Replies to the comments on "**Effects of precipitation seasonality, vegetation cycle, and irrigation on enhanced weathering**".

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COMMENTS FROM THE REVIEWERS: REVIEWER #1

General comments:

Referee: *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.*

Response: We thank the Reviewer for his/her in-depth analysis of this work. Please find below our responses to the comments.

Specific comments:

Referee: *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.*

Response: We thank the Reviewer for his/her very useful comments. Based on the Reviewer's suggestions, we provided more details in the manuscript, which can also be found in the responses to the comments below.

Technical corrections:

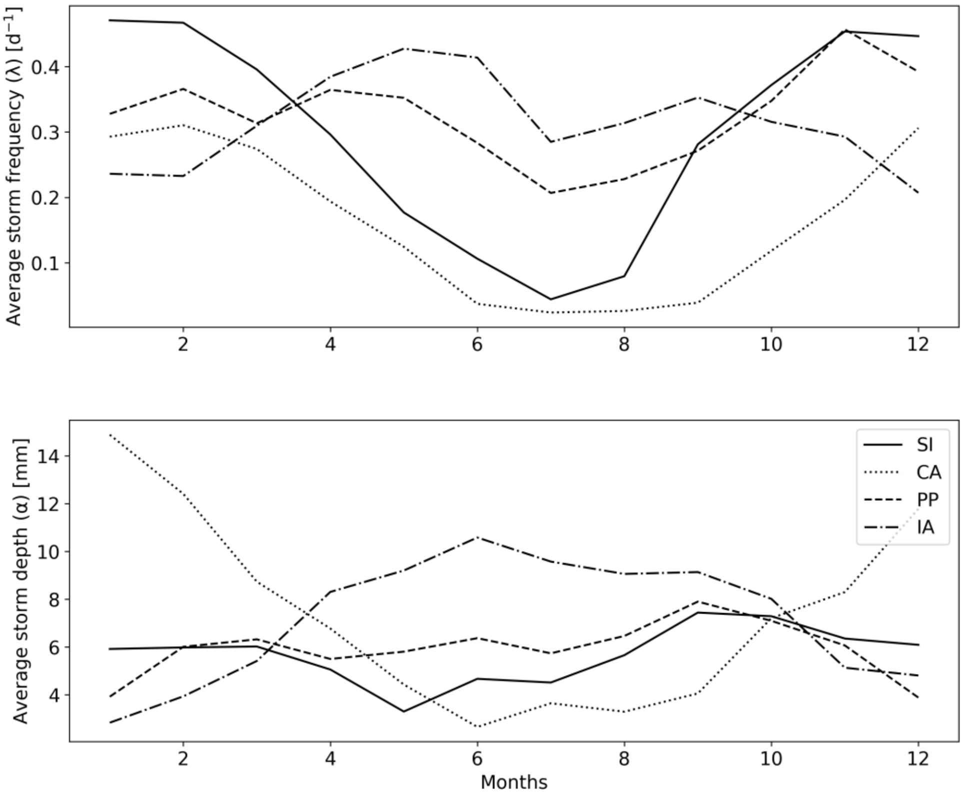
Figures

Referee: *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.*

Response: Good point. We tried to make the figures clearer. Please find below our new arrangements.

Referee: ***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.*

Response: This is a very good suggestion. We modified the figure as follows.

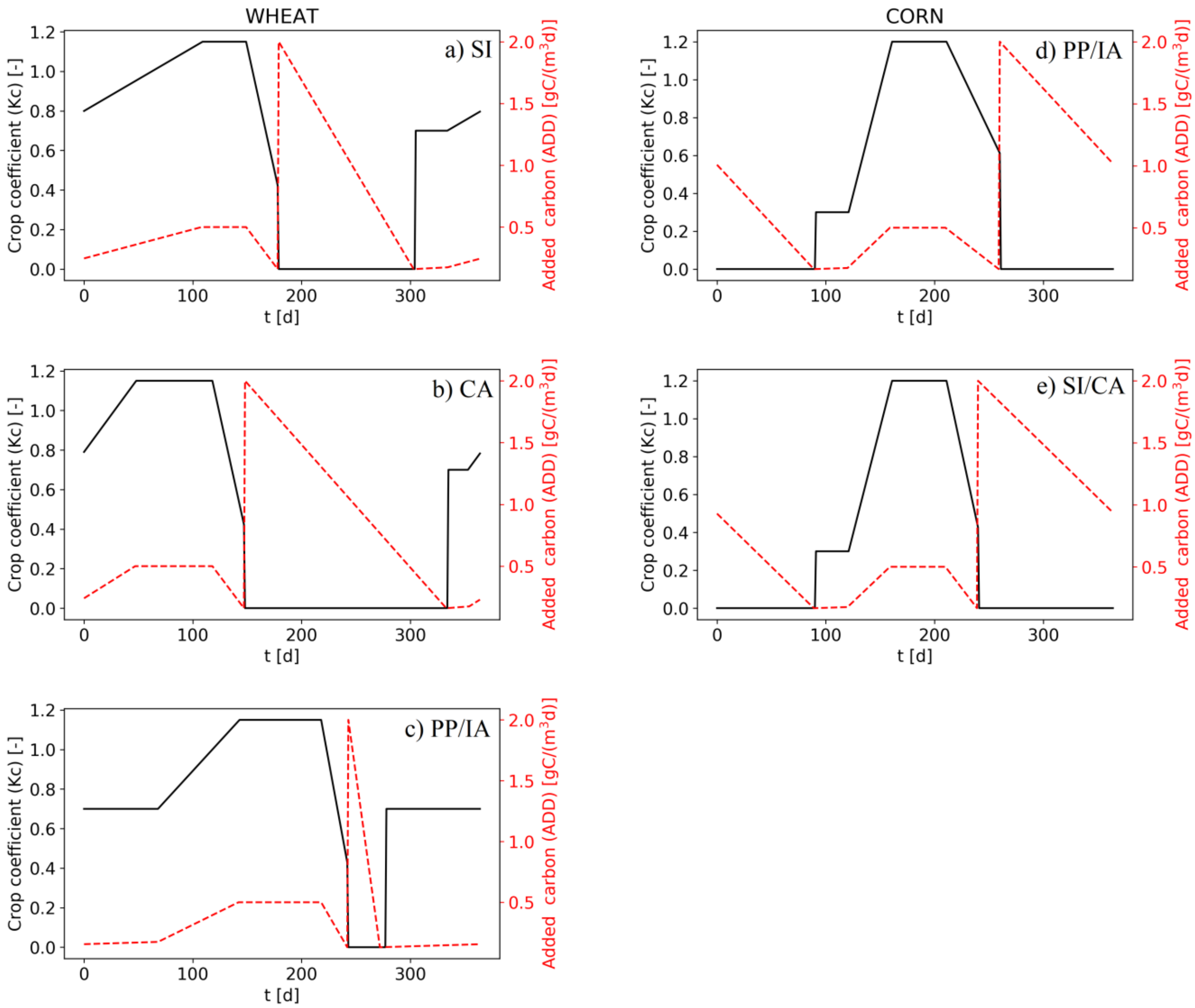


Furthermore, we modified the caption:

Values of the average rainfall depth (α) and frequency (λ) from January (i.e., month indicated with 1) to December (i.e., month indicated with 12). The reader is referred to Section 2.2.1 for more details.

Referee: ***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.*

Response: Following this suggestion, we modified the figure:

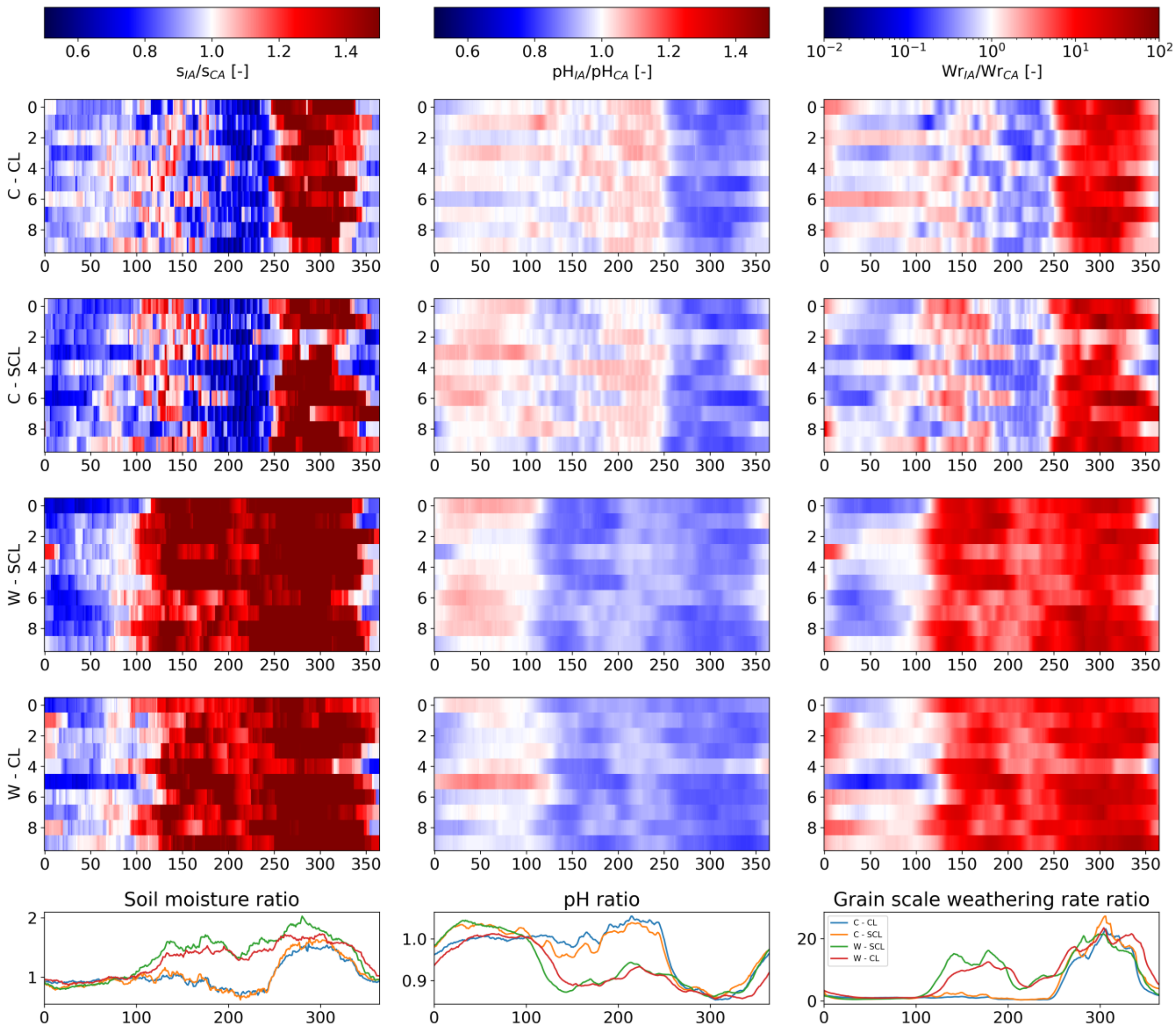


Furthermore, we modified the caption:

Seasonal variability of the crop coefficient (Kc) and the added carbon from vegetation (ADD) for a) wheat in Sicily, b) wheat in California, c) wheat in the Padan plain and Iowa, d) corn in Padan plain and Iowa and e) corn in Sicily and California. The reader is referred to Section 2.2.3 for more details.

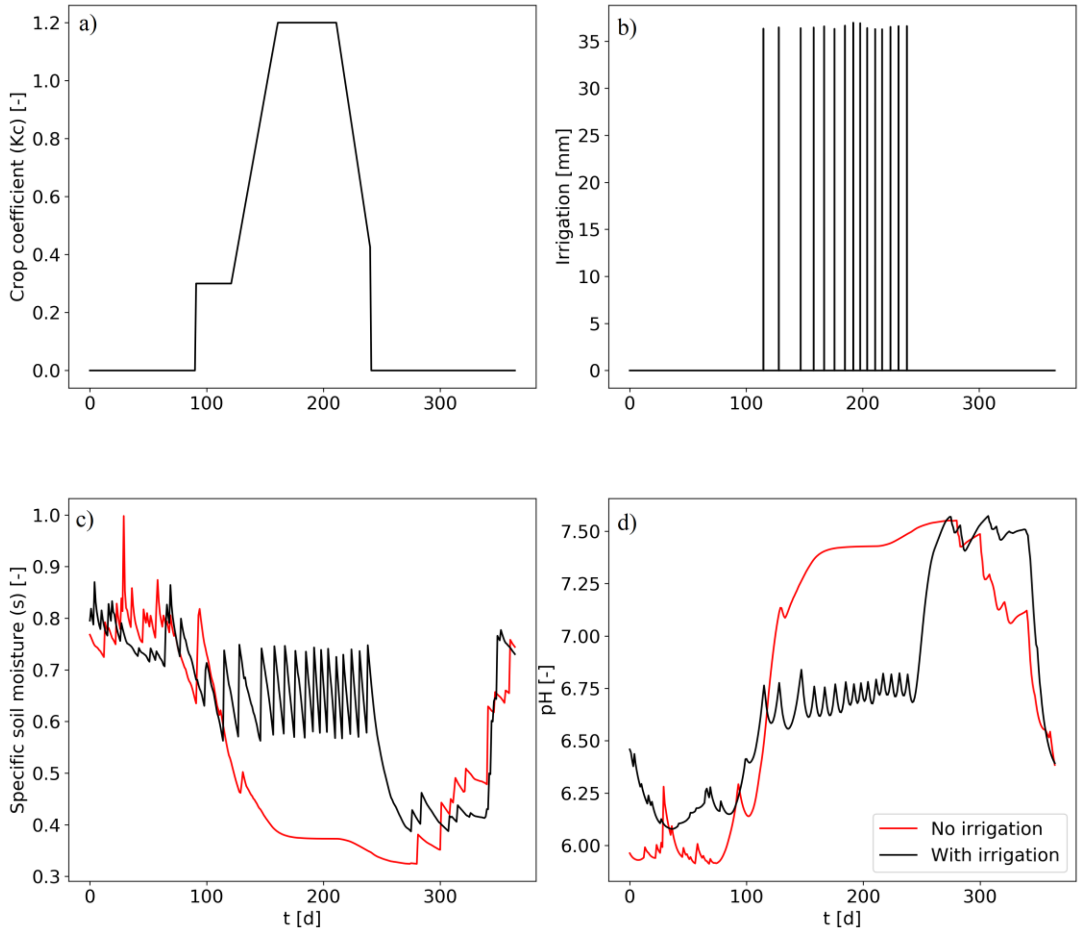
Referee: ***Figure 5***. *It would be helpful if the labels "Corn Clay Loam" etc could appear on the left of each row of this figure, the individual panel colorbars were removed, and a single colorbar with legible label e.g. "soil moisture ratio SIA/SCA" appeared at the top of each row. Not all software 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. "SIA/SCA". The label for column 3 should make it clear what scale is being considered, e.g. "Grain scale weathering rate ratio".*

Response: Figure modified as follows:



Referee: ***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".*

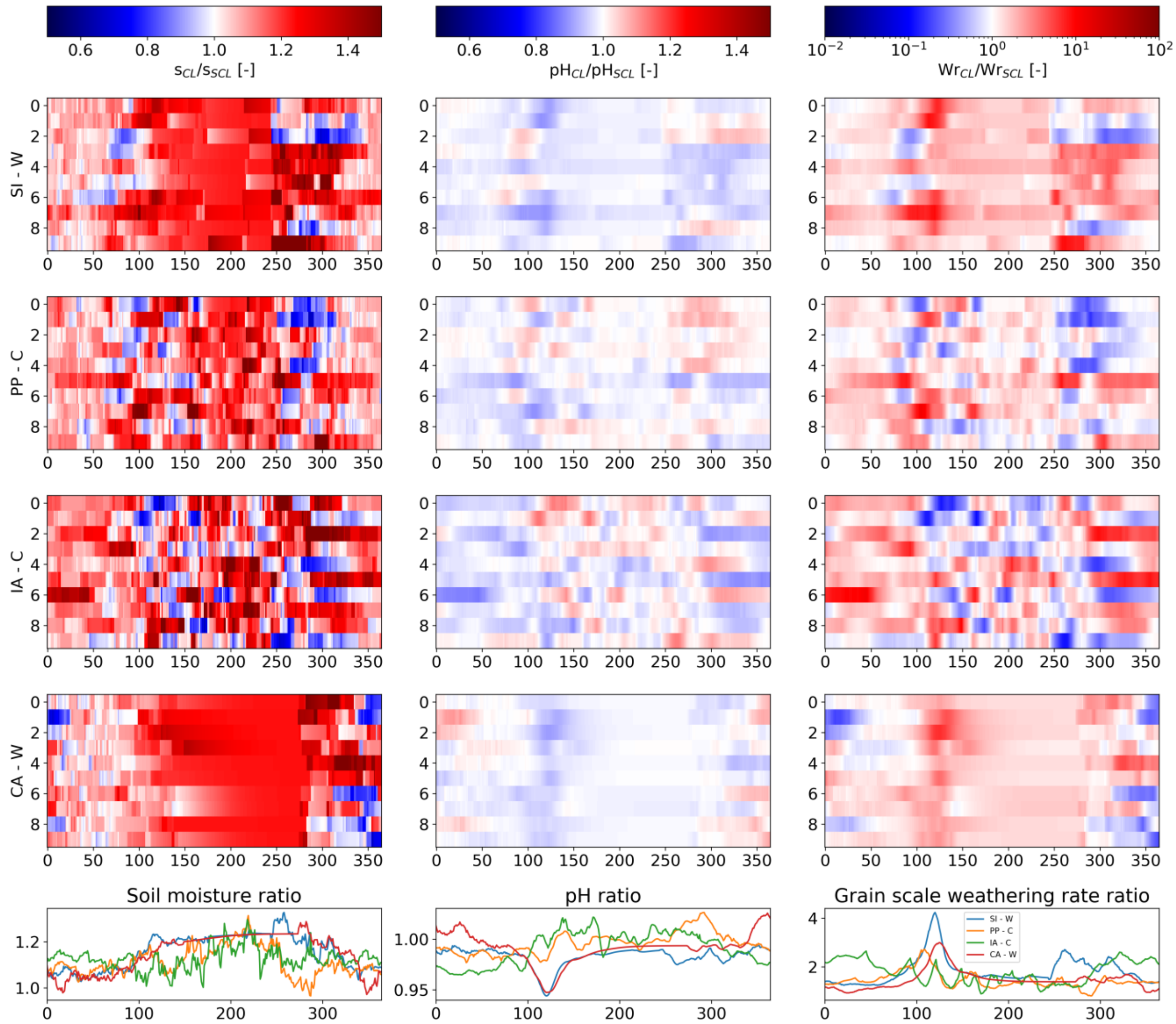
Response: Thanks for highlighting this. Please find below the new version of the figure:



Referee: ***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/(m2 olivine s) in Figure 10.*

Response: We improved Figures 7, 8, and 10.

Figure 7:



s_{CL}/s_{SCL} [-]
pH_{CL}/pH_{SCL} [-]
Wr_{CL}/Wr_{SCL} [-]
SI - W
PP - C
IA - C
CA - W
Soil moisture ratio
pH ratio
Grain scale weathering rate ratio

Figure 8:

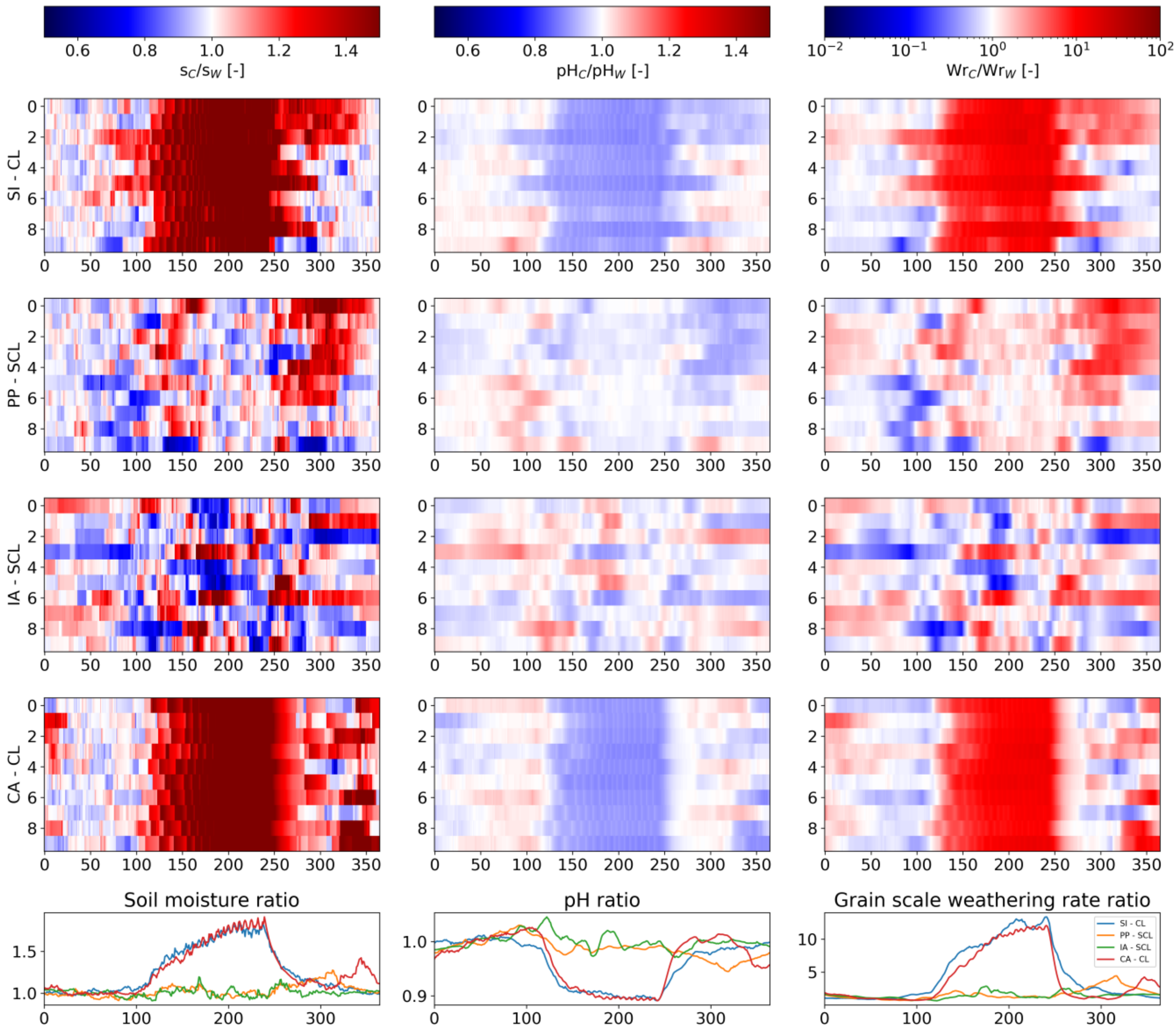
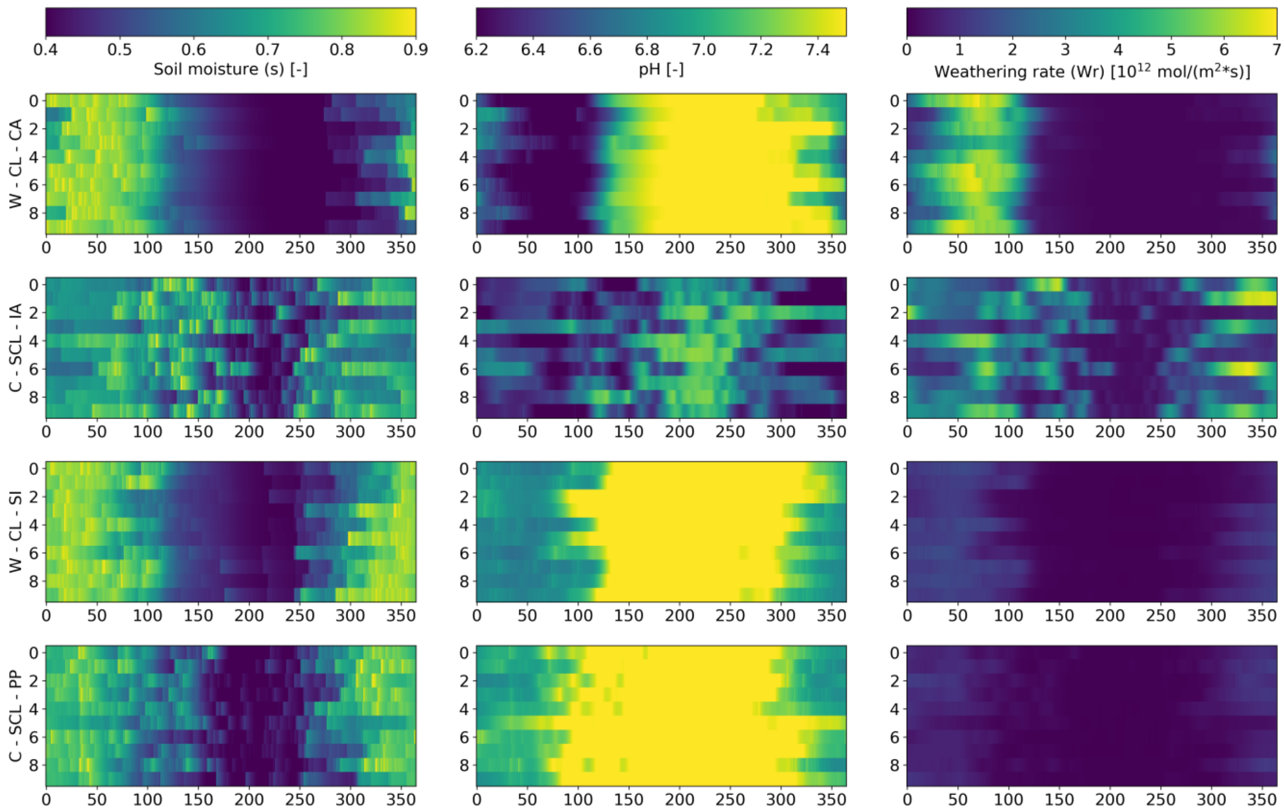


Figure 10: In this figure, we included the heatmaps regarding the Italian case studies that were previously in the supplementary materials, in order to provide a better comparison between the four selected sites.



Referee: ***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.*

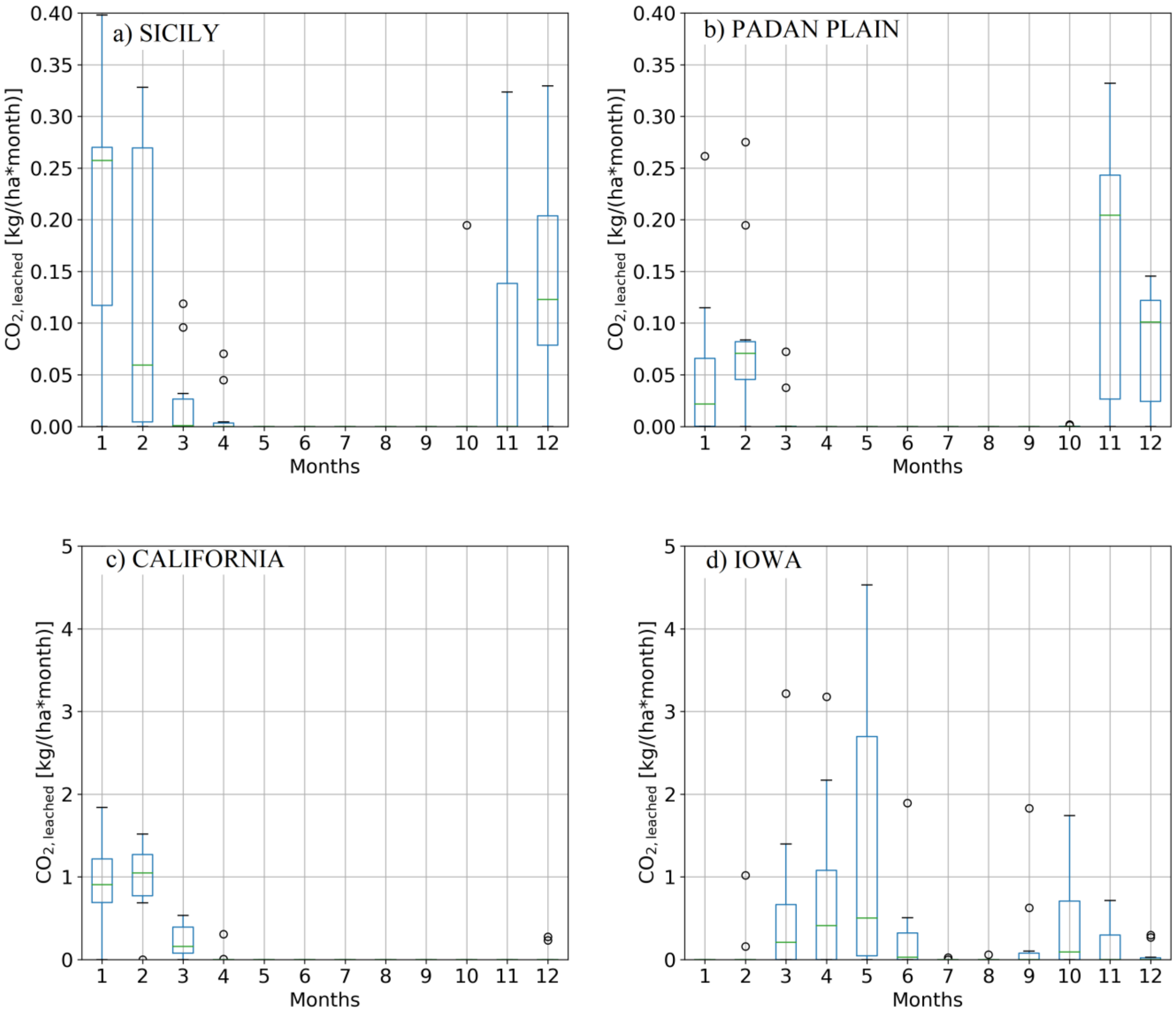
Response: Thank you for pointing this out. The boxplots of sequestered CO_2 are related to single runs of the four case studies, considering the single climatic conditions, along with the most frequent crop and soil type. Basically, these simulations are those referred to as "Most frequent scenario (Section 3.4)" shown in table 1. We better clarified this aspect at the beginning of Section 3.4 with this sentence:

The results here analyzed are those related to the simulations referred to as "Most frequent scenario (Section 3.4)" as shown in table 1, where the considered climatic condition is analyzed along with the most frequent crop and soil type.

We also clarified this aspect in the caption:

Box plots representing the seasonality of the CO_2 sequestered by leaching of extra HCO_3^- and CO_3^{2-} produced by olivine dissolution, computed over the 10 years subsequent to olivine amendment. The plots are related to a) Wheat in clay loam soil for Sicily (W - CL - SI), b) Corn in silty clay loam soil for Padan plain (C - SCL - PP), c) Wheat in clay loam soil for California (W - CL - CA) and d) Corn in silty clay loam soil for Iowa (C - SCL – IA).

Finally, the names of the locations were added to the panels as suggested. The new figure is the following:



Introduction

Referee: *line 20: allows*

Response: Thank you for highlighting this mistake. We corrected it.

Referee: *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 endmember forsterite Mg2SiO4) ..." Olivine is a solid solution series between forsterite (Mg2SiO4) and fayalite (Fe2SiO4), but the common ones tend to be more Mg-rich and rate laws for forsterite dissolution are freely available.*

Response: Good insight! We modified the sentence as follows:

Many studies discuss using olivine (often modeled as the end-member forsterite Mg2SiO4) in EW applications…

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

Response: Thank you for highlighting this mistake. We corrected it.

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

Response: Thank you for this suggestion. We now clarified this aspect. The modified sentence is the following:

The achieved weathering rates, expressed in moles of dissolved olivine per unit of specific surface of the mineral and time, were on the order of…

Referee: *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."*

Response: Thank you for your suggestion. We modified the sentence as suggested.

2.1 Methodology

Referee: *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.*

Response: This is a worthwhile comment, thank you! We modeled a one-time olivine application with a rate of 10 kg m^{-2} of cropland area. The olivine is assumed to be mixed for the whole active root zone depth of the considered crops. All the particles dissolve according to the same rate since the presence of preferential flow paths is not here considered. The dissolution of olivine particles is modeled according to the shrinking core model of Lasaga (1984) (https://doi.org/10.1029/JB089iB06p04009), considering particles as perfect spheres having an initial diameter of 200 μm, since the model considers a single effective diameter, defined as the mean diameter of a particle size distribution, in the name of simplicity. In fact, to consider the actual particle size distribution, the model would need to include partial differential equations, which would greatly increase the computational costs. This effective diameter is meant to be representative of the whole particle size distribution, in that its weathering rate represents the average weathering rate found by integrating over the particle size distribution.
The dissolution rate law is the one presented in Cipolla et al., (2021a), (https://doi.org/10.1016/j.advwatres.2021.103934), where the weathering rate, expressed in number of moles of dissolved olivine per unit of reactive surface of the mineral and per unit of time, is a function of soil moisture, pH and the ion activity product, which expresses the products of olivine dissolution reaction (i.e., magnesium and silicates) with respect to soil water pH. We better clarified all these aspects at the end of Section 2.1 of the manuscript. The added part is the following:

For all the analyzed scenarios, a one-time olivine amendment with a rate of 10 kg m^{-2} (i.e., 100 t ha^{-2}) was considered. The olivine is assumed to be mixed for the whole active root zone depth of the *considered crops. All the particles dissolve according to the same rate since the presence of preferential flow paths is not considered. The dissolution of olivine particles is modeled according to the shrinking core model of Lasaga (1984), considering particles as perfect spheres having an initial diameter of 200 μm, since the model considers a single effective diameter, defined as the mean diameter of a particle size distribution, in the name of simplicity. The dissolution rate law is the one presented in Cipolla et al. (2021a), where the weathering rate, expressed in number of moles of dissolved olivine per unit of reactive surface of the mineral and per unit of time, is a function of soil moisture, pH and the ion activity product, which expresses the products of olivine dissolution reaction (i.e., magnesium and silicates) with respect to soil water pH.*

2.1.1 Soil type and composition

Referee: *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).

Response: Thank you for the very in-depth analysis of the work. We actually extracted the most frequent lithological class for the sites under study and look at its basic dissolution rate constant. Then, we calibrated these constants to achieve the reported steady-state pH for the analyzed soils, as explained in lines 176-181. We actually did not consider a proper stoichiometry of the bedrock since our main idea is to incorporate different possible consumptions of H^+ in a single term, without considering the dissolution of individual minerals (too cumbersome computationally). This is a simplified way to look at these aspects but is effective since it easily allows obtaining the typical soil pH values by calibrating a single constant.

We also extended table 3 adding the typical pH, CEC, and the achieved dissolution rate constants for the soils under study, which better represent the background weathering component. The table in its new form is reported as follows:

Table 3 - Initial organic carbon content in the litter and humus pools (C_0) and the biomass pool (C_b) for the four sites under study. Typical values of pH, CEC and background dissolution rate constants are also reported.

	Sicily	Padan plain	California	Iowa
$C_0 [kg\ m^{-3}]$	13.19	18.19	19.74	11.58
$C_b [g\ m^{-3}]$	131.9	181.9	197.4	115.8
pH [-]	7.2-8.8	7.2-8.8	7	6
CEC [cmol kg^{-1}]	50	50	35	25
k_{bg} [mol $m^{-2}\ s^{-1}$]	10^{-5}	10^{-5}	10^{-6}	10^{-8}

2.2.3 Crop cycle

Referee: *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.*

Response: Thank you for highlighting this aspect. We clarified the meaning of the crop coefficient and its use after mentioning it in the manuscript for the first time. We also cited the FAO guidelines and tables, from which we extracted the Kc and crop stage length values. The sentence is the following:

To investigate the role of the crop cycle on the EW dynamics, the monthly variation of the crop coefficient (Kc), commonly defined as the ratio of the crop evapotranspiration over the reference evapotranspiration, was considered. It is here used as a proportionality constant relating actual evapotranspiration to potential evapotranspiration, depending on the crop and stage of growth. For each development stage, a single crop coefficient per crop type and the climatic area was obtained following FAO guidelines (tables 11 and 12 in Allen et al. 1998) (solid lines in Figure 5).

Referee: *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 ..."*

Response: Thank you for the useful suggestion. We rephrased the sentence as follows:

Root exudation products consist of carbon-based compounds deriving from plant metabolic activity that are released from living roots (Shen et al., 2020). Their contribution to the carbon input to the soil can be modeled as a slight linear increase from the background ADD (i.e., the starting point of the ADD axis in Figure 5) to a minimum ADD value during the initial growing stage…During the development phase, it can be modeled with a more relevant growth…

Results

3.1 The role of rainfall seasonality on EW dynamics

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

Response: Thank you. We removed the comma as suggested.

Referee: *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 pCO2? 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.*

Response: Thank you for your useful comment. We moved the part where we explain the application of irrigation contributions (lines 242 – 257 of the old submitted version of the manuscript) to a specific subsection of the methodology section, named *2.2.2 Irrigation*. Basically, we did not consider a specific chemistry of the irrigation water; green water (irrigation) and blue water (rainfall) lowers the pH since it increases soil moisture and wet environments are characterized by a more acidic soil water. Then, the equilibrium between soil water (which derives from rain and irrigation, when applied) and atmospheric pCO_2 is set in the formation of the carbonic acid (H_2CO_3), which is produced by the part of the dissolved CO_2 that reacts with water (refer to Section 2.3 of Cipolla et al., 2021a, https://doi.org/10.1016/j.advwatres.2021.103934, for more details). Answering your last question, when soil moisture is too low (i.e., close to the hygroscopic point) the olivine weathering rate is basically low because it linearly depends on soil moisture (see equation 3 in Cipolla et al., 2021a, https://doi.org/10.1016/j.advwatres.2021.103934). At that point, the saturation state of olivine is low as well given the slow dissolution of the mineral and of the consequent release of reaction products (i.e., magnesium and silicates).

Referee: *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."*

Response: Thank you for the useful suggestion. We rephrased the sentence as follows:

Weathering rate dynamics are affected by soil moisture and pH. Due to irrigation, which leads to higher soil moisture and lower pH, weathering rate is higher in California with respect to Iowa during summer.

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

Response: Thank you for the useful suggestion. We rephrased the sentence as suggested.

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

Response: Thank you for the useful suggestion. We rephrased the sentence as suggested.

Referee: *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."*

Response: Thank you for the useful suggestion. We rephrased the sentence as suggested.

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

Response: Thank you for the useful suggestion. We rephrased the sentence as suggested.

Referee: *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 ...".

Response: Thank you for the suggestion. The rephrased sentence is:

For the same soil and vegetation, higher rainfall leads to an olivine dissolution considerably faster in Iowa than in California due to higher soil moisture driven by higher seasonal rainfall frequency (λ). For the Italian case studies…

Referee: *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?*

Response: We actually meant "larger differences" in terms of MAP. We provided to replace "More relevant differences" with "larger differences" to better clarify the sentence.

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

Response: Thank you for the useful suggestion. We rephrased the sentence as suggested.

3.2 The role of soil type on EW dynamics

Referee: *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.*

Response: We totally agree with the Reviewer in this statement. However, we took only these two soil types into account since they are the most frequent textures in the areas under study. We are aware that considering more extreme textures would emphasize the differences in EW dynamics due to soil types, but since we are referring all the analyses to four specific locations, we would prefer to maintain the presented structure in this manuscript. However, what is suggested by the Reviewer is certainly a useful aspect to be analyzed in a future work.

3.3 The role of vegetation on EW dynamics

Referee: *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 ..."*

Response: Thank you for the insight. We rephrased the entire period as follows:

As visible in Figure 10, when either of the two crops is in the rest phase (for example, in DOY 180-300 for wheat in Sicily and in DOY 0-100 and 250-365 for corn in Padan plain), water losses due to bare soil evaporation are similar in magnitude to transpiration for the other crop. It happens that, when corn is in the rest phase and at the same time wheat is in its initial or mid-season stage, in the first case water losses are mainly governed by bare soil evaporation, while in the other one by crop transpiration. The fact that wheat and corn cycles are not in phase does not affect much water balance and, in turn, pH and weathering rate dynamics.

3.4 EW case studies

Referee: *Line 322: Is this a one-time application of 10kg/m2 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.*

Response: Thank you for the insight. We actually moved this information to the end of Section 2.1, where an outline of the various simulation scenarios is presented. You can refer to the response to the comment related to section *2.1 Methodology* for more details about the olivine amendment.

Referee: *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."*

Response: Thank you for the useful suggestion. We rephrased the sentence as suggested.

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

Response: Thanks for noticing the mistake. We corrected it.

Referee: *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.*

Response: Thank you for this suggestion. We split the paragraphs in a better way. Indeed, the sentence "*A similar situation can be observed from the comparison between Sicily and the Padan plain (Figure S2 of the supplementary material).*" belongs to the previous paragraph, where a comparison between Iowa and California is presented. The new paragraph, where a comparative analysis between Sicily and California and Padan plain and Iowa is reported, now starts with the sentence "*Because of the similar rainfall seasonality*....".

Referee: *Line 338: Remove "thus"*

Response: Thanks. Removed!

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

Response: Thank you for the useful suggestion. We rephrased the sentence as suggested.

Referee: *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, m2 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.

Response: Thank you for your valuable comment! The dissolution rates we achieved here are faster than those of Amann et al., (2020), despite the similar annual rainfall rate and soil pH, although this could depend on many other factors, such as CEC and rainfall seasonality that affects the soil moisture signal. We tried to rephrase our sentence, specifying that our dissolution rates are more typical of a field environment rather than those obtained in laboratory conditions, also indicating some characteristics of the experiment presented in Amann et al., (2020), such as soil type, pH and average annual rainfall. Regarding the information about the olivine application, please refer to the response to the comment related to section *2.1 Methodology*. The modified sentence in Section 3.4 is reported below:

The order of magnitude of weathering rates provided by our model is more typical of the field environment with respect to those achieved in laboratory conditions. Indeed, we achieved weathering rate values similar to those presented in the mesocosm experiment of Amann et al., (2020), which used a loamy sandy soil with pH equal to about 6.6 and a total amount of annual rain of 800 mm y^{-1}*, similar to the annual average soil pH for Iowa.*

Referee: *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.*

Response: Thank you for the in-depth analysis of the manuscript. As we stated in Section 2.2.2, the CEC was not calibrated, but just extracted from measurements related to the cropland areas under study. We understand this may be a bit confusing in line 348, where we stated to have calibrated both the CEC and the background weathering flux. We thus rephrased the sentence as follows, also explaining how these components affect EW dynamics:

This aspect stresses the importance of using measurements of soil properties (e.g., CEC, pH) for calibrating the background weathering flux, allowing to obtain more realistic estimates of olivine dissolution dynamics. Indeed, CEC and background weathering strongly affect pH levels before olivine amendment and, in turn, dissolution rates which are faster under more acidic conditions (e.g., the case of Iowa that has the slowest background weathering flux).

Referee: *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.*

Response: The experiment we cited is that of Amann et al., (2020) since about the double rate of olivine is added to the soil, with respect to our setup. We corrected the amount of forsterite added to their mesocosm experiment to make a more reliable comparison with our results. The forsterite application rate in our setup (i.e., 10 kg m^{-2}) is here specified at the end of Section 2.1, along with other details of olivine application (refer to the response to the comment related to section *2.1 Methodology*). The modified sentence is the following:

The values achieved for the US locations are lower but still comparable with those of Amann et al. (2020), that derived a sequestered CO_2 within the range 23 - 49 kg ha^{-1} y^{-1}, amending more than the double olivine with respect to our study (i.e., 22 kg m^{-2} of dunite corresponding to about 18 kg m^{-2} of olivine) in his mesocosm experiment with conditions similar to the field environment.

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

Response: Thank you. We replaced the word as suggested.

Referee: *Line 395: Here olivine-derived CO2 leached and CO2 in soil water are distinguished, with leached CO2 being the preferred metric of carbon sequestration. If EW were rolled out on a large scale and CO2 consumption then calculated based on river water samples, then the leached DIC (dissolved inorganic carbon), alkalinity or HCO3 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 flow paths 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 flow paths. Following the carbon all the way to the ocean where it is believed to be sequestered on 105-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.*

Response: Thank you for the very important comment. We totally agree with the Reviewer that assessing EW carbon sequestration is not an easy task. There is, indeed, much uncertainty in the scientific community about the path that carbonates and bicarbonates, produced by the weathering reaction, may follow on the way to the oceans where, as you stated, they are assumed to be sequestered on long timescales. We here distinguished the HCO_3^- and CO_3^{2-} produced by the reaction of olivine with CO_2 and remain dissolved in soil water from those that are leached away from the reference domain. Indeed, the former can easily react with H^+, making the carbonic acid (i.e., H_2CO_3) which, since it is in equilibrium between the liquid and the gas phase, can thus lead to a CO_2 release to soil pores and, in turn, to the atmosphere (refer to Cipolla et al., 2021a, https://doi.org/10.1016/j.advwatres.2021.103934, and Cipolla et al., 2021b, https://doi.org/10.1016/j.advwatres.2021.103949, for more details). The mass of HCO_3^- and CO_3^{2-} that goes to groundwater is therefore computed as the product between the leaching rate and the concentration of these ions in soil water at each time step, taking into account only the extra amount produced by weathering reaction. The boxplots reported in Figure 11 are representative of the seasonal variability of the sequestered CO_2 and are derived by means of 10 values (one per year in the considered time window) of leached mass of CO_2 per month. We better clarified this aspect in Section 3.4 of the manuscript. The added part is the following:

In particular, for each time-step, the loss of HCO_3^- and CO_3^{2-} to groundwater is derived by the product of the leaching rate and the concentration of these ions in soil water, taking into account only the extra amount produced by weathering reaction.

Referee: *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.*

Response: We rephrased the sentence as suggested. It was a mistake to cite Hartmann and Moosdorf (2012) here since, as you correctly state, the role of nitrogen on weathering is not mentioned. We meant to cite Hartmann et al., (2013) (https://doi.org/10.1002/rog.20004), where this aspect is clearly defined (i.e., see Figure 1).

Referee: *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"*

Response: Thank you for the useful suggestion. We rephrased the sentence as suggested.
