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Interactive comment on "Iron and aluminum association with microbially processed organic matter via meso-density aggregate formation across soils: organo-metallic glue hypothesis" *by* Rota Wagai et al.

Rota Wagai et al.

rota@affrc.go.jp

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Thank you for the thorough review, encouragement, and valuable constructive comments. We responded to each of your comments below.

Rota Wagai (on behalf of the authors)

– It is impossible to verify the calculations proposed by the authors, it is impossible to know what are the losses during densimetric fractionation for individual soils, it is impossible to re-use the data for other works. My recommendation is to provide a

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Discussion paper

summary table of all the data as additional information.

—> We did not provide individual data as we tried to limit the volume of the study. In revision, we can provide the values for individual soil density fractions as a table (Table A1).

– The title needs to be changed because evidence that the organic matter of "nanocomposites" has been biotransformed by the microbial compartment is “light”. This argument is only based on the value of C:N which shows quite variable values (shown only in the supplementary informations graphs). Even if the trend is probably true, the authors have not investigated enough to state it with certainty.

—> We agree that we have to be careful on this point. We think the OM in the meso-density fractions is the mixture of both plant- and microbially-derived OM as depicted in Fig. 7b (glad that you liked this!). That was why we did not call it "microbially-derived".

However, we think it is appropriate to call it “microbially-processed” for three reasons: (i) the C:N ratio of the meso-density fractions was consistently lower by 2-23 units relative to the lowest-density fraction which mainly consists of plant detritus. (ii) SEM observation showed decreasing abundance of plant detritus with increasing density, (iii) delta N-15 analysis also showed that the meso-density OM was always more enriched relative to the OM in the low-density fraction by ca 2-6 per mil (unpublished data). The N-15 analysis of the density fractions was done for 14 out of the 23 soil samples. This result is more complex to interpret and will be incorporated into our next work. For these reasons, we think it is more appropriate to interpret that the majority of the OM present in the meso-density fractions is “microbially-processed”.

We will revise the abstract section related to this topic as follows (L21-24). “The OM in meso-density fractions showed 2-23 unit lower C:N ratio than the lowest-density fraction of respective soil and thus appeared microbially processed from the original plant material.”

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- In the introduction, last paragraph: the scientific questions asked must be made explicit.

—> We think the questions asked are clear enough (one hypothesis and several related questions). Or do you mean the last paragraph where we define the terminology?

- Abstract : - Indicate the regions of origin of the samples.

—> the information will be added in Abstract as follows: “We identified density fraction locations of major metal phases and OM using 23 soil samples from 5 climate zones and 5 soil orders (Andisols, Spodosols, Inceptisols, Mollisols, Ultisols) from Asia and North America, including . . ...”.

- L 24 -27 Sentence too long

—> We agree that it is a long sentence. We thought hard but we cannot think of a good way to shorten it at this moment.

- L27 : remove stable : there’s no argument that the OM is "stable."

—> “OM” is now removed.

Methods - The soils choices could be better justified.

—> We now explained the rational for the soil samples we selected in L134-136.

- Give more details on the fractionation protocol.

—> Now the protocol is fully described.

- Explain why the number of fractions is different from one soil to another?

—> Now it is explained.

- SPT is very acidic (pH down to 2); what could be the effect of such a pH on the “nanocomposites” ?

—> We will add the following sentence at the end of density fractionation section under

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Method. “We also assume little impact of sodium polytungstate on the extractability of Fe and Al phases or the nature of soil microaggregates as the SPT solution after the density fractionation typically had the pH value similar to bulk soil pH.”

Chemical analyses - Instead of adding weight bas concentration (Al+1/2 Fe) to approximately normalize the atomic mass difference between Al and Fe, working with atomic concentration would be more rigorous!

—> We debated on this point. We agree that the expression in the atomic mass is more rigorous and facilitate the comparison with other materials (e.g., experimental mixture, sediments). But the major interest is the comparison between these metals and C. And the C concentration in soils and soil physical fractions is almost always expressed on a weight basis. So we would like to keep our unit as is. But we now also provide these values on atomic mass in appropriate sections (figure legend in Fig. 1 and 3, the main text where we explain OC:metal ratios) to allow such comparison.

So, in the revised manuscript, we plan to add a following sentences in Method section (L189). “This allows us to compare the metal values with C on a weight basis. We also reported some values including the stoichiometric relationships among the target elements (e.g., Al:Si ratio) on molar basis.”

Peak density determination - To my mind, “peak density” is a term which is not really appropriate

—> We cannot think of any better ways to explain this. Sorry... We will add more explanation the corresponding parts in the main text to enhance the clarity to on this expression.

Result - I would suggest to first present the data (see previous remark) before presenting the recovery

—> We think it is better to show the recovery data first. If the recovery is now good, then the quality of the rest of results becomes questionable. So we prefer to put this

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information upfront as previous studies did (e.g., Swanston et al., 2005, Geoderma) https://www.sciencedirect.com/science/article/pii/S0016706104003258

- L230 Fig A1 : left panel - L290 : I may add an additional sentence to be sure that the reader understand properly the difference between fig 2 concentration and fig 5 distribution

—> Thank you for the advice. We will add some words to improve the clarity of this first sentence.

Discussion - L326 what are the “non-centrifugeable colloidal Fe/ Al oxide phases?

—> This refers to the colloids that were too small to spin down by the centrifugation used. This corresponds to the limitation of PP extraction discussed 11 lines above this sentence in the same paragraph.

- L328-331 : no data on Si were provided in the result part. (I think you may add a short section also on Si in the results)

—> Thank you for pointing out. We now will report Si results in Result section by reporting (1) Al:Si molar ratios of each density fraction for each soil, and (2) the proportion of total extractable Si in pyrophosphate- and oxalate-extractable phases.

- L345 : Is a graph Al:Fe as a function of pH interesting? I would have enjoyed to see it !

—> I agree that it would be interesting! But we don’t have pH of each density fraction. So we cannot do this.

- “nanocomposite” could also have been called “nanoCLICS” as proposed by Tamrat et al. 2019.

—> We will add this acronym where we cited this work in Discussion section.

- Fig 7a : position of the peaks is not consistent with fig 5

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—> The Y axis in Figure 7a is the concentration of metal per fraction where as Fig. 5 showed the metal distribution. So Fig. 7a matches with Fig. 2.

We realized that it is easy for readers to get confused with two expressions (concentration and distribution). So we plan to add some sentences to clearly distinguish the two expressions in Method as well as Results/Discussion sections.

- I do enjoy Fig 7 b ! —> Very glad that you liked it!

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