Reviewer#1

The authors have responded to and addressed the prior comments effectively, resulting in a marked improvement in the manuscript's overall quality. With only a few minor adjustments still needed to polish specific details.

1. The Introduction is contained within a single overly long paragraph. It is recommended to split it into 2-3 paragraphs according to the logical structure of the content.
We divided the introduction in three paragraphs: mechanisms and potential of enhanced rock weathering, interactions between inorganic sinks and organic carbon dynamics, and hint on the approach used in the study.

2. The authors indicated they had adopted the suggestion from the previous review, yet the revision has not been implemented in the main text: "Comment #55: Results 3.1 'Climate condition' should be moved to Section 2.3 'Climate simulations' in Materials and Methods."
We are ready to do that, but we want to be sure to understand what is the rationale behind? For us the section in the M&M should focus on describing our targets (choice of climate scenario). Which environmental conditions were actually achieved in the ecotron is technical validation and therefore seems to be more suited for the results section?

3. The unit "$mL^{-1} \cdot h^{-1}$" in the legend of Figure 3 is incorrectly formatted.
Corrected.

4. The font of Equations 1–3 is not Times New Roman, which is inconsistent with the font used in the main text.
Corrected.

Reviewer#2

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5. Abstract: Could you please be more specific about 'organo-mineral' interactions? Which minerals are you referring to? Are carbonates likely to be involved in such interactions? Given that the initial pH is around 7 (see L125-130), it is not surprising that the contribution of chemical weathering is very small at this pH range (neutral), since silicate dissolution kinetics are low. Please add a comment on this issue in the abstract.
Indeed, this is a crucial point that was not previously addressed in the manuscript. We appreciate you highlighting this; it further strengthens the hypothesis of increased root exudation, as these exudates can lower soil pH and subsequently enhance Enhanced Rock Weathering (ERW) processes. We added the following sentence in the abstract: “Moreover, the observed carbon sequestration surpassed the levels expected from weathering processes alone. This is notable because the near-neutral soil pH environment was not favourable to EW kinetics.”.

6. Macrocosm: It would be good if you could also provide the specific surface area of the powder (L120–125). The same applies to the soil CEC and exchangeable cations, since they are an important factor in CDR by chemical weathering.
Unfortunately, as written in the previous response to reviewers, we cannot get the surface area information (“Unfortunately we couldn’t retrieve more information on the particle size distribution and the batch used in the experiment is now gone.”, see reply to comment #74). However, we could try to make an estimate based on the manufacturer standards: Fine Grade, lower than 50 µm, with 70–80% of the material under 20 µm size. We added this information in the Material and Methods, and flagged it as manufacturer data, not actually measured data: “The basalt was of fine grade (less than 50µm particle size, with 70-80% of the particles under 20µm, according to manufacturer data)”.

Providing the exact soil CEC and exchangeable cations are also not feasible, as the experiment was terminated in 2022, and all of the soil samples have been used for TOC and PIC measurements, so we don’t have any material left. However we can make an estimation based on the DOV (Databank Ondergrond Vlaanderen) database, where this type of soil (Zbf1t) ranges between 3 – 8 cmol(+)/kg, with the main origin of the cations being the soil organic matter, as these sandy soils are very low in clays. Taking into account the 10t/Ha $CaCO_3$ treatment in the year prior and the subsequent +1 pH unit increase, we assume that i) most of this CEC was attributable to Ca^{2+} ions; ii) the total CEC has been raised by 1.5-2.5 cmol(+)/kg. We therefore estimate the CEC of this soil to be 4.5-10.5 cmol(+)/kg. We now clarify this in the material and methods section: “Based on the DOV database (https://www.dov.vlaanderen.be/), the CEC (Cation Exchange Capacity) of this type of soil (classified as Zbf1t) is expected to range between 3 and 8 cmol(+)/kg. “, and “This liming increased the pH to 7, which we estimate increased CEC by 1.5-2.5 cmol(+)/kg, resulting in a soil CEC estimated at 4.5-10.5 cmol(+)/kg and largely dominated by Ca^{2+} ions before the basalt application.”.

7. Conclusion: L510 double parenthesis.
Corrected.

Reviewer#3

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8. Re. Comment 67:

I think the strength of this study is that it offers the opportunity to compare a whole-system flux budget (as presented in Figure 1) with independent measurements of the size of C pools (soils [SIC, SOC], and plants). This is important, because the central argument of the paper is that you suggest a 1.2% boost in total soil carbon (Line 447) due to the impact of ERW. In this context, the SOC data should be a central pillar of this work, given that it constitutes the largest C pool. It is regrettable that the change in SOC and SIC cannot be calculated given the lack of baseline measurements. This means that you cannot independently verify the "1.2% boost" claimed based on the C flux data. Nevertheless, even in the absence of such a calculation, I still think that the SIC and SOC data you do have should be given greater prominence in the paper than it currently is.

You suggest that there is no discrepancy between the SOC data presented here (end-of-experiment control v basalt) and the C fluxes measured across the whole experiment in Figure 1, because a) the latter is a measurement based on the difference between time-integrated fluxes and the former is an absolute value at one time-point; b) the difference in SOC is not statistically significant between "control" and "basalt" mesocosms; c) that measured C fluxes were not large enough to result in an increase in SOC in either treatment that would be significant over measurement uncertainty.

To your point a): I agree that comparing the difference between time-integrated fluxes and a value at a single time point is comparing apples with oranges – but in the absence of a t=0 SOC measurement, it's the only way to reasonably test the impact of the treatments on SOC pool size. On reflection, I do agree with your point though: given the lack of t=0 measurements, the only thing you can reasonably do is assess whether or not the control and basalt data for SOC can reasonably be derived from the same population (i.e. if the variability in SOC in the original "marginal" soil is large enough that the difference between the control and basalt treatments at the end of the experiment is not significant – which is what you state in point (b)). If this is the case, then you need to demonstrate this with an appropriate statistical test (i.e. present test statistics, not just state that the difference is not significant); for instance a two-sample t test or Welch's t test, depending on your assumptions on the distribution of SOC in the "marginal" soil you're using. The null hypothesis here should be that the soils are drawn from the same population. Please also present all SOC data in the supplement, and produce a box plot (similar to Figure S2) for the SOC and SIC data. I don't think this can be published without those data being included (rather than just the mean and standard error).

We now include a graph annotated with the t.test statistics for both POC and PIC data in the supplementary materials (Figure S9).

Note that if you're claiming a "1.2% boost in total soil carbon", then the average difference between the initial soils at the start of the experiment must be even greater than that measured at the end of the experiment; i.e. control at t=0 >> basalt at t=0, such that the 1.2% increase in soil carbon in the basalt treatment relative to the control is accounted for. I suspect that if you do a back-of-the-envelope calculation for this using a simple C mass balance, and then run the same statistical test, then the null hypothesis of the soils being drawn from the same population will almost certainly be violated; though it is impossible to tell without the data. This is my major concern here – that it is fundamentally a mass-balance problem, rather than a variability or uncertainty problem, which goes to the heart of the main conclusion of the paper.

I do see the merit in argument c), though I would not call this "measurement uncertainty" – rather, it's the variability in OC between soils I think you're talking about.

Indeed, this is what we wanted to point out in the manuscript: that this flux-calculated 1.2% increase in SOC is smaller than the noise in actual SOC measurements, and therefore we cannot conclude on SOC increase. We agree that this noise comes from SOC variability, not measurement uncertainty. We tried to make this clearer in the new version of the manuscript. We especially paid attention to clarify that our main conclusions are based on fluxes, not on pool size change. See also our reply on the next point that partly integrates this.

Regardless, two points, which I think should be explicitly stated in the discussion:

1) The means show a difference in OC between the soils of the different treatments at the end of the experiment that is larger than the total difference in C pool size between treatments, as calculated by the difference in C fluxes – even if this difference might not be significant above background variability.

2) In fact, and in addition to this, the variability in OC between replicates of the same treatment is larger than the total C pool size difference.

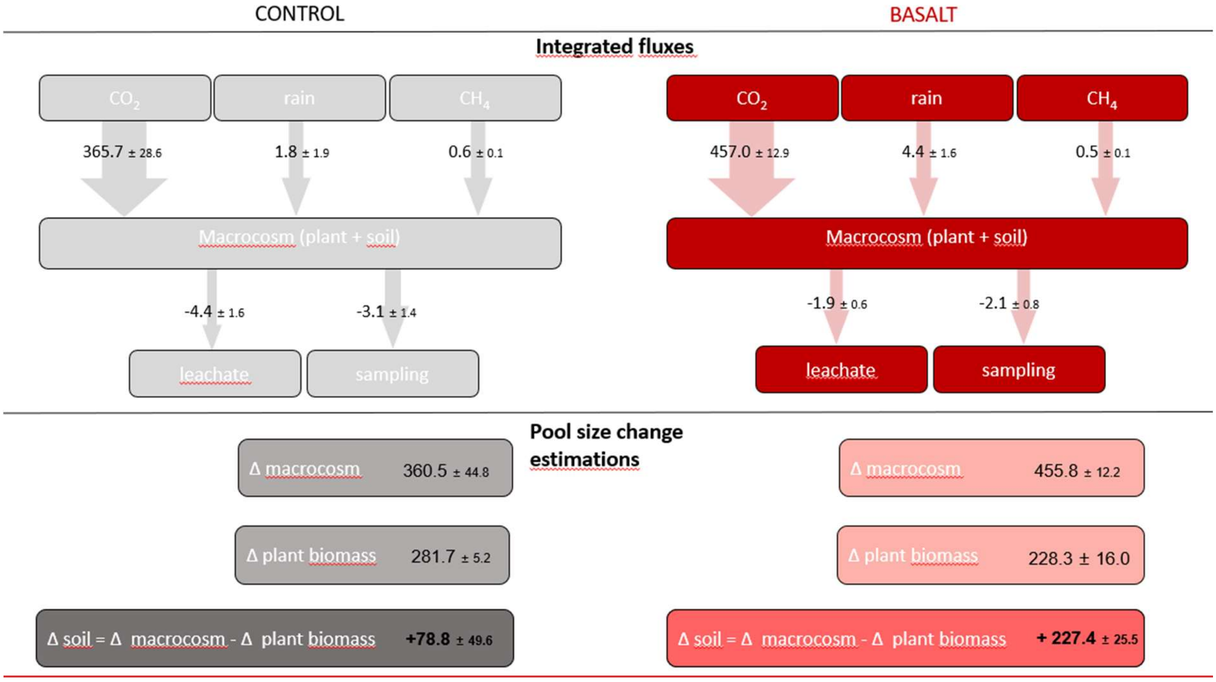
The additional text you have supplied is good, and it goes some way to addressing these points; but it does not mention the mass balance problem or the variability at all (in the discussion). I think a major part of the discussion needs to be devoted to the implications of these points, and this actually is a really positive impact of your work: because the CDR observed via the difference in fluxes is greater than that that can purely be attributed to carbonic acid weathering using the Steinour / Renforth formulations, this MUST be due to organic C storage in soils; but the variability of SOC is such that this can only be measured by flux difference. This speaks to the point that measuring SOC is fraught with problems due to large background heterogeneity. There is a lot of relevant literature on this that you should cite – for example Bradford et al., 2025 in Nature Climate Change.

Indeed, we realize that this point was almost not developed in the discussion. We therefore added the following paragraph in the discussion: “At the conclusion of the experiment, the observed differences in soil organic carbon pools between treatments exceeded the increase in size predicted by our integrated flux budgets (NEE). Because i) the observed NEE reaches the stoichiometric limit of basalt weathering alone—as defined by the Steinour and Renforth formulations— and ii) we did not detect significant increases in the inorganic C pools, we assume that the end destination of this NEE flux must be attributed to organic carbon storage. Yet, the spatial variability of the SOC pool (even within a single lysimeter) is such that this sequestration signal is effectively obscured by biological variation. The difficulty in reconciling these values highlights the 'signal-to-noise' problem inherent in soil carbon monitoring. Because the magnitude of sequestration over several years is typically small compared to the large and variable background SOC pool, detecting a 1.2% shift is analytically challenging (Bradford et al., 2015). In fact, under the sampling constraints of the current study (one year and limited soil sampling possible), these small-scale accruals would remain statistically indistinguishable from natural soil variability (Smith, 2004). Consequently, integrated flux-based measurements, such as Net Ecosystem Exchange (NEE), emerge as a more sensitive and reliable metric for detecting short-term carbon drawdown (Conant et al., 2011). We therefore proceed with the following discussion by evaluating the observed carbon sequestration through the lens of NEE.”

9. I also think Figure 1 is misleading because the “soil pool” is presented as if it is an absolute value in g C/m^2, whereas it is a difference (Δ g C /m^2), the way you have calculated it – and given the size of the OC pool, this difference is a fraction of the standard error of the absolute value, in both

treatment and control. It is important to change Figure 1 to reflect this. I would suggest to change the description of plant and soil values to Δs, and to include a box to represent the total calculated sizes of the soil C pool at the end of the experiment in the figure to explicitly show the relative size of the difference.

We agree with the reviewer that the distinction between pool estimations and integrated fluxes could be better conveyed in Figure 1. We reworked the figure accordingly:



In summary: given the DIC and SIC data, your the results of your work imply that the CDR from EW is mostly due to SOC stock increase. That stock increase cannot be determined from soil samples due to the lack of t=0 measurements; and due to variability. Currently, the data on that variability isn't presented in full or rigorously tested enough. The discussion does not adequately handle the implications of the results on C pools and SOC calculation difficulties for the field of EW. Therefore, in my opinion the manuscript still requires major revisions.

We hope that the new figures & tests and the new paragraph in the discussion address this comment properly.

Other comments:

10. In abstract, mention that you are using simulated future "warmed" climate conditions in this experiment. Also mention this at the end of the introduction.

We added this information in the abstract "Here, we evaluated the effects of EW on a crop ecosystem under future climate change conditions within a macro-scale ecotron—an enclosed facility enabling complete quantification of carbon fluxes among the atmosphere, vegetation, soil, and leachates." Note that in the previous versions of the manuscript we intentionally omitted the climate scenario from the abstract to maintain a tight focus on the main biological question. Introducing secondary project-wide variables at this stage would likely invite unnecessary questions regarding climate scenarios that the present study was not designed to resolve.

11. Line 38: change wording to "solely be attributed to basalt weathering." (delete "rates").

Corrected.

12. Section 4.3 (and in response to my comment #69): As I previously commented in my earlier review, MAOM and microbial entombment changes are still OC and should therefore be measurable as part of SOC or DOC budgeting. Separately, there is still no explicit discussion of the discrepancy between supposedly greater soil C storage as OC in the basalt treatment, and the higher microbial activity in this treatment, which should result in greater respiration of OC.

We agree with the reviewer that Mineral-Associated Organic Matter (MAOM) and microbial products are indeed components of the total Soil Organic Carbon (SOC) pool and should, in theory, be captured in a mass balance. Our point, however, is not that these fractions are 'invisible' to chemical analysis, but rather that the magnitude of change in these pools over the study period remains several orders of magnitude smaller than the pre-existing background variability of the total soil C stock. Even if a significant fraction of the atmospheric flux was 'entombed' as MAOM, the total shift in the SOC pool (calculated at 1.2%) remains well below the statistical detection limit in a high-heterogeneity field setting (as discussed above).

Regarding the second part of the comment: indeed we did not explicitly address this discrepancy between higher microbial activity but higher soil C storage. We would speculate the following that is also linking to another paragraph of the discussion: the basalt treatment, instead of immediately reacting with soil CO_2, leads to a considerably higher root exudation from plants, stimulating microbial activity, which, if coupled with higher CUE, could cause more C ending up in MAOM. We tried to make it clearer in the discussion: “While we observed no significant change in total plant biomass, the increased NEE in the basalt treatment suggests a strategic shift in carbon allocation toward below-ground fluxes. Although root exudates alone are unlikely to fully account for the total magnitude of carbon storage observed, we speculate that they could have served as a primary substrate for the enhanced microbial activity (FDA) measured during the growing season. This stimulation may have been responsible for the initially higher soil CO2 emissions; however, if the Microbial Carbon Use Efficiency (CUE) is higher under basalt treatment, this metabolic throughput leads to a net accrual of microbial necromass that exceeds respiratory losses. Under this 'microbial carbon pump' hypothesis, the improvements in soil biochemical conditions and the abundance of fresh mineral surfaces provided by the basalt facilitate the rapid transfer of metabolized carbon into stable associations with soil minerals. Such 'microbial entombment' produces highly recalcitrant carbon forms (Xiao et al., 2023) and is consistent with our observation of increased enzymatic potential alongside a net carbon sink. If microbial turnover is particularly rapid, this pathway could explain the significant organic carbon sequestration measured in our experiment even in the absence of standing biomass gains or inorganic carbonate accumulation. This suggests that microbial entombment may represent a carbon storage pathway for enhanced rock weathering that is comparable in importance (and perhaps more immediate in detection) to carbonate formation”

13. Comment #76: Note that the Renforth formulation is originally from a paper by Steinour (this is cited in Renforth’s paper). Please also cite the original source.

Interestingly we couldn’t trace it back to the original source. The Renforth paper doesn’t cite any paper from Steinour, it refers to a paper from Gunning et al (2010) instead, that doesn’t seem the most relevant to support the use of this formula. After some search in different databases for papers from Steinour, we could only find an obscure reference to a paper from 1959 (H.H. Steinour, Some effects of carbon dioxide on mortars and concrete-discussion. J. Am. Concrete I, 30 (1959), p. 905), which in fact is a 3 page report from two other authors without any formula in there. We therefore decided to keep this citation, but we definitely can use the Gunning one if the reviewer finds it more relevant.

14. Comment #79: That’s fine, I don’t think you need to include this.

Checked.

15. Comment #98: I like the first of these figures.
This figure has been added as Figure S8.

Reviewer#4

This revised MS has a well improved introduction and discussion section, and contains an elaborate discussion about the possibilities and mechanisms on why CO2 uptake was larger in basalt amended soils. We have some minor remarks which, when taken into account, will warrant publication of this MS.

16. Abstract: Please also update the abstract such as not to frame ERW as mitigation but as a carbon dioxide removal technology. (see also comment 19 in the first round of review);
Corrected.

17. Abstract: Please also state in the abstract that the marginalized soil was limed and fertilized prior to basalt amendment and was at near neutral pH (to avoid the misinterpretation that this is a study about basalt application on acid soils)
Corrected. See also reply to comment #5 above.

18. Removing topsoil: 'marginalization'. Please specify how many cm was removed?
It was the top 20cm. We now mention it in the material & methods section.

19. Can the authors specify which feldspars/plagioclases are present?
Unfortunately we did not have this level of detail in the basalt analysis. We also could not trace back this information at the manufacturer.

20. Line 125: space is missing between 10 and t/ha
Corrected.

21. Table 1: Please explain color coding in the caption.
Corrected.

22. In Table 2 you use SIC and SOC, in other places in the manuscript PIC & SIC, and there is also mixed use of both abbreviations (line 214). Please make the use of these terms consistent throughout the manuscript
Corrected. We refer now to PIC and POC consistently throughout the manuscript.

23. Section 2.7 Rainwater flux. Please explicitely state that you ignore DIC in rainwater, perhaps with a reference as support.
Corrected. As we could not identify a literature reference for a study with an identical experimental design, we have justified the omission of this flux by demonstrating that its magnitude is negligible compared to the primary carbon flows analyzed.

24. L180-200 Adapt formulae such that it is clear that you compute a difference in soil carbon stock (Δ(SOC+SIC)) as a sum/integration of gas-, leachate-, and sampling fluxes (Σ(F_CO2 + F_CH4 – F_leachate – F_sampling) and a difference of the carbon stock in plant (Δ(plant_C)). Particularly equation 3 L199 reads as an equation solely in carbon stocks, while some are integrated fluxes. Also in the text there is sometimes confusion between (difference in) stock and fluxes.
We modified sections 2.4 and 2.10 accordingly, and tried to clarify as much as possible what were fluxes, pools, and integrated fluxes. See also replies to reviewer#3, that address this issue in the figures.

25. L185 Please motivate why DIC in leachates should be negligible, as it was only measured in pore waters throughout the depth profile. You could refer to the smaller DIC relative to DOC at pore water at 20 cm depth (Table 2) and extrapolate this to the leachate. As leachate DOC had a negligible effect in the total C mass balance and we expect smaller DIC, changes in leachate DIC among treatments should thus be negligible in the total mass balance.
We would appreciate further clarification on this point, as there may be a misunderstanding. The paragraph at line 185 is intended to justify our selection of an upper threshold for DOC concentrations. Regarding Dissolved Inorganic Carbon (DIC), we did measure this parameter; the data is presented in Table 1 and has been added to Figure S8 for better visibility (see comment #15). Please let us know if there is a specific aspect of the DIC analysis we should further address.

26. L214 Add a "." between SOC and PIC.
Corrected.

27. L216 "carbonates ions" -> "carbonate ions"
Corrected.

28. L250 Soil Ca and Mg concentrations as proxy for the EW reaction -> change to "Ca and Mg concentrations as proxy for weathering". See also comment below about the conclusion section.
Corrected.

29. L251 "To account for the carbonates which might have initially formed in the soil but later degassed as CO_2 during sample storage prior to analysis, we measured divalent cations (Ca^{2+}, Mg^{2+}), resulting from silicate mineral dissolution." Please delete this sentence, degassing of CO2 due to a change in CO2 pressure does not change Ca and Mg concentrations and vice versa.
We think it's important that the reader understands the rationale behind this cation measurement. We therefore reformulated this sentence as follows: "Cation weathering products (Ca^{2+}, Mg^{2+}) were measured to provide a mass-balance estimate of carbon sequestration. This approach compensates for potential CO_2 losses via degassing during storage by focusing on the stable mineral-derived cations that drive the sequestration reaction."

30. L416 (equivalent to 10 tonnes per hectare) please remove this part between brackets or make it cleare that this is the application rate of basalt, and not the carbon removal rate.
Corrected.

31. L511 "We had no evidence that the EW reaction took place" – please be more specific. In general this conclusion section can be strenghtened by pointing out that you don't measure inorgnic carbon sequestration but you find significant organic C sequestration. This means that the indirect effects of weathering (such as the ones that you have already discussed) are much more important than the direct/geochemical effects.
We re-wrote the conclusion accordingly: "Our results demonstrate that basalt application significantly enhances the capacity of crop ecosystems to act as carbon sinks, tripling soil carbon sequestration over a single growing season. Basalt treatment shifted the macrocosms from marginal (0.5 tC/ha) to substantial (1.5 tC/ha) carbon sinks, successfully overcoming the initial carbon losses associated with soil disturbance during application. While we found no direct evidence of inorganic carbon sequestration via carbonate formation, potentially due to an incomplete dissolved inorganic carbon (DIC) budget, the total carbon accrued exceeded what could be attributed to weathering

alone. These findings suggest that the indirect effects of weathering, such as the stimulation of mineral-associated and particulate organic matter, could be the primary drivers of carbon storage in this system.”