as despite the similarities with the US sites with/without irrigation, these maps are sufficiently different. Supplementary material is often a separate document from the main paper containing raw data, so it would be better if this Figure S1 would become the second figure in the subsection 3.1 after the time-series heat maps for the US sites. The explanation written in the Supplementary material along with Figure S1 is the exact same text as what is described here in this section, showing that text and figure best go together (in the main paper).

- lines 270-271: Please rephrase this sentence as it is awkward to read and not very clear.

- lines 275-276: Please rephrase this sentence as it is awkward to read and not very clear.

- <u>lines 278-279</u>: Please rephrase/rewrite these important conclusions regarding the modelled effect of rainfall seasonality and irrigation on EW dissolution rates as the text is difficult to read and unclear. Thereby keep in mind to replace 'significantly' with 'distinctly' (significantly usually refers to statistically verified differences between values).

- <u>line 281-283</u>: "Larger differences in mean annual precipitation would likely result in bigger changes of EW dynamics (Cipolla et al., 2021b), emphasizing the important effect of rainfall seasonality and climatic conditions on olivine dissolution and EW."

### 3.2 The role of soil type on EW dynamics

- <u>line 286</u>: ... and silty clay loam soil,...

- <u>lines 288-290</u>: Add the parameter symbols please: ...soil moisture (S), pH and weathering rate (Wr) ... clay loam soil (CL) ... silty clay loam soil (SCL).

- <u>lines 294-295</u>: ...weathering rates obtained with the clay loam soil <u>tends</u> tend to be about twice as high as those obtained with the silty clay loam soil... Where is this 2X higher weathering rate for CL compared to SCL derived from? The average daily ratio of Wr in Figure 7? Please clarify where this number comes from.

## 3.3 The role of vegetation on EW dynamics

- <u>line 300</u>: ... of H<sup>+</sup> to balance ...

- <u>lines 301-302</u>: "Brady, 2017). Vegetation furthermore provides the organic matter that, once decomposed, is one of the CO<sub>2</sub> sources in the soil system..."

- <u>line 305</u>: ... about four times higher than ... for wheat... Where is this 4X higher weathering rate for CL compared to SCL derived from? The average daily ratio of Wr in Figure 8? Please clarify where this number comes from.

- <u>line 306-307</u>: " and fourth row of the figure). When both crops are planted in a silty clay loam soil in the Padan plain and lowa (second and third row of the figure), the olivine dissolution dynamics are very similar. An annual average weathering rate daily ratio equal to about 1.5 might reflect slightly higher weathering rates for corn."

- <u>lines 311-312</u>: ' when any of the two crops is in the rest phase' please specify which exact periods these are to make it easier to spot them in Figure 9. For example by: '... in the rest phase (from about day as to day bb for wheat and from about day xx to day yy for corn)'

### 3.4 EW case studies

- <u>lines 320-321</u>: 'The time dynamics of soil moisture, pH and weathering rate across the four locations in Italy and the USA are shown in Figure 10. In all scenarios...'

The time series heat maps for the Italian sites now in Supplementary material Figure S2 should be brought to this section of the main text to illustrate it. As Figures 5, 7, 8 each have 4 rows of three heat maps and an extra bottom row with the average daily ratio, it should be possible to add the Italian heat maps in Figure S2 to those of the US sites in Figure 10.

- <u>line 322</u>: The information regarding the olivine application rate should already have been given in a subsection of section 2.2 on olivine amendment. Why was this rather high application rate of 10kg/m<sup>2</sup> chosen? Practically, farmers apply lime and other rock dusts annually at a rate of 1-4 tons/ha.

- line 324: ...(i.e., before day 100) ... (i.e., from day 300 onwards)...

- line 326: values from day 100 to about day 250 mainly ...

- <u>lines 333-336</u>: Where can the annual average values for soil moisture, pH and weathering rate for Iowa and California be found? The start of the sentence with 'Comparing the annual average values..., one can observe...' suggests that this can be seen in a figure or table? If these data are only presented here within this paragraph, then please rephrase. "Annual average values of the three variables calculated for California and Iowa suggest that faster olivine dissolution occurs at the latter site (2.13X10<sup>-12</sup>mol/m<sup>2</sup>s) than at the former (1.61X10<sup>-12</sup>mol/m<sup>2</sup>s). This is in accordance with a lower annual average pH (6.61 in Iowa and 7.03 in California) and higher mean annual soil moisture (0.62 in Iowa and 0.57 in California). "

Whereas pH seems indeed different between the two US sites, soil moisture shows a smaller difference. How meaningful is the difference between the Iowa and California olivine dissolution rates? Any estimation of the uncertainty on these calculated values?

- <u>line 337</u>: Please add the heat maps of Figure S2 to Figure 10 and add the description of them which is currently in S2 here in the main text of the manuscript. "A similar situation is observed from the comparison of the two Italian sites as Sicily and Padan plain present only small differences in terms of the seasonality of soil moisture, pH, and , in turn, weathering rate. ... (i.e., before day 110) and the last (i.e., from day 300 onwards) ...with respect to the two sites in the USA."

- <u>line 338</u>: 'Because of the similar rainfall seasonality...' seems to be the start of a new paragraph where now Italian sites are being compared to US ones.

- <u>lines 341-343</u>: No need to repeat the pH and dissolution rates calculated for the Italian sites here if it is already mentioned in the previous paragraph which used to be the text of S2.

- <u>lines 346-347</u>: 'the achieved order of magnitude of weathering rate reflects the values presented in the mesocosm experiment of Amann et al. (2020), which present a condition very similar to the field.' What exactly are the weathering rates presented in Amann et al. (2020)? How do the conditions of their mesocosm experiment compare to those of the models discussed in this paper? What 'olivine' type used, application rates, which crop in the mesocosm, irrigation scheme, soil type and composition? A comparison of the results of the current study with those of a published paper benefits from some info on the published study.

- <u>lines 348-349</u>: 'suitable calibration' seems odd in this sentence, perhaps rewrite as "the importance of site representative model input data for the background flux, ..."

Rainfall seasonality, irrigation scheme, CEC, soil type, main soil properties and crop phenology have indeed been determined as representative as possible for the four respective sites. In comparison, the soil mineralogy, another very important parameter influencing the olivine dissolution dynamics, chosen for the models is much less site specific or realistic.

- <u>lines 352-353</u>: "The overall rather low monthly values of sequestered CO<sub>2</sub> for all case studies are due to the generally low leaching rate, which reflects the low MAP values for all considered sites."

- <u>line 354</u>: "The annual average sequestered CO<sub>2</sub> equals 0.62 kg/ha for Sicily, …"

- <u>lines 355-358</u>: The difference between the Amann et al. (2020)  $CO_2$  sequestration values and the ones of this study are on a scale of 1 to 2 orders of magnitude – yet considered comparable to one another. The weathering ratio values obtained for the US and Italian sites differ only 1 order of magnitude from one another – yet deemed different (and this difference explained by the least site representative/realistic model input parameter of soil mineralogy). Please be consistent with interpreting the difference between values. It is true that Amann et al. (2020) added 22kg/m<sup>2</sup> whereas in these models 10kg/m<sup>2</sup> was applied, but the rock dust of the former only contains about 90% olivine. How does the soil moisture throughout the year compare between both these studies? How was the  $CO_2$  sequestration value calculated in Amann et al.

(2020)? What other factors might play a role in the difference of CO<sub>2</sub> sequestration rates obtained for these two different study approaches (mesocosm experiment and model)?

- <u>line 362</u>: ... with a corresponding increase of HCO<sub>3</sub><sup>-</sup>...

#### 4 Discussion and conclusions

- <u>line 372</u>: Analyzing the interactions between rainfall and crop properties...

- line 378: ... with a corresponding increase of HCO<sub>3</sub>...

- <u>lines 379-380</u>: "... by olivine reaction with CO<sub>2</sub>. Higher soil water contents also mean higher leaching rates and hence better transport of the (bi)carbonate anions away from the active olivine dissolution zone."

- line 395: ... the one we called CO<sub>2,sw</sub> ... Please shortly define/explain this parameter instead of just giving the symbol and referring to a previous publication. ...In effect, even in this <del>our</del> previous work we obtained...

Although I understand the reasoning that (bi)carbonates formed by olivine dissolution but which stay in the 'EW zone' are seen as a risk to recombine to carbonic acid releasing CO<sub>2</sub> back to the atmosphere, and that hence (bi)carbonates leached out from EW zone are interpreted as more reliable measure for CO<sub>2</sub> sequestration, it is not so straightforward. Some of the olivine dissolution sourced carbonate anions might precipitate in solid carbonate minerals within the soil (calcite) which is then stable carbon sequestration that can not be traced back in the leached groundwater below. On the other hand, (bi)carbonates dissolved in leached groundwater and hence taken into account for CO<sub>2</sub> sequestration calculations, might recombine to carbonic acid and degas CO<sub>2</sub> when they resurface or mix with water of different composition, temperature,... The permanence of CO<sub>2</sub> sequestered as (bi)carbonates in groundwater through olivine dissolution is difficult to estimate and probably varies from one context to the next. Maybe this suggests that (bi)carbonate anions and DIC are not the best parameters to estimate the amount of captured CO<sub>2</sub>. Another product from olivine dissolution is Mg<sup>2+</sup> cations. In general, weathering of silicate rocks will release base cations into the soil water as well as (bi)carbonates. Please see what is written about this in literature and assess the pros and cons of using (bi)carbonates or cations to estimate sequestered carbon. Is there a possibility within your model to obtain values for cations resulting from olivine dissolution, and to use these data for an alternative calculation of CO<sub>2</sub> sequestration?

- <u>line 414</u>: Good to come back to possible more complexity in future models regarding the silicate rock dust that can be used for EW.

- <u>line 415</u>: basalt is NOT a mineral, it is a rock and hence an assemblage of minerals. See comments for lines 22-25. The reason that basalt has lower Ni and Cr contents compared to olivine is because basalt only partially consists of olivine. Since the topic of potential Ni and Cr contamination resulting of EW of olivine rich rocks is touched upon here, please add a sentence explaining that both these heavy metals occur in olivine crystals and are thus released when the latter are dissolved.

Another reason to use basalt is that the other minerals it contains release plant nutrient cations upon dissolution, effectively being a natural fertilizer.

All in all, using silicate rock powder consisting of different minerals, instead of just one mineral (olivine, wollastonite) would greatly improve the model's representation of realistic field situations.

- <u>line 417</u>: The wollastonite EW field trials of Haque et al (2020) are NOT across the world but at three different locations in Canada.

- <u>lines 417-718</u>: Wollastonite is a calcium silicate –  $CaSiO_3$  – that upon reaction with water and  $CO_2$  dissolves and forms, among other products,  $Ca^{2+}$  and  $CO_3^{2-}$ . This cation and anion can combine within the EW zone to form secondary, pedogenic calcite which is then an easy measure to assess how much wollastonite dissolved, and hence  $CO_2$  was sequestrated into this new calcite. In case of olivine dissolution, the released cations are less likely to form new carbonate minerals within the EW zone, only under certain chemical conditions they might. This is one of the reasons why  $CO_2$  sequestration from olivine and other silicate rock dusts dissolution is more difficult to measure. (see comments line 395).

- <u>line 421</u>: Indeed, most lab, pot and mesocosm experiments are carried out under continuous (near) saturation of soil moisture which is not representative of the real life situation. The detailed incorporation of rainfall seasonality and irrigation in the here presented model is therefore one if its greatest merits and strengths towards more realistic EW potential predictions.

- <u>line 424-426</u>: Precipitation of secondary minerals as pedogenic carbonates from products of silicate rock powder dissolution in the field is far from well understood and likely not the most common scenario. A more relevant improvement of the here presented excellent EW model would therefore be to go from single mineral olivine (which in reality is never applied as it is not available) to a realistic assemblage of minerals (for example resembling that of a mantle dunite, or a basalt) that takes into account the dissolution rates of the individual minerals and their relative presence in the silicate rock dust.

### **Figures**

<u>Figure 3</u>: To allow easier comparison between the average rainfall depth  $\alpha$  and rainfall frequency  $\lambda$  (please label both fully on the vertical axes) between the 4 different areas, maintain the same scale for all four diagrams (i.e.  $\lambda$  up to 0.47 and  $\alpha$  up to 13.5). Putting the location names in each of the 4 plots would also make it easier to interpret this figure at a glance. In a black and white print out it is not clear which of the two lines is which, so perhaps make one a dotted line and either put in a small legend, or describe in the figure caption which parameter is represented by the full, and which one by the dotted, line. Also fully describe what the  $\alpha$  and  $\lambda$  "rainfall parameters" exactly represent.

<u>Figure 4</u>: Please write full name and symbol for both parameters (crop coefficient Kc and added carbon ADD) in both figure caption and alongside the vertical axes. To interpret more easily the graph, perhaps put a) b) c) vertically below one another in the first column, writing 'wheat' above it and the site name in each of the 3 graphs. And then have d) and e) vertically below one

another in the second column, writing 'corn' above this column and the names of the sites in the respective graphs.

<u>Figure 5</u>: Please add the respective symbols for the different variables to the figure caption: soil moisture (S), weathering rate (Wr), Iowa (IA), California (CA), corn in clay loam soil (C-CL), etc. To make this figure easier to read it would be good to have the colour legend just once, write the full parameter name above each column, and the full crop/soil type combination in front of every row.

<u>Figure 6</u>: Please add the symbol to the parameter name in the figure caption, and the full name to the symbol along the Y-axis of the specific graph (crop coefficient Kc, soil moisture S). Letters a), b). c) and d) are missing in the respective panels.

<u>Figure S1</u>: Please make a main manuscript figure occurring after the time-series heat maps for the US sites, and make the same corrections/changes as detailed above for Figure 5. The resolution of the current S1 figure needs to be improved as the Y-axis labels are poorly readable both in print and in the pdf on screen.

Figure 7: Please adjust in the same way as suggested for Figure 5.

Figure 8: Please adjust in the same way as suggested for Figure 5.

<u>Figure 9</u>: Please add the symbol to the parameter name in the figure caption, and the full name to the symbol along the Y-axis of the specific graph (bare soil evaporation (E), crop transpiration (T)). To allow easier comparison of these parameters between a) wheat sand b) corn, please have both Y-axis the same length (4.25 mm).

<u>Figure 10</u>: As before, please write the parameter codes in the figure caption and the full parameter name along with its code above the respective column of heat maps. (soil moisture (S), weathering rate (Wr)). To the left of each of the rows, add the codes of the case studies, and in the figure caption write the case study code with the full description (wheat in clay loam soil in California (W-CL-CA), corn in silty clay loam soil in Iowa (C-SCL-IA).

<u>Figure S2</u>: Include these two rows of each 3 heat maps for the Italian sites in Figure 10, with both full reference and code (wheat in clay loam soil in Sicily (W-CL-SI), corn in silty clay loam in Padan plain (C-SCL-PP)).

<u>Figure 11</u>: Please put the site name in each of the plots to make this figure easier to read. Do these plots reflect the model outcomes from the input parameters used in section 3.4 (last 4 rows in table 1)? Please write again in the figure caption which soil type and crop, with or without irrigation, is presented here for each of the 4 sites.

#### Tables

<u>Table 2:</u> Please have the same number of digits after the separation point for values of the same parameter (they are different for soil moisture at field capacity and saturation hydraulic conductivity). Please also write which of the four study sites are represented by which soil type.

<u>Table 3</u>: According to the text (lines 153-155), biomass pool  $C_b$  is defined as 1% of the abovedefined carbon input ( $C_0$ ). Yet in the table the values for  $C_b$  are 10 times higher than those for  $C_0$ ? Maybe  $C_b$  is here expressed as g/m<sup>3</sup> instead of kg/m<sup>3</sup> as  $C_0$  above?

Please also add to this table the model input values for the different study sites of CEC, derived soil mineralogy, soil pH and calculated dissolution rate constants.

Perhaps it is possible to combine tables 2 and 3 into one table with all soil type and composition data used in the models?

## References

All references in text are in the references list and vice versa. The only irregularity is the first entry in the references list 'Critical Review ..., 1979' which seems to lack authors but might coincide with the incomplete reference in the text in line 170 '44 (1979)'.

## Supplementary material

Please include this in the main manuscript text as suggested in comments above