

## Author's response to reviewer's comments:

Thanks for the thorough and thoughtful response to review! I especially appreciate the additional explanation of distinctions between nanoCLICs and other phases and supporting literature in the introduction text. However, I agree with the editor's concern about the interpretation of land use conversion or change effects. The changes to wording (e.g., use of softer wording such as “may”) that address the limitations of the ability to test a land-use conversion/change is appreciated, but as described in the authors' response, in the revised title, and in the manuscript, the changes do not fully address the concern as highlighted by reviewers and the editor. In my view, it is not necessarily a situation where softening the language is primarily what's needed: it is the framing of the study around forest-to-crop conversion as the main focus, given the lack of statistical power, representation of spatial variation, and direct before/after comparison of a conversion process or land-use change. The revised aims do make this more clear ("under both forest and cropland conditions"), but (1) keeping forest-to-crop conversion in the title and in some important conclusions elsewhere and (2) the revised sentences in point (ii) still imply that forest-to-crop conversion is a key focal area of the manuscript and the primary conclusions of the study.

Overall, I think additional revision of the overall framing (and a shift towards the novelty of compositional and structural insights, as suggested by the editor) is still needed. In my response below, I identified a few areas where I suggest statements need further consideration:

Author's answer: We thank the reviewer for this thoughtful review. The requested changes have been made, including:

- A change of title to focus on microscale characterization of mineral-organic associations: *“Interplay of coprecipitation and adsorption processes: deciphering amorphous mineral-organic associations under both forest and cropland conditions”*.
- A refinement of the objectives in the abstract specifically focusing on the identification of the types of mineral-organic associations, as follows : *“Identifying the types of mineral-organic associations present within a single soil is already a known challenge and detecting those susceptible to disruption during forest-to-crop conversion is even more complex. Yet, addressing this identification challenge is essential for devising strategies to preserve organic matter in croplands. Here, we aimed to identify the predominant mineral-organic associations within an Andosol (developed on Fe-poor parent material) under both forest and cropland conditions.”*
- Concerning comments on land-use change, we have addressed all the reviewer's points by focusing on the “differences between the forest and crop Andosol profiles” rather than the “impact of forest-to-crop conversion.” Moreover, we acknowledged the limitations of comparing forest and cropland conditions in the abstract and conclusion to provide context for the study's findings (L64: “Although the sample size for comparing land-use types is limited, ...”). Additionally, we included a detailed discussion on these limitations in section 4.4 and further referenced them in section 4.5 to ensure comprehensive coverage of this critical aspect. See L420-426 *“Ultimately, these results suggest that the cropland conditions mainly affected the*

*amount, rather than the type, of mineral-organic associations in the studied Andosol. However, it is important to consider that the representativeness of a forest-to-cropland conversion in this study has some limitations: firstly, the samples come from only one soil profile taken at a single point in time, which means they may not fully capture the spatial variability within a plot. Secondly, the quantitative differences in organic carbon content and extractable pools are based on a single observation per depth. Although multiple depths within a profile contribute valuable information to the overall pattern, they do not function as replicates per se."*

Please, find below a detailed answer to Reviewer's comments.

Lines 1-2 (Title): Keeping "forest-to-crop conversion" may over-emphasize the interpretations regarding this element of the study, and downplays the stronger microscale insights (such as secondary interactions), in my opinion.

Author's answer: As suggested by the reviewer, we changed the title and emphasized the microscale insight and the secondary interactions. Here is the modified title: *"Interplay of coprecipitation and adsorption processes: deciphering amorphous mineral-organic associations under both forest and cropland conditions"*

Lines 32-33: While the inclusion of a depth profile does strengthen the contrast, the comparison of bulk soil characteristics is still limited by the single profile sampled per land use type. A quantitative assessment here (i.e. 75%) should be much more constrained, especially in the abstract which communicates this without the limitations of sample size being mentioned.

Author's answer: The sentence was split, and the limitation of sample size is now mentioned in the abstract as follows: *"While the presence of similar amorphous coprecipitates in both the forest and crop Andosols was confirmed, the crop soil had 75 % less C in mineral-organic associations (in the 0-30 cm depth). Although the sample size for comparing land-use types is limited, these results suggest that the nature of mineral-organic associations remains identical despite quantitative differences."*

Lines 35-36: In the response to review, the argument is made that the aims were revised to focus on a contrast to prior observations: "...Fe-poor parent material (andesite) are similar to mineral-organic associations in a forested Andosol developed on Fe-rich parent material (basalt; from Jamoteau et al., 2023)." This doesn't come through in the abstract, which remains focused on forest and cropland differences as the primary focus of the study. Further refinement of this focus is needed to clearly identify the takeaway from the study.

Author's answer: The aim has been modified as follows in the abstract: *"Here, we aimed to identify the predominant mineral-organic associations within an Andosol (developed on Fe-poor parent material) under both forest and cropland conditions"*. Knowing that the challenge of identifying mineral-organic associations has been clarified in the abstract (*"Identifying the types of mineral-organic associations present within a single soil is already a known challenge and detecting those susceptible to disruption during forest-to-crop conversion is even more complex. Yet, addressing this identification challenge is essential for devising strategies to preserve organic matter in croplands."*), we choose to keep the main objective of the paper and not to detail the secondary objective in the

abstract for fluidity of reading and space constraint. All objectives are clearly detailed in the introduction.

Line 47: “Transitions from...” This is another area where the text implies the primary focus is on a forest to crop conversion/transition; revision of the framing here is needed.

Author’s answer: This sentence has been modified as follows: *“In order to maintain agricultural productivity in cultivated soils, it is essential to preserve mineral-organic associations in croplands.”*

Line 60: “refuted...” don't think there is enough evidence from these observations to refute the role of SROs in general. Suggest softening the language here from "refuted" to "raised questions about the stabilizing role of short-range order minerals alone in..."

Author’s answer: This sentence has been modified as follows: *“In Andosols, i.e. soils with high concentrations of mineral-organic associations, microscopy and spectroscopy analyses raised questions about the stabilizing role of short-range order minerals in the form of imogolite or allophane for C (Levard et al., 2012).”*

Lines 102-103: It should be stated here that two Andosol topsoils from single profiles were compared, with a brief explanation of why this is a useful comparison (e.g., for high-intensity imaging, limited sample sets are often necessary and enable direct evidence/visualization etc., but site-level or broader comparisons are limited).

Author’s answer: This sentence has been added: *“Although only single profiles per land use type were compared (limiting broader site-level or land-use comparisons), this approach was chosen to enable high-resolution imaging and direct visualization of mineral-organic associations.”*

Lines 203-204: I think here you could state that these measurements were conducted primarily as context for the microscale measurements. This particular contrast doesn't really "probe" this process (forest-to-crop conversion) or statistically testable contrasts between forest and cropland Andosols. In general, I might also suggest keeping this information in the supplementary only and referencing it for context as needed in the discussion.

Author’s answer: We agree with the reviewer and constrained our aims to show the depth profiles, as follows: *“To investigate the differences between the forest and crop soil profiles and select the appropriate horizon with quantitative differences in mineral-organic associations for micro and nanoscale mappings, we compared key physicochemical parameters between the forest and crop Andosol profiles (Fig. 2).”*

Figure 2 caption: The inclusion of the depth profiles does add a lot to the context, but it still remains single profile comparisons; as mentioned above, I suggest moving this information to the SI to use as context for observations but downplay the numerical estimation contrasts which lack any representation of error/spatial variation.

Author’s answer: Following the clarification of the objective of these measurements (see response to the comment above: *“... and select the appropriate horizon with quantitative differences in mineral-organic associations for micro and nanoscale mappings”*), we

believe that showing the profile data is necessary. Additionally, these data provide context, as the reviewer mentioned.

Lines 393-394: Very interesting! I thank the authors for emphasizing this result more strongly in the abstract.

Author's answer: We thank the reviewer for their comment.

Lines 420-422: The limitation of this contrast is two-fold: first, whether or not it represents a "conversion" (since it's a space-for-time substitution) but also that the quantitative differences in OC content and extractable pools is based only on one observation per depth (multiple depths within a profile do add information to the overall pattern, but do not serve as replicates per se, and the issue of single point comparisons for a given depth or the depth profile across two horizons stands). As such, this statement is not sufficient to account for the limitation in this contrast. In general, I think it's good that the analyses covered two land use types and a range of parent materials (compared to prior studies) to get a sense of the variability in these microscale characteristics, but the overall emphasis on forest-to-crop conversion still needs further adjustment in all sections of the manuscript, including the title and abstract. To clarify, I think it's reasonable (and common, given the high time investment) to compare these two land uses with respect to microscale measurements; it's (1) the bulk measurement contrasts (e.g., 75% decrease) and (2) the argument for "conversion" (rather than differences across systems) that requires further consideration and a bit more revision in the text, in my opinion.

Author's answer: We thank the reviewer for recognizing the importance of using the contrast between forest soil and cultivated soil. Regarding paragraph 4.4, we have revised the term "conversion" at the beginning of the paragraph, as follows: *"This substantial C difference, with showed up to 75% less C in mineral-organic association in the crop topsoil compared to the forest topsoil, suggest a destabilizing effect of agricultural conversion on C in mineral-organic associations.").*

Moreover, we now discuss the limitations of sample representativeness in lines 420 to 424. Specifically, we have addressed the two main limitations: *"However, it is important to consider that the representativeness of a forest-to-cropland conversion in this study has some limitations: firstly, the samples come from only one soil profile taken at a single point in time, which means they may not fully capture the spatial variability within a plot. Secondly, the quantitative differences in organic carbon content and extractable pools are based on a single observation per depth. Although multiple depths within a profile contribute valuable information to the overall pattern, they do not function as replicates per se."*

Lines 427-430: Comment above applies here as well.

Author's answer: In paragraph 4.5, we have avoided the term "forest-to-crop conversion" and instead focused on "decreases between forest and crop soil" to better reflect the study's limitations. We have also added a reference to the limitations discussed in section 4.4 to provide context for the representativeness of the samples: *"In this study, if the two analyzed Andosol profiles represent decreases between forest and crop soil (see the*

*limits of sample representativeness of forest-to-cropland conversion in 4.4), our results suggest that...".*

Minor comments:

Line 29: Revise to "i.e."

Line 30: Would "secondary" be more appropriate here? Not sure what is meant by "subsequent".

Line 60: Revise "analyzes" to "analyses"

Line 67: Suggest revising to "These results suggest that in some situations"

Line 76: Suggest revising to "mineral-organic associations in Andosols"

Line 93: Suggest revising to "Andosol"

*Author's answer: we thank the reviewers for their minor comments. All suggested changes have been implemented.*