

## Point-by-point response to anonymous Referee#1 comments

Dear Referee#1,

We would like to thank you for your time and thorough evaluation of our manuscript “*The role of geochemistry in organic carbon stabilization in tropical rainforest soils*”, (<https://doi.org/10.5194/soil-2020-92>). We are very pleased that you positively assessed our work and recognized its relevance. Your comments helped us to significantly improve our manuscript and we want to sincerely thank you for the constructive and valuable insights.

We have addressed all comments and suggestions to the best of our ability. Please find below a point-by-point response to all the concerns raised and how we addressed them. Reviewer original comments are highlighted in grey. New text to be added or modified in the manuscript has quotation marks and is blue-colored in the response.

We hope you find our response and changes to the manuscript satisfying and we are looking forward to hearing from you.

Yours sincerely,

The authors

*REVIEWER#1 COMMENT 1: “The authors use the term “organic carbon stabilization” in the title and in many other parts of the manuscript. It could be stabilization against decomposition, temperature, erosion, dispersion, oxidation, or all these together. However, the authors do not provide a context in which the word “stability” is used.”*

**Our response:** We agree with the reviewer that the term “organic carbon stabilization” needs more context. In our manuscript, we use this term as “carbon stabilization against microbial decomposition” Our study and the project it is implemented in aims to understand soil microbial activity, C cycling and C stabilization against decomposition in tropical soils and their feedback between geology, geomorphology and pedogenesis (Doetterl et al. 2021a; 2021b). In this context, we focus on C stabilization against microbial decomposition in the manuscript. We have accordingly changed this term throughout the manuscript where appropriate and also in the title:

“The role of geochemistry in organic carbon stabilization **against microbial decomposition** in tropical rainforest soils.”

*Used literature:*

Doetterl, S., Asifiwe, R.K., Baert, G., Bamba, F., Bauters, M., Bukombe, B., Cadisch, G., Cizungu, L., Cooper, M., Hoyt, A., Kabaske, C., Kalbitz, K., Kidinda, K.L., Maier, A., Mainka, M., Mayrock, J., Muhindo, D., Mujinya, B., Mukotanyi, S.M., Nabahungu, L., Reichenbach, M., Rewald, B., Six, J., Stegmann, A., Summerauer, L., Unseld, R., van Oost, K., Verheyen, K., Vogel, C., Wilken, F., Fiener, P. Organic matter cycling along geochemical, geomorphic and disturbance gradients in forests and cropland of the African Tropics - Project TropSOC DATABASE\_v1.0, Earth System Science Data DISCUSSIONS (pre-print), <https://doi.org/10.5194/essd-2021-73>, 2021a.

Doetterl, S.; Bukombe, B.; Cooper, M.; Kidinda, L.; Muhindo, D.; Reichenbach, M.; Stegmann, A.; Summerauer, L.; 36 Wilken, F.; Fiener, P. TropSOC Database. Version 1.0. GFZ Data Services. <https://doi.org/10.5880/fidgeo.2021.009>, 37, 2021b.

*REVIEWER#1 COMMENT 2: “In the introduction (section 1.2 Environmental and geochemical controls on SOC dynamics in tropical forests), you never talk about very important environmental*

*controls such as temperature, moisture, pH, redox potential, and oxygen diffusion. You should explore the optimum conditions (e.g., temperature and moisture) for enzymatic activity in the tropics, which ultimately will determine organic matter decomposition rates”*

**Our response:** The authors agree that it is necessary to highlight the importance of environmental conditions on enzymatic activity in the tropics, since it is an important component in C cycling. We therefore changed the paragraph accordingly:

“Climatic factors such as temperature and precipitation are also strong drivers of soil environmental conditions, which can greatly influence soil microbial activity and hence C mineralization and turnover (Davidson and Janssens, 2006; Zhang et al., 2011; Feng et al., 2017). For example, decomposition rates increase in general with temperature, but soil microbial communities adapted to high temperatures are less sensitive to warming (Blagodatskaya et al., 2016). Microorganisms in humid tropical soils with varying soil moisture content are exposed to fluctuating redox potentials but remain active (DeAngelis et al., 2010). C-depleted tropical subsoils contain small but metabolically active microbial communities contributing to C cycling (Kidinda et al., 2020; Stone et al., 2014). Low soil pH in combination with high clay content dominated by pedogenic oxides show a high potential to stabilize enzymes on mineral surfaces which will affect microbial C acquisition (Dove et al., 2020; Allison and Vitousek, 2005; Liu et al., 2020). These climate-driven factors also influence SOC dynamics but more indirectly through the interaction with soil geochemical factors (Doetterl et al., 2015).”

*Used Literature:*

Allison, S.D., Vitousek, P. M.: Responses of extracellular enzymes to simple and complex nutrient inputs, *Soil Biol. Biochem.*, 37, 937–944, <https://doi.org/10.1016/j.soilbio.2004.09.014>, 2005.

Blagodatskaya, E., Blagodatsky, S., Khomyakov, N., Myachina, O., and Kuzyakov, Y.: Temperature sensitivity and enzymatic mechanisms of soil organic matter decomposition along an altitudinal gradient on Mount Kilimanjaro, *Scientific Reports*, 6, 10.1038/srep22240, 1-11, 2016.

Davidson, E. A., and Janssen, I. A.: Temperature sensitivity of soil carbon decomposition and feedbacks to Climate Change, *Nature*, 440, 165-173, <https://doi.org/10.1038/nature04514>, 2016.

DeAngelis, K. M., Silver, W. L., Thompson, A. W., and Firestone, M. K.: Microbial communities acclimate to recurring changes in soil redox potential status, *Environ. Microbio.*, 12, 3137-3149, doi:10.1111/j.1462-2920.2010.02286.x, 2010.

Doetterl, S., Stevens, A., Six, J., Merckx, R., van Oost, K., Pinto, M. C., Casanova-Katny, A., Muñoz, C., Boudin, M., Venegas, E. Z., and Boeckx, P.: Soil carbon storage controlled by interactions between geochemistry and climate, *Nat. Geosci.*, 8, 780–783, <https://doi.org/10.1038/ngeo2516>, 2015.

Dove, N. C., Arogyaswamy, K., Billings, S. A. Botthoff, J. K. Carey, C. J. Caitlin, C., DeForest, J. L., Dawson, F., Fierer, N., Gallery, R. E., Kaye, J. P., Lohse, K. A., Maltz, M. R., Mayorga, E., Pett-Ridge, J., Yang, W. H., Hart, S. C., Aronson, E. L.: Continental-scale patterns of extracellular enzyme activity in the subsoil: an overlooked reservoir of microbial activity, *Environ. Res. Lett.*, 15, 1040a1, <https://doi.org/10.1088/1748-9326/abb0b3>, 2020.

Feng, W., Liang, J., Hale, L. E., Jung, C. G., Chen, J., Zhou, J., Xu, M., Yuan, M., Wu, L., Bracho, R., Pegoraro, E., Schuur, E. A. G., and Luo, Y.: Enhanced decomposition of stable soil organic carbon and microbial catabolic potentials by long-term field warming, *Global Change Biol.*, 23, 4765–4776, <https://doi.org/10.1111/gcb.13755>, 2017.

Kidinda, L. K., Olagoke, F. K., Vogel, C., Kalbitz, K., and Doetterl, S.: Patterns of microbial processes shaped by parent material and soil depth in tropical rainforest soils, *SOIL DISCUSSIONS*, in review, <https://doi.org/10.5194/soil-2020-80>, 2020.

Liu, J., Chen, J., Chen, G., Guo, J., Li, Y.: Enzyme stoichiometry indicates the variation of microbial nutrient requirements at different soil depths in subtropical forests, PLoS ONE 15, e0220599, <https://doi.org/10.1371/journal.pone.0220599>, 2020.

Stone, M. M., DeForest, J. L., and Plante, A. F.: Changes in extracellular enzyme activity and microbial community structure with soil depth at the Luquillo Critical Zone Observatory, Soil Biology & Biochemistry, 75, 237-247, <http://dx.doi.org/10.1016/j.soilbio.2014.04.017>, 2014.

Zhang, L., Zeng, G., and Tong, C.: A review on the effects of biogenic elements and biological factors on wetland soil carbon mineralization, Acta Ecol. Sin., 31, 5387-5395, 2011.

*REVIEWER#1 COMMENT 3: “It is not clear in your hypothesis ii, the proxy for the amount of “stabilized” SOC. Is it the carbon content adsorbed on clay minerals? Does it also consider the carbon occluded in microaggregates that are physically stable against dispersion?”*

**Our response:** We thank the reviewer for this comment. The proxy for the stabilized SOC is the amount of mineral-associated carbon adsorbed on clay minerals and pedogenic oxides (C mass of the free silt and clay fraction) and the carbon occluded in the microaggregates (C mass of the microaggregate fraction). Since the amount of unprotected carbon (coarse particulate organic matter fraction) is negligible, this manuscript focuses on the carbon associated with microaggregates and the free silt and clay fraction. Note that in that sense we consider the small amount of particulate organic matter in aggregates also to be protected by minerals. We have now changed the first sentence of hypothesis (ii) for better clarification:

“Chemical variability in parent material will result in contrasting geochemical soil properties that affect the formation of C stabilization mechanisms **against microbial decomposition** (organo-mineral associations and soil aggregates) and hence govern pattern of SOC stocks and stability **as a function of C associated with stable microaggregates and the free silt and clay fraction (mineral-associated C).**”

*REVIEWER#1 COMMENT 4 “How many soil profiles were sampled? What is the classification of these soil profiles that you sampled? It needs to be shown in the methods section “2.2 Study design and soil sampling.”*

**Our response:** The reviewer is right that this important information is indeed missing in the manuscript. The following paragraph has thus been amended to section “2.2 Study design and soil sampling”:

“**In total, 36 soil cores were sampled (12 soil cores per geochemical region across four topographic positions in triplicate) on which the soil analysis was conducted. In addition, one soil pit down to at least 100 cm per topographic position was dug in the center in one of the three replicate plots in each region and described according to FAO guidelines (FAO, 2006). The soils were classified after World Reference Base (WRB) soil classification (IUSS WRB, 2015). Soils in the mafic region can be described as umbric, vetic and geric ferralsols and ferralic vetic Nitisols. Soils in the felsic regions are classified as geric and vetic Ferralols. The mixed sedimentary region shows geric and vetic Ferralols along plateaus and slopes, whereas soils at the valley bottoms are described as fluvic Gleysols.**”

*Used Literature:*

FAO: Guidelines for soil description, 4th edition, 4th ed., FAO, Rome. Available from: 950 <http://www.fao.org/3/a-a0541e.pdf>, 2006.

FAO: World references base for soil resources 2014. International soil classification system for 953 naming soils and creating legends for soil maps. Update 2015. Food and Agriculture Organization 954 of the United Nations, Rome, Italy, 203 pp., 2014.

*REVIEWER#1 COMMENT 5: “L22: I think you meant “changes in hydraulic conditions.”*

**Our response:** The authors thanks for pointing this out to us. In the context of our study, the term “changes in hydraulic conditions” is the better choice. We explained the terms “fluvial and hydraulic conditions” in more detail in the context of our study sites. The sentences have now been corrected and amended accordingly:

“Instead, fluvial dynamics and changes in soil moisture conditions had a secondary control on SOC dynamics in valley positions. Valley bottoms might be affected not only by sediments derived from associated hillslopes but also from material redistribution in the entire catchment during flood events (Douglas and Guyot, 2005). In addition, soil moisture conditions at the valley bottoms might be affected not only from interflow from the hillslopes but also from temporarily high ground water levels (Bonell, 2005). These fluvial and hydraulic conditions lead to higher SOC stocks there than at the non-valley positions.”

*Used Literature:*

Bonell, M.: Runoff generation in tropical forests, in: Forests, Water and People in the Humid Tropics, edited by: Bonell, M., and Bruijnzeel, L. A., Cambridge University Press, New York, USA, 314-406, 9780521829533, 2005.

Douglas, I., and Guyot, J. L.: Erosion and sediment yield in the humid tropics, in: Forests, Water and People in the Humid Tropics, edited by: Bonell, M., and Bruijnzeel, L. A., Cambridge University Press, New York, USA, 407-421, 9780521829533, 2005.

*REVIEWER#1 COMMENT 6: “L23/24: What is “fossil organic carbon (FOC)”? Is it defined by a specific organic compound (e.g., polycyclic aromatic hydrocarbon) or by a certain “age” (e.g., >10000 years)? The authors should define “fossil organic carbon” when they first mentioned it in the abstract.”*

**Our response:** This is an important comment. Fossil organic carbon (FOC) is of geogenic origin. It is organic carbon deposited during sedimentation and undergoes coalification or kerogen transformation during diagenesis (Buseck and Beysacc, 2014). In our study, FOC is characterized by high C / N ratios, depleted in N and free of <sup>14</sup>C due to the high age of rock formation. We have now defined the term FOC more precisely in the according sentence in the abstract:

“At several sites we also detected fossil organic carbon (FOC), which is organic C of geogenic origin and is characterized by high C / N ratios, depletion of N and free of <sup>14</sup>C. Here, FOC constitutes up to 52.0 ± 13.2 % of total SOC stock in the C depleted subsoil.”

*Used Literature:*

Buseck, P. R. and Beysacc, O.: From organic matter to graphite: Graphitization, Elements, 10, 421–426, <https://doi.org/10.2113/gselements.10.6.421>, 2014.

*REVIEWER#1 COMMENT 7: “L29: What depth interval are you considering for this observation about SOC stocks? 0-20 cm, 0-50 cm, 0-70 cm, 0-100 cm?”*

**Our response:** The reviewer addresses an important point regarding the missing information about the considered depth intervals. The regression models considered three depth intervals for all dependent variables: 0 - 10 cm (topsoil), 30 - 40 cm (shallow subsoil) and 60 - 70 cm (deep subsoil). These depth intervals were selected as they differ in C input and biogeochemical soil factors. Even though the full soil core down to 100 cm was sampled in 10 cm depth increments, only these three selected depth intervals were analyzed in the laboratory to keep the workload at a feasible level. We added this information in the according sentence in the abstract:

“Regressions **models, considering depth intervals of 0 - 10 cm, 30 - 40 cm and 60 - 70 cm**, showed that variables affiliated with soil weathering, parent material geochemistry and soil fertility, together with soil depth, explained up to 75 % of the variability of SOC stocks and  $\Delta^{14}\text{C}$ .”

*REVIEWER#1 COMMENT 8: “L66: What do you mean for “stabilize C in the soil”? stabilize against decomposition? For how long? 100, 1000, 10000 years?”*

**Our response:** The authors agree that the term “C stabilization” should be used with more precision. The sentence was changed to give more information to the reader in following way:

“The accessibility of C for mineralization, however, is predominantly driven by several interacting mechanisms that can stabilize C in soils **against decomposition on a decadal up to a millennial time scale** (Trumbore, 2000; Trumbore, 2009).”

*Used Literature:*

Trumbore, S.: Age of soil organic matter and soil respiration: radiocarbon constraints on belowground C dynamics, *Ecol. Appl.*, 10, 399-411, [https://doi.org/10.1890/1051-0761\(2000\)010\[0399:AOSOMA\]2.0.CO;2](https://doi.org/10.1890/1051-0761(2000)010[0399:AOSOMA]2.0.CO;2), 2000.

Trumbore, S.: Radiocarbon and soil carbon dynamics, *Annu. Rev. Earth Planet. Sci.*, 37, 47-66, [10.1146/annurev.earth.36.031207.124300](https://doi.org/10.1146/annurev.earth.36.031207.124300), 2009.

*REVIEWER#1 COMMENT 9: “L66-68: Please, provide the reasoning for why pyrogenic carbon may remain in the soil for centuries. Does it have to do with the following? The degradation of condensed aromatic carbon compounds is an energy-demanding process. For degradation to occur, the biological capacity for specific degradation pathways must exist in the soil [Baldock et al., 2004]. It is likely that certain microorganisms can produce the (costly) enzymes required for pyrogenic carbon degradation and their presence or absence from soils is thus a crucial control on pyrogenic carbon accumulation. However, images of fungal hyphae encasing charcoal particles in soils [Hockaday et al., 2007] indicate that fungi can very well decompose pyrogenic carbon.”*

**Our response:** The authors thank the reviewer for suggesting to be more specific about the controls of pyrogenic carbon persistence. The provided literature already points out that pyrogenic carbon cannot simply be classified as an inert C pool but is in contrast dynamic and interacts with soil organisms and the mineral matrix. We therefore changed the sentence in following way:

“For example, certain C compounds such as pyrogenic C **show biochemical resistance since the decomposition of its complex molecular structure is an energy demanding process and microbes will preferentially consume more easily available organic C forms** (Czimczik and Masiello, 2007; Knicker, 2011).”

*Used Literature:*

Czimczik, C. I., and Masiello, C. A.: Controls on black carbon storage in soils, *Global Biogeochem. Cycles*, 21, 1-8, 10.1029/2006GB002798, 2007.

Knicker, H.: Pyrogenic organic matter in soil: its origin and occurrence, its chemistry and survival in soil environments, *Quat. Int.*, 243, 251-263, 10.1016/j.quaint.2011.02.037, 2011.

*REVIEWER#1 COMMENT 10: L69: Are you sure that the word “recalcitrant” is the best one to describe what you meant here? Maybe using terms like “long turnover times” may help you to overcome this issue. I highly recommend the authors to read the following publications for further ideas and suggestions:*

*Mikutta, R., Kleber, M., Torn, M.S. and Jahn, R., 2006. Stabilization of soil organic matter: association with minerals or chemical recalcitrance?. Biogeochemistry, 77(1), pp.25-56.*

*Kleber, M., Nico, P.S., Plante, A., Filley, T., Kramer, M., Swanston, C. and Sollins, P., 2011. Old and stable soil organic matter is not necessarily chemically recalcitrant: implications for modeling concepts and temperature sensitivity. Global Change Biol., 17(2), pp.1097-1107.”*

**Our response:** We agree with the reviewer that the term “recalcitrant” in the context with fossil organic carbon is misleading. Our study, together with other recent studies, show that fossil organic carbon is dynamic and decomposable (Bukombe et al., 2021; Hemmingway et al., 2018). Hence, the authors have agreed to use the term “long turnover times” instead:

“Another C fraction that is **characterized by long turnover times** is fossil organic carbon (FOC) (...)”

*Used Literature:*

Bukombe, B., Fiener, P., Hoyt, A. M., Doetterl, S.: Controls on heterotrophic soil respiration and carbon cycling in geochemically distinct Afrcian tropical forest soils, *SOIL DISCUSSIONS*, in review, <https://doi.org/10.5194/soil-2020-96>, 2021.

Hemmingway, J. D., Hilton, R. G., Hovius, N., Eglinton, T. I., Haghypour, N., Wacker, L., Chen, M.-C., and Galy, V. V.: Microbial oxidation of lithospheric organic carbon in rapidly eroding tropical mountain soils, *Science*, 360, 209–212, <https://doi.org/10.1126/science.aao6463>, 2018.

*REVIEWER#1 COMMENT 11: “L116: Define “geogenic carbon”? Is it the same as “fossil carbon”?”*

**Our response:** The reviewer is right in recognizing this inconsistency. It is meant to be “fossil organic carbon”. We have now changed this through the manuscript to the latter.

*REVIEWER#1 COMMENT 12: “L117: You said “more active microbial communities”. However, you didn’t conclude the comparison. Is it more active than what or where?”*

**Our response:** Thank you for pointing this out. We will conclude the comparison in the following way:

“Similarly, **fossil organic carbon** that is brought to the surface might become increasingly decomposed when brought in contact with more active and abundant topsoil microbial communities **compared to subsoil environments.**”

*REVIEWER#1 COMMENT 13: “L140: Check the concordance of the preposition “on” and the verb “affect” in this sentence: “our current understanding on how geochemistry and topography in highly weathered tropical soils affects SOC stocks and stabilization mechanisms is still limited.”*

**Our response:** Thanks for this comment. The preposition will be corrected in following way:

“In summary, our current understanding of how geochemistry and topography in highly weathered tropical soils affects SOC stocks and stabilization mechanisms against microbial decomposition is still limited.”

*REVIEWER#1 COMMENT 14: “L225: What method did you use for “texture” determination? Hydrometer, pipette?”*

**Our response:** We used the Bouyoucos hydrometer method (Bouyoucos, 1962) modified following Beretta et al. (2014). To keep the “2.3 Soil analysis” section streamlined, we decided to focus on the SOC fractionation and pedogenic extraction scheme. A more detailed description of all other soil analysis listed in the manuscript can be found in the project-related database publication (Doetterl et al., 2021a; 2021b). However, if the reviewer thinks that all used methods should be described in detail, we will expand the according section.

*Used literature:*

Beretta, A. N., Silbermann, A. V., Paladino, L., Torres, D., Bassahun, D., Mussell, R., and García-Lamohte, A.: Soil texture analysis using a hydrometer: modification of the Bouyoucos method, *Cien. Inv. Agr.*, 42, 263-271, 10.4067/S0718-16202014000200013, 2014.

Bouyoucos, G. J.: Hydrometer method improved for making particle size analyses of soils, *Agronomy Journal*, 54, 464-465, 10.2134/agronj1962.00021962005400050028x, 1962.

Doetterl, S., Asifiwe, R.K., Baert, G., Bamba, F., Bauters, M., Bukombe, B., Cadisch, G., Cizungu, L., Cooper, M., Hoyt, A., Kabaske, C., Kalbitz, K., Kidinda, K.L., Maier, A., Mainka, M., Mayrock, J., Muhindo, D., Mujinya, B., Mukotanyi, S.M., Nabahungu, L., Reichenbach, M., Rewald, B., Six, J., Stegmann, A., Summerauer, L., Unseld, R., van Oost, K., Verheyen, K., Vogel, C., Wilken, F., Fiener, P. Organic matter cycling along geochemical, geomorphic and disturbance gradients in forests and cropland of the African Tropics - Project TropSOC DATABASE\_v1.0, Earth System Science Data DISCUSSIONS (pre-print), <https://doi.org/10.5194/essd-2021-73>, 2021a.

Doetterl, S.; Bukombe, B.; Cooper, M.; Kidinda, L.; Muhindo, D.; Reichenbach, M.; Stegmann, A.; Summerauer, L.; 36 Wilken, F.; Fiener, P. TropSOC Database. Version 1.0. GFZ Data Services. <https://doi.org/10.5880/fidgeo.2021.009>, 37, 2021b.

*REVIEWER#1 COMMENT 15: “L248: Did you subtract the rock fragment to calculate SOC stocks?”*

**Our response:** The samples were free of rock fragments or the amount was negligible (all soils were deeply weathered). We added this information to the section “Soil C fractionation and nutrient analysis”:

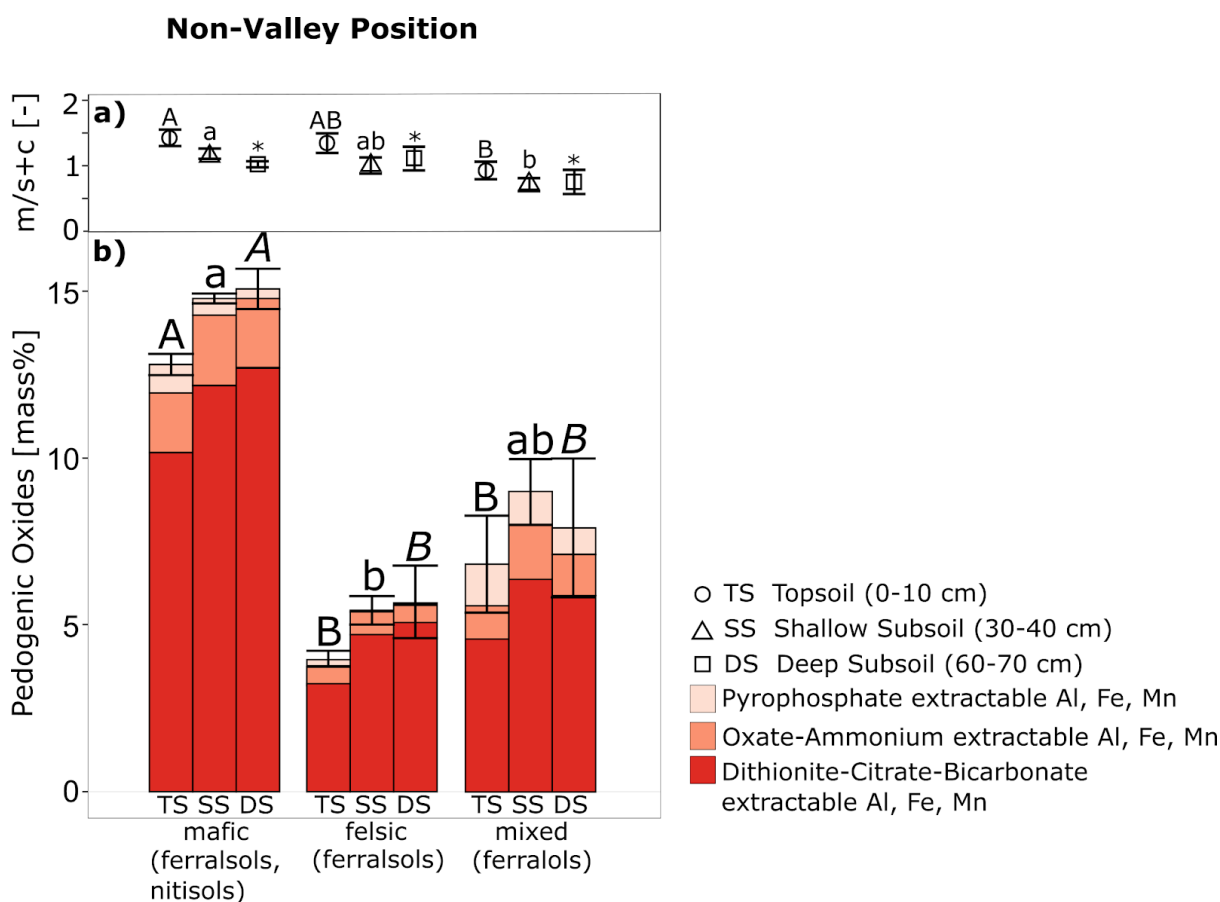
“Since the content of rock fragments of all samples were negligible, the SOC stock of the bulk soil (SOC<sub>bulk</sub>) was calculated by just multiplying the SOC concentration with the bulk density and thickness of the depth increment (10 cm).”

REVIEWER#1 COMMENT 16: “L329: Please, check if this statement is necessary: “For non-valley positions, pyrophosphate extractable oxides (0.02 to 1.93 mass%) and oxalate extractable oxides (0.32 to 2.33 mass%) were low compared to DCB extractable oxides”. Assuming that DCB extracts most of the oxides (including oxalate and pyrophosphate extractable oxides), then, DCB extractable oxides will always be the highest regardless of the landscape position.”

**Our response:** Thanks for this comment. The statement is still valid, since we used a sequential pedogenic oxide extraction scheme. In the first step, sodium-pyrophosphate was used to extract organically complexed metals. In the second step, ammonium-oxalate-oxalic acid was used on the same sample to extract short-range order oxides and in the last step, DCB was used again on the same sample to extract crystalline oxides. Therefore, the DCB extract will not contain any pyrophosphate or oxalate extractable oxides. We presented the results of all three oxide fractions separately, since we used each of them in the rotated principal component analysis, in which they loaded on different components.

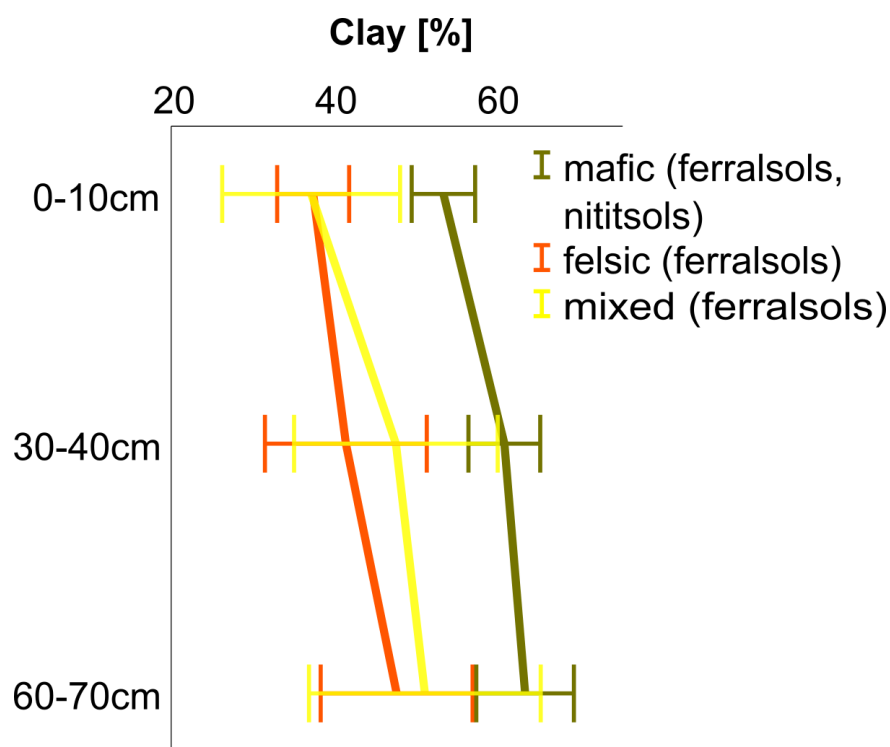
REVIEWER#1 COMMENT 17: “Figure 3. It would be helpful if we know the soil types. Are they all ferralsols? Or the soils derived from “mafic” parent material are lixisol and nitisol as well? Also, it would be nice to see in another figure and table the average clay content as a function of soil depth in each group: mafic, felsic, mixed.”

**Our response:** Thank you for your suggestion. We have now added the soil types based on our soil profile descriptions for each region in Figure 3. We also followed the advice of the reviewer to plot the clay content against soil depth for each group with an additional table for reviewers and editors information (Figure R1, Table R1). It shows that the clay content increases with soil depth and is highest in mafic soils. To be consistent, we would also add the soil types to Figure 4, Figure S1 and Figure S2 in the same way.





**Figure 3:** (a) m / s+c ratio (n = 9 per soil depth) and (b) pedogenic oxide fractions (n = 3 per soil depth) of the sequential extraction across geochemical regions in non-valley positions. Letters indicate significant differences between geochemical regions per soil depth for m / s+c ratio (a) and total pedogenic oxide mass (b). Asterisks indicate no significant differences in means (p > 0.05). Error bar represents standard error.



**Figure R1:** Mean clay content as a function of soil depth across geochemical regions in non-valley positions (n = 9). Error bar represents standard deviation.

**Table R1:** Mean and standard deviation of clay content (%) across geochemical regions in non-valley positions (n = 9).

depth increment	mafic (ferralsols, nitisol)	felsic (ferralsols)	mixed (ferralsols)
0 - 10 cm	53.51±3.9	37.47±4.43	37.23±10.95
30 - 40 cm	61±4.42	41.5±9.95	47.66±12.53
60 - 70 cm	63.56±6	47.75±9.35	51.22±14.26

*REVIEWER#1 COMMENT 18: "L539: Iron concretions are found, but I am not sure about aluminon concretions. Check whether all these soils contain Fe-concretion or nodules because these concretions are not always found even in microaggregates."*

**Our response:** Thanks for this important comment. We addressed this topic in the discussion as followed:

“Fe-oxides like hematite can incorporate large amounts of Al in their crystal structure by substitution, especially within a kaolinite-rich soil matrix (Tardy and Nahon, 1985). Such (hydro)oxides of Al and Fe act as a cementing agent in the formation of pseudosands and their chemical composition is controlled by parent material geochemistry. In general, (hydro)oxides dominated by Fe are more abundant on mafic rocks, whereas Al is more abundant on rocks with low amounts of Fe (e.g. quartz-rich sedimentary rocks) (Martinez and Souza, 2020). In our study both, the parent material and soil geochemistry show considerable amounts of Al even though the Fe-content exceeds that of Al. The dominating soils are ferralsols and nitisols, which are dominated by kaolinitic clays. Given the above mentioned observations, it is likely that Fe- and Al-concretions are present in the studied soils, even though the elemental composition of the concretions were not directly measured.”

*Used Literature:*

Martinez, P., and Souza, I. F.: Genesis of pseudo-sand structure in Oxisols from Brazil – a review, *Geoderma Regional*, 22, <https://doi.org/10.1016/j.geodrs.2020.e00292>, 2020.

Tardy, Y., and Nahon, D.: Geochemistry of laterites, stability of Al-goethite, Al-hematite, and Fe<sup>3+</sup>-kaolinite in bauxites and ferricretes: an approach to the mechanism of concretion formation, *Am. J. Sci.*, 285, 865-903, 1985.

*REVIEWER#1 COMMENT 19: “L540: The citation Martinez and Souza, 2019 is actually from 2020. Check it in other parts of the manuscript.”*

**Our response:** The reviewer is right. Thank you for pointing this out. The manuscript was citing the pre-proof version of this publication. The citation is now updated throughout the manuscript.

*REVIEWER#1 COMMENT 20: “L543: Check the following publication to obtain more information about the effects of soil depth and texture on carbon retention in Ferralsols. They may help you with new insights and allow comparisons with your data. Soil depth and texture seem to regulate the content of carbon retained by the mineral matrix in Ferralsols. Make sure you addressed it in the discussion.*

*Souza, I.F., Almeida, L.F.J., Jesus, G.L., Kleber, M. and Silva, I.R., 2017. The mechanisms of organic carbon protection and dynamics of C-saturation in Oxisols vary with particle size distribution. European Journal of Soil Science, 68(5), pp.726-739.*

*Souza, I.F., Almeida, L.F., Jesus, G.L., Pett-Ridge, J., Nico, P.S., Kleber, M. and Silva, I.R., 2018. Carbon sink strength of subsurface horizons in Brazilian oxisols. Soil Sci. Soc. Am. J., 82(1), pp.76-86.”*

**Our response:** We thank the reviewer for the literature suggestion as it provides further insights in our data. To address the effect of soil depth on C retention we conducted a correlation analysis using the rotated components and SOC stocks separately for each depth increment. The result is comparable to Souza et al. (2018), where C stabilization via interaction with the mineral soil matrix is minor in topsoils but becomes more important in subsoils (Table R2). Please note, that Table R2 is for reviewer and editor information only and we would not include it in the manuscript. We addressed the impact of soil depth on C content in the discussion section in the following way:

“On the contrary, the C stabilization with the geochemical predictors is more effective in shallow and deep subsoils compared to topsoils as indicated by insignificant correlations in the topsoils but significant correlation in subsoils (data not shown). This indicates that the capacity to stabilize C is reached due to high C input in contrast to subsoils receiving less C hence less saturated mineral surfaces (Souza et al., 2018).”

**Table R2: Pearson correlation between SOC<sub>bulk</sub> and rotated components for each depth increment.**

Soil depth		$rPC1_{nv}$ - solid phase	$rPC2_{nv}$ - liquid phase	$rPC3_{nv}$ - silt content	$rPC4_{nv}$ - organic-metal complexes
0 - 10 cm	Pearson Correlation	0.366	-0.149	0.09	0.162
	Sig.	0.332	0.701	0.818	0.678
	N	9	9	9	9
30 - 40 cm	Pearson Correlation	0.681*	-0.927**	-0.791*	0.931**
	Sig.	0.043	0	0.011	0
	N	9	9	9	9
60 - 70 cm	Pearson Correlation	0.509	-0.822**	-0.719*	0.816**
	Sig.	0.162	0.007	0.029	0.007
	N	9	9	9	9

\* Correlation is significant at the 0.05

\*\* Correlation is significant at the 0.01

The effect of texture as a function of clay content on the potential to stabilize C (especially in the fraction < 53  $\mu\text{m}$ ) as shown by Souza et al. (2017) could not be detected in our data. No significant correlations were found between clay content, SOC<sub>bulk</sub> or C associated with the free silt and clay fraction (< 53  $\mu\text{m}$ ). This can be probably explained by the very different experiment design. In the study of Souza et al. (2017), the particle size distribution was manipulated to investigate the effect of changing soil texture on the C saturation deficit in tropical soils. They argue that the manipulation of the particle size distribution leads to large C losses. Further, reactive pedogenic oxide surfaces became exposed by the experiment treatment, which were previously involved in stabilizing aggregates. This difference in the experimental design may explain why clay is not that important in our study. We therefore would just add the reference and only slightly adjust the corresponding sentence about clay content:

“Clay content, identified as a major factor for stabilizing SOC in temperate soils (Angst et al., 2018) and also in tropical soil systems (Quesada et al., 2020; Souza et al., 2017) was not identified as a major control for our soils.”

*Used Literature:*

Angst, G., Messinger, J., Greiner, M., Häusler, W., Hertel, D., Kirfel, K., Kögel-Knabner, I., Leuschner, C., Rethemeyer, J., and Mueller, C. W.: Soil organic carbon stocks in topsoil and subsoil controlled by parent material, carbon input in the rhizosphere, and microbial-derived compounds, *Soil Biol. Biochem.*, 122, 19–30, <https://doi.org/10.1016/j.soilbio.2018.03.026>, 2018.

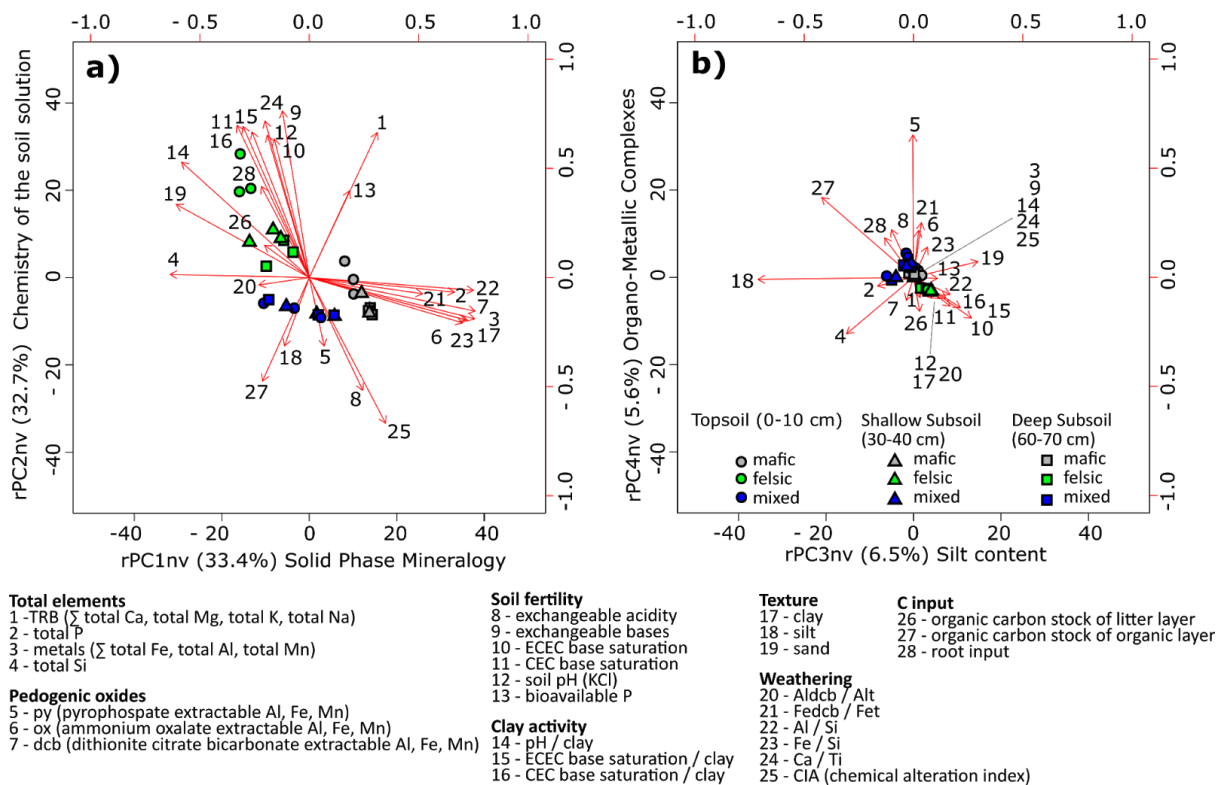
Quesada, C. A., Paz, C., Mendoza, E. O., Phillips, O. L., Saiz, G., and Lloyd, J.: Variations in soil chemical and physical properties explain basin-wide Amazon forest soil carbon concentrations, *SOIL*, 6, 53–88, <https://doi.org/10.5194/soil-6-53-2020>, 2020.

Souza, I. F., Almeida, L. F. J., Jesus, G. L., Kleber, M., and Silva, I. R.: The mechanisms of organic carbon protection and dynamics of C-saturation in Oxisols vary with particle-size distribution, *European Journal of Soil Science*, 68, 726-739, 10.1111/ejss.12463, 2017.

Souza, I. F., Almeida, L. F. J., Pett-Ridge, J., Nico, P. S., Kleber, M., and Silva, I. R.: Carbon sink strength of subsurface horizons in Brazilian Oxisols, *Soil Sci. Soc. Am. J.*, 82, 76-86, 10.2136/sssaj2017.05.0143, 2018.

**Errata found in manuscript**

The legend in Figure 5, explaining the meaning of color and shape of the dots, is missing. It will be corrected as follows:



**Figure 5: Biplots of the varimax rotated principal component analysis. (a) rPC1nv and rPC2nv and (b) rPC3nv and rPC4nv of non-valley positions (n = 27). Observations cluster together based on similarities within geochemical regions and their distinction to other geochemical regions. Vector length indicates how strongly variables influence a specific rPC. The angles between vectors display the degree of auto-correlation between variables. Small angles represent positive correlations and high degree of autocorrelation, diverging angles represent negative correlations and high degree of autocorrelation, high angles indicate no correlations between variables and/or rPCs.**

We hope we have addressed all concerns and look forward to hearing from you.

Best regards,

The authors