

Dear editors and referees,

We have now adjusted our initially submitted manuscript SOIL-2022-13 as per the proposed changes set out in our response letter that was uploaded into the online Discussion area on 11th of July 2022.

- This accompanying letter does not fully report on all changes made but summarizes our actions to the raised three main points of criticism (see 1-3° below). For smaller changes to the text, we would refer back to our initial response letter and to the track-changes version of this revised manuscript. An exception being the Editor's feedback onto our 11/07-response letter, which has been literally reiterated below under 4° and complemented with our new replies.
- All figures were updated and for convenient reading changes to figures have already been accepted in the track-changes version of our revised manuscript.
- Line numbers referred to in the below report apply to the manuscript version with track-changes still visible.

We thank the referees and associate editor for all their attention to our manuscript. We believe that accommodation of raised constructive comments strongly led to an improved analysis and manuscript. We hope that with these changes its quality now meets the criteria for publication in SOIL.

Yours sincerely,

The authors

1° Used soil fractionation scheme:

Both the 2nd referee and the AE questioned the effectiveness of the applied Zimmermann et al. (2007) method to subdivide SOM into meaningful fractions for the concerned set of volcanic soils. Specifically, separation of a stable aggregate associated OM fraction with sieving and just weak ultrasonic dispersion was deemed unsuccessful. In our response letter, we agreed to this point and proposed to add in a further fractionation step to “deconvolute” the in our case sizable and composite S+A (sand + aggregate OM) fraction originally obtained after 22 J ml⁻¹ ultrasonic dispersion and wet sieving at 63 µm. This further soil fractionation has been carried out for all 54 soil samples. Adaptations to the original ‘Zimmermann et al. (2007)’ fractionation scheme are now described in L144-151 (7 lines added). In supplementary material, we now also present a schematic of the fractionation procedure (Supplement 2). Specifically, the S+A fraction has now been further subdivided by a combination of ultrasonication at 400 J ml⁻¹ and sieving at 63 µm. A 100% disruption of > 63 µm aggregates was not achieved, as was in fact also not expected for these volcanic soils. In our previous Geoderma paper (<https://doi.org/10.1016/j.geoderma.2022.115963>) we found that full dispersion of these soils requires use of Na⁺ resin, very strong ultrasonication at 1200 J m⁻¹ and maintenance of pH at 8-9. Such conditions would be far too disruptive for SOC fractionation with likely dissolution of mineral-bound OC and diminution of POC. We tested lower energies (200 & 300 J ml⁻¹), only to find that then a substantial remnant macroaggregate fraction was left, while at 400 J ml⁻¹ this was no longer the case – though as explained above a full dispersal of > 63 µm aggregates was not achieved. Nevertheless, we at least succeeded in isolating OM contained in highly stable soil aggregates from dispersible ones. As described below, the SOC proportion of the former fraction was strongly dependent on land-use and related to levels of important binding agents. We therefore believe this adjustment of the soil fractionation procedure was meaningful and the further interpretation and discussion of found results benefitted strongly from this revision.

Results reported in Table 2 and Fig. 1 and in text in 3.1 have accordingly been updated. Interestingly we now detect clear differences between both considered land-uses with more SOC contained in S+A and less in s+c under agricultural than under forest land-use. The proportion of SOC in the S+A fraction also correlates well with the content of amorphous minerals ($r = 0.76$ and $p < 0.01$) and to a certain extent as well with Al_o (or $Al_o + 1/2Fe_o$, $p < 0.01$), exchangeable Ca^{2+} ($r = 0.6 - 0.67$ and $p < 0.01$). These observations now in fact strengthen our initial interpretation that strikingly more OC is occluded within stable soil aggregates under agricultural land use in our soils. Particularly, enhanced formation of pedogenic oxides (more Al_o) under agriculture could be identified as a candidate cause for enhanced soil aggregation and occlusion of OC therein. We now also found a negative correlation between the % of SOC in S+A and the relative degradability of the SOC. Liming caused lifting of exchangeable Ca^{2+} might also have contributed to improved soil aggregate strength, but as per the AEs advice we nuance this reasoning now in the text. As these results were all in line with our initial interpretation (support is now actually much stronger) just an all-in-all relatively limited update of the discussion section was required. Particularly changes were made to L339-340, L343-345, L377-405, L431, L445-450, and the conclusion section. Small changes were made to the abstract as well.

The referees also requested presentation of the C:N ratio and contents of pedogenic Fe and Al of both S+A and s+c fractions to further appreciate the nature of these fractions and the OM contained therein. We now present these data in a newly inserted Table 3. Using these new data, we also comment on the in part still overlapping nature of the derived s+c and S+A fractions (9 lines inserted L353-363).

2° Inquiries on C-inputs as part of the SOC balance:

Referee 1 asked to provide SOC stock per considered depth increment instead of SOC content – this has been implemented in Table 1. Referee 1 also requested to present soil C inputs, which would serve to interpret why there were particularly high SOC stocks in the included agricultural soil profiles, in fact comparable to those under pine forest and native tropical forest. As explained in our response to the referee, it is practically very hard to quantify C inputs from vegetation to the forest sites, while for the agricultural sites this is more straightforward (except for below-ground C inputs, that is). We thus proposed to at least provide estimates of annual plant and organic fertilizer C-inputs, based on literature estimates of Indonesian tropical forest C-inputs and inquiries to the agricultural management. Annual C-inputs on a per ha basis have been added to Table 1. We also commented on this now in newly inserted L447-450 in the discussion and changed the conclusion (L484-488), as also suggested in our response letter.

3° Similarity of the parent materials:

As explained in our response letter, detailed soil mineralogical on the six sites can be found in our recent publication (Anindita et al., 2022). Geoderma has granted us to reproduce data from that paper – we inserted soil mineralogical and weathering data in a new Supplement 1 Table S1. We do recognize that there was a discrepancy between the mineralogical composition of our Native Forest (NF) site and the other sites. However, the detailed comparison of mineralogy and weathering stage presented previously (Anindita et al., 2022) does demonstrate the likeness of the included pine forests and agricultural sites' parent material, validating to use these sites to investigate the impact of land-use on soil mineralogy and soil carbon quality. Throughout the discussion we also comment on found differences in SOC fractions and decomposability between the pine forest sites and agricultural sites, thus excluding the native forest site. We did the same for correlations with soil geochemical properties (e.g., in L354-355). As explained in L343-345 excluding the NF site from our analysis did not fundamentally change our interpretation. Hence, we feel strengthened that our conclusions are sufficiently robust.

4° Reply to the Editor's feedback & extra comments:

My additional suggestions / comments for the revision of the manuscript:

I agree with the authors that the parent material might be similar for the pine forests and the horticulture soils but not for the soil with the primary forest. Therefore, you should avoid to use the term "forest" sites / soils if you want to keep the results of the primary forest. Instead, I would recommend to use "pine forests" in order to clearly differentiate the primary forest from the pine forests. I would further recommend to add the weathering indices from your Geoderma paper to table 1 of this manuscript as well. Looking in your Geoderma paper I recognized that the "AG-i" showed some larger differences in comparison to the other sites, i.e., more amorphous Al and less content of 1:1 clay minerals (and clay content). I wonder what kind of consequences that might have. I wonder why you used different acronyms for the same sites in the Geoderma paper and the SOIL manuscript. That makes the live of potential readers not easy.

The acronyms of study sites have been harmonized with the Geoderma paper.

Weathering indices have now been entered in Supplementary 1.

AG-i indeed has more Al_o and amorphous materials and less clay than the other two agricultural sites. However, for the partitioning of SOC into fractions this soil actually displays similarity with the AG-y site (please see Fig. 1). The AG-o site on the other hand had just very little OC contained in the S+A fraction and much more in silt and clay even though its physicochemical properties resembled those of AG-y. If anything, this goes to show that soil aggregation and occlusion of OC is not just function of soil properties like mineralogy but also of other here not investigated factors. The $\delta^{13}\text{C}$ and C:N ratio of the AG-o bulk topsoil were -24.0‰ & 10.5, opposed to -15.4‰ & 14.7 at AG-i and 13.6‰ & 18.2 at AG-y. These SOC signatures suggest a closer correspondence between SOM quality in the AG-o site and pine forest sites then between the other two agricultural sites (Ag-y and Ag-i) and the pine forest sites. Such was not illogical given that these two sites were already longer in agricultural use. In sum, we see no grounds to specifically point out deviances in the mineralogy of the AG-i site, when actually overall there was much variation in either SOC quality or soil properties between replicate fields within the same land-use class. We would not want to further scrutinize causes for such 'within-group variation' with just 3 sites per land-use hampering a statistical analysis that is otherwise needed to render any interpretation less speculative.

As reviewer 2 and 3, I have some critical comments regarding your results from the density fractionation. Therefore, I highly appreciate to include a second ultrasonication step. Please explain / show that the 400 J ml⁻¹ is sufficient using e.g. the approach recommended by Cerli et al. (2012). A large part of your story is built on microaggregates and their potential role for C stabilization. Please discuss the limitations of the Zimmermann approach in relation to the role of microaggregates in your soils. I totally agree with reviewer 2 about additional analyses of the S+A and S+C fraction for oxalate extractable Al and Fe in order to allow mechanistic interpretations of your results. In addition, please report C/N ratios of your fractions. Taking all of these additional analyses into account, please rewrite your results and particularly your interpretation / discussion of the results.

Please see our responses under **1°**.

I also recognized that your POM is not always POM because of C contents smaller than 150-200 g kg⁻¹. However, I consider that as a minor problem because of the small contribution of POM to soil organic C. Nevertheless, you should mention that.

Indeed, a pertinent observation and we recognize that POM was not entirely well isolated by the used density fractionation. This inability to rid of smaller mineral particles from POM is not surprising for these volcanic soils (fine texture, high levels of pedogenic oxides and amorphous materials). At 1.8 g cm^{-3} (as used in the original Zimmerman et al method) we could clearly visually observe much remnant mineral matter in the 'POM' fraction, which is why we adapted the procedure to 1.5 g cm^{-3} . Because of the small mass proportion of the POM (median across soils just 0.1%) this minor contamination of the POM-fraction by probably s+c C would hold negligible impact on the derived distribution of SOC across soil fractions and our interpretation. We comment on this point in L225–227 (2 sentences inserted). The 20-40cm subsoil of NF-y site could have formed an exception, with a POM-C SOC proportion of 16% but its C-concentration was 39.3%C, demonstrating that it did largely consist of POM. We did not further comment on this final point in the text.

A further minor comment: The correlations using exchangeable Ca are not very convincing because of data distribution (Fig. 2). Therefore, I would not really interpret such correlations.

We recognize that the exchangeable Ca^{2+} data were not entirely evenly distributed. But in the discussion we already commented that in spite of the found positive correlation between S+A C and exchangeable Ca, we anyhow interpret this covariation to be unimportant given the far greater soil contents of Al_o and amorphous materials (L347-350). We therefore did not further comment on this point to the text.

In line 348 you wrote "... a larger biological lability". Do you mean "stability"?

Indeed, now corrected to 'biological stability' (L433)