

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 #2

General comments:

Referee: *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 CO₂ 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 CO₂ 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.

Response: We thank the Reviewer for the in-depth analysis of this work. The comments provided many valuable suggestions about soil mineralogy, highlighted some aspects that needed to be explained more in detail (i.e., the characteristics of the olivine amendment), and pointed out possible future developments of the model. The multi-mineral EW represents certainly an aspect worth to be analyzed, either from the perspective of putting it into practice, or understanding which of the various minerals can provide the best carbon sequestration rates.

Specific comments:

Referee: *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.

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

Title:

Referee: *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'.

Response: Good point. We revised the title as: "Effects of precipitation seasonality, irrigation, vegetation cycle and soil type on enhanced weathering – Modelling of cropland case studies across four sites".

Abstract:

Referee: *- lines 9-10: ... 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.*

Response: Thank you for the recommendation. We replaced “*pre-EW soil pH*” with “*background weathering flux*”, which is the component of the model that mostly determines soil pH before olivine amendment.

Referee: - lines 10-11: *Looking at the numbers presented here for sequestered CO₂, 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.*

Response: Good point! We now better clarified the sentence as suggested. You can find below the modified part of the manuscript.

Regarding the US case studies, Iowa sequesters the greatest amount of CO₂ if compared to California (4.20 and 2.21 kg ha⁻¹ y⁻¹, respectively), and the same happens for Sicily with respect to the Padan plain (0.62 and 0.39 kg ha⁻¹ y⁻¹, respectively).

1 Introduction:

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

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

Referee: - lines 21-22: *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.*

Response:. We rephrased the sentence as follows:

...which can precipitate as carbonates in the soil or at any stage during their transport from land to ocean.

Referee: - lines 22-25: *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 (Mg₂SiO₄) and fayalite (Fe₂SiO₄), where the Mg richer varieties are more common and also more reactive with CO₂ and H₂O. (Generally, it is the Mg-Ca-silicates that have the most potential for CDR - forsterite and wollastonite CaSiO₃.) So for ease of representation/calculation Mg₂SiO₄ is often used to represent an olivine mineral with real formula (Mg_{1-x},Fe_x)₂SiO₄.

- 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)

Response: Thank you for your clarifications. The rephrased sentence is as follows:

Many studies discuss using olivine (often modeled as the end-member forsterite Mg_2SiO_4 or fayalite Fe_2SiO_4 , although the former is the most common mineral that dissolves and reacts faster with CO_2) in EW applications (Köhler et al., 2010; ten Berge et al., 2012). This mineral can be extracted from igneous rocks, such as volcanic (i.e., basalt), plutonic (i.e., gabbro) and mostly from ultramafic rocks, which can have up to 95 % of olivine and are widely distributed across the globe. Additionally, olivine is characterized by relatively fast dissolution rates if compared to other silicate minerals, such as albite and orthoclase (Hartmann et al., 2013).

Referee: - lines 28-29: *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.*

Response: We agree with the reviewer. Therefore, we replaced the “White & Barley (2003)” reference with a more recent one (<https://iopscience.iop.org/article/10.1088/1748-9326/aaa9c4/meta>) that mentions the uncertainties of field weathering rates.

Referee: - line 34: *“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 -> “such as basalt or wollastonite”*

Response: Good suggestion. We specified that silicate minerals containing calcium and magnesium are usually used for EW. The modified sentence is the following:

To begin to address these uncertainties, several experimental approaches have been carried out to characterize olivine or any other Ca-Mg-silicate mineral (such as wollastonite) used for EW dissolution dynamics.

Referee: - line 35: *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 [https://doi.org/10.1016/S0016-7037\(00\)00434-8](https://doi.org/10.1016/S0016-7037(00)00434-8) ; Oelkers et al 2018 <https://doi.org/10.1016/j.chemgeo.2018.10.008> ...)*

Response: Thank you for highlighting this aspect. We replaced the references as suggested, also adding a work about an experimental setup used to extract wollastonite dissolution rates (10.1023/B:BIOG.0000015787.44175.3f).

Referee: - lines 35-36: 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 <https://doi.org/10.5194/bg-18-169-2021>).

Response: Thank you. We better distinguished the state-of-the-art of EW experiment. The adjusted part of the manuscript is the following:

These are mainly based on laboratory experiments conducted on single mineral particles (Oelkers et al., 2018; Pokrovsky and Schott, 2000; Peters et al., 2004), laboratory experiments involving soil cores/columns amended with silicate rock powder (SRP), under controlled temperature and irrigation conditions, without biological processes (Renforth et al., 2015; Dietzen et al., 2018), 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) and field trials where SRP is added outdoors to a field, grassland or forest soil representing complex open system of real life conditions (Taylor et al. (2021); Haque et al. (2020) using wollastonite).

Referee: - line 41: "magnesium and silica concentrations"

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

Referee: - lines 42-43: Please clarify that weathering rates of $10^{-13} \text{ mol}/(\text{m}^2.\text{s})$ refers to the surface of the mineral grain in contrast to sequestration rates in $\text{kgCO}_2/(\text{ha}.\text{year})$ which refers to land surface on which mineral dust is spread on.

Response: Good point. We modified this part of the manuscript as follows:

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 $10^{-13} \text{ mol m}^{-2} \text{ s}^{-1}$ corresponding to carbon sequestration rates of 23 and 49 $\text{kgCO}_2 \text{ ha}^{-1} \text{ y}^{-1}$, where ha refers to land surface over which mineral dust is spread.

Referee: - line 46: 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?

Response: Thank you. We added a reference (<https://doi.org/10.1016/j.gca.2014.10.013>) describing the gap between laboratory and field weathering rates for albite, which is a silicate mineral containing calcium, also used for EW applications. However, we prefer to cite “White and Brantley (2003)” in this part of the manuscript since it makes a very clear distinction between intrinsic (e.g., shape and roughness of mineral surface particles) and extrinsic (e.g., pH, temperature and soil water content) factors responsible for the differences between lab and field weathering rates.

Referee: - line 60: *“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)*

Response: Thank you for highlighting this aspect. We modified the sentence in the manuscript as follows:

By introducing stochasticity in rainfall and connecting ecohydrological with biogeochemical processes, the model presented in Cipolla et al. (2021a) leads to carbon sequestration rates of the same order of magnitude as those in the mesocosm experiment of Amann et al. (2020), that represents conditions similar to those in the field, despite not being in the full extent of their complexities.

Referee: - lines 63-64: *“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”?*

Response: Good suggestion. We modified the sentence as indicated.

Referee: - line 66: *The acronym “MAPs” is used here without introducing/explaining it.*

Response: Thank you for raising this aspect. “MAPs” was a mistake. We replaced it with “MAP”, which stands for “Mean Annual Precipitation”.

2.1 Methodology:

Referee: - line 88: *Long sentence which might be more easily readable by splitting as “... to which we refer for details. It links ecohydrological and ...”*

Response: We split the sentence as suggested.

Referee: - line 90: *“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?*

Response: The four components are presented as i) “Organic matter”, ii) “DIC system”, iii) “CEC” and iv) “Dissolved minerals” in Cipolla et al., (2021a) (<https://doi.org/10.1016/j.advwatres.2021.103934>). Since here only a short description of the model is provided, we referred the reader to this manuscript for more details.

Referee: - line 93: “... of soil water **ions** released by olivine dissolution...” as you refer both to silicates which are anions and magnesium which forms cations

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

Referee: - line 94: Mg^{2+} can be removed here as base cation as it is already referred to as one of the main ions formed upon olivine dissolution

Response: Good point! We removed it.

Referee: - line 95: “the ... (CEC) accounts for the process between”: which process?

Response: Thank you for raising this unclear aspect. We were referred to the adsorption process, which is now clearly expressed in this sentence.

2.2 Study areas and data:

Referee: - line 116: there seems to be a mistake with the web link: there is a ‘c’ in subscript <https://ipad.fas.usda.gov/rssiws/al/globalcropprod.aspx> and I get an error message when trying this link

Response: Thank you. It was a LaTeX typo. We corrected it.

Referee: - line 119: “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?

Response: We specified in this part of the manuscript the active root zone depths of the considered crops. The modified version is the following:

As in Cipolla et al. (2021b), all simulations are related to a unit ground area of homogeneous soil, vegetation and rainfall characteristics, vertically delimited by the active root zone depth of the involved crop, i.e., 40 and 60 cm for the corn and wheat, respectively (Fan et al., 2016).

2.2.1 Rainfall seasonality:

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

Response: Replying to the comment related to line 66, we introduced the explanation of the acronym MAP before this point of the manuscript. So, the reader at this point can understand its meaning.

Referee: - line 126: 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.

Response: That is correct. We extensively wrote what the acronym ARPA stands for.

Referee: - lines 133-134: “... are two months out of phase, ...” If ‘out of phase’ refers to different trends for α and λ , 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).

Response: We actually meant that the trend of α presents a time shift of about two months with respect to the trend of λ . We better clarified this aspect in this way:

For the latter, the monthly time series of the two parameters are shifted by about two months...

2.2.2 Soil type and composition:

Referee: - line 151: 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?

Response: This is a very valuable consideration! We actually derived C_0 and C_b values from the GSOC map, so they are related to the first 30 cm soil layer. However, the application domain is characterized by a unit surface area and a depth equal to the active root zone, which is higher with respect to the carbon data availability, for both the considered crops. Despite we are fully aware that soil organic carbon is not homogeneously distributed, we assumed, because of limited data availability, that the carbon stock over the first 30 cm is distributed over the whole 40-60 cm rooting depth.

Referee: - line 163: ... consume H^+ ions...

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

Referee: - line 165: “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.

Response: Thank you for this comment. Even if not well described in the original version of the manuscript, we are aware that soil mineralogical input data are not as representative as meteorological input data for the four analyzed locations. Indeed, our aim is not to describe an exact location (with specific coordinates), but is more devoted to describe a generic geographical area. For this reason, the background component of the model, which affects the baseline soil pH (i.e., before olivine amendment), is characterized by calibrating the background dissolution rate constants on the base of pH measurements. This has also been carried out to incorporate other factors contributing to the consume of H^+ (i.e., the action of microorganism or other less present minerals). As can be seen in Section 2.2.2, at the end of the calibration of the background weathering flux, we achieved dissolution rate constants values very different to those typical of calcite and quartz minerals, that mainly compose carbonate and siliciclastic sedimentary rocks, respectively. The availability of pH data and the possible calibration of the background weathering component are therefore the main reasons why we decided to use these “raw” data to describe the mineralogical characteristics of the soils under study. To make this clear in the manuscript, we modified this part in the following way:

...This weathering flux can be estimated on the basis of the mineral composition of the soil and the type of the existing bedrock, but also depends on the action of various other factors that consume H^+ . As a preliminary indication of the mineralogical composition of the soil, the lithological map presented in Hartmann and Moosdorf (2012) was used to extract the nature of bedrock at the cropland areas for the four considered sites. Sicily and the Padan plain are prevalently characterized by carbonate sedimentary rocks (e.g., limestone, dolostone mainly composed of carbonate minerals, such as calcite or dolomite), while the other two sites in the USA mainly present siliciclastic sedimentary rocks (e.g., sandstone, conglomerate mainly composed of silicate minerals, such as quartz or feldspars)....

...

However, soil pH depends on other factors that are not considered in the EW model (i.e., the presence of fertilizers, the action of microbes, fungi and bacteria or the action of other minerals that may release or take up H^+ ions).

Referee: - lines 168-170: 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.

Response: Thank you for highlighting this aspect. We corrected the sentence of the manuscript as indicated:

Sicily and the Padan plain are prevalently characterized by carbonate sedimentary rocks (e.g., limestone, dolostone mainly composed of carbonate minerals, such as calcite or dolomite), while the other two sites in the USA mainly present siliciclastic sedimentary rocks (e.g., sandstone, conglomerate mainly composed of silicate minerals, such as quartz or feldspars).

Referee: - line 170: “Lasaga (1984) and 44 (1979)” Please correct the later reference and perhaps also check more recent references on dissolution rate constants for carbonate and siliciclastic rocks.

Response: Thank you for highlighting this typo. We corrected it in this way:

...considering Lasaga (1984) and Plummer et al. (1979),...

Referee: - line 171: *... "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.*

Response: As in the response to the comment related to *line 165*, we are aware that this is a simplistic view of the mineralogical characteristics of the soil under study. Indeed, we only initially looked at the type of bedrock to have an idea of the order of magnitude of the background dissolution rate constants; then, we calibrated these parameters based on soil pH data. Please refer to the response to the comment related to *line 165* for the modified part of the manuscript.

2.2.3 Crop cycle:

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

Response: We added the meaning of FAO and the reference related to the crop coefficient values and length of the corresponding stages. The modified part of the manuscript is the following:

For each development stage, a single crop coefficient per crop type and the climatic area was obtained following the Food and Agriculture Organization (FAO) guidelines (tables 11 and 12 in Allen et al., 1998).

Referee: - lines 190-198: *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...*

Response: We specified the meaning of all the variables in equations (1) and (2) as suggested. Below you can find the modified part of the manuscript:

The effects of the seasonal pattern of the crop coefficient on transpiration losses, $T(s)$, were computed as,

eq(1)

where s refers to varying soil moisture, s_w is soil moisture at wilting point, while s^ is soil moisture at the incipient stress...*

The bare soil evaporation, $E(s)$, is evaluated as,

eq(2)

where s_h is soil moisture at the hygroscopic point and s_{fc} is soil moisture at the field capacity.

Referee: - lines 208-218: *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.*

Response: Thank you for your comment. We believe that the information regarding the crop cycle's different stages and lengths can be easily found in tables 11 and 12 in Allen et al. (1998). For this reason, dedicating an entire figure of the manuscript to this aspect may be redundant. We believe that how this aspect is presented in the current version, and also looking at Figure 4 of the manuscript, the reader can get a complete picture of the seasonal variability of the crop coefficient and the length of different stages.

3 Results:

Referee: - lines 226-228: *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-sitespecific 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.*

Response: As in the response to the comment related to *line 165*, we are aware that this is a simplistic view of the mineralogical characteristics of the soil under study, since we are not describing a specific location with its own coordinates but rather a generic geographical area. We stated in Section 2.2.2 that the considered mineralogical characteristics of the soils under study were derived in a simplistic way, but this was done given that the dissolution rate constants of the background weathering were then calibrated on the basis of pH data. Please refer to response to the comment related to *line 165* for more details.

Referee: - line 229: *Another major control factor of EW dynamics is the silicate rock dust powder (SRP) applied for EW. Its mineral composition greatly determines CO₂ sequestration potential (for example whether it is mostly olivine in ultramafic matle rocks, or olivine along with feldspars and volcanic glass in basalt). CO₂ 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 m²? 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”.

Response: Thank you for the very in-depth comment. For the presented analysis, we considered the same 100% forsterite rock dust since, as you correctly affirm, the main scope of our work is to explore the role of different factors (i.e., rainfall seasonality and irrigation, soil type and composition and crop cycle) on EW dynamics.

In particular, we modeled a one-time olivine amendment 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 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.

3.1 The role of rainfall seasonality and irrigation on EW dynamics

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

Response: We modified the title of Section 3.1 and added the symbols of the considered variables as suggested.

Referee: - line 236: *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.*

Response: This is a good suggestion. We added what the blue and red colors represent in the heatmaps.

Referee: - line 240: *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?*

Response: That is correct. Given that results are always expressed at each year from day 0 to day 365, we do not need to specify “Julian” day. We replaced it with DOY (Day Of the Year) in the text.

Referee: - line 241: *As before, Julian day needed or is “some days around day 300” also ok?*

Response: The response to this comment follows the one to the comment related to line 240.

Referee: - line 254: *Soil moisture time-series in the figure 6 caption is referred to as panel b , not c)*

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

Referee: - line 255: *Please rewrite as “the field capacity in the days from about day 100 up to day 250”.*

Response: Done, thank you.

Referee: - lines 242-257 until “... is provided.”: *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.*

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), along with Figure 6 that became Figure 4, to a specific subsection of the methodology section, named 2.2.2 *Irrigation*. Discussions on the effects of irrigation contributions on EW dynamics (i.e., heatmaps in Figure 5 of the old submitted version) remained in Section 3.1 instead.

Referee: - line 240: 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**”.

Response: Right! Thank you.

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

Response: Good point! In general, the presence of a high soil water content is certainly due to greater rainfall and/or irrigation contributions. If you look, for instance, at equation (21) in Cipolla et al., (2021a) (<https://doi.org/10.1016/j.advwatres.2021.103934>), you can notice that higher precipitation leads to a higher input of H^+ since rain is characterized by a slightly acidic pH (about 5.6). Furthermore, a greater soil water availability leads to a greater transpiration rate, which reflects into a higher nutrient cations uptake by plants (i.e., Mg^{2+} , Ca^{2+} and K^+). This is translated in a higher input of H^+ by plants, given that they tend to maintain a neutral charge.

Referee: - line 261: 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.

Response: We replaced the word “Julian” with DOY, as indicated before.

Referee: - line 263: Please replace “the Julian day 300” with “the 300th day” or “around day 300”.

Response: We replaced the word “Julian” with DOY, as indicated before.

Referee: - lines 267-269: 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.

Response: We better clarified that, in lines 267-269, we were referring to wheat and that the seven times higher weathering rate for Iowa with respect to California comes from averaging the grain scale weathering rate ratio. At the same time, we specified in lines 259-261 that the results described here are related to corn. The modified part is the following:

Lines 267-269:

Averaging the grain scale weathering rate ratio achieved considering wheat, over the considered 10 years, weathering rates derived for Iowa are about seven times higher than those in California under the two considered soil types, resulting from slightly higher average soil moisture and slightly lower pH.

Lines 259-261:

Due to irrigation, which leads to higher soil moisture and lower pH, weathering rate is higher in California during summer. The average daily weathering rate ratio with corn assumes values higher than one...

Referee: - *lines 270-276 where the role of rainfall seasonality on EW dynamics is discussed for the Italian sites: 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).*

Response: We originally put this figure in the supplementary material for the sake of length of the manuscript. However, as you affirm, it makes sense to add it within the main paper since it describes different results with respect to the US case studies. We therefore provided to add it to the main paper, along with the related explanations of results.

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

Response: We rephrased the sentence as follows:

Similar considerations apply to corn grown in Italy (Figure S1 of the supplementary material). In summer, corn requires irrigation in Sicily, given the scarcity of precipitation, but not on the Padan plain.

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

Response: We rephrased the sentence as follows:

For the rest of the year, the weathering rate ratio between the Padan plain and Sicily, tends to be slightly less than 1, translating to slightly more favorable olivine dissolution dynamics in Sicily.

Referee: - *lines 278-279: 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).*

Response: We rephrased the sentence as follows and replaced “significantly” with “considerably”:

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, rainfall seasonality leads to small differences in EW dynamics, given the similar distribution of precipitation across the year.

Referee: - line 281-283: “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.”

Response: We rephrased the sentence as indicated.

3.2 The role of soil type on EW dynamics

Referee: - line 286: ... and silty clay loam **soil**, ...

Response: Word “soil” added!

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

Response: We added the parameters symbols as suggested (lower case *s* letter for soil moisture).

Referee: - lines 294-295: ...weathering rates obtained with the clay loam soil tends 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.

Response: We better clarified that, in lines 294-295, the two times higher weathering rate for CL with respect to SCL soil comes from averaging the grain scale weathering rate ratio. The modified part is the following:

Lines 294-295:

Apart from some spikes, occurring on some specific days, averaging the grain scale weathering rate ratio, we achieved that the clay loam soil results in a weathering rate about twice as high as what is obtained with the silty clay loam soil, at all four locations.

3.3 The role of vegetation on EW dynamics

Referee: - line 300: ... of **H⁺** to balance ...

Response: Corrected!

Referee: - lines 301-302: “Brady, 2017). Vegetation furthermore provides the organic matter that, once decomposed, is one of the CO₂ sources in the soil system...”

Response: Sentence rephrased.

Referee: - line 305: *... 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.*

Response: We better clarified that, in line 305, the four times higher weathering rate for corn with respect to wheat, if planted in a clay loam soil in Sicily and California, comes from averaging the grain scale weathering rate ratio. The new part of the manuscript is:

Looking at the panels in Figure 9, averaging the grain scale weathering rate ratio, it is evident that corn leads to a weathering rate, on average, about four times higher than the one achieved for wheat when planted on clay loam soil in Sicily and California...

Referee: - line 306-307: *“ and fourth row of the figure). When both crops are planted in a silty clay loam soil in the Padan plain and Iowa (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.”*

Response: Sentence rephrased as suggested.

Referee: - lines 311-312: *‘ 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 aa to day bb for wheat and from about day xx to day yy for corn) ’*

Response: Good suggestion. We rephrased the sentence adding, for example, the rest days for wheat and corn crop in California and Iowa.

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.

3.4 EW case studies

Referee: - lines 320-321: *‘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...’*

Response: Sentence rephrased.

Referee: *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.*

Response: We modified Figure 10 as suggested. You can find the new version of the figure below in your comment related to *Figure 10*.

Referee: - line 322: *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/m² chosen? Practically, farmers apply lime and other rock dusts annually at a rate of 1-4 tons/ha.*

Response: We moved the olivine amendment information at the end of Section 2.1 of the manuscript, as already suggested by the Reviewer. We have chosen this slightly higher application rate to get a relevant signal of olivine application. We also considered the application rate in the experiment of Renforth et al., (2015) (<https://doi.org/10.1016/j.apgeochem.2015.05.016>), in which 100 g of olivine are added to a soil sample within a cylindrical pot characterized by a diameter of 10 cm. This amount of olivine corresponds to about 13 kg/m², higher than our amendment rate.

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

Response: “Julian day” replaced with “DOY”.

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

Response: “Julian day” replaced with “DOY”.

Referee: - lines 333-336: *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 (2.13X10⁻¹²mol/m²s) than at the former (1.61X10⁻¹²mol/m²s). 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?

Response: Thank you for suggesting a way to rephrase the sentence.

Regarding your second point, the main difference in weathering rate between Iowa and California is due to the background weathering flux, which is greater in California with respect to Iowa. For this reason, olivine is amended in a more acidic soil in Iowa and its dissolution is faster.

Referee: - line 337: *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.”

Response: We added the heatmaps related to the Italian case studies to Figure 10 and their description was moved to the main text. The added part to the manuscript, that in the previous version was in the supplementary material, is the following:

A similar situation can be observed by comparing the two Italian sites as Sicily and Padan plain, which present only small differences in terms of the seasonality of soil moisture, pH, and, in turn, weathering rate. The highest soil moisture values for Sicily occur in the first (i.e., before DOY 100) and the last (i.e., from DOY 300 onwards) part of the year since, during those days, the greatest part of the total annual rainfall occurs. Low soil moisture values from the DOY 100 to about 250 are due, as in California, to the scarcity of precipitation in this period.

Referee: - line 338: ‘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.

Response: That is correct. We moved this part as the starting point of a new paragraph.

Referee: - lines 341-343: 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.

Response: Corrected, thank you.

Referee: - lines 346-347: ‘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.

Response: By using a loamy sandy soil with pH equal to about 6.6, which is similar to the annual average soil pH for Iowa, and a total amount of annual rain of 800 mm y⁻¹, Amann et al., (2020) achieved weathering rates of 10^{-13.12} and 10^{-13.75} mol m⁻² s⁻¹ for coarse and fine dunite, respectively. These rates are a bit lower than those achieved by our study (the annual average weathering rate for Iowa is 2.13 x 10⁻¹² mol m⁻² s⁻¹). This may be due to the fact that EW dynamics can depend on many other factors, such as CEC and the seasonality of rainfall that affects the soil moisture signal. Given these considerations, we modified this part of the manuscript specifying that our dissolution rates are more typical of a field environment rather than those obtained in laboratory conditions, also stating some characteristics of the experiment presented in Amann et al., (2020), such as soil type, pH and average annual rainfall:

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: - lines 348-349: ‘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

Response: We rephrased the sentence in lines 348-349 in the following way:

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.

Regarding the second point, as expressed before, we are aware that mineralogical information of soils is less realistic with respect to the other factors, but the calibration of the background weathering flux based on soil pH data should indirectly resolve this aspect, as described in the response to the comment of line 165.

Referee: - lines 352-353: “The **overall rather** low monthly values of sequestered CO₂ for all case studies are due to the generally low leaching rate, which **reflects** the low MAP values for all considered sites.”

Response: Sentence rephrased as suggested.

Referee: - line 354: “The annual average sequestered CO₂ **equals** 0.62 kg/ha for Sicily, ...”

Response: Sentence rephrased as suggested.

Referee: - lines 355-358: *The difference between the Amann et al. (2020) CO₂ 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/m² whereas in these models 10kg/m² 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 CO₂ sequestration value calculated in Amann et al. (2020)? What other factors might play a role in the difference of CO₂ sequestration rates obtained for these two different study approaches (mesocosm experiment and model)?*

Response: Amann et al. (2020) derived an amount of sequestered CO₂ higher than the one we estimated for Iowa, that is the site having closer MAP and pH to the experiment condition (i.e., a range of 23 - 49 kg ha⁻¹ y⁻¹ of the mesocosm experiment against 4.2 kg ha⁻¹ y⁻¹ obtained by our model). This can be explained by various reasons:

- Amann et al. (2020) applied 22 kg dunite m⁻² to their mesocosms. 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, which is almost the double of our application rate.
- The sequestered CO₂ in Amann et al., (2020) is calculated on the base of the chemical characteristics of the outlet water, thus on the base of the extra dissolved Mg²⁺ and DIC (Dissolved Inorganic Carbon), while we refer to the leached extra bicarbonates and carbonates produced by olivine dissolution.
- Furthermore, the experiment considers two precipitation regimes, with daily and weekly rainfall, delivering the same total annual precipitation volume, thus scheduled irrigation interventions, which certainly do not reproduce the stochasticity in the temporal distribution of precipitation.

We summarized these reasons in Section 3.4 of the manuscript. The added part is the one below:

Apart from this aspect, the differences in the achieved carbon sequestration rates may be due to the way in which this is computed. Indeed, while Amann et al., (2020) considers the dissolved magnesium produced by olivine dissolution, we refer to the leached of the extra bicarbonates and carbonates after olivine dissolution. Furthermore, the mesocosm experiment considers two different rain regimes, namely a daily and weekly rainfall, delivering the same total annual precipitation volume. These may be considered as scheduled irrigation interventions, which certainly do not reproduce the stochasticity in the temporal distribution of precipitation.

Referee: - line 362: ... with a corresponding increase of HCO₃-...

Response: Corrected.

4 Discussion and conclusions

Referee: - line 372: Analyzing the interactions between rainfall and crop properties...

Response: Corrected.

Referee: - line 378: ... with a corresponding increase of HCO₃...

Response: Corrected.

Referee: - lines 379-380: "... by olivine reaction with CO₂. 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."

Response: Sentence rephrased. Thank you.

Referee: - line 395: ... the one we called CO_{2,sw} ... Please shortly define/explain this parameter instead of just giving the symbol and referring to a previous publication.
...In effect, even in this our 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 CO₂ back to the atmosphere, and that hence (bi)carbonates leached out from EW zone are interpreted as more reliable measure for CO₂ 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 CO₂ sequestration calculations, might recombine to carbonic acid and degas CO₂ when they resurface or mix with water of different composition, temperature,... The permanence of CO₂ 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 CO₂. Another product from olivine dissolution is Mg²⁺ 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 CO₂ sequestration?

Response: We explained the meaning of the parameter CO_{2, sw} in the manuscript, as reported below:

...the one that we called CO_{2, sw} in Cipolla et al., (2021b), that represents the amount of extra bi(carbonate) anions dissolved in soil water due to olivine weathering.

We agree with you in all you affirm about uncertainties in carbon sequestration definition. Scientific literature, indeed, presents various studies that define it in different ways, representative of the fact that there is not a unique way to assess it. Sometimes, Mg²⁺ released from olivine dissolution is used (Amann et al., 2020, <https://doi.org/10.5194/bg-17-103-2020>; ten Berge et al., 2012, 10.1371/journal.pone.0042098; Renforth et al., 2015, <http://dx.doi.org/10.1016/j.apgeochem.2015.05.016>), while other approaches use extra DIC in soil water produced by olivine dissolution as a carbon sequestration metric (Beerling et al., 2020, <https://doi.org/10.1038/s41586-020-2448-9>). We agree with the fact that even leached bi(carbonate) anions may recombine to carbonic acid and release CO₂ into the atmosphere, depending on the chemical and mineralogical characteristics of the soils on their way to the oceans, as well as on biogeochemical processes that can occur along their path.

Our model is certainly able to compute the concentration of Mg²⁺ released by olivine dissolution, as you can see from Cipolla et al., (2021b). However, relying only on the dissolved magnesium in soil water may provide an uncertain estimation of carbon sequestration, given that this cation is removed from soil water by many processes. Indeed, cations such as Ca²⁺ and Mg²⁺ are essential macronutrients for living organisms (White et al., 2010, [10.1093/aob/mcq085](https://doi.org/10.1093/aob/mcq085)). Plant and bacteria uptake them when they are available in the water solution and from soil colloid surface exchange sites. For this reason, leached bi(carbonate) anions provide the most reliable carbon sequestration metric, at least with reference to the domain we adopt in the simulations, since they express the carbon that comes from the reaction of olivine with CO₂ and is taken away from the considered domain by the leaching process.

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

Response: Thank you. This is a promising challenge for future works.

Referee: - line 415: 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.

Response: We specified that basalt is a rock that may be also used for EW applications. Furthermore, we stated that the risk of heavy metals release occurs upon olivine dissolution since its crystals contain Ni and Cr. One of the future goals of our research is indeed to study the EW capabilities of other silicate minerals or assemblages of them (i.e., basaltic rocks) since, as you correctly stated, it would represent a more realistic situation and this rock is certainly easier to be found, crushed and used as amendment, with respect to single minerals. The modified sentence can be found below:

Indeed, many EW experiments have been conducted with wollastonite or using basaltic rocks, among various aspects to avoid or simply reduce the high Ni and Cr content potentially released by olivine during dissolution. These heavy metals are present, in fact, in olivine crystals and are then released upon olivine dissolution.

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

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

Referee: - lines 417-718: 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 sequestered 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).

Response: We totally agree with you in the sense that carbon sequestration from olivine is more uncertain to quantify with respect to wollastonite. You can refer to our response comments of line 395 for some considerations about carbon sequestration assessment.

Referee: - line 421: 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 of its greatest merits and strengths towards more realistic EW potential predictions.

Response: Thank you for appreciating and valuing this aspect of our work. Whether the seasonality of rainfall and, in turn, soil moisture is important or not in this process is one of the most debated aspects in scientific literature. We strongly believe, as you affirmed, that considering near saturation conditions with stationary water fluxes is a lot far away from what happens in reality. As we demonstrated with our results, in fact, hydroclimatic fluctuations are one of the most important aspects to predict EW dynamics.

Referee: - line 424-426: *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.*

Response: Once again we thank you for appreciating our work and providing very useful suggestions. We added this important aspect to our discussions, modifying the sentence as follows:

Therefore, a possible development of this work may consist of a comparison of EW yields under the amendment of different assemblages of silicate minerals (i.e., basaltic rocks) in various areas of the world, taking into account the dissolution rates of the individual minerals and their relative presence in the silicate rock dust, thus providing a more reliable prediction of EW dynamics

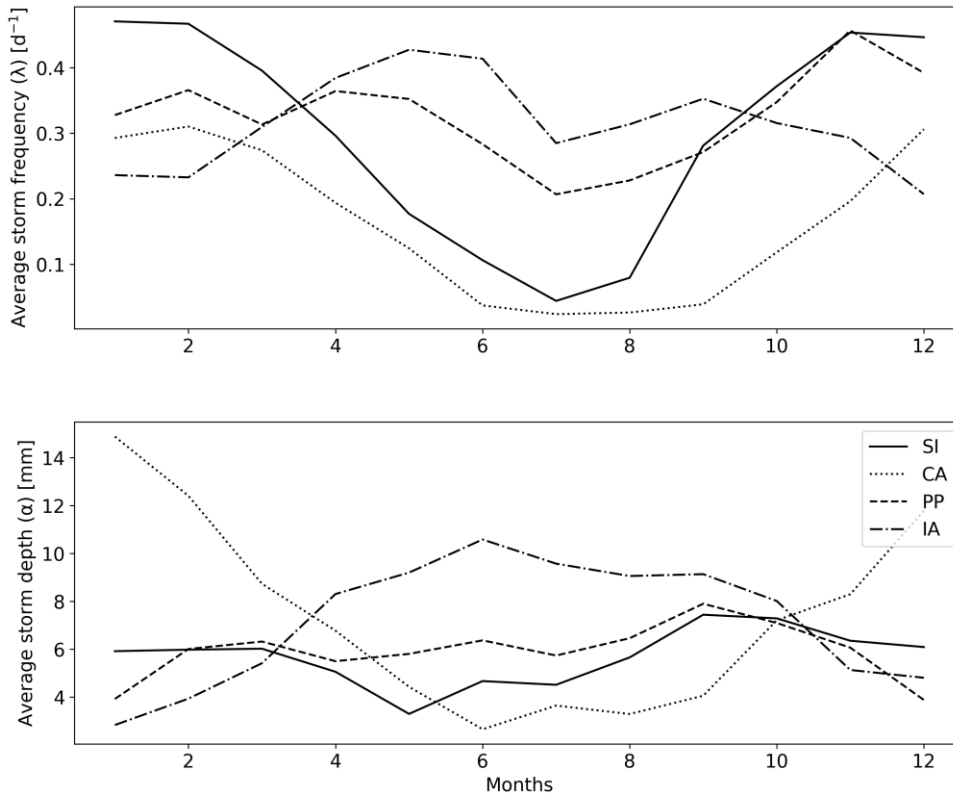
Figures

Referee: *Figure 3: To allow easier comparison between the average rainfall depth α and rainfall frequency λ (please label both fully on the vertical axes) between the 4 different areas, maintain the same scale for all four diagrams (i.e. λ up to 0.47 and α 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 α and λ “rainfall parameters” exactly represent.*

Response: We agree with you in the sense that the same scale is needed for both the rainfall parameters to easily compare them across the four selected places. For a better comprehension of the figure, we decided to plot the average rainfall depth (α) and frequency (λ) of the four sites under study in a single plot, showing them with different line styles, in order to have a clear plot even in a black and white print out. We also modified the caption, clarifying the meaning of the two rainfall parameters:

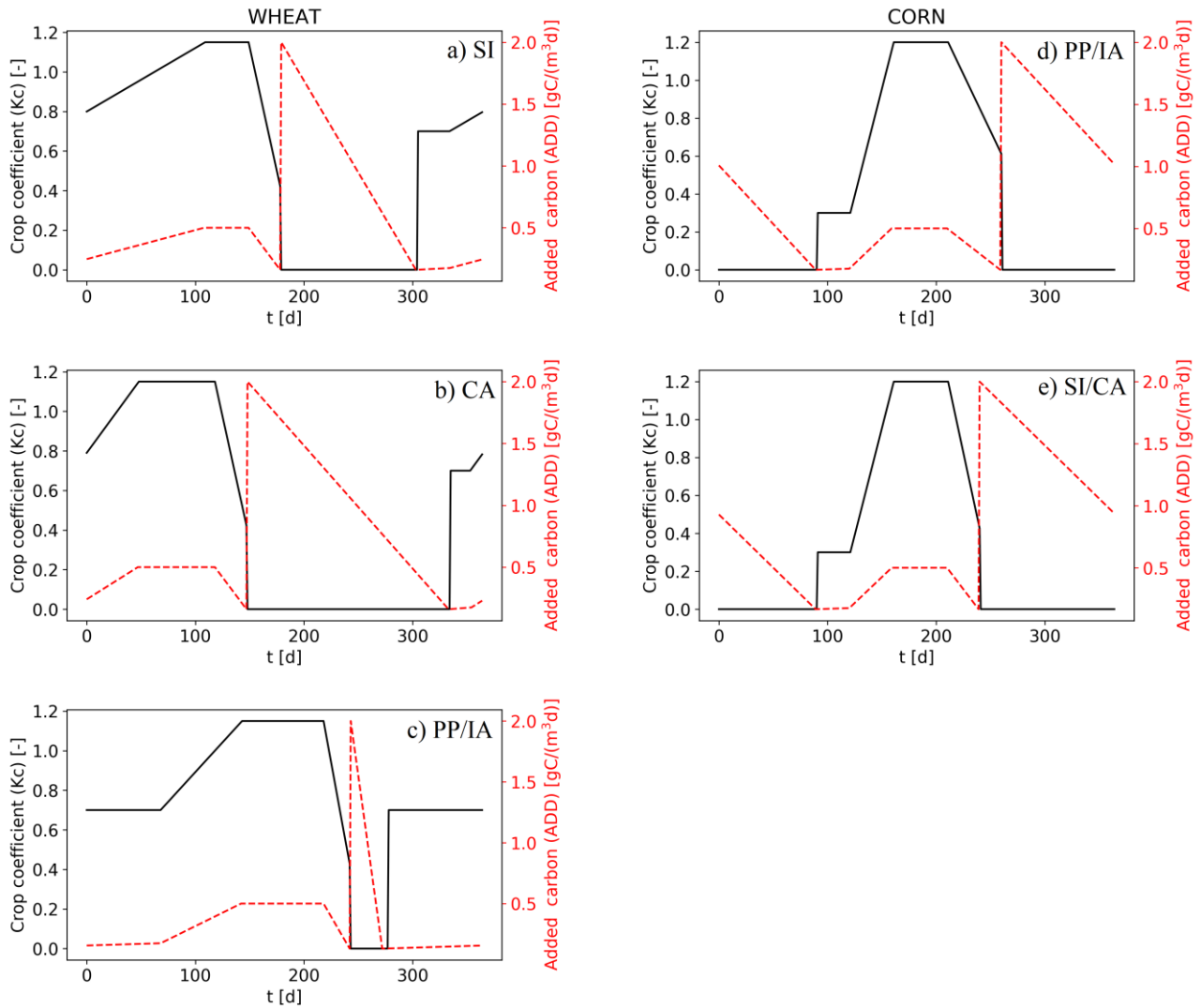
Values of the average rainfall depth (α) and frequency (λ) from January (i.e., month indicated with 1) to December (i.e., month indicated with 12). The reader is referred to Section 2.2.1 for more details.

You can find the modified figure in the following:



Referee: Figure 4: Please write full name and symbol for both parameters (crop coefficient K_c 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.

Response: Following your suggestions, we modified the figure:

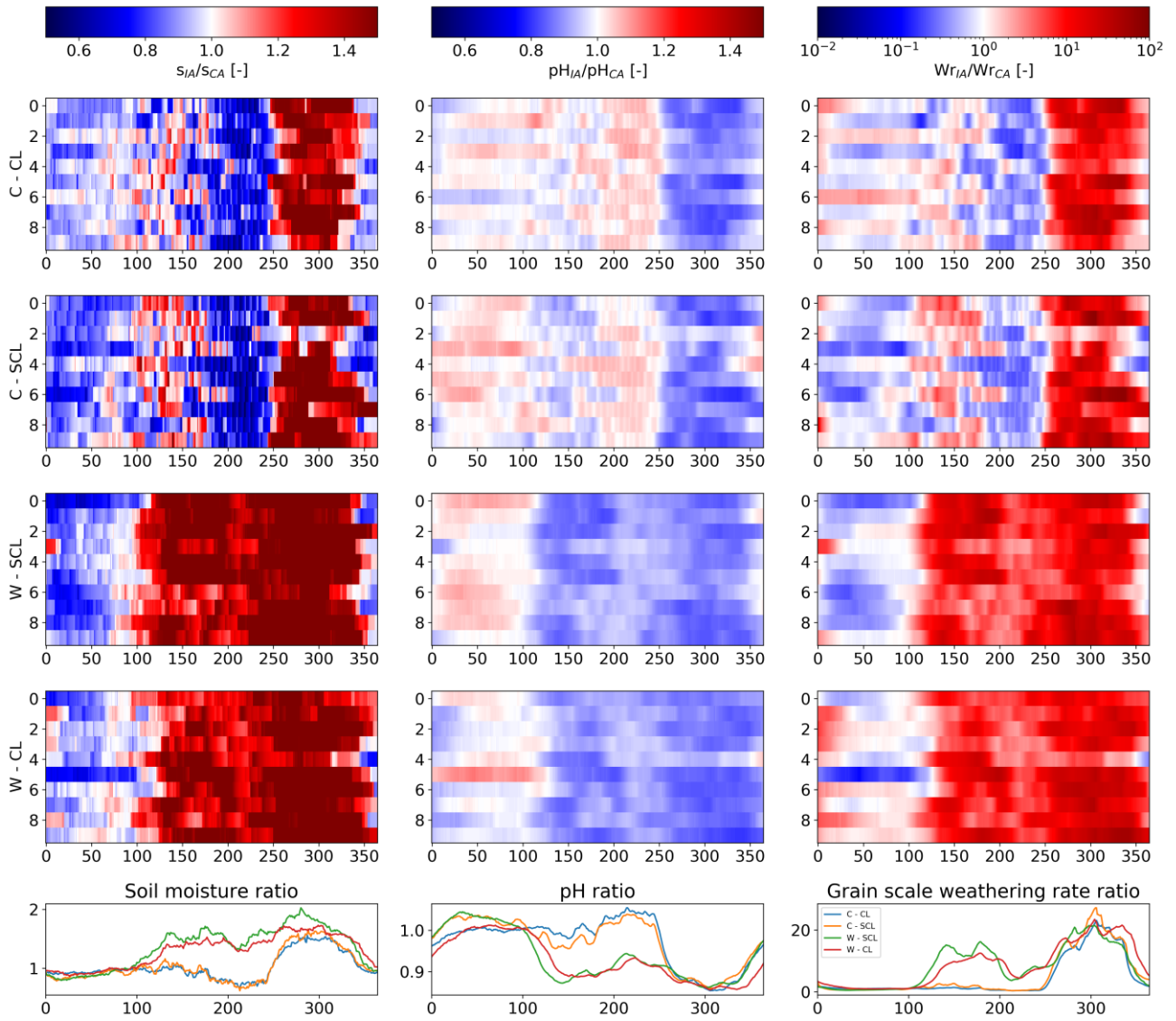


Furthermore, we modified the caption like this:

Seasonal variability of the crop coefficient (K_c) 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.

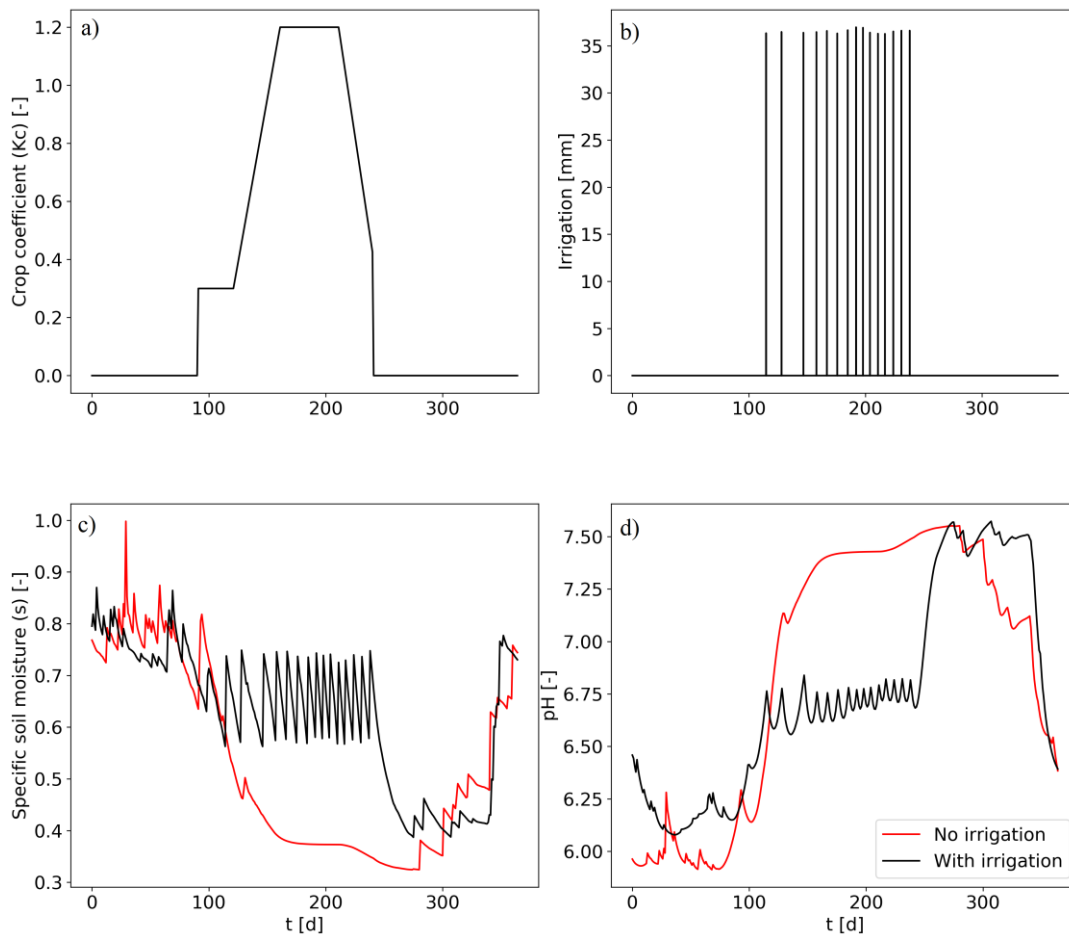
Referee: *Please add the respective symbols for the different variables to the figure caption: soil moisture (S), weathering rate (Wr), Iowa (IA), California (CA), corn in clay loam soil (C-CL), etc. To make this figure easier to read it would be good to have the colour legend just once, write the full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

Response: The figure is now clearer to read. This is the modified version:



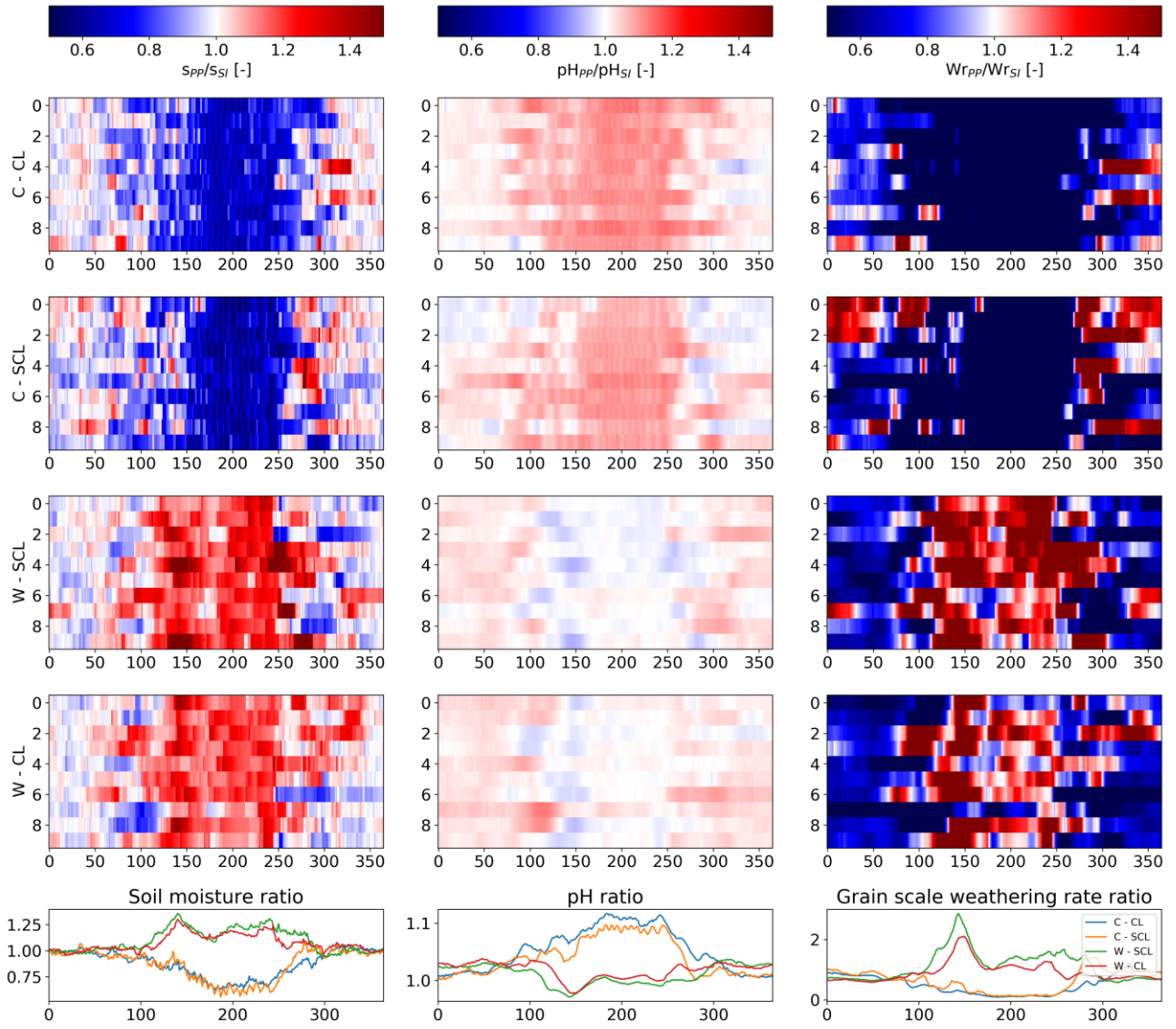
Referee: Figure 6: Please add the symbol to the parameter name in the figure caption, and the full nameto the symbol along the Y-axis of the specific graph (crop coefficient Kc, soil moisture S). Letters a), b). c) and d) are missing in the respective panels.

Response: Done! The modified version of the figure is shown below:



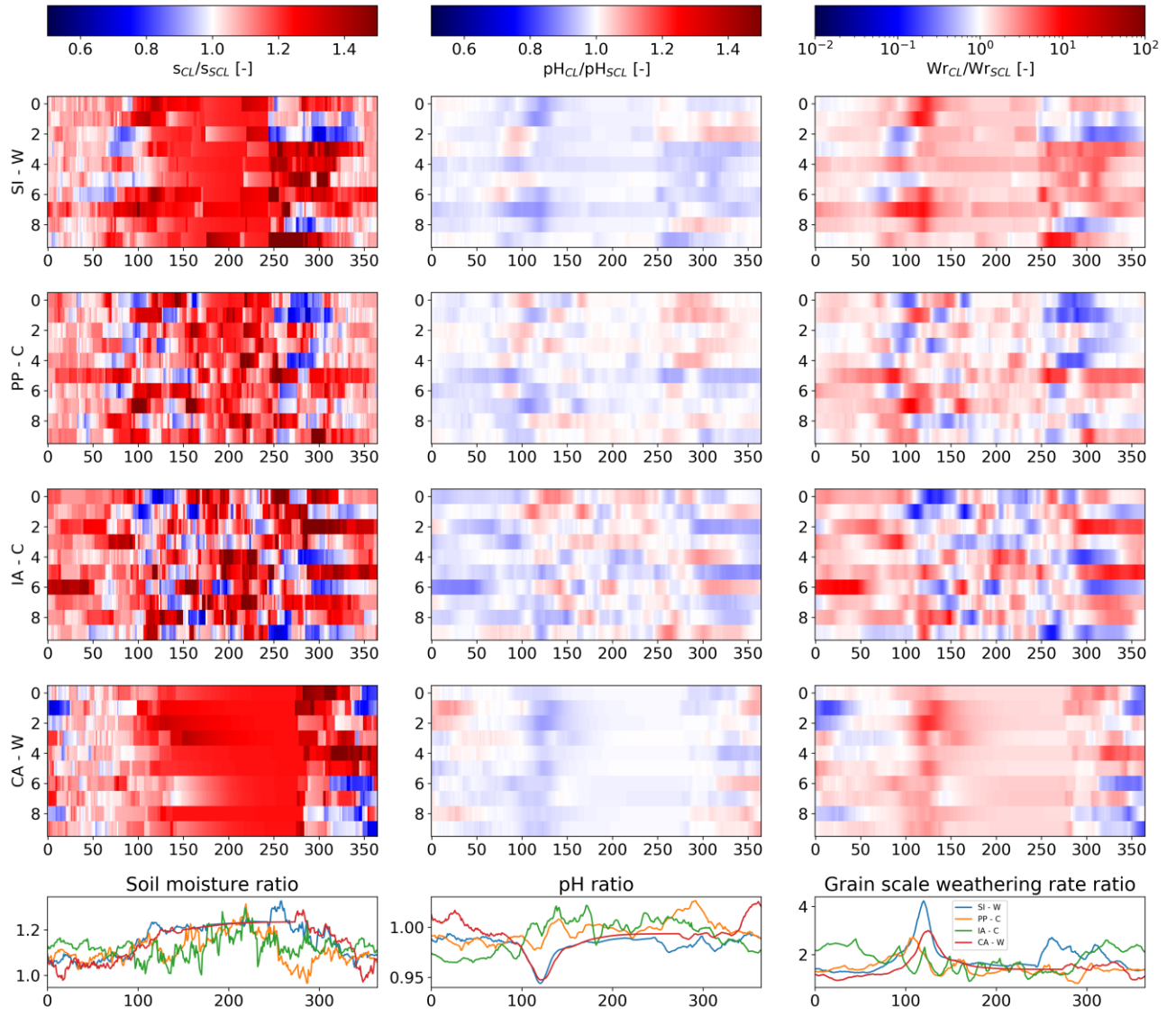
Referee: *Figure S1: Please make a main manuscript figure occurring after the time-series heat maps for the US sites, and make the same corrections/changes as detailed above for Figure 5. The resolution of the current S1 figure needs to be improved as the Y-axis labels are poorly readable both in print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

Response: We made the same corrections suggested for Figure 5. Now this figure is in the main manuscript after that related to the US case studies. The new version is the following:



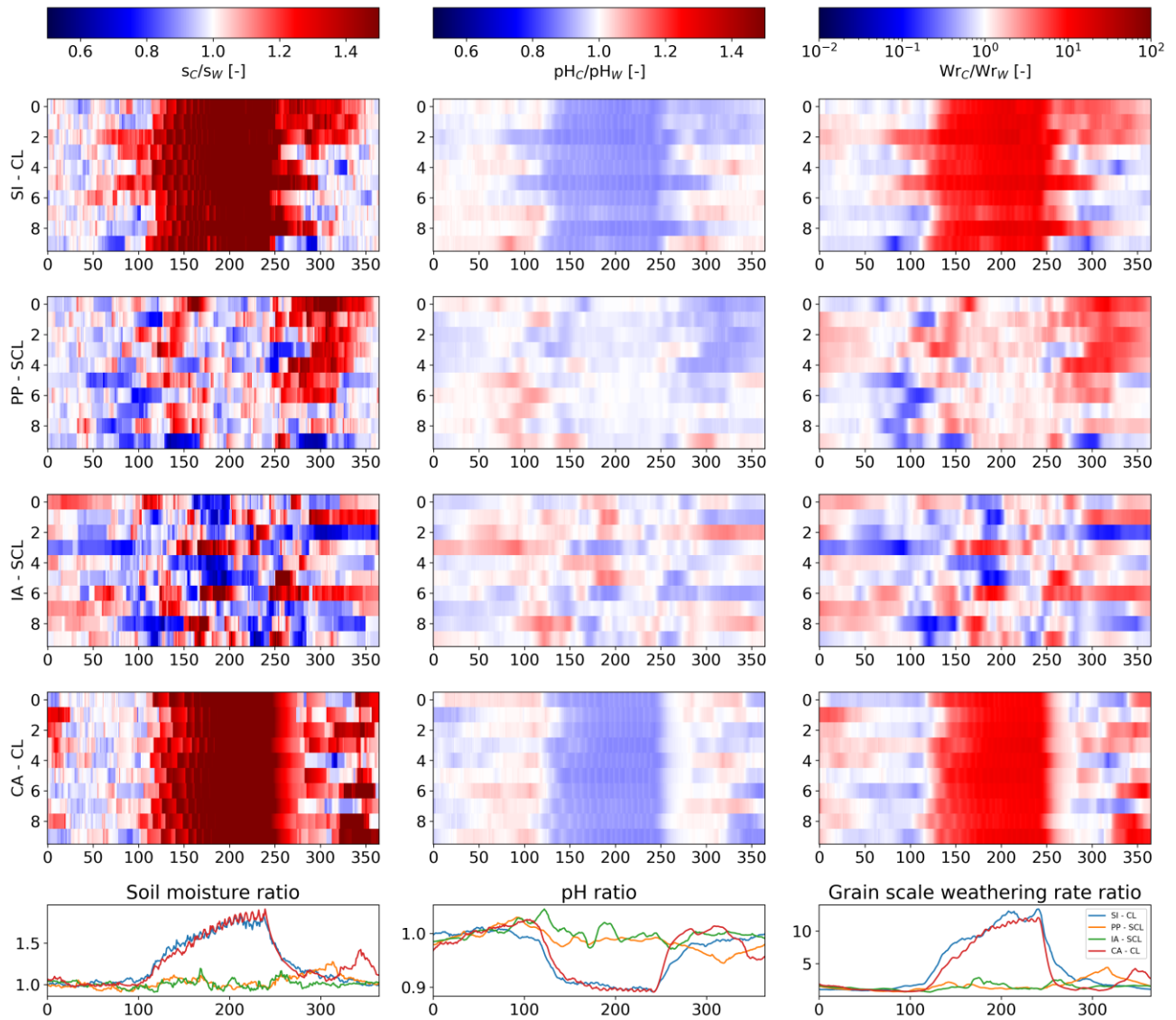
Referee: Figure 7: Please adjust in the same way as suggested for Figure 5.

Response: Same corrections as in Figure 5:



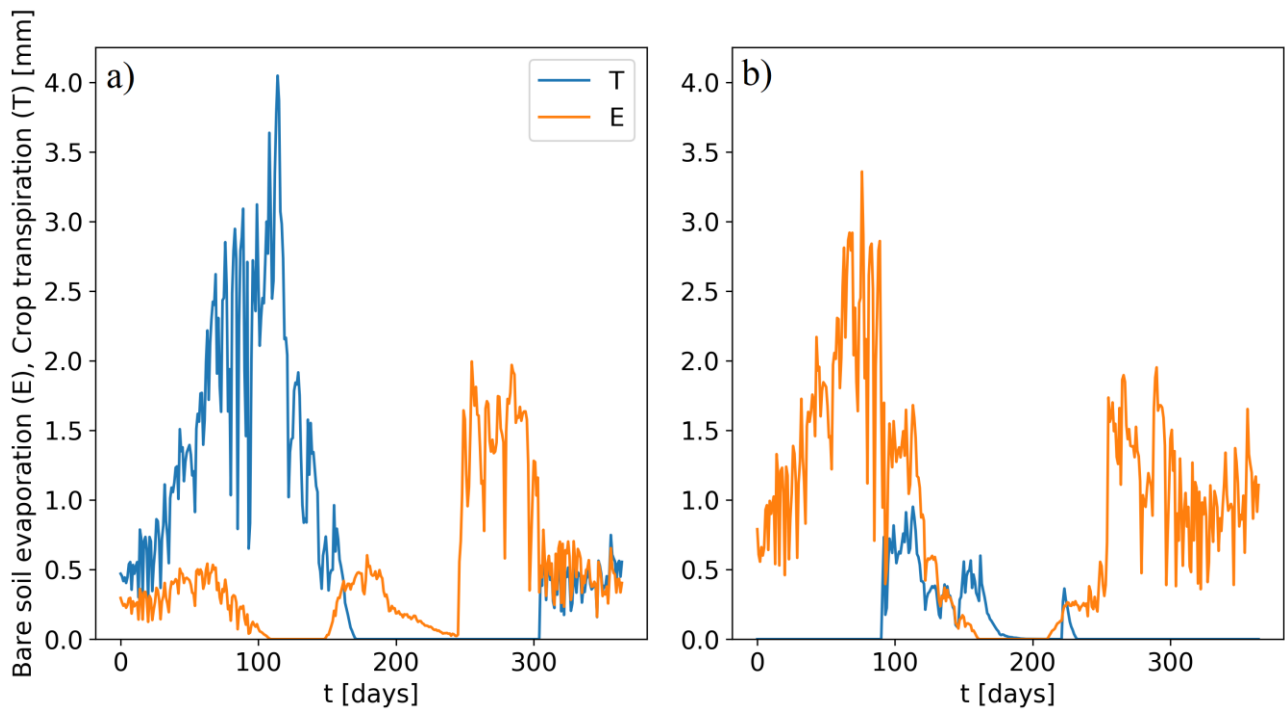
Referee: *Figure 8: Please adjust in the same way as suggested for Figure 5.*

Response: Same corrections as in Figure 5:



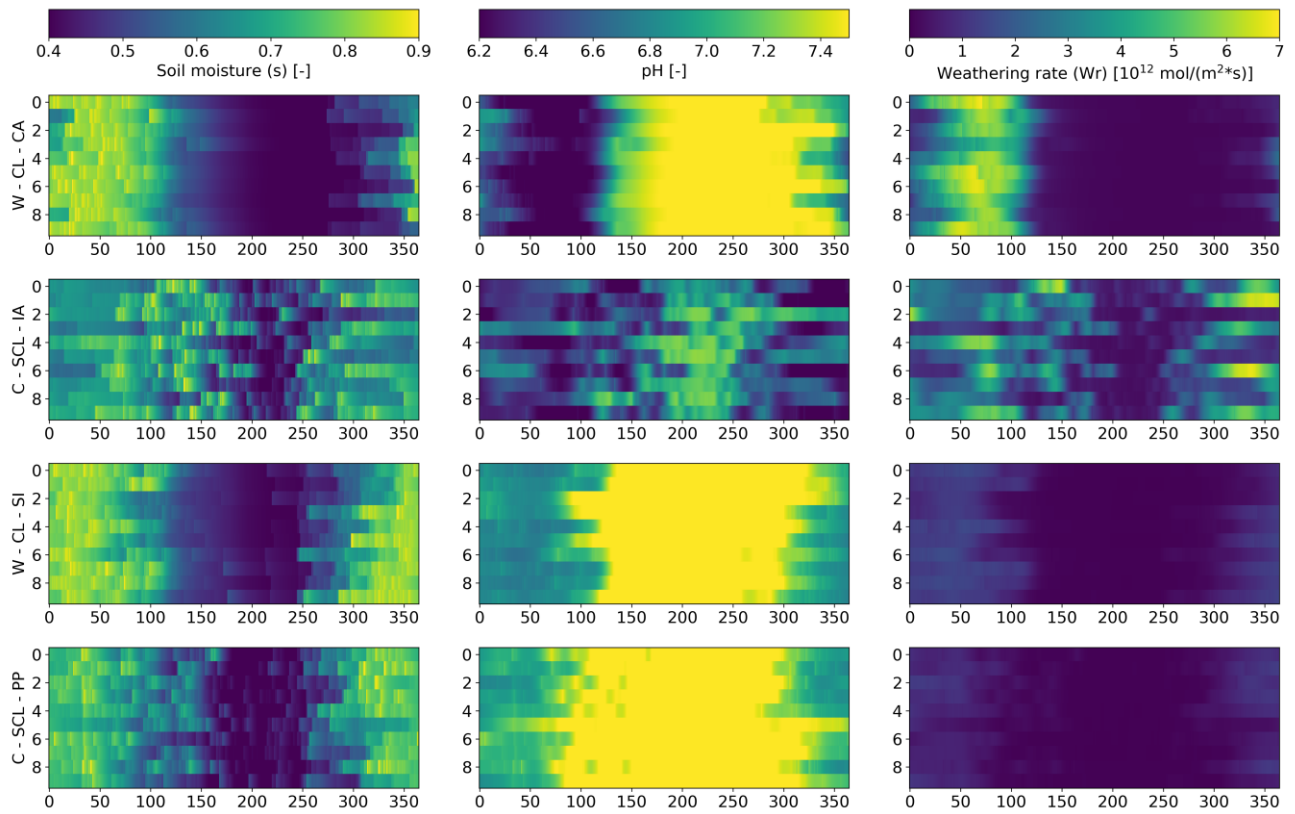
Referee: Figure 9: Please add the symbol to the parameter name in the figure caption, and the full name to the symbol along the Y-axis of the specific graph (bare soil evaporation (E), crop transpiration (T)). To allow easier comparison of these parameters between a) wheat sand b) corn, please have both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

Response: Figure and caption have been edited as suggested:



Referee: *Figure 10: 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)).*

Response: Figure has been edited as suggested by this and the following comment:



Referee: Figure S2: 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)).

Response: This figure does not exist anymore. All its contents have been reported to Figure 10 in the main manuscript, following the Reviewer's suggestions.

Referee: Figure 11: 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.

Response: 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 provided to better clarify this aspect at the beginning of Section 3.4 with this sentence:

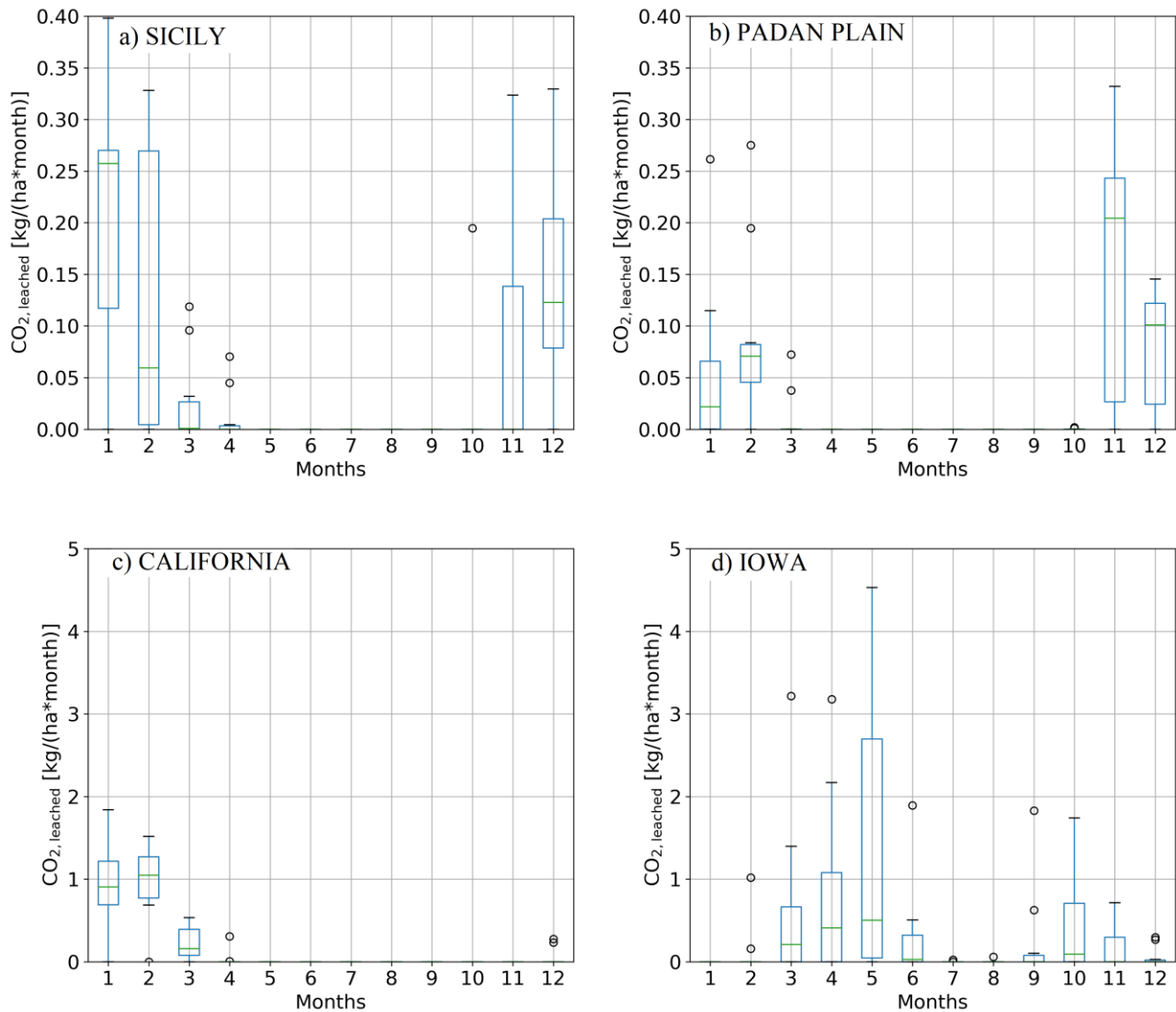
The results here analyzed are those related to the simulations referred to as "Most frequent scenario (Section 3.4)" as shown in table 1, where the considered climatic condition is analyzed along with the most frequent crop and soil type.

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 place names to the panels were added as suggested. The new figure is the following:



Tables

Referee: Table 2: 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.

Response: Done. The new version of the table and the related caption can be found below:

Table 2 - Properties of the clay loam (Sicily and California) and silty clay loam (Padan plain and Iowa) soils used in the model.

		Clay loam (SI and CA)	Silty clay loam (PP and IA)
Soil porosity	n	0.476	0.477
Soil moisture at the hygroscopic point	s_h	0.394	0.319
Soil moisture at wilting point	s_w	0.453	0.373
Soil moisture at incipient stress	s^*	0.64	0.56
Soil moisture at field capacity	s_{fc}	0.821	0.750
Saturation hydraulic conductivity	Ks	0.212	0.147
Pore size distribution index	b	8.52	7.75
Bulk density of soil	ρ_b	1450	1500

Referee: Table 3: According to the text (lines 153-155), biomass pool C_b is defined as 1% of the above defined 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^3 instead of kg/m^3 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?

Response: We corrected the unit of measurement of C_b since it is expressed as g/m^3 . 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. We prefer to keep separated tables 2 and 3 since the latter presents some parameters that depend on the site under study, rather than only on the soil type (i.e., pH differ from Padan plain and Iowa despite the most frequent soil type is the same). The table in the new form is reported in the following:

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}

References

Referee: 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)'.

Response: We corrected it adding the authors to that reference.

Supplementary material

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

Response: All elements of the supplementary material have been included in the main text.
