

Response to reviewer and editor: “Tropical Andosol organic carbon quality and degradability in relation to soil geochemistry as affected by land use” (SOIL-2022-13).

We would like to thank reviewer and editor for their positive evaluation and valuable comments in this manuscript. Below, please find the answer, presented in the same order as in the review. The comments we received are in black and our response to them were written with a blue font after each comment. Line numbers refer to the track-changes version.

Comments from the reviewer:

Regardless of the editor’s decision, congratulation to the lead scientist and the entire team as their study brought to the table interesting data on the impact of the land use change on SOC contents in Andosols. The underlying assumption that land use history has an indirect impact on SOC stabilization via changing soil geochemistry, which in turn affects soil mineralogical properties/reactivity, is very exciting.

General comments:

1° I would like to ask the authors to double-check their calculations. I noticed inconsistencies in some places which I marked in the attached pdf file (no claim to completeness). Work should be done on the clarity of the presented tables (e.g. table 2). Furthermore, the authors should clearly formulate what exactly is being compared in the respective statements. It is often unclear whether defined depth increments were compared on average with each other, or whether the mean is compared across all depth increments of a land use.

We have thoroughly checked the calculations and found mistakes made during carrying out some of the statistical tests. Namely, it emerged that some t-tests were conducted with data from different depth layers jointly, inflating the p-values. When omitting such erroneous pseudo-replication, the comparison per depth increment is based on a total of 5 observations only. All p-values and report thereof in the results and discussion sections have now been corrected. The description of statistical methods (section 2.5) and the results and discussion (sections 3.1, 3.2, 3.3, 4.1, and 4.2) have been revised. We sincerely apologize for this obvious mistake. We also checked the text and more clearly indicated each time what depth layer and which groups are being compared.

2° The lack of mineralogical data in the main text of the work and the restrained use of mineralogical data in relation to changing geochemical parameters of the individual locations limit possible explanations for the measured data. + L. 321-323: “*With high to very high levels of Al_o, ... prominent binding agents in our set of soils.*” A further step could be taken here, and explanatory models should be set up using the mineralogical data and the geochemical data of the soils examined. Since the PF-o has very similar values to the agricultural soils.

Soil mineralogical characteristics had been reported before in Anindita et al. (2022). We did not copy these findings here but agree with the reviewer that such information would serve to get a more developed interpretation of found results on SOC fractionation and decomposability. **Accordingly, we now added some 15 lines on how XRD-spectrum or mass-proportions of minerals differed between the studied land-use categories and how these co-varied with degree of aggregation (indicated by the mass proportion of the S+A fraction) and containment of SOC within the S+A fraction.** This

discussion on soil mineralogy was in part complementary to the original 4.1 section but also in part replaced existing discussion parts and the original text was thoroughly reorganized into:

“We expected a lesser share of SOC to be occluded inside aggregates in the tilled agricultural soils, as absence of tillage results in less disruption of macroaggregates with increased formation of microaggregates and occlusion of POM (Six et al., 2000b; Zheng et al., 2018). However, tillage is not the only determinant of soil aggregate formation and breakdown as aggregate stability also depends on presence of binding agents that cluster mineral and organic particles into aggregates as well as size and mineralogy of the less reactive ‘skeletal’ particles. A further analysis of the soil mineralogy could prove useful to explain the observed variation in storage of OM in the S+A fraction between both land-uses. With respect to the ‘skeletal’ particles, main identified constituents were from the quartz group (Tridimite, Quartz, Cristobalite) (7-14%) and 1:1 clay minerals (5-35%) (Anindita et al., 2022), which, because of their inherent low permanent negative charge may be considered as relatively unreactive as well. In addition, smaller shares of Hornblende, Andesine, Albite and Rutile were present as well. Unsurprisingly, the XRD-estimated levels of these primary minerals and 1:1-clays did not correlate to the mass proportion of the S+A fraction, indeed suggesting that instead mainly other soil constituents acted as binding agents. Moreover, as contents of these relatively unreactive phases did not differ between the PF and AG soils, their variation could not explain the strong contrast in S+A contained OC between both land-uses. (Lines 340 – 353)

Major binding agents considered to work at micron to submicron scales are instead poorly-crystalline minerals, redox-sensitive minerals and microbial products (detritus and metabolites) (Oades & Waters, 1991). Particularly XRD amorphous materials constituted the bulk of the mineral phase (up to 65%). The limited oxalate extracted Si suggested allophane, imogolite and volcanic glass levels to be minor (Anindita et al., 2022). Hence a large pool of unidentified Fe and Al (oxy-)(hydr-)oxides instead likely formed the chief part of the XRD amorphous mineral phase. Particularly, minerals associated with Al_o would appear to be relevant colloids as bulk soil Al_o level correlated well with the bulk soil micropores volume (Anindita et al., 2022) and specific surface area. Wagai et al. (2018) indeed also found reactive metal phases to be important binding agents in Allophanic Andisols. As NH_4 -oxalate extraction is not entirely selective towards poorly-crystalline minerals (Rennert et al., 2018) it is probable that Al_o partly derived from crystalline Gibbsite as well. The same goes for Fe_o and crystalline pedogenic Fe. All soils contained rather large proportions of XRD detectable Gibbsite (3-15%) and with the exception of AG-y also 1-3% of Magnetite (Anindita et al., 2022). However, with no consistent differences in their XRD-spectrum proportions between both land-uses, and no correlation to the S+A fraction mass proportion, it is unlikely that variation in these secondary crystalline minerals led to contrasting soil aggregation and distribution of OC between the isolated S+A or s+c fractions between the PF and AG land-uses. Instead, the S+A mass proportion (in $g\ kg^{-1}$ bulk soil) correlated to S+A concentrations (in $g\ kg^{-1}$ S+A fraction) of both Al_o ($r = 0.86$; $p < 0.05$) and OC ($r = 0.73$; $p < 0.05$). It is thus likely that the ‘glue’ between ‘skeletal’ mineral particles consisted of a mixture of OM and poorly crystalline non-allophane/immogolite Al-containing minerals.” (Lines 355 – 370)

Furthermore, some of the conclusions drawn by the authors are untraceable for the reader from the data presented, please check this.

We have revised the sentences pointed out by the reviewer – please see our response to comments on L439-441 and 445-447 below.

3° What I see critically in the established discussion threads of the work is the strict separation between aggregation and direct interaction between reactive mineral surfaces among themselves and with organic compounds. Especially, aggregation is determined by the interaction of single minerals and more complex structures (also aggregates in the narrower sense) in the soil. These in turn change their properties due to the soil environment (geochemical properties) and generate their own geochemical setting within the complex structure. Nevertheless, such spatial units should still be characterized by the interaction of their individual mineral and organic components. This fact is often not considered in sufficient depth by the authors. From my point of view, the prerequisites for this are given if the individual soils of both land uses are considered in addition to a generalized consideration by averaging. Studying these details could make an important contribution to our understanding of carbon stabilization for Andosols.

+ L. 306: "4.1 Land use effects on free and aggregate protected OC" In this section, apart from the effect of the mineralogical composition in quality and quantity and its impact on the aggregation, the discussion of the direct interaction of reactive mineral surfaces within and on the surfaces of aggregates as a function of the geochemical changes between the investigated soils is missing.

We accept this criticism: The discussion of the nature of organo-mineral interactions and spatial arrangement of organo-metallic phases into aggregate-like structures was rather limited. We welcome the AE's and reviewer's suggestion to contemplate further on how Al-oxides and MAOM may 'shape' aggregates and in doing so also impact occlusion of POM or any OM for that matter. At the same time, we feel that we must do so rather sparsely because the applied fractionation proved rather inefficient in further subdividing the soil (micro)aggregates fraction into its various distinct constituents. Also, we did not employ spectroscopic methods or microscopy to ascertain the biochemical composition or physical arrangement of OM contained in aggregate fractions here. Nevertheless, we fully agree that assuming that a strict subdivision exists in either silt and clay- associated and aggregate-occluded OM is too simplistic. Indeed, aggregates themselves are largely composed of silt and clay and our results in fact do not suggest a substantially different OM or mineral composition of the s+c and S+A fractions. **We moved original discussion parts on the separation of POM and MAOM into a separate new section 4.5, in which we further contemplate more on whether or not there would be a causal relation between entrapment of OM inside microaggregates (that is 400 J ml⁻¹ resistant but > 63 μm) and the observed negatively correlated relative degradability of the SOM:**

" 4.5 Importance of physical occlusion of POM for SOC stabilization

As an important side mark to the interpretation of the SOC fractionation data it should be borne in mind that there is no guarantee that isolated SOM fractions coincide with distinct SOM pools, neither in terms of bioavailability nor intrinsic degradability. By expanding the Zimmerman et al. (2007) fractionation procedure with a strong (400 J ml⁻¹) ultrasonic dispersion step, we attempted to isolate s+c associated OC from POM contained in the S+A fraction. This ultrasonication step proved to result in incomplete disruption of the apparently very stable > 63 μm sized aggregates. With high levels of both OM as well as amorphous materials and poorly crystalline metal oxides, this is perhaps not surprising. The insignificant difference in C:N ratio between the S+A and s+c fractions suggests that while they obviously differ in their physical organization there is probably no strong contrast in the biochemical composition of the OM. Previous studies also found no clear difference of C composition from sand to clay fractions in Andosols (Baldock et al., 1992). Regardless, the here achieved subdivision of the SOM into OM present inside (S+A) or not (s+c) microaggregate structures would yet still appear relevant as the S+A SOC proportion correlated negatively to SOC mineralization, suggesting indeed that such physical organization of soil grants 'physical protection' against microbial breakdown, thought to result from reduced accessibility of microbes, exo-enzymes, and/or e-acceptors (Keil and

Mayer, 2014; Lehmann et al., 2007). The overall rather low C:N ratio of the S+A fraction (13.0 ± 4.2) indicates that it mainly contained mineral associated OM and probably little POM, complicating interpretation of mechanisms for a particular S+A SOC storage in the agricultural sites. Still, in the 0-20 cm layer at least a higher S+A fraction C:N ratio of the agriculture (17.0) vs. pine forest (11.0) S+A fraction suggests that at least to some extent enhanced SOC storage in aggregates under agricultural land-use might have resulted from extra occlusion of POM. However, our data do not provide unequivocal proof that the physical protection of SOM in the S+A fraction causes relative protection against mineralization. In addition, the absence of any land-use effect on protection of freshly added plant litter (see 4.4) leads to question this. Extra aggregation of OM into the S+A fraction in the agricultural soils might just be but a concomitant effect of the larger presence of tightly intermixed organo-metallic phases, while such smaller scaled structures themselves are instead zones of SOC stabilization. On the other hand, the fact that there was no negative correlation between Al_o or Fe_o and the relative SOC mineralization while Al_o did positively relate to S+A SOC proportion would instead suggest that indeed the physical arrangement of OM into stable aggregates led to a relative stabilization of SOC. From the limited presence of POM in the S+A fraction it logically emerges that mainly occlusion of smaller sized OM and its protection into aggregates was important. To further elucidate this, however, more detailed investigation of the formation of organo-mineral phases and their organization into microstructures is required, building on the recently forwarded organo-metallic glue hypothesis and proposed methodologies by Wagai et al. (2020).” (lines 511-539)

Detailed comments from the reviewer:

L. 7-8: Wording difficult to understand, please revise

We revised the sentence as follows:

“Young volcanic soils in tropical areas are subject to much faster pedogenesis, noticeable already on the centennial or even decadal scale. As land use is a recognized factor for soil formation, it is thus conceivable that even relatively recent land-use conversion in such areas would already bear significant impact on the resulting formed soils, e.g. in terms of content of pedogenic oxides.” (lines 7-11)

L. 11-14: Wording and sentence structure difficult to understand, please revise

We revised the sentence as follows:

“The content of oxalate-extracted Al (Al_o) correlated well with OC associated with sand-sized aggregates, particularly in subsoil. The proportion of SOC in sand-sized $400 J ml^{-1}$ ultrasonication resistant aggregates was also higher under agricultural than pine forest land-use and a likewise contrasts existed for Al_o . These combined observations suggest that enhanced formation of Al-(hydr)oxides promoted aggregation and physical occlusion of OC.” (lines 13-17)

L. 14-16: *“This was importantly ...was not found to depend on land use.”* This statement should be modified so that it refers to the secondary forest (pine afforestation) and not to the semi-natural forest (related to Fig. 4 right panel)

We revised the sentence as follows:

“This was importantly also consistent with a relatively lesser degradability of SOC in the agricultural sites, though we found no likewise difference in degradability of added ^{13}C -labelled ryegrass or in native SOC priming between the pine forest and agricultural land uses.” (lines 17-20)

L. 36-37: *"While soil mineralogy ... stabilization,"* It would be good if a few references were given for this central statement.

We added following references:

"While soil mineralogy itself is thus a well-recognized controller of SOC stabilization (Basile-Doelsch et al., 2005; Hernández et al., 2012; Lyu et al., 2021; Tamrat et al., 2019), surprisingly ..." (line 41)

L. 38-39: *"how land use history, through its impact on soil geochemistry ..."* I think land use history very directly controlled what kind of OM the soil received

Land use of course directly determines the kind of OM-inputs, but as we already spelled that out in L 29 – 32, we did not change this sentence.

L. 48-49: *"just very few of have considered the potential indirect impact of land use on SOC stability and its effect on the soil mineral phase."*

Impact of land use on soil mineral phases → indirect impact on soil mineral phases on SOC stability. Soil mineral phases and their reactivity central part of the study.

Since we did not really analyse the soil mineral phase and reactivity of minerals in detail, but only more general geochemical properties we revised this sentence as follows:

"just very few have considered the potential indirect impact of land use on SOC stability through its effect on the geochemical properties." (lines 53-55)

L. 58-59: *"Introduction of tillage and removal of permanent ... protection."* Needs reference because this statement does not go well with the references in the next sentence.

We added the following reference:

"Introduction of tillage and removal of permanent vegetative coverage usually adversely affect intra-microaggregate particulate OM- induced physical protection (Besnard et al., 1996)." (lines 64-65)

L. 67: *"how Andosol development"* What does the authors mean by this with the above-mentioned central object of investigation (land use effect on mineralogy <-> indirect impact on SOC stability)? The neoformation of soil minerals during weathering, change in reactivity of functional groups, etc. This should be defined more precisely.

We agree with the referee that our central point of research is the indirect impact of land use on SOC stability via moderation of geochemical properties. A robust investigation of neoformation of soil minerals or change in reactivity of functional groups across time is out of scope as that would requires analysis of an established chronosequence. Thus, we revised the sentence as follow:

"The co-occurrence of native forest, secondary forest, or agricultural land use forms a useful means to understand how land use would influence geochemical properties and impact SOC storage and stability in volcanic soils." (Lines 72-74)

L. 147: *"The S+A and s+c fractions"*

Revised.

L. 186: “2.5 Statistic” In their response letter, the authors point out that they exclude the native forest site from certain comparisons and tests due to its very different mineralogical properties. The procedure should be described here so that the reader can understand what was tested.

We revised the description of the statistics as follows:

“Two-way ANOVA with depth increment and land-use as fixed factors was used to compare soil properties and the distribution of OC over soil fractions. For individual depth increments (0 – 20, 20 – 40, and ± 60 – 80 cm) additional t-tests were used as well to test for specific differences between pine forest and agricultural land use. Total, ryegrass- and native SOC cumulative OC mineralization and net cumulative priming of native SOC were only assessed for the 0-20cm depth increment and the comparison of means between the pine forest and agriculture land-uses was made by independent samples t-tests. The primary forest was excluded from this comparison as it emerged that the parent material from which it developed differed from the other sites (Anindita et al., 2022). Pearson correlation coefficients were used to investigate relations between soil fraction SOC proportions, soil OC mineralization, and geochemical properties for the group of pine forest and agricultural soils. Analyses were completed in IBM SPSS Statistics 27.” (Lines 196-207)

L. 203: “pine forest” What is the history of the pine forests? It would be a good addition to the description of the study sites so that the reader can get a better understanding of the SOC development.

Secondary pine forest was planted around 1960 for the purpose of reforestation. Along the period 1960 – 2020, for the PF-i, the status of pine forest changed from production forest (local farmers can take pine resin and woods) and unmanaged plantation (with coffee) to protected forest in 2007. At PF-o, for 5 years pine trees were intercropped with horticulture and coffee and this forest has likewise received a protected status.

This history was already mentioned in (Anindita et al., 2022) and is referred to in the footnote of Table 1.

L. 203: I’m really surprised that exactly the same mean values and standard deviation of total SOC were calculated

An apparent mistake in our reported data. We revised SOC content of soil fractions thoroughly: the means total SOC in agricultural and pine forest soils were 29.6 ± 15.1 and 16.3 ± 7.7 , respectively (Lines 218-220)

L. 208-209: “The amount of OC in the S+A fraction ...” If you are talking about the mean values here, please write it like this, ‘..The mean OC content in the S+A fraction ...’ Otherwise, you should be more specific about what soils and depth increments you are writing about.

We meant the content of OC present in that fraction on a bulk soil basis: sentence revised into “The mean OC content in the S+A fraction was higher in the agricultural (11.3 ± 10.3 g C kg⁻¹ soil)...”.

L. 208: “ 11 ± 10 g C kg⁻¹ soil” I cannot derive this OC amount (g/kg soil) from the data presented in Table 2. It doesn’t matter whether I calculate the mean value from all depth increments or only a specific depth increment. Please clarify this. For the forests, the value can be derived by calculating the mean for the top depth increment. This should be compared to the same depth increment of agricultural soils

The value of $11.3 \pm 10.3 \text{ g C kg}^{-1}$ soil was derived by taking the average of AG-y, AG-i, and AG-o across all depths. Likewise, the content SOC in S+A fraction in pine forest ($1.5 \pm 0.6 \text{ g C kg}^{-1}$ soil) was the average of PF-i and PF-o across all depths. We revised the p-values after recalculation using two-way ANOVA.

L.208: " $11 \pm 10 \text{ g C kg}^{-1}$ soil" Fig. 1

Revised

L. 211: " $AG-y (\pm 13 \text{ g C kg}^{-1} \text{ soil})$ " Please use the value given in Table 2 or round the value in Table 2. +
L. 211: " $AG-i (\pm 31 \text{ g C kg}^{-1} \text{ soil})$ " However, this is now the value of the depth increment 20-40 cm and no longer the value of 0-20 cm as in the previous comparison. What is being compared in each case must be structure more clearly.

We revised the sentence as follow:

"Particularly in AG-y at depth 0 – 20 cm (13 g C kg^{-1} soil) and in AG-i at depth 0 – 20 and 20 – 40 cm (22 and 31 g C kg^{-1} soil, respectively), a large share of the SOC was in the S+A fraction" (lines 227-229)

L. 217: "*(Table 2) ~~indicate that some mineral matter~~ was indicate that some mineral matter was ..."*

The sentence was revised as follow:

"The occasional low free POM C-concentrations (Table 2) indicate that some mineral matter was left in the isolated POM fraction." (Lines 233-234)

L. 222-223: Insertion of lines between the different areas in Table 2. Otherwise, the table is difficult to read. Please also include a column with the total SOC content of the soil in the 'amount of C' category → This will be also a quality indicator of your fractionation scheme (recovery rate).

Revised. The bulk soil SOC content of was also added.

L. 227: "*Fig. 1 (0-60 cm)*" Does that mean that the depth increments specified for the figure do not match Table 2?" Please clarify this.

We revised Figure 1's caption as follows:

"Figure 1. The distribution of OC over soil fractions obtained by a modified version of the Zimmerman et al. (2007) procedure for 6 soil profiles sampled at 0 – 20, 20 – 40, ± 60 – 80 cm depth in the Sunda Volcanic complex in West Java, Indonesia. Error bars indicate standard deviations for three lab-replicates (n = 3)"

L. 231: "*6 - 20 in S+A and s+c fractions*" Please use the values given in your table or adjust them. It's a little bit confusing to have these differences in the tables and the text.

The section 3.2 has been revised (lines 249-256). In the revised text we used the value as mentioned in Table 3.

L. 233: "*9.6*" Here you give the exact value and did not round it.

Revised

L. 236: “*pine forest (20 mg g⁻¹)*” The authors should also state the mean \pm the standard deviation here, since they have done so before.

The sentence was revised as follows:

“The mean Al_o content in S+A fraction was generally higher under agriculture (28.9 \pm 20.8 g kg⁻¹) than pine forest (20.9 \pm 14.6 g kg⁻¹) (though only at p = 0.09).” (Lines 254-255)

L. 236-237: “*The s+c fraction Fe_o content was higher ...*” the authors should be precise about what is being compared, whether the mean values across all depth increment or defined depth increments among each other. Otherwise, it is difficult to understand.

The depth has been specified and the sentence has been revised as follow:

“The Fe_o contents in S+A and s+c fraction was generally higher under pine forest than agricultural soils, particularly at depth 20 – 40 and \pm 60 – 80 cm, with no significant difference.” (lines 255-257).

L. 252-253: “*Figure 2*” Why is the number of data points in the top right figure different? Please include the number of repetitions in the figure caption. Corresponding abbreviations in the different figure (e.g. ‘a’, ‘b’, ...) would also be useful to guide the reader through the overall figure.

Figure 2 has been revised after recalculation, the proportion of OC in S+A fraction correlated only with Al_o content (in bulk soil and the S+A fraction) and the content of amorphous materials. The number of samples (n) per depth is 5. Accordingly, section 3.3 was also revised (lines 253-275):

“The proportion of several of the isolated SOC fractions correlated to some of the geochemical soil properties (Fig. 2). By considering only pine forest and agricultural soils, the SOC proportion of the S+A fraction was positively correlated with Al_o (in bulk soil as well as in the S+A fraction) and content of amorphous materials, particularly at depth of \pm 60 – 80 cm (p < 0.05), and C:N ratio of S+A fraction at all depths (p < 0.05). These soil properties were also positively correlated with the amount of OC in S+A fraction (g C kg⁻¹ soil) and OC concentration in S+A fraction (g C kg⁻¹ fraction). Positive correlations existed between SOC proportion in s+c fraction with the amount of Fe_o s+c fraction at depth of \pm 60 – 80 cm (p < 0.05), but the amount of OC (g C kg⁻¹ soil) and OC concentration (g C kg⁻¹ fraction) in s+c fraction correlated negatively to Fe_o s+c fraction at all depths. The proportion of rSOC in s+c fraction (i.e. rSOC / s+c \times 100) correlated positively (p < 0.05) to Al_o content at depth 60 – 80 cm (r = 0.9), SSA at 20 – 40 cm (r = 0.9), and the micropores volume at 0 – 20 and 20 – 40 cm (r = 0.66 and r = 0.86, respectively).”

L. 262: “*Pine forest soils displayed higher cumulative C mineralization than agricultural soils (p < 0.05)*” Author should also indicate whether there are significant differences between the soils of similar land use

The research was focus on the comparison between agricultural and pine forest land use. Since we did not have field replicates (we do have lab replicates) for each site (so n per site across depth = 1), we could not do the required statistical tests to explore variation between sites under similar land use.

L. 286: “*A larger % of SOC*” What does this % mean?

The “%” in the sentence “refers to the relative proportion of native SOC that was mineralized. We rephrased this: “A larger relative share of SOC was mineralized in the pine forest soils (2.9 ± 0.1 % of SOC) than in the agricultural soils (1.2 ± 0.3 % of SOC) ($p < 0.01$).” (lines 313-314)

L. 311-313: “We expected a lesser share of SOC to be occluded ...” However, the mean values given for all agricultural soils in comparison with the forests point in a different direction. So, you assume that there is a direct surface interaction of OM with the reactive minerals contained in the S+A fraction? Please clarify.

No, with “We expected a lesser share of SOC to be occluded inside aggregates in the tilled agricultural soils, as absence of tillage results in less disruption of macroaggregates with increased formation of microaggregates and occlusion of POM (Six et al., 2000b; Zheng et al., 2018)” we were referring to that tillage is well known to induce breakdown of aggregates – speeding up macroaggregate turnover. Consequently, less POM becomes occluded within aggregates. In this study we found more stable aggregates in the tilled agricultural soils than in the pine forest soils. In the revised discussion on this part, we considered causes for this promoted aggregation and apparent larger proportion of S+A, i.e. aggregate occluded SOC under agricultural land-use (please see our response above to the major referee comment).

L. 356: “~~Thus~~ association” should be ‘A’

Revised

L. 359-362: Why shouldn’t that be an indication of a causal relationship? The pH value influences the variable charge carriers in the soil and their content represents the basis of reaction partners. Similar mechanisms can work inside aggregates (previous paragraph).

Regarding the negative relation between the proportion of SOC and s+c fraction and soil pH, after correction of our calculations, their correlation now proved to be insignificant. Thus, we removed the description about this relationship from section 4.2.

L. 363-365: “But, again as s+c SOC content did not differ between the agricultural and the forest sites... unlikely.” Maybe a closer look at the individual soils under both land uses would be useful in the course of the paragraph.

Due to the limited samples and no field replicates for each site, we could not run the required statistical analysis to compare individual sites and thus prefer to focus on found differences between both land-uses.

L. 369-371: “These correlations suggest that the oxidizable (Mikutta et al., 2006).” The data shows a really great connection between the mineralogy and the associated SOC. Furthermore, the data show that in soils with high content of variable charge carriers, the surface interactions are strongly dependent on the pH value. When oxidizing with NaOCl, the pH is raised to 8. At this point, the different soils with their geochemical properties should be discussed in detail again.

See our answer above, when correcting our statistical, we now no longer see this trend between pH and SOC in the s+c fraction and share of that is oxidizable by 6%NaOCl. We can follow the referee's line of thought: in a soil with more Fe_o or Al_o relatively more SOC could be 'bound' to Al-bearing sesquioxides, and in such soils a larger share of the colloid surface charge is pH-dependent. If the pH is lifted during 6% NaOCl oxidation to 8.5, the net surface charge of these sesquioxides becomes less positive (or more negative) and mineral associated SOC could be released, and oxidized. If so, this 6%NaOCl treatment fractionation step then gives a false image of the degree of how organo-mineral-association vs. other mechanisms play a role in SOC stabilization. Such artefact would gain importance in the soils with more pedogenic Fe and Al. While a good point to take along, we prefer to not elaborate on this in the current paper as no relation existed between the rSOC resistance of the s+c fraction SOC to 6%NaOCl oxidation and measured SOC mineralization.

L. 439-441: *"Our study thus indicates ... compared to forest land use."* This was not derived in the work.

Indeed, we stand corrected and changed it into *"Our study thus indicates the larger abundance of Al_o in the agricultural soils promoted soil aggregation and physical occlusion of OC compared to pine forest."* (lines 545-547)

L. 445-447: *"Based on the present study ... compared to primary and secondary forest."* This cannot be derived in this way, since no time series was actually examined. Possible differences in e.g. redox/oxidation cycles, small differences in parent material, differences in acidification, etc. may have previously influenced the soil.

Yes, we agree and added *"Moreover, confirmation that agricultural land-use promotes Al_o formation vs. forest will require time series analysis of such sites."* (lines 556-557)

Comments from the editor:

Thank you very much for your revision and particularly for the additional analyses you carried out. That improved the database of the paper quite a lot. Although the manuscript is much better and clearer there are still some issues, which needs attention during a next (hopefully final) round of revision. It is important to recognize that the data might not solely show a clear effect of changes in mineralogy (Al_{ox}) on aggregation and organic matter (OM) occluded within aggregates. Aggregates are built by minerals with OM sorbed or occluded. I really miss a discussion how minerals and mineral-associated OM (MAOM; and the minerals) might affect aggregation and therefore an apparent protection of OM occluded in such aggregates. What do you think about a continuum between MAOM and OM in aggregates? The small differences in the C/N ratios between the S + A and the S + C fraction would support such an interpretation. Effects of changes in mineral surfaces and charges might be useful to include in this discussion. Please consider the very helpful comments of the reviewer for this discussion.

We fully agree with your analysis – please see our response to the referee general comment nr. 3

What I see critically in the established discussion threads of the work is the strict separation between aggregation and direct interaction between reactive mineral surfaces...”

As indicated in our response we now in more detail discuss binding agents responsible for soil aggregation (in 4.3) and we added a new section 4.5 in which we critically discuss the (lack of) contrast between separated s+c and S+A fractions.

Furthermore, the authors should clearly differentiate between micro- and macroaggregates if possible. There is a big difference in their formation and in the C stabilizing effects of micro- and macroaggregates. Sometimes the reader might get the impression that different aggregate size fractions were analysed.

As the reviewer, I struggle a little bit with direct and indirect effects of land use changes on SOC because it is impossible to disentangle them. It is more about OM input, management and weathering with interactions between these factors. As the reviewer, I think that soil mineral phases and their reactivity are the central parts of the study.

Indeed, we recognize that direct and indirect effects cannot be deconvoluted with the limited set of sites available. Such was also recognized in discussion part 4.3. In response to some of the referee comment we also further recognize this now in the conclusion section.

A detailed comment to your sampling: How many subsamples did you use for a composite sample? Did you sample different plots?

For the sampling, we used three sub-samples for a composite sample and did not sample different plots per site (so various sites per land-use = field replicate). As responded to already in our response to likewise referee comment on sampling and statistics, we have revised the statistics description as well as recalculated the p-values of some of the erroneously run t-tests and amended the results and discussion sections 3.1, 3.2, 3.3, 4.1 and 4.2. We once more apologize for some mistakes made in the previously submitted version.

Please consider all of the very helpful comments of the reviewer (particularly in the annotated manuscript)

We deeply thank the referee and editor for his/her very detailed and well-founded comment to our study and hope our revised manuscript can meet standards for publication in SOIL.