To Editor:

We appreciate the editor for your time to thoroughly assess and for your comments to improve our manuscript. We addressed all points raised by the editor. Below, we respond to each of the comments.

A few small pending comments:

1. There is no need to add a justification related to limited beam time with respect to STXM analysis of the fine basalt.

➔ We deleted the sentence related to limited beam time with respect to STXM analysis of the fine basalt in the revised manuscript.

2. Obviously, do update the reference to your recently accepted paper in SBB

➔ We now cite the reference (Yang et al., 2026; copied below) in the manuscript.

Yang, P. T., Kurokawa, K., Nakao, A., Matsumura, E., and Wagai, R.: The potential of enhanced rock weathering for CO_2 removal and soil organic carbon storage via organo-mineral aggregation: the trade-off induced by basaltic rock particle size. Biogeochemistry, 169, 9, https://doi.org/10.1007/s10533-025-01296-7, 2026.

3. Should the authors agree, than it seems indeed a good idea to still add a schematic diagram clarifying how the forwarded organo-metallic glue hypothesis applies to the findings. On top, such a graphical presentation should aid in making that the message of this interesting contribution gets picked up more readily by a wider part of the soil science community.

➔ We have added Figure 10 (copied below) as a conceptual diagram in the main text.

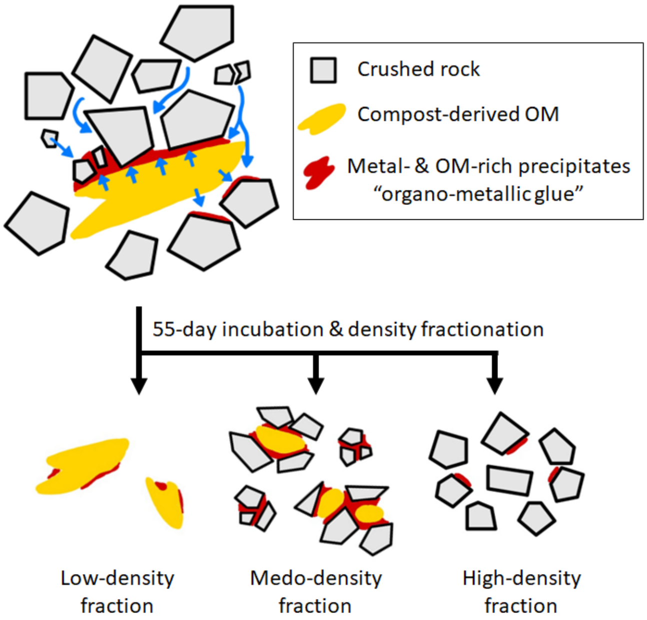

Figure 10. Conceptual synthesis of the experimental results in the context of the organo-metallic glue hypothesis. Metals (primarily Fe, Al) released from crushed rock under wet conditions preferentially associated with dissolved and particulate OM, forming organo-metallic precipitates on mineral and organic particle surfaces upon drying. Following eight wet-dry cycles with physical mixing, these precipitates became enriched in the meso-density fraction, supporting their role as "glue" to bind mineral and organic particles to form physically stable organo-mineral aggregates.

To Editor:

Now we submit the revised manuscript, figures, and tables. We thank the two reviewers for their time to thoroughly assess our manuscript and for their valuable comments. Some of the comments were substantial, which gave us an opportunity to further analyze our results and improve our discussion. We believe we were able to address all the points raised by the two reviewers.

Below we respond to each of the comments by Reviewer 1 and 2. But, first, we'd like to point out the main changes in the revised manuscript.

1) Figures and Tables: We now provide more supplementary tables following the request by Reviewer 2, which resulted in 14 supplementary tables. To help readers to navigate, we newly added Table S7 which shows the list to show which figures/tables correspond to major quantitative data (mass, carbon, metal before and after the incubation).

   Reference: Yang et al. (under review) was just accepted last week in Biogeochemistry. Title/author info is copied below. Both our current study and Yang et al. study examine the relation between rock weathering and organo-mineral associations using the same basaltic rock (particle size range and incubation conditions are not exactly the same). We cite this reference to provide more in-depth mineralogical data of the rock material, and to discuss the complex relationship between basalt weathering and OM-basalt aggregation.

   "The potential of enhanced rock weathering for CO2 removal and soil organic carbon storage via organo-mineral aggregation: the trade-off induced by basaltic rock particle size"

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2) We extensively edited and revised all sections of the manuscript by removing many redundant points, clarifying our conceptual framework. Discussion sections (esp. 4.1, 4.2, 4.3) were extensively edited. In section 4.1., we now more carefully discussed why organo-metal coprecipitation is more likely to be the dominant form of organo-metal materials accumulated in

the meso-density fraction rather than sorptive association of OM with metal oxides, based on Fe and Al distribution across the density fractions before and after the incubation. This point is further discussed in section 4.2 from another aspect (chemical composition).

RC1: 'Comment on egusphere-2025-2840', Anonymous Referee #1, 15 Sep 2025

General comments

This manuscript reports results of a well-designed incubation experiment aimed at quantifying the formation of organo-mineral aggregates from leaf compost and different rock powders. The manuscript is generally well-written and the amount of analyses performed is impressive, ranging from classical soil fractionation and extractions, through microbial community analyses, to nanoscale spectroscopy.

The introduction is effective, and the methods are well-described and largely appropriate (see comments below). The results section is very rich, but I did not find it overly clear. In particular, the figures' legend need to be more explicit (see comments below). The discussion is generally well-written and interesting. I thought that the largest shortcoming was the lack of an explicit conceptual framework for the organo-mineral associations. Sorptive association, complexation, cation bridging, co-precipitation are mentioned, but the relation of these mechanisms among one another and with the « organo-metallic glue » hypothesis is not made clear. Perhaps a diagram would help? Co-precipitation usually has a different definition than the one implied here - a mixture of adsorption and complexation, if I understood correctly (see for instance https://doi.org/10.1038/s41467-025-61273-4). Furthermore, it is not clear how the results can discriminate between these mechanisms. In fact, I am not sure that they can.

I would recommend that the authors streamline the results section to bring the most important ones to the forefront, and re-focus the discussion on substantiated trends rather than speculative ones. There are a lot of exciting findings here – rapid mineral weathering, formation of reactive secondary phases, association with microbially processed organics, etc.!

- ➔ Thank you so much for your time going through our manuscript very carefully and pointing out weakness as well as strength. Now the manuscript is extensively edited. Result section is shortened a bit by improving the connections and clarity. We now more clearly showed our conceptual framework and removed more speculative parts of the discussion.
- ➔ "Coprecipitation" mechanism: we originally regarded coprecipitate as a material having intermediate OC:metal molar ratio between sorptive association and complexation. While it is generally true, we agree that the use of this term in our original discussion was confusing. In

the revised manuscript, we focused more on actual process of coprecipitation. Strictly speaking, we cannot distinguish complexation from coprecipitation for the materials we examined. Thus, we decided to interpret as follows: Fe and Al released from rock appeared to complex or coprecipitate with organic compounds in the meso-density fraction (Please see Discussion 4.1-4.3).

- ➔ The relationship between the mineral-organic association mechanisms and “organo-metallic glue (OMG)” hypothesis: we agree that this was also not clearly discussed in the original version. Our general conceptual model of organo-metallic glue hypothesis is the following. First, organo-metallic associations (via sorption, complexation, and/or coprecipitation) occur at molecular scale, say at nano to tens of nano meter scale. These nanoscale associations eventually grow (cluster, coagulate) to form slightly bigger materials enriched in reactive metals and organics (perhaps the size of submicron to tens of microns). These organo-metallic materials act as glue to bind less reactive solids into physically stable microaggregate (tens to hundreds of microns). This concept is more clearly discussed in the revised main text (Abstract and Discussion 4.1, 4.2, & 4.3). If you think it is helpful, we are happy to prepare a conceptual framework figure (a schematic diagram).
- ➔ Discussion section 4.1 (and 4.2, 4.3) was significantly changed by putting an emphasis on the distribution of extractable Fe and Al across density fractions before and after the incubation (new Fig. 9). During the revision process, we realized that the results of extractable metal increase in MF (and LF) and slight decline in HF have an important mechanistic implication. If released Fe and Al during 55-day basalt weathering largely precipitated to form “pure” secondary metal oxides, it should be coated on basalt surfaces. Basalt particles with such coating must have much higher chance to fall into high-density fraction (HF) even when DOM was fully adsorbed on the oxide coating later. The fact that most increases in Fe and Al was found in MF thus strongly suggests that released metals were more likely complexed or coprecipitated with low-density material (i.e. organic compounds) to form MF material. We think this is more convincing argument than the original OC:metal ratio argument. We shuffled and revised all discussion sections (esp. 4.1) to reflect this new point.

Specific comments

L 102+: Your wet sieving procedure probably induced some mineralogical fractionation compared to the initial rock. For instance, for granite, I assume that the micas were preferentially lost in the discarded < 38 um particle size class and quartz was preferentially retained. I don’t think that it is necessarily a problem, but it should probably be discussed somewhere.

- ➔ We agree. We now add this possible artefact in Line 108-110 in the revised text as follows: “Preferential loss of some fine-grain minerals (e.g., mica) and enrichment of coarser ones (e.g., quartz) during the sorting are possible. Mineralogical data, nevertheless, showed the sizesorted materials fall within a common compositional range (Fig. S1).“ New Fig. S1 is shown below.

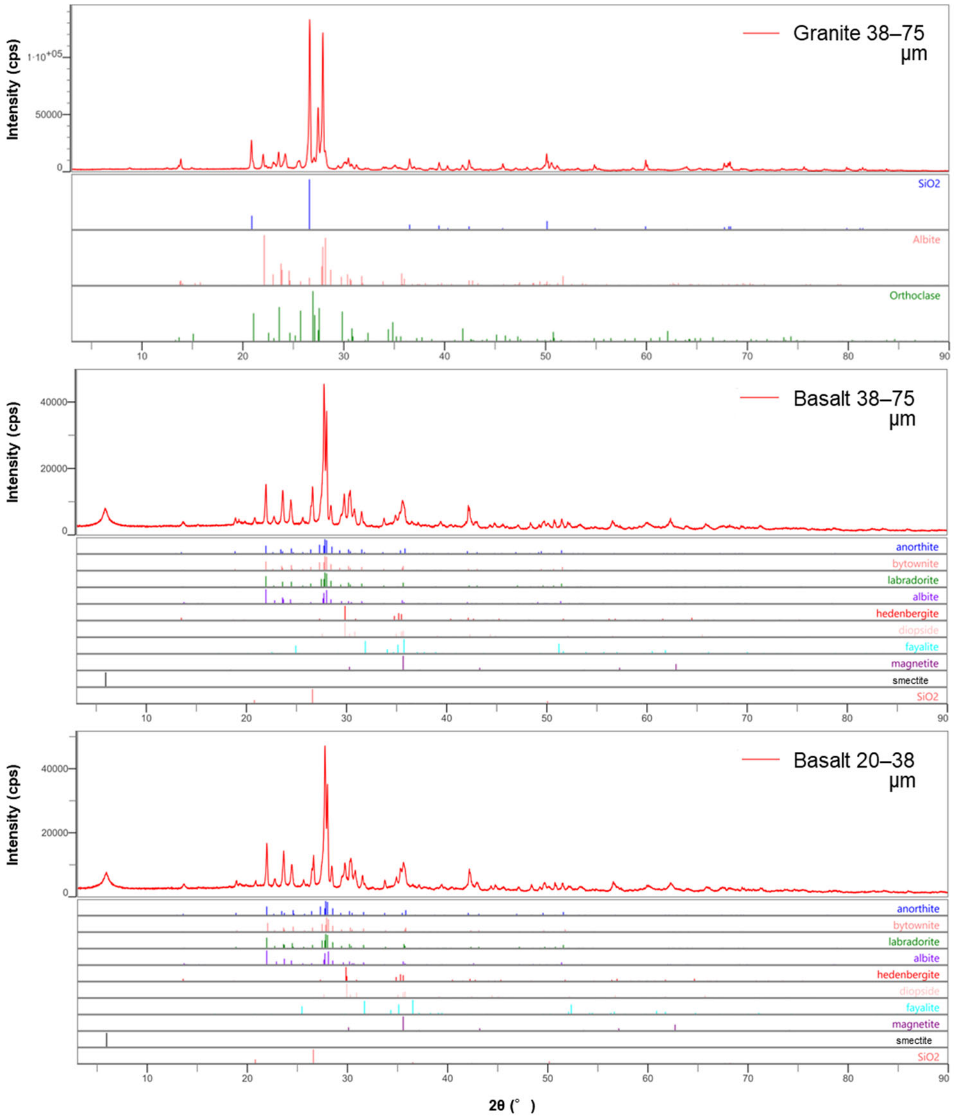

Figure S1. Mineralogical composition of the crushed rocks (granite, coarse basalt, and fine basalt) used in this experiment.

L 111: A C content of 0.3 mg C / g rock is not a lot, but still very significant compared to your reported C content in the middle density fraction (0.6 – 1.5 mg C / g rock). For basalt, this is likely to consist of secondary carbonates. Did you consider the possible effect on your ^{13}C results? For granite, I am not sure where this C could be coming from.

➔ Thank you for pointing out. TC content in the original rock was not clearly presented. As shown in Table S8-1 now, we detected 0.2-0.3 mgC/g for the fine and coarse basalts. This can be due to carbonate or organic C contamination in the original rock (e.g., via dust) and during density fractionation. In our companion study (Yang et al., accepted), we did XANES analysis and no calcite was detected. So, at least, not so much calcite was present in the initial basaltic rocks. Now we mention this point in Line 120-122.

- ➔ After the incubation, carbonate formation is possible, but the carbonate mass formed appeared to be minor. If carbonate formation accounted for large enough mass, we would not see the consistent density-dependent changes in C:N ratio, C-13 and N-15. We now discuss this point in L590-592.

L 187: I don't think that it is appropriate to analyse Ca in these extracts, as Ca-pyrophosphate, Ca-oxalate and Ca-citrate salts are only very sparingly soluble.

- ➔ We agree. We removed the discussion on the extractable Ca as well as Fig. S3 in the previous version.

L 213: « The subsets of MF from selected treatments (granite and coarse basalt) ... »: I might have missed it, but why was the fine basalt omitted?
This sentence is also missing a conjugated verb.

- ➔ Our original intention was to analyze the fine basalt mixture as well as the other two. But we ran out of STXM beam time. Getting a beam time is always challenging. Given the limited beam time we had, we had to focus on the granite and coarse basalt samples (one region of interest per sample, only). We thus gave up on detecting the difference between the rock treatments. Instead, we focused on identifying some features of the organo-mineral assemblage formed in the meso-density fraction. We briefly mentioned the lack of beamtime issue in Result section (line 418) now.
- ➔ We added a conjugated verb to this sentence.

L 244: « The effect of mineral type on the measured variables was tested by one-way ANOVA » followed by a Tukey test, if I understand the legend of your figures correctly – to clarify.

- ➔ We have now added the description of a Tukey test.

This is a minor point, but I would argue that it is not coherent to use a family-wise correction (Tukey) for the effect of rock, whereas you used simple t-tests (4 times, once for each rock) for the effect of incubation. The inflation in type I error is close to the same in both cases. I would recommend t-tests everywhere. If you are concerned about type I error, you can simply decrease your alpha level (e.g., to 0.01).

- ➔ Thank you for the advice. We agree that it would be more coherent. However, for the results shown in Fig. 3 and Fig. 5, we cannot think of a good graphical representation of t-test results if we compare the four rock treatments. Instead, we improved the explanations of these figures to minimize readers' confusion.

L 246-249: For me this approach is ok, but the explanation is not entirely convincing, since the portion used for density fractionation was sub-sampled again prior to C and N analysis, right? Or did you not like the results you got from the bulk samples?
Your recovery rates do look very nice.

- ➔ Thank you for pointing out the important details. We actually did not really use or discuss the total C at Day 55 in the main text except for reporting the recovery in Table 1. However, we used the total extractable metal at Day 55 in Discussion (section 4.1). So, we modified the sentences accordingly (Line 255-259 in revised text).
- ➔ We now also discussed briefly that we were not able to detect the expected increase in Feox from Day 0 to Day 55 bulk samples due presumably to well-known Fe colloid problem in extracts (Line 477-483). Nevertheless, Alox showed the expected increase from the bulk sample comparison. Furthermore, we clearly detected the significant increase in mass, extractable metals, and C in MF after the incubation for the basalt treatments. So, we believe our discussion has solid basis.
- ➔ Why we did not save enough mass for bulk sample chemical analyses? For the bulk chemical analysis after the incubation, we should have used much greater portions of the bulk material. But we had to split the bulk sample to conduct both size and density fractionation, in addition to the bulk chemical analysis. Unfortunately, the size fractionation did not work out well and thus no report on this.

L 346-349: See previous comment – your extractions are inappropriate for Ca; I don't think that the results are interpretable. The salts of Ca with oxalate, citrate and pyrophosphate have a very low solubility. In addition, Ca pyrophosphate solubility decreases with increasing free Ca ions. I would recommend removing this part.

- ➔ We agree. We removed the discussion on the extractable Ca as well as Fig. S3 in the previous version.

L 396, Table 5: It is noteworthy that the amount of base cations leached from the fine basalt was about the same as from the coarse basalt, except for Na, which was 3 x greater. This suggests preferential weathering of Na-phases?

- ➔ Yes, we think it is likely due to Na plagioclase dissolution. We added this point now (Line 393-395). Yang et al. (accepted), mentioned in page 1 on this letter, used the same basalt but slightly different size classes, and showed clear Na-plagioclase dissolution during 6-month incubation.

L 501: In the absence of soil respiration data, I find hypotheses about relative heterotrophic activity highly speculative.

➔ We agree. We now removed this speculative argument. Due to the lack of respiration data, we also changed “heterotrophic activity” to “microbial activity” throughout the texts.

L 520+: This section seems quite nebulous to me. Co-precipitation first « cannot be excluded » (even if sorptive associations alone could account for the observations). I don’t understand why the associations are « best characterized as … coprecipitates » in the next sentence. Based on which evidence?

➔ We agree that the argument had weak basis. Now we have fully revised this part of discussion. Please see our first response above for the detail.

L 525+: I agree that the increase in Fe phases suggest a predominance of sorptive associations. I don’t quite understand how this relates to the « organo-metallic glue hypothesis » (4.1). Overall, I think that the discussion is missing an explicit conceptual framework.

➔ We agree that this part of our discussion lacked solid basis. Now we have fully revised this part of discussion. Please see our first response above for the conceptual framework. If you think it is helpful, we are happy to prepare a conceptual framework figure (schematic diagram).

L 564+: This discussion of the relevance of the study for enhanced rock weathering is potentially interesting but it is not sufficiently grounded in the authors’ results, in my opinion. The nearly one-unit pH increase in the fine basalt treatment, together with Na leaching, points to significant mineral dissolution; this contradicts the idea that the higher abundance of aggregates « likely slows down the rate of basalt weathering ».

➔ Thank you for catching the important point. We agree that our results don’t support this statement. The slow-down of basalt weathering by basalt-OM aggregation was shown in Yang et al. (accepted) study, but not in the current study. We removed this statement. We thus modified the sentences (Line 594-596 in revised text).

Similarly, I don’t think that this study supports the « increase in soil OM upon the mixing of basic rock powders ». It does not contradict it either, but you did not see changes in bulk C. It is possible that you had more heterotrophic microbial activity, thus enhancing both C mineralisation and formation of organo-mineral associations between microbial compounds and reactive phases.

➔ We agree. Our results showed the increase in organo-mineral assemblage formation and C in MF (i.e., mineral-associated OM) but not that in total C. We thus removed the sentence.

Technical corrections

L 48-49: SRO aluminosilicates are not oxides, strictly speaking. Please rephrase.

➔ We changed from “oxides” to “mineral phases”.

L 87: … linkages BETWEEN

➔ We revised to « … linkages between».

L 170: centrifuged

➔ We revised to « centrifuged ».

L 215: Other subsets (plural) or Another subset (singular)

➔ We revised to « Another subset ».

L 243: Replace ‘mineral type’ with ‘rock type’ (or equivalent). You did not look at individual minerals. Also, L 260, 289

➔ Strictly speaking, the sand we used as one of the four treatment is not a rock. But, broadly speaking, we can consider it as a rock. We thus replace “mineral type” and “mineral treatment” to “rock type” and “rock treatment” throughout the manuscript.

L 285: Clarify the Fig. 2 legend. « The same letters … are not significantly different » does not mean much. What did you compare?

➔ Thank you for pointing out. We revised it for clarity.

L 305: Same comment for Fig. 3. Yes, the same letters are not different, but what is the comparison? The differences represented by letters and stars are not clear.

➔ Thank you for pointing out. We revised it for clarity.

Also L 308, for Fig. 4. It looks like the letters are for different things in part a and b (part a, for rock type, part b, for fractions?)

➔ Thank you for pointing out. We revised it for clarity.

L 366: « C was MORE enriched for Fe than Al… »

➔ We revised to “C was more enriched for Fe than Al… “.

L 566: Monovalent base cations (Na and K) are released too.

➔ Yes. We simplified the corresponding sentences as follows.
“The application of crushed basalt and other mafic rocks to soils has drawn increasing attention as a strategy for CO2 removal through enhanced weathering (Beerling et al., 2020)..”

Citation: https://doi.org/10.5194/egusphere-2025-2840-RC1

RC2: 'Comment on egusphere-2025-2840', Anonymous Referee #2, 24 Sep 2025

This article addresses the formation of organo–mineral associations during the early stages of pedogenesis. The study is based on a 55-day incubation experiment of a mixture of leaf compost with crushed rocks. After incubation, a density fractionation step separates the mixtures into three fractions. These fractions were then subjected to various protocols, including chemical extractions (PP, Ox, DC). A broad range of analytical methods was applied to the samples (at day 55 and day 0 for comparison), covering chemical compositions, isotopic (C and N) compositions, bacterial analyses, as well as SEM and STXM imaging. Overall, the results converge towards supporting the "organo-metallic glue hypothesis," with variations depending on the lithology (granite / basalt / sand) and grain size of the tested rocks.

The experimental and analytical work presented in this article is truly substantial. I believe it is important to emphasize this at the beginning of the review: the dataset is impressive, and these results deserve publication. That said, I also believe the manuscript could be significantly improved in several respects.

➔ Thank you so much for your time going through our manuscript very carefully and pointing out weakness as well as strength. We now conducted XRD analysis and also provide further information on mineralogical aspects as much as we could. We responded to each of your comment below.

Major comments

1. **Mineralogical approach**

   In my opinion, the manuscript lacks a sufficiently solid mineralogical perspective. In particular, there is a major conceptual confusion: the authors state that the weatherability of a mineral is directly linked to its chemical composition. This is not entirely correct. Weatherability also depends strongly on (1) the crystallinity of the mineral, and (2) its crystallization temperature (for igneous rocks, as is the case here; see for instance Bowen's reaction series in geology textbooks). Point (1) is especially critical given the comparison between granite (fully crystallized) and basalt (largely composed of an amorphous glassy phase, hence much more weatherable). I recommend the authors revisit these theoretical aspects of mineral weatherability before revising the sections that address this topic. This is particularly important because the current mineralogical arguments appear somewhat unconvincing.

➔ Thank you for the valuable points. We acknowledge that our mineralogical characterization was insufficient. We thus conducted XRD for the basaltic rock and granite materials (new Fig. S1, copied below). We also paid more attention to the description of mineralogical aspects including the crystallinity-mineralogy linkage (Line 94-98 in revised text) and provided further information on the source rock in the revised texts (section 2.1).

➔ In the basaltic rock used in the current study, we did not detect amorphous mineral phase according to XRD (Yang et al., accepted). According to the Geological Survey of Japan (2022), the lithology of the mining site for the basaltic rock is mainly dolerite (basaltic intrusive rock), which could potentially explain the low amorphous phase presented (i.e., less amorphous phase than extrusive basalt). Therefore, the granite and basaltic rock used in this study are mainly composed of crystalline minerals that formed under different temperatures, which have different weatherability depending on their chemical composition and crystal structure.

➔ In this study, we selected two rocks that were expected to release Fe and Al at different rates so that we can test the effect of metal supply on the formation of organo-mineral assemblage, a key aspect of “organo-metallic glue” hypothesis. The degree of rock weathering during 55-day incubation was assessed by leaching of rock-forming elements and by the formation of secondary phases that are approximated by sequential metal extractions. The results were consistent with the differences in the relative abundance of weatherable primary minerals in the two rocks.

2. **Mineralogical composition of the samples**

   Several aspects would benefit from clarification:

- What is the mineralogical composition of the granite? Which ferromagnesian minerals, which feldspars, and in what proportions respective to quartz?

➔ We conducted XRD of the crushed granite used. Major minerals identified were feldspar (albite and orthoclase) and quartz. The XRD patterns (shown below) were now added in the Supplementary Information as Fig. S1.

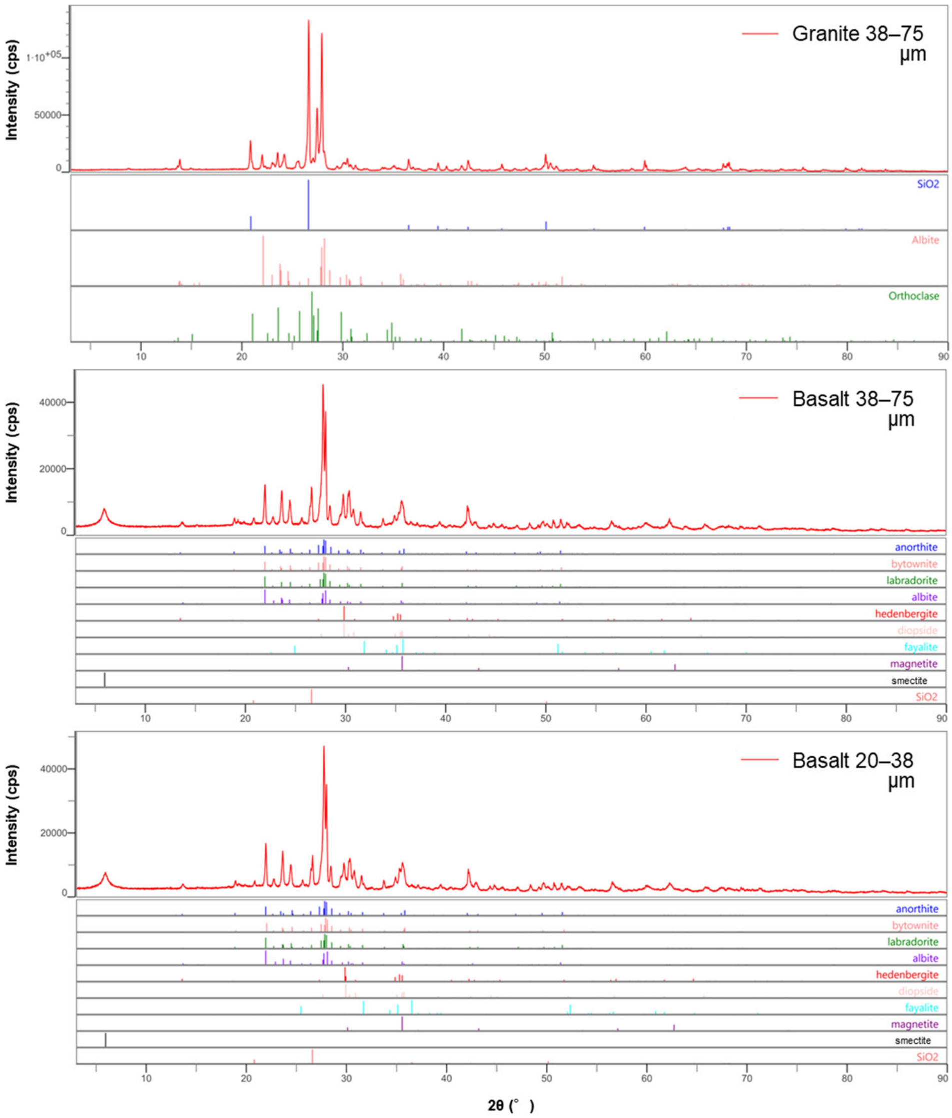

Figure S1. Mineralogical composition of the crushed rocks (granite, coarse basalt, and fine basalt) used in this experiment.

- Why do the hypotheses about the formation of ferrihydrite, goethite, and allophane not rely on direct mineralogical analyses (e.g., comparing MF at D0 vs. D55)? Such information is crucial for a mechanistic understanding.

➔ We agree that would be ideal. The main challenge was the mass limitation, esp. of MF. Out of 10 g mineral-OM mixture, MF was only 0.057 g for Granite, 0.017 g of Sand, and 0.68−1.2 g for the two basalts on Day 55 (new Table S6 in Supplementary Information, also shown below). For this reason, direct mineralogical characterization by XRD was not feasible for MF, and the metal extraction was conducted only for the two basalt treatments (Fig. 5).

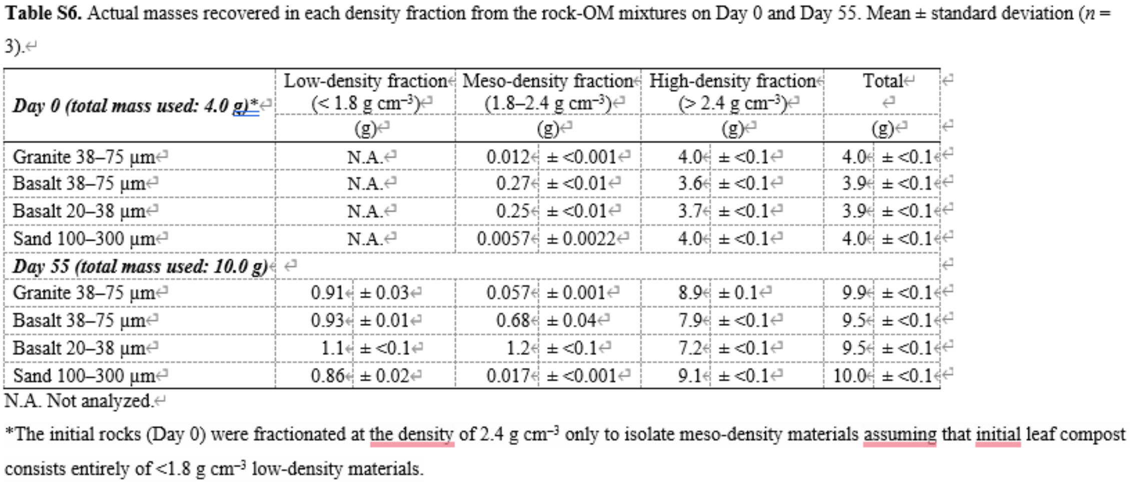

Table S6. Actual masses recovered in each density fraction from the rock-OM mixtures on Day 0 and Day 55. Mean ± standard deviation (n = 3).

Day 0 (total mass used: 4.0 g)*	Low-density fraction (< 1.8 g cm^{-3}) (g)	Meso-density fraction (1.8–2.4 g cm^{-3}) (g)	High-density fraction (> 2.4 g cm^{-3}) (g)	Total (g)
Granite 38–75 µm	N.A.	0.012 ± <0.001	4.0 ± <0.1	4.0 ± <0.1
Basalt 38–75 µm	N.A.	0.27 ± <0.01	3.6 ± <0.1	3.9 ± <0.1
Basalt 20–38 µm	N.A.	0.25 ± <0.01	3.7 ± <0.1	3.9 ± <0.1
Sand 100–300 µm	N.A.	0.0057 ± 0.0022	4.0 ± <0.1	4.0 ± <0.1
Day 55 (total mass used: 10.0 g)				
Granite 38–75 µm	0.91 ± 0.03	0.057 ± 0.001	8.9 ± 0.1	9.9 ± <0.1
Basalt 38–75 µm	0.93 ± 0.01	0.68 ± 0.04	7.9 ± <0.1	9.5 ± <0.1
Basalt 20–38 µm	1.1 ± <0.1	1.2 ± <0.1	7.2 ± <0.1	9.5 ± <0.1
Sand 100–300 µm	0.86 ± 0.02	0.017 ± <0.001	9.1 ± <0.1	10.0 ± <0.1

N.A. Not analyzed.

*The initial rocks (Day 0) were fractionated at the density of 2.4 g cm^{-3} only to isolate meso-density materials assuming that initial leaf compost consists entirely of <1.8 g cm^{-3} low-density materials.

- What is the mineralogical composition of the sand that justifies its use as a control? It is unfortunate that the mineral characterization was not performed more thoroughly, as this would provide real added value compared to previous studies on the organo-metallic glue hypothesis.

➔ The sand (called "Toyoura sand") used in this study is a well-characterized river sand in Japan. We now ran XRF for the sand and now included in Table S1 (copied below). The XRD data of the sand, based on another previous study, showed that it was entirely composed of quartz (Kimura et al., 2015, Energies, 8, 7202–7223, https://doi.org/10.3390/en8077202).

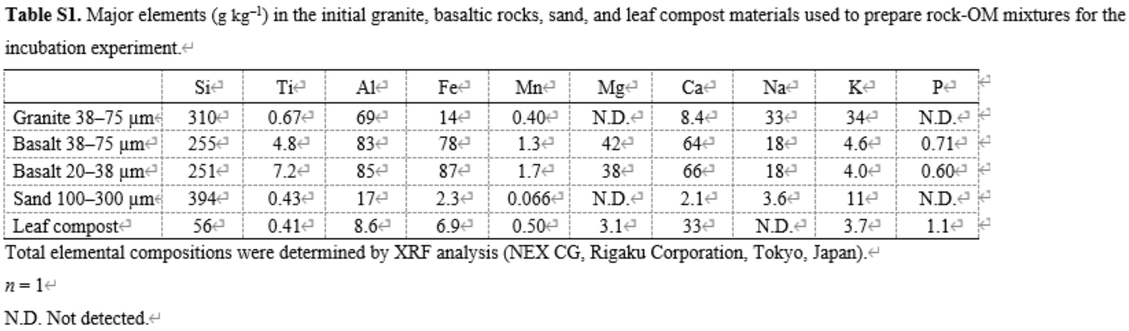

Table S1. Major elements (g kg^{-1}) in the initial granite, basaltic rocks, sand, and leaf compost materials used to prepare rock-OM mixtures for the incubation experiment.

	Si	Ti	Al	Fe	Mn	Mg	Ca	Na	K	P
Granite 38–75 µm	310	0.67	69	14	0.40	N.D.	8.4	33	34	N.D.
Basalt 38–75 µm	255	4.8	83	78	1.3	42	64	18	4.6	0.71
Basalt 20–38 µm	251	7.2	85	87	1.7	38	66	18	4.0	0.60
Sand 100–300 µm	394	0.43	17	2.3	0.066	N.D.	2.1	3.6	11	N.D.
Leaf compost	56	0.41	8.6	6.9	0.50	3.1	33	N.D.	3.7	1.1

Total elemental compositions were determined by XRF analysis (NEX CG, Rigaku Corporation, Tokyo, Japan).

$n = 1$

N.D. Not detected.

1. **Coherence of Methods and Results**

   The "Materials and Methods" and "Results" sections are not always fully consistent, which sometimes makes the reading confusing. It is essential that these two sections match precisely, especially in a manuscript with such a large dataset. Otherwise, readers may easily lose track.

➔ We now extensively revised to improve readability. Also, to help readers to navigate, we now newly added Table S7 which shows where one can find key quantitative data (mass, carbon, metal before and after the incubation).

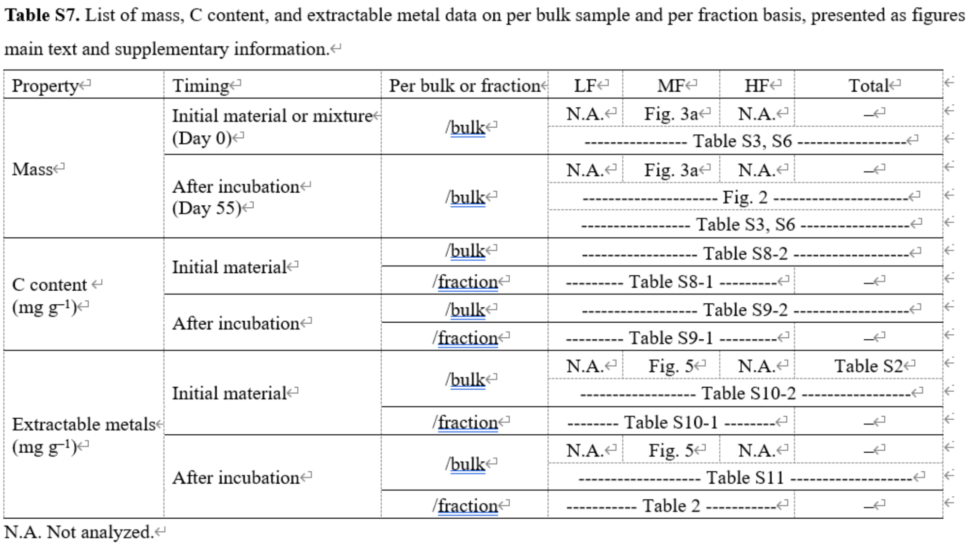

Table S7. List of mass, C content, and extractable metal data on per bulk sample and per fraction basis, presented as figures and tables in the main text and supplementary information.

Property	Timing	Per bulk or fraction	LF	MF	HF	Total
Mass	Initial material or mixture (Day 0)	/bulk	N.A.	Fig. 3a	N.A.	–
			---------------- Table S3, S6 ----------------			
	After incubation (Day 55)	/bulk	N.A.	Fig. 3a	N.A.	–
			-------------------- Fig. 2 --------------------			
			---------------- Table S3, S6 ----------------			
C content (mg g^{-1})	Initial material	/bulk	----------------- Table S8-2 -----------------			
		/fraction	--------- Table S8-1 ---------			–
	After incubation	/bulk	----------------- Table S9-2 -----------------			
		/fraction	--------- Table S9-1 ---------			–
Extractable metals (mg g^{-1})	Initial material	/bulk	N.A.	Fig. 5	N.A.	Table S2
			----------------- Table S10-2 -----------------			
		/fraction	-------- Table S10-1 --------			–
	After incubation	/bulk	N.A.	Fig. 5	N.A.	–
			------------------ Table S11 ------------------			
		/fraction	----------- Table 2 -----------			–

N.A. Not analyzed.

2. **Figures**

   It may be strategic to prioritize the most informative figures for the main text, while moving others to the Supplementary Information. This would reduce the number of figures in the core manuscript and allow those retained to be shown at a larger, more legible scale.

→ Thank you for your advice. We had 9 figures and 5 tables in the original manuscript. We kept those in the revised manuscript. In the Supplementary Information, we now have 4 figures and 14 tables, which include 4 additional tables to the Supplementary Information to accommodate your comments, as shown below. We intended to select the most informative figures and tables in the main text.

3. **Density fractionation yields**

   I am not fully convinced by the explanations regarding the quantitative mass balance of the density fractionations. The calculations do not seem to compare to pre-fractionation values. This point needs to be clarified (and perhaps discussed, since SPT extractions are often not neutral in terms of yields).

→ We provided the mass, C, and N recovery of the density fractions in Table 1. We realized that the C and N in density fractions presented in supplementary tables were confusing. In revised Table S8-1 and S8-2, we explained how we obtained LF, MF, HF data in more detail.

→ Now we have also added the mass of each fraction before and after the density fractionation, including the summed total mass as a new table in the Supplementary Information (Table S6, copied above). In addition, the mass distribution across the density fractions is shown in Fig. 2. Because MF was our focus, we compared the change in MF mass between pre- and post-incubation (Day 0 vs. Day 55) in Fig. 3(a).

4. **STXM images**

   I am not fully convinced that the STXM chemical maps demonstrate co-precipitates. The colocalization of C, Al, and Fe—which at this resolution should be observed pixel by pixel—is not clear. Moreover, this colocalization is not shown for basalt (which should provide the most favorable conditions for the glue hypothesis). It is also unclear why Al and Fe were not measured in the basalt samples; at the very least, this should be discussed.

➔ Thank you for pointing out important details. Overall, we are aware of the limitations in our STXM analysis. But, given the limited beam time we had, this was the best we could because it takes lots of time and some luck to find the region of interest. We sometimes found particles or composites that are almost entirely composed of OM or minerals, or those too thick to be analyzed by STXM.

➔ As explained to Reviewer 1 response, we wanted to characterize “fine basalt treatment”, not just “corase basalt” and “granite” MF materials. But we ran out of time. This point is added in Line 418.

➔ Whether the images taken represent coprecipitates or not is clearly an important question. Our answer is “possibly yes, possibly no”. The dominant Fe-rich zone in the coarse basalt image (Fig. 7B) may represent short-range-order Fe oxides (e.g., ferrihydrite) but the lower part adjacent to C and Al hot spots are more likely to be coprecipitate. Given the high heterogeneity even in such artificial soil system, we think it is difficult to identify specific phase. For this reason, we did not suggest that STXM images represent coprecipitate in the main text.

The question of sample thickness is also not addressed: as drop deposits, the sample thickness varies across pixels, and signal intensity is directly linked to thickness. Furthermore, it would be valuable to provide a C spectrum for the leaf compost at D0 and for the LF at D55 as references. These would allow the reader to assess whether the organic matter of the MF fraction underwent transformations detectable by STXM.

➔ Thank you for the comments. We now addressed the thickness heterogeneity issue in the revised text as follows (Line 229-231):
“Deposited materials inevitably have sample thickness variation. Interpretation of the spectral results therefore requires cautions because obtained signal from each pixcel is intensified in thicker region of the deposited sample (Wan et al., 2007)”.

Finally, I find the discussion of the nature of the OM in the MF somewhat contradictory:

- On one hand, the authors argue for the presence of “entrapped, less microbially processed OM” (which in my opinion corresponds more to leaf debris from the compost).
- On the other, they argue for results “in line with microbially processed OM in mineral

soils."

➔ Thank you for pointing out the potentially contradictory discussions. Given the heterogeneity of the material even within the MF, we think both types of OM may be present in MF. This view is consistent with the results of C:N and delta C-13, N-15 where the OM in MF showed intermediate values between LF and HF. We tried to discuss this point more thoroughly in discussion section 4.3 in the revised text.

Other remarks

To aid the reader, I suggest providing the following in Supplementary Information in tables:

- Masses of the mixtures before and after incubation.

➔ A new table has been added to the Supplementary Information (Table S3, copied below). We have also shown the mass of each component —crushed rocks, sand, and leaf compost —to prepare the mineral-OM mixtures in the main text (Line 132-133).

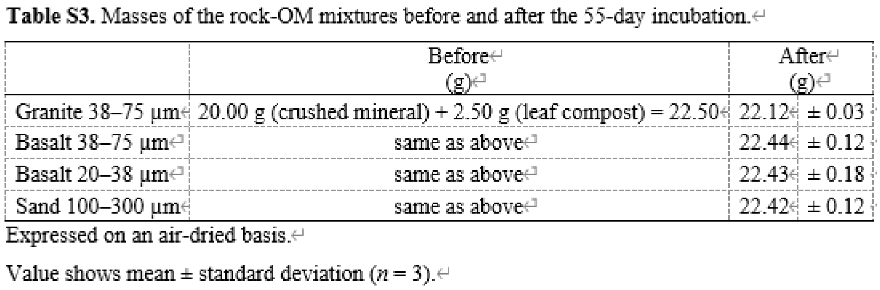

Table S3. Masses of the rock-OM mixtures before and after the 55-day incubation.

	Before (g)	After (g)	
Granite 38–75 µm	20.00 g (crushed mineral) + 2.50 g (leaf compost) = 22.50	22.12	± 0.03
Basalt 38–75 µm	same as above	22.44	± 0.12
Basalt 20–38 µm	same as above	22.43	± 0.18
Sand 100–300 µm	same as above	22.42	± 0.12

Expressed on an air-dried basis.

Value shows mean ± standard deviation (*n* = 3).

- Mass distributions across each analysis (density fractions, microbial analyses, chemical extractions), and how homogenization was performed before splitting the samples.

➔ The mass distribution across each analysis is now shown as Table S5 (copied below).

➔ The homogenization step is now described in Materials and Methods (Line 157 in the revised text) as follows.

"After incubation, the rock-OM mixtures (Table S3) were gently disaggregated, sieved (2 mm) using a spatula, homogenized in a plastic bag, and stored for further analysis (Table S5)."

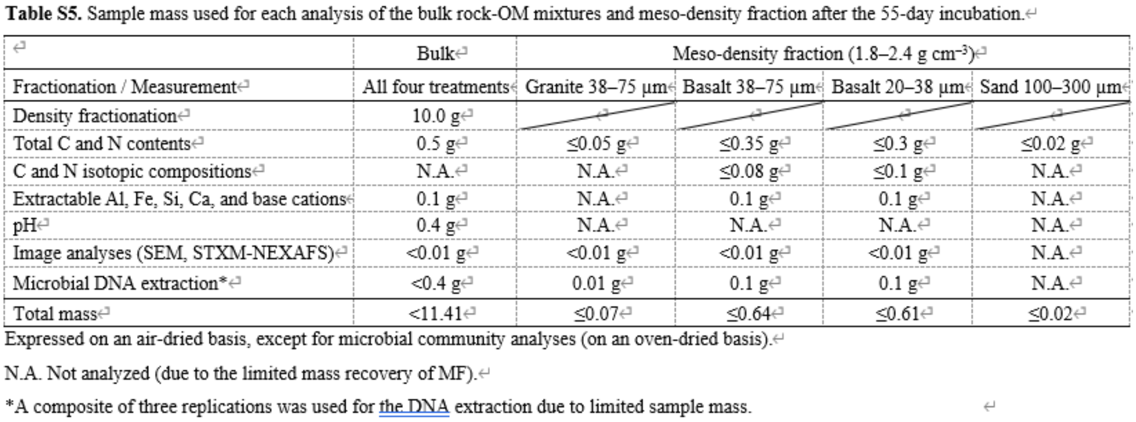

Table S5. Sample mass used for each analysis of the bulk rock-OM mixtures and meso-density fraction after the 55-day incubation.

	Bulk	Meso-density fraction (1.8–2.4 g cm^{-3})			
Fractionation / Measurement	All four treatments	Granite 38–75 µm	Basalt 38–75 µm	Basalt 20–38 µm	Sand 100–300 µm
Density fractionation	10.0 g				
Total C and N contents	0.5 g	≤0.05 g	≤0.35 g	≤0.3 g	≤0.02 g
C and N isotopic compositions	N.A.	N.A.	≤0.08 g	≤0.1 g	N.A.
Extractable Al, Fe, Si, Ca, and base cations	0.1 g	N.A.	0.1 g	0.1 g	N.A.
pH	0.4 g	N.A.	N.A.	N.A.	N.A.
Image analyses (SEM, STXM-NEXAFS)	<0.01 g	<0.01 g	<0.01 g	<0.01 g	N.A.
Microbial DNA extraction*	<0.4 g	0.01 g	0.1 g	0.1 g	N.A.
Total mass	<11.41	≤0.07	≤0.64	≤0.61	≤0.02

Expressed on an air-dried basis, except for microbial community analyses (on an oven-dried basis).

N.A. Not analyzed (due to the limited mass recovery of MF).

*A composite of three replications was used for the DNA extraction due to limited sample mass.

- Compositions of the incubated samples before density fractionation.

➔ We are not sure what the compositions mean here. We used a subset of the homogenized post-incubation samples for the density fractionation. As mentioned above, we now add the homogenization step in Materials and Methods section.

- Actual masses collected in LF, MF, and HF fractions (so readers do not need to reconstruct mass balances from the bar chart in Fig. 2). This point is very important to my mind.

➔ Now shown as Table S6. See above.

- C and N concentrations in fractions expressed as mg C (or N) per g fraction, not only per g bulk.

➔ New tables (S8-1, S9-1) are added in the Supplementary Information (copied below).

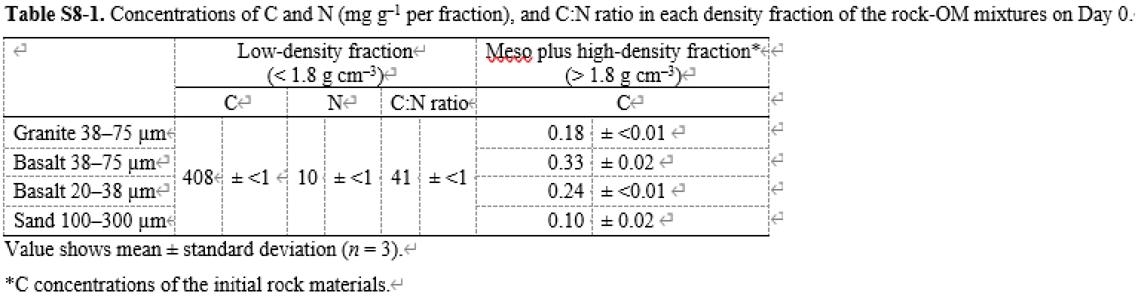

Table S8-1. Concentrations of C and N (mg g^{-1} per fraction), and C:N ratio in each density fraction of the rock-OM mixtures on Day 0.

	Low-density fraction (< 1.8 g cm^{-3})			Meso plus high-density fraction* (> 1.8 g cm^{-3})
	C	N	C:N ratio	C
Granite 38–75 µm	408 ± <1	10 ± <1	41 ± <1	0.18 ± <0.01
Basalt 38–75 µm				0.33 ± 0.02
Basalt 20–38 µm				0.24 ± <0.01
Sand 100–300 µm				0.10 ± 0.02

Value shows mean ± standard deviation ($n = 3$).

*C concentrations of the initial rock materials.

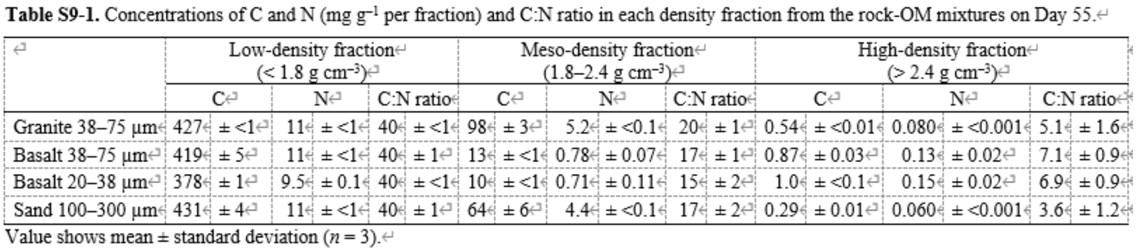

Table S9-1. Concentrations of C and N (mg g^{-1} per fraction) and C:N ratio in each density fraction from the rock-OM mixtures on Day 55.

	Low-density fraction (< 1.8 g cm^{-3})			Meso-density fraction (1.8–2.4 g cm^{-3})			High-density fraction (> 2.4 g cm^{-3})		
	C	N	C:N ratio	C	N	C:N ratio	C	N	C:N ratio
Granite 38–75 µm	427 ± <1	11 ± <1	40 ± <1	98 ± 3	5.2 ± <0.1	20 ± 1	0.54 ± <0.01	0.080 ± <0.001	5.1 ± 1.6
Basalt 38–75 µm	419 ± 5	11 ± <1	40 ± 1	13 ± <1	0.78 ± 0.07	17 ± 1	0.87 ± 0.03	0.13 ± 0.02	7.1 ± 0.9
Basalt 20–38 µm	378 ± 1	9.5 ± 0.1	40 ± <1	10 ± <1	0.71 ± 0.11	15 ± 2	1.0 ± <0.1	0.15 ± 0.02	6.9 ± 0.9
Sand 100–300 µm	431 ± 4	11 ± <1	40 ± 1	64 ± 6	4.4 ± <0.1	17 ± 2	0.29 ± 0.01	0.060 ± <0.001	3.6 ± 1.2

Value shows mean ± standard deviation ($n = 3$).

- If possible, the total amount of C mineralized during the 55-day incubation.

➔ We did not measure the C mineralization (respiration).

Section-specific remarks

Introduction

- Granulometric details are overly precise here and belong in Material & Methods section. The rationale behind the chosen grain sizes should be better justified.

➔ We agree. We removed the particle size information from Introduction now.

- Justify the use of sand as a control, and explain why its granulometry differs from that of the crushed rocks. Add the control to Table S1.

➔ Sand used largely consists of quartz. Please check the XRD data available online (Kimura et al., 2015, Energies, 8, 7202–7223, https://doi.org/10.3390/en8077202). The XRF data of the sand were now included in Table S1.

- Clarify why "heterotrophic activity" is specified—were autotrophic processes expected?

➔ Organic matter added drives microbial activity in this type of incubation experiment. Thus, heterotrophs are the dominant players. But we removed "heterotrophic" to make it simple.

Materials and Methods

- L111: avoid citing a reference "under review," as it is neither accepted nor accessible.

➔ Please see our general response (2) to the editor above. We will provide more detailed info as soon as the DOI is created.

- Grain size analyses can be moved to SI.

➔ We moved the description of grain-size analyses to Supplementary Information, where we provided the grain size distribution figure (Fig. S2).

- L121-122: unnecessary here; should be moved to L143.

➔ We moved this description.

- L132: report C concentration at D0.

➔ The C and N concentrations in fractions expressed as mg C and N per g fraction (new Table S8-1, copied above) and per g bulk (new Table S8-2, copied below) on Day 0 were provided in the Supplementary Information.

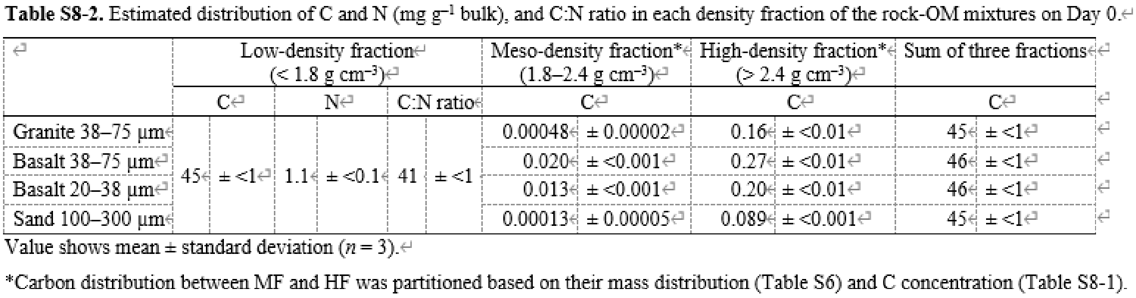

Table S8-2. Estimated distribution of C and N (mg g^{-1} bulk), and C:N ratio in each density fraction of the rock-OM mixtures on Day 0.

	Low-density fraction (< 1.8 g cm^{-3})			Meso-density fraction* (1.8–2.4 g cm^{-3})	High-density fraction* (> 2.4 g cm^{-3})	Sum of three fractions
	C	N	C:N ratio	C	C	C
Granite 38–75 µm	45 ± <1	1.1 ± <0.1	41 ± <1	0.00048 ± 0.00002	0.16 ± <0.01	45 ± <1
Basalt 38–75 µm				0.020 ± <0.001	0.27 ± <0.01	46 ± <1
Basalt 20–38 µm				0.013 ± <0.001	0.20 ± <0.01	46 ± <1
Sand 100–300 µm				0.00013 ± 0.00005	0.089 ± <0.001	45 ± <1

Value shows mean ± standard deviation ($n = 3$).

*Carbon distribution between MF and HF was partitioned based on their mass distribution (Table S6) and C concentration (Table S8-1).

- L154-155: unclear step.

→ This is an important step. So, we have now added a more detailed description as follows (Line 152-155). "Columns were then incubated in the dark at 30 oC for 5 days until near complete drying, forming partially cemented material. Before re-wetting each time, the hardened material was gently loosened with a spatula to approximately half the column depth to facilitate infiltration of rainwater and inoculum. ".

- L158-160: was the leachate filtered before analysis?

→ Yes, we added this information to the text now (Line 158)

- L161: the section title implies density fractionations were performed only post-incubation, but results show they were also performed at D0. Please clarify.

→ Thank you for pointing out. We realized it is rather confusing. We revised the section title to include the pre-incubation. The density fractionation we conducted for the initial material is slightly different from what we did Day 55 material because leaf compost was assumed to fall into low-density fraction entirely. This is now clearly explained on the footnotes in Table S6, S8, and S10.

- L178: isotopic analyses appear suddenly; their rationale should be included in the Introduction.

→ The rationale of isotopic analyses is now included in Method section 2.4 as we cannot find a good place to add that in Introduction (Line 183-184).
"The degree of microbial alteration of OM can be assessed by C and N isotopic compositions. Thus, initial leaf compost and density fractions under the two basalt treatments after the incubation were analyzed by…"

- L215: clarify the meaning of "shaking-resistant microaggregates." Does this mean aggregates not dispersed by US? Please define precisely.

→ We used this term because the meso-density materials isolated after the density fractionation experienced mechanical shaking (not ultrasonic dispersion) during the fractionation procedure. The meso-density materials isolated are thus organo-mineral assemblages (as OM is too light and rock minerals alone are too heavy) and thus typically aggregated at microscale.

- Justify why the focus is on bacteria only, not fungi.

➔ We addressed this limitation in Method section 2.5 as follows (Line 234).
"Bacteria may exert stronger effect on rock weathering than fungi in our short-term condition with repeated physical mixing. We thus assessed the changes in bacterial community 16S amplicon sequencing…."

Results

- Check the order of SI tables cited.

➔ We checked the order of SI tables.

- L294: Table S4 should include the total value discussed here.

➔ New table S9-2 includes the sum of density fractions now.

- L322: explain why LH and MH are pooled.

➔ Our apologies for the insufficient explanation. To obtain the values for the Day 0 mixture, we assumed that the leaf compost is 100% LF and the crushed rock was fractionated at the density of 2.4 g cm^{-3} only. However, we measured the CN and extractable metals of the original leaf compost as well as crushed rock, separately. Using that data, we were able to calculate the mass and elemental concentration of LF, MF, and HF for the Day 0 mixtures as well as Day 55 mixtures. Now Table S5 is changed to Table S10 by separating the metal concentration of LF and MF.

➔ Because this point was unclear in the original text in the Method section (2.3), we have now added the following sentence.
"For the pre-incubation samples, crushed rocks and sand were fractionated by density at 2.4 g cm–3 to obtain the fraction (< 2.4 g cm–3) and HF (> 2.4 g cm–3). The initial leaf compost was assumed to be < 1.8 g cm–3."

- Use clear indices for chemical parameters (e.g., Al_tot, Al_pp, Al_ox, Al_DC, or Al_extract for the sum). This will help readers follow text, figures, and tables.

➔ We agree. We attempted to improve the readability and clarity in the revised version.

- Add decimals when uncertainties are 0.0.

➔ We changed the expressions to e.g., ± <0.1.

- Fig. 6: add an arrow to highlight fungi; also show surface states for D0 particles (10 µm image). Increase image size—they are currently very hard to interpret. Provide control images of sand samples as well as additional aggregates in SI to demonstrate representativeness.

➔ We highlighted fungi (Fig. 6d) and (Fig. 6, copied below).

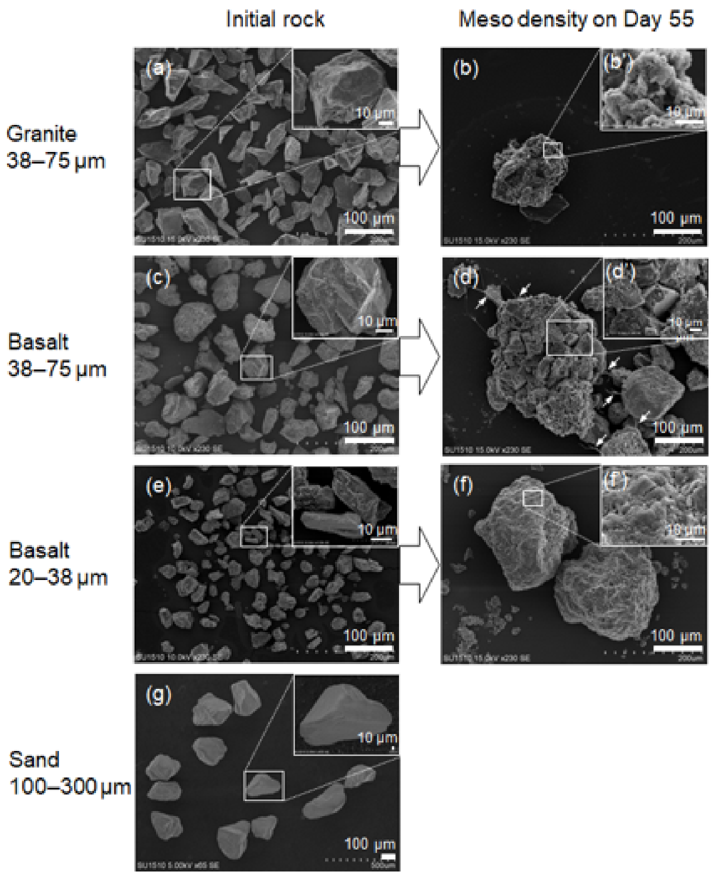

Figure 6. SEM images of the crushed rock minerals and the isolated density fraction. (a) initial granite, (b) MF of granite, (c) initial coarse basalt, (d) MF of coarse basalt, (e) initial fine basalt, (f) MF of fine basalt, and (g) initial sand. Fungal hyphae were shown with arrows (d).

- L421: is Fig. 7B coarse basalt? « Fine » is mentioned in the text

➔ Thank you for pointing out. We replaced it with “coarse basalt”.

- L424: why were Al and Fe not measured successfully?

➔ As responded above, we ran out of STXM beam time, unfortunately. Switching X-ray energy for specific element takes time. Plus, finding the same location after switching energy is sometime a challenge esp. for Al due to the zone plate used in the KEK STXM beamline. We think it is still valuable to report Al and Fe map for just one sample/ROI. If it is not informative and rather confusing for readers, we are willing to delete them.

- Figure S4 legend: specify SEM images.

➔ We improved the clarity of the figure title and legend.

- Fig. 7: include S4b and d images to directly show STXM-analyzed samples. Label maps (a1, a2, a3…) directly on images to improve readability.

➔ Thank you for advice. Now we have included the SEM image in Fig. 7 and added labels on the maps (copied below). We also improved the figure legend description for readability.

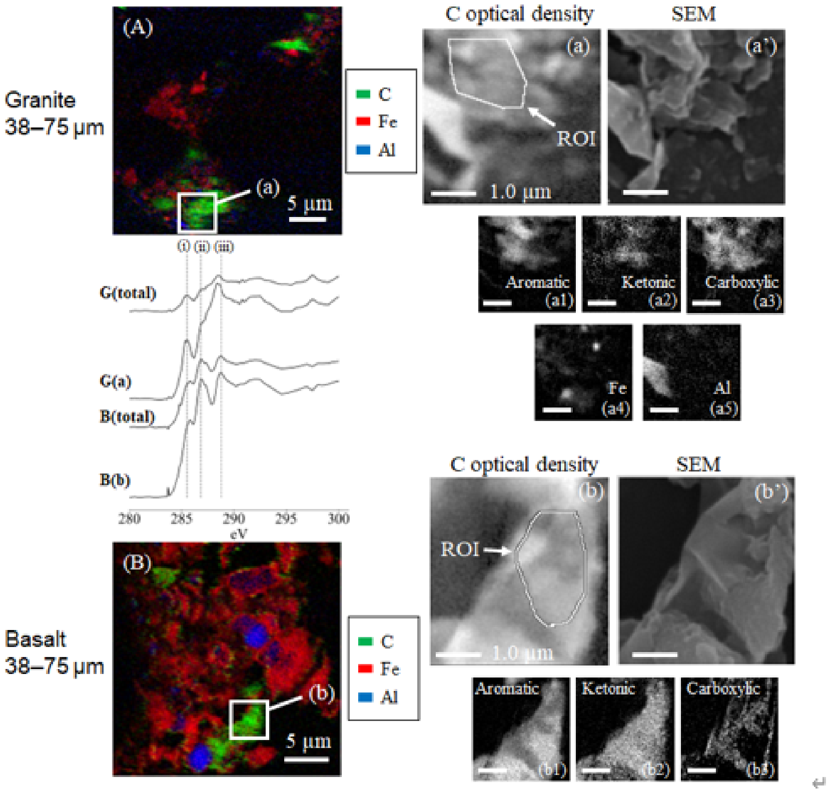

Figure 7. The elemental maps obtained with STXM-NEXAFS analysis. The images show the distribution of elements in the 35 x 35 µm region of interest for MF of granite (A) and 30 x 30 µm region of interest for coarse basalt treatments (B) after the 55-day incubation, respectively. C optical density (a, b) and SEM (a', b') images for a selected C-rich region at higher resolution (5 x 5 µm and 4 x 4 µm, respectively). The C K-edge NEXAFS spectra at middle left panel show the image stacks for the 35 x 35 µm region of granite, G(total), and coarse basalt, B(total), and for the C-rich regions of granite, G(a), and coarse basalt, B(b). Dashed lines (i), (ii), and (iii) represent the energy levels at 285.5, 286.8, and 288.8 eV corresponding to aromatic C, ketonic C, and carboxylic C. The images (a1), (a2), and (a3) and (b1), (b2), and (b3) show the spatial distribution of specific C type, aromatic C, ketonic C, carboxyl C, respectively, in the higher resolution images. The distribution of Fe (a4) and Al (a5) was measured only for the granite sample due to beamtime limitation.

- §3.6: why was the inoculum (meant to introduce microbial diversity) not characterized?

➔ In retrospect, we should have done that. But our main goal was to compare the community composition of leaf compost with that of rock-OM mixtures. By the water extraction from the compost, the community diversity is expected to decrease. It would be interesting to see how the low-diversity community changes upon inoculation to the mixture. However, that was beyond the scope of the current study.

- L446: a genus is mentioned but not illustrated; conversely, Fig. 8b shows the phylum but is not cited or discussed. Fig. 8c is also not referenced in the text.

➔ Thank you for pointing out. We cited Fig. 8b, c in the text now.

Discussion

- L461: “along with associated changes in OM”—please clarify which changes are meant.

➔ Now the discussion is extensively edited, which we believe increased the clarity and readability as mentioned above (our response to the editor).

- L465: why switch to “mg C/g rock” when all results are presented as “mg C/g bulk”?

➔ We now changed to “per gram bulk” basis.

- L488: “described” seems missing? Please check.

➔ Sorry for the typo. It is now corrected.

- L496: aggregation—could it be quantified? This is very important regarding the glue theory and should be discussed.

➔ Yes, this is very important question in this study. We quantified it by isolating meso-density material which is, by operational definition, organo-mineral assemblage (=organo-mineral aggregate). We think this point became more clear in the revised discussion (4.1-4.3).

- L589-590: statement is unclear.

➔ We revised to increase clarity (Line 525-532).

“We made a first-order approximation of the organo-metallic material, recognizing that the calculation relies on simplified assumptions that Fe and Al occurred as ferrihydrite and allophane and OM consisted of 50% C. This component accounted for approximately a quater of the total MF mass, suggesting that resultant Fe-Al-OM coprecipitates may be abundant enough to bind the remining solids (mainly basalt particles with minor POM) into stable organo-mineral assemblages. Other major elements released from rocks (esp. Si and Ca) may synergestically contribute to aggregation (Oades 1988; Jamoteou et al, 2025). These results indicate that organo-metallic glue formed in the current study is best described as Fe- and Al-rich coprecipitates, rather than OM-dominated complexes.”