

Dear Prof. Dr. Yakov Kuzyakov,

We highly appreciate your comments, as well as the three referee comments and the short comment. We considered and discussed all suggestions and remarks and they substantially contributed to improve our manuscript.

As we already explained and discussed in detail all our changes and revisions in our responses to the referee comments and the short comment, we would now like to draw your attention to the fact that we also followed your suggestion to include hypotheses in our manuscript. We did so in the final part of our introduction and referred to them again in the conclusion.

Below, you find all comments on our manuscript and our responses combined with the reworked and marked-up version of our manuscript.

We thank you very much for considering our manuscript for publication in Biogeosciences!

Yours sincerely,

Isabel Prater on behalf of all authors

Comment from Referee

Interactive comment on "From fibrous plant residues to mineral-associated organic carbon – the fate of organic matter in Arctic permafrost soils" by Isabel Prater et al.

Anonymous Referee #1

Received and published: 19 March 2020

The present manuscript by Prater et al. reports new data on various physical fractions of soil organic matter (aggregated, small occluded, mineral associated) in Arctic permafrost-affected environments in the Lena River Delta, Siberia. Several chemical analyses (C and N content, $\delta^{13}\text{C}$, ^{13}C NMR) were jointly used on the different fractions to better understand the fate of fibrous plant residues in permafrost soils. The manuscript is well written and the presented results and discussion are important for understanding the soil organic matter fate in changing Arctic regions. The authors should consider some minor comments before accepting the manuscript for publication with Biogeosciences.

Specific minor comments and suggestions:

- line 45: please check the reference (should be Frank et al., 2012)
- lines 58-59: the sentence should come before, the warming climate is already mentioned lines 38-39 for example
- lines 75-78: please clarify your objectives, the "physical fractionation" is an approach and not an objective
- line 81: add "Siberia" somewhere
- line 98: add "electric conductivity (EC)" to be consistent with line 117
- line 162: "to detect correlation": check the sentence, statistically a correlation is quantified and not "detected" using a plot
- line 179: the data could be presented with cumulative area charts for each profile and each element (C and N) to illustrate the proportions of each fraction by depth (in supplement)
- lines 221-222: move to the discussion
- line 271: "considerable amount of N" compared to what?
- line 283: change "C:N" to "C/N"
- line 340: change "dinitrogen" to "N₂"
- figure 3: use "I" and "II" instead "a" and "b" to be consistent with the figure 2
- figure 4: the quality is too bad, be consistent with fig. 2-3, use indication of x log-scale and not just 10 and 100 that are not indicative, add minor gridlines for example), use the same color code as in figure 5
- figure 5: change the labels in the PCA to be in agreement with the text (C/N, a/o a ratio, etc.)
- figure 7: same comments as for the Excel plot in figure 4
- figure 8: I do not understand the point of adding both PCA and correlation matrices. I suggest to keep either the PCA, including individuals (as done in figure 5), or correlation matrices only

Author's Response

Response to *Interactive comment on "From fibrous plant residues to mineral-associated organic carbon – the fate of organic matter in Arctic permafrost soils" by Isabel Prater et al. from Anonymous Referee #1* by the authors

Dear Referee #1,

We thank you for your kind and helpful comments on our manuscript and we really appreciate that they helped to further improve it. Please find our answers below in *italics*, we also added the respective line numbers of the updated manuscript to improve the traceability:

- line 45: please check the reference (should be Frank et al., 2012)

Thank you for this hint, the reference reads now Frank et al, 2012 (l. 51).

- lines 58-59: the sentence should come before, the warming climate is already mentioned lines 38-39 for example

As recommended, we moved the sentence (l. 43/44).

- lines 75-78: please clarify your objectives, the “physical fractionation” is an approach and not an objective

We rephrased the sentence to clarify that the detailed insights into chemical composition and stabilization mechanisms of SOM are the objectives and not the fractionation itself (l.80-86).

- line 81: add “Siberia” somewhere

We added "Siberian" (l. 89).

- line 98: add “electric conductivity (EC)” to be consistent with line 117

Thank you for the remark, we added (EC) (l. 109).

- line 162: “to detect correlation”: check the sentence, statistically a correlation is quantified and not “detected” using a plot

We changed "detect correlations" to "identify interrelations" (l. 173).

- line 179: the data could be presented with cumulative area charts for each profile and each element (C and N) to illustrate the proportions of each fraction by depth (in supplement)

We thank you for this remark; it is always good to explore better ways to visualize data. We tested the cumulative approach, but decided to stick to the table as we think that a graphical depiction would be redundant.

- lines 221-222: move to the discussion

We moved this part of the sentence to l. 338/339.

- line 271: “considerable amount of N” compared to what?

We changed "considerable" to "noteworthy" (l. 284) as this expression works without comparison. We do not aim at comparing the values; we want to show that the N content should not be neglected.

- line 283: change “C:N” to “C/N”

Thank you for this hint, we changed it accordingly (l. 296).

- line 340: change “dinitrogen” to “N₂”

To meet both this recommendation and the recommendations of the co-authors, we changed this part to "nitrous oxide (N₂O) and dinitrogen (N₂)" (l. 365).

- figure 3: use “I” and “II” instead “a” and “b” to be consistent with the figure 2

We reworked all our figures according to your suggestions and we appreciate that you helped to clearly improve them. We changed the captions and labels accordingly, used a consistent color code for all figures (that should also work for people with color vision deficiency) and took care of a much better quality.

- figure 4: the quality is too bad, be consistent with fig. 2-3, use indication of x log-scale (and not just 10 and 100 that are not indicative, add minor gridlines for example), use the same color code as in figure 5

We changed it accordingly, see comment above (figure 3).

- figure 5: change the labels in the PCA to be in agreement with the text (C/N, a/o a ratio, etc.)

We changed the figure, see comment above (figure 3).

- figure 7: same comments as for the Excel plot in figure 4

We changed the figure accordingly, see comment above (figure 3).

- figure 8: I do not understand the point of adding both PCA and correlation matrices. I suggest to keep either the PCA, including individuals (as done in figure 5), or correlation matrices only.

We decided to follow your suggestion and left only the correlation matrices in the manuscript as we are convinced that those help best to illustrate the differences between large POM fractions and oPOMs fraction.

Comment from Referee

Interactive comment on “From fibrous plant residues to mineral-associated organic carbon – the fate of organic matter in Arctic permafrost soils” by Isabel Prater et al.

Anonymous Referee #2

Received and published: 20 March 2020

The manuscript entitled “From fibrous plant residues to mineral-associated organic carbon – the fate of organic matter in Arctic permafrost soils” analysed four Cryosol soil cores for quantity and quality of organic matter to understand stabilization mechanisms and mineralization potential under climate change. The manuscript is very well written, within the scope of Biogeosciences and informative. No major methodological flaws were detected. The novelty of the results is somewhat limited, the results are close to what should have been expected. However, every new dataset on soils from these remote land masses that are warming up rapidly is valuable per se and the mix of methods is strong, of course. My most important major concern is that I was not able to understand how the authors exactly did the 1 m stock extrapolation when only some selected layers were fractionated. There is no information on that specific issue and looking at the reference publication reveals that soil profiles (usual for river terraces) were extremely heterogeneous with depth. This depth dependency and spatial heterogeneity in general, is of major importance for upscaling, which is again of specific importance for that vast and SOC rich region. This starts from how single soil profiles are averaged which should thus be done and described with care. A second major issue is related to the data shown in Fig. 4a: How do the authors explain this positive correlation of $\delta^{13}\text{C}$ and C/N ratio within the fractions, I think the opposite would be expected (the more decomposed, the more positive $\delta^{13}\text{C}$)? Still, my recommendation is publish after some revisions. Please find specific comments below:

General: You might want to consider to call oPOM oPOMI (I for large), this would be more consistent when you also have oPOMs as individual fraction.

I.16: “the permafrost region” appears too unspecific. Is it the Northern Circumpolar Permafrost region or do you include the high mountain ranges here?

I.74: Why Ping et al. at the end?

I.94-97: Please be more specific about the sapling locations, or the selection of these 4 cores. Why those, any criteria to ensure that they are representative for the most likely very heterogeneous area? Can't really find information on that in the cited literature. Also, please mention the soil types. In Zubrzycki et al.2013, different soil types are mentioned. Was it all cryoturbated soils?

I.102: Which selected layers, how many samples were fractionated?

I.102: Table 1 is missing, but would be extremely important, also to judge about some interpretations of the authors.

I.117: During the washing... → How did you separate MAOM and oPOMs? Another density step? This is not clear.

I.126: Projected to 1m: How was this done exactly, and why is there no information about the depth distribution of fractions if the authors state that different depths were analysed? This is a bit confusing.

I.164: find correlations with what?

I.188: give ranges for the silt and sand-sized MAOM as well, otherwise the sentence reads incomplete.

I.217: This is something that is not clear to me: How do you explain this trend? Usually it should be the other way around. Why is $\delta^{13}\text{C}$ more negative when C/N ratios are decreasing?

Results in general: I was missing a depth distribution of the fractions, all results are depth independent and it is unclear, how homogeneous the profiles were.

I.268: It is not clear to me, if really cryoturbation caused the depth distribution of POM, but again this can only be judged if some depth information is included. Especially in river terraces, it could also be successive growth of the soil profile via sedimentation, and potentially even growth of organic layers.

I.286: Title sounds like POM fractions do also dominate N stocks, which doesn't seem to be the case (table 1). Maybe consider to rephrase.

I.293: “release vast amount of N” → This is a contradiction to what has been said before and also the title (not much N in POM fractions)

I.313: Where does this information come from that fibres act as hot spots for microbial decay?

I.356: Where is this comparison per single soil layers?

Fig.4: X-axis: Why did you put the x-axis on top of the graph and show only 10 and 100? Is there any reason for that. Readability would improve when numbers are at the bottom and more continuous.

Author's Response

Response to *Interactive comment on "From fibrous plant residues to mineral-associated organic carbon – the fate of organic matter in Arctic permafrost soils" by Isabel Prater et al. from Anonymous Referee #2* by the authors

Dear Referee #2,

We thank you for your valuable and very helpful comments on our manuscript! We appreciate that they supported us substantially with further improving it. Please find our answers to your remarks below in *italics*, we also added the respective line numbers of the updated manuscript to improve the traceability:

General: You might want to consider to call oPOM oPOMI (I for large), this would be more consistent when you also have oPOMs as individual fraction.

We agree that your suggestion could imply some consistency in the naming. We discussed this and decided to keep our naming as the use of oPOMI and oPOMs could imply the existence of at least one more oPOM fraction between oPOMI and oPOMs and could therefore cause confusion. Furthermore, there are numerous publications that use the same terminology and we would like to maintain comparability for the reader.

I.16: "the permafrost region" appears too unspecific. Is it the Northern Circumpolar Permafrost region or do you include the high mountain ranges here?

We agree that this term is relatively broad. In l. 18 we mentioned the Arctic and according to your suggestion we additionally added "Northern circumpolar" to avoid further misunderstandings (l. 19).

I.74: Why Ping et al. at the end?

We placed Ping et al. 2015 here (l. 79), as we think this paper provides a great overview on the facts that we stated before.

I.94-97: Please be more specific about the sapling locations, or the selection of these 4 cores. Why those, any criteria to ensure that they are representative for the most likely very heterogeneous area? Can't really find information on that in the cited literature. Also, please mention the soil types. In Zubrzycki et al. 2013, different soil types are mentioned. Was it all cryoturbated soils?

You are right that this is a very important point in our study. We further specified the sampling area (l. 104), and referred here directly to the Holocene river terrace and also added another reference with more details on the sampling area (l. 108). We also added information on the soils (l. 93-95).

I.102: Which selected layers, how many samples were fractionated?

I.102: Table 1 is missing, but would be extremely important, also to judge about some interpretations of the authors.

We thank you very much for this crucial hint. We moved the respective table to the Supplement, but at this point (l. 113) we did not change the reference. The information on the samples as the depth layers etc. is now given in table S1. We also added the number of the selected layers we fractionated (l. 113).

I.117: During the washing... → How did you separate MAOM and oPOMs? Another density step? This is not clear.

As this step is very fundamental for our study we are thankful for your remark. To better clarify the procedure, we added more detailed information (l. 128/129). We followed a standard density fractionation approach, where we separated the POM fractions (including oPOMs) from the MAOM by density fractionation, which we describe in l. 116-129. The oPOMs was not separated from the MAOM, but from the oPOM fraction as described in l. 128/129.

I.126: Projected to 1m: How was this done exactly, and why is there no information about the depth distribution of fractions if the authors state that different depths were analysed? This is a bit confusing.

The stocks for the respective sampled and analyzed soil depths were taken as a bulk and projected to one cubic meter. As common practice, we report the stocks based on the sampled and analyzed material. The depth distribution and all according information can be found in table S1.

I.164: find correlations with what?

We rephrased this sentence (l. 173) to better clarify the statement.

I.188: give ranges for the silt and sand-sized MAOM as well, otherwise the sentence reads incomplete.

Thank you, we totally agree and added the missing information (l. 200/201).

I.217: This is something that is not clear to me: How do you explain this trend? Usually it should be the other way around. Why is $\delta^{13}\text{C}$ more negative when C/N ratios are decreasing?

You are pointing to a very interesting aspect of our work that nicely demonstrates the specificity of the studied soil systems. We discuss this in more detail in the discussion section 4.3 (l. 338-356). We rewrote this section to better emphasize the differences in the ^{13}C abundance for different SOM fractions.

Results in general: I was missing a depth distribution of the fractions, all results are depth independent and it is unclear, how homogeneous the profiles were.

You are right that the depth distribution of fractions is an important fact. We are now providing all according information in table S1.

I.268: It is not clear to me, if really cryoturbation caused the depth distribution of POM, but again this can only be judged if some depth information is included. Especially in river terraces, it could also be successive growth of the soil profile via sedimentation, and potentially even growth of organic layers.

You are right, in areas that are flooded regularly a burial of organic matter can be expected. This was the reason we avoided such areas and did not take samples from the floodplain. We only sampled the terrace that is only very rarely flooded, which is thought not to lead to the burial of pockets of organic matter as the ones discovered. Related information on the depth distribution is given in table S1.

I.286: Title sounds like POM fractions do also dominate N stocks, which doesn't seem to be the case (table 1). Maybe consider to rephrase.

In the studied soils, the particulate OM represents an important N storage pool. While the C stock is dominated by the large POM fractions, the N stock is dominated by the fPOM and the oPOMs fraction – especially the fPOM fraction plays a crucial role for both stocks. We discuss this in l. 302-306.

I.293: "release vast amount of N" → This is a contradiction to what has been said before and also the title (not much N in POM fractions)

We found relatively high amounts of N in fPOM, oPOMs and clay-sized MAOM, therefore, we assume an increased release of this N under ongoing warming. With the title we want to indicate that the POM fractions are more important for the C stock than for the N stock, not that the POM fractions are negligible.

I.313: Where does this information come from that fibres act as hot spots for microbial decay?

Thank you for this hint, we added a source for this information (l. 329) and rewrote parts of the respective section. The fibrous OM particles represent relatively undecomposed plant residues, as we were able to demonstrate using NMR spectroscopy. Thus, these particles represent detrital material

that most likely provides highly bioavailable OM sources for microbial activity. This hot spot effect of the detritusphere for microbial activity is well known and was addressed in numerous other studies (e.g. Beare et al. 1995, Poll et al. 2006, 2008, Sanaullah et al. 2016).

I.356: Where is this comparison per single soil layers?

Thank you for this hint, we added the respective information (l. 382).

Fig.4: X-axis: Why did you put the x-axis on top of the graph and show only 10 and 100? Is there any reason for that. Readability would improve when numbers are at the bottom and more continuous.

Thank you for this remark, we reworked all of our figures and changed fig. 4 according to your suggestion.

Comment from Referee

Interactive comment on "From fibrous plant residues to mineral-associated organic carbon – the fate of organic matter in Arctic permafrost soils" by Isabel Prater et al.

Anonymous Referee #3

Received and published: 22 April 2020

This is a very well-written manuscript that describes organic matter content and composition of physically-isolated density and particle size fractions collected from ice-wedge polygon centers in the Arctic. The objective of the paper is to characterize degree of decomposition of organic matter in permafrost soils with varying degrees of association with mineral surfaces to better understand potential bioavailability of this organic matter pool to warming and thawing. The authors present a thorough chemical characterization of particulate and mineral associated organic matter pools through C and N elemental analysis, stable isotopes and C13-NMR spectroscopy. The results interestingly reveal large contributions of potentially chemically bioavailable POM to the bulk soil C pool, whereas mineral-associated fractions contribute more to the soil N pool. This work has implications for predictions of the response of similar permafrost-affected soils to warming.

Abstract:

L. 25: "We demonstrate that" It would be helpful in this sentence to operationally identify the fraction being discussed (that is, how was it isolated physically?) to better understand how it is being interpreted as "bioaccessible." Can you define the term bioaccessible? Is it synonymous with the more common "bioavailable" or does it specifically refer to physical accessibility?

Methods: The methods indicate soil drill cores are taken but do not highlight what depths are analyzed and presented. The text states in L. 102 : "Our analyses focused on selected layers only, as shown in Table 1" but Table 1 does not include this information. One would expect that the contribution of POM vs MAOM and the state of decomposition may vary with soil depth (perhaps not in the traditional predictions) yet the paper does not describe what depths are being analyzed.

Discussion:

The discussion is quite long with extensive paragraphs that have multiple ideas, which makes it sometimes a little difficult to follow all the ideas. Consider where the discussion can be streamlined and how paragraphs could be split into smaller blocks of text.

Section 4.1- The section heading is perhaps not the most informative of the text, as permafrost processes (other than one mention to cryoturbation) are not discussed in depth here. Consider renaming the section or including more information on processes. It may also be helpful to separate the text into a paragraph on C and N stocks and another one on composition of SOM, mainly C:N ratios.

Section 4.2: It is very interesting that the POM and MAOM fractions play such different roles in C and N storage in these soils.

Section 4.3: Consider starting the paragraph l. 332 with summarizing results of N dynamics or 15N and their implication as the first sentence on N fixation seems to have no context. This paragraph could also be moved after the NMR paragraph which flows better after the 13C paragraph.

Minor edits:

Introduction, paragraph starting l. 58-78 is too long with too many different ideas. Should be broken up into smaller paragraphs, one on effects of climate change on SOM, one on SOM methods, then the research objectives.

Spell out abbreviations for symbols in the Table legends. For example, fPOM, MAOM...

Also indicate whether data reported are means and standard error or means and standard deviation.

Table 2. Should a/o-a ratio be O-a ratio? (capital O)

Figure 1. May be helpful to indicate what the white and blue colors are on the image.

Ice and open water? Unclear because the ocean is black.

l. 240: add ppm after 70-75 ppm /52-57 ppm

Author's Response

Response to *Interactive comment on "From fibrous plant residues to mineral-associated organic carbon – the fate of organic matter in Arctic permafrost soils" by Isabel Prater et al. from Anonymous Referee #3* by the authors

Dear Referee #3,

We are very grateful for your helpful and supporting comments on our manuscript that help to further improve it. Please find our answers to your remarks below in *italics*, we also added the respective line numbers of the updated manuscript to improve the traceability:

L. 25: "We demonstrate that" It would be helpful in this sentence to operationally identify the fraction being discussed (that is, how was it isolated physically?) to better understand how it is being interpreted as "bioaccessible." Can you define the term bioaccessible? Is it synonymous with the more common "bioavailable" or does it specifically refer to physical accessibility?

We agree that these terms are often used in a confusing way. When we aim at emphasizing the spatial inaccessibility of the OM, we use "bioaccessibility". When we refer to the microbial availability determined by the chemical composition of a substrate, we use "bioavailability". We made some changes in our manuscript according to our remark.

Methods: The methods indicate soil drill cores are taken but do not highlight what depths are analyzed and presented. The text states in L. 102 : "Our analyses focused on selected layers only, as shown in Table 1" but Table 1 does not include this information. One would expect that the contribution of POM vs MAOM and the state of decomposition may vary with soil depth (perhaps not in the traditional predictions) yet the paper does not describe what depths are being analyzed.

We thank you very much for this crucial hint that we were not aware of. We moved the respective table to the Supplement, but at this point (l. 113) we did not change the reference. The information on the samples like depth layers etc. is now given in table S1 and we corrected the reference accordingly.

Discussion:

The discussion is quite long with extensive paragraphs that have multiple ideas, which makes it sometimes a little difficult to follow all the ideas. Consider where the discussion can be streamlined and how paragraphs could be split into smaller blocks of text.

According to your suggestion, we restructured the discussion to increase the readability of the manuscript and split the paragraph discussing stable isotopes and NMR results into two paragraphs.

Section 4.1: The section heading is perhaps not the most informative of the text, as permafrost processes (other than one mention to cryoturbation) are not discussed in depth here. Consider renaming the section or including more information on processes. It may also be helpful to separate the text into a paragraph on C and N stocks and another one on composition of SOM, mainly C:N ratios.

According to your suggestion we changed the heading to "Cryoturbation determines bulk soil organic matter distribution". As we do not widely discuss the C and N stocks, the aim of the manuscript is clearly on the composition of the SOM fractions, thus we would like to stick to the current paragraph.

Section 4.2: It is very interesting that the POM and MAOM fractions play such different roles in C and N storage in these soils.

We are happy that you acknowledge that this is an interesting finding in our study.

Section 4.3: Consider starting the paragraph l. 332 with summarizing results of N dynamics or ^{15}N and their implication as the first sentence on N fixation seems to have no context. This paragraph could also be moved after the NMR paragraph which flows better after the ^{13}C paragraph.

We followed your suggestion above and separated the paragraph further. We have now one paragraph (4.3) discussing $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ and we slightly rearranged this paragraph. Another paragraph (4.4) is now only focusing on the NMR discussion.

Minor edits:

Introduction, paragraph starting l. 58-78 is too long with too many different ideas. Should be broken up into smaller paragraphs, one on effects of climate change on SOM, one on SOM methods, then the research objectives.

We slightly restructured the Introduction according to your suggestion.

Spell out abbreviations for symbols in the Table legends. For example, fPOM, MAOM... Also indicate whether data reported are means and standard error or means and standard deviation.

Thank you for this remark, we added the missing information to the captions of the tables.

Table 2. Should a/o-a ratio be O-a ratio? (capital O)

This ratio relates to functional groups that consist of O/N-alkyl-C and Alkyl-C, which is normally given as "Alkyl-C to O/N-alkyl-C ratio". To make it easier to read, we defined the ratio of alkyl C to O/N alkyl C as a/o-a ratio in the method section (2.4) and we kept this wording throughout the manuscript and in the tables and figures as well.

Figure 1. May be helpful to indicate what the white and blue colors are on the image. Ice and open water? Unclear because the ocean is black.

Thank you for this important remark, we added the information according to your suggestion. The white color mainly in the western part is the unvegetated sandy sediment of the floodplain and the blue spots indicate water: larger water bodies and shallow water on the terrace.

l. 240: add ppm after 70-75 ppm /52-57 ppm

We introduce the ratio according to Bonanomi et al. (2013) in the methods section (2.4), where we clearly state the chemical shift regions that are considered for this decomposition proxy. To increase readability we use a reduced naming of the ratio which is in accordance with the "a/o-a ratio" term. Thus, we would like to be consistent in the form that we use to express NMR-derived decomposition proxies.

Short Comment

Interactive comment on “From fibrous plant residues to mineral-associated organic carbon – the fate of organic matter in Arctic permafrost soils” by Isabel Prater et al.

Marijn Van de Broek and master students

Received and published: 3 April 2020

The manuscript by Prater et al. provides new data on the different physical fractions of soil organic matter from the Lena River Delta in the Arctic. The area on Samoylov Island is characterized by permafrost. The authors investigated soils with respect to the composition and distribution of organic C among differently stabilized SOM fractions, in order to gain knowledge on the mechanisms stabilizing organic C in Arctic soils, besides impaired decomposition due to low temperatures. The methods consists of the use of sophisticated approaches, separating SOM into different fractions, allowing for a detailed understanding of the stabilization mechanisms of organic carbon in soils. The study is relevant, as there are still rather few analytical approaches to the stabilization mechanisms to assess the variability of C stocks in tundra soils. The research question is particularly relevant as the study deals with a region where permafrost occurs and where soils are both an important store of carbon and other greenhouse gases and are affected by global warming. The authors did not formulate a clear hypothesis or the expected results. Consequently, they did not make a comparison between their results and their original expectations, which makes it difficult to compare the results of the research with other studies.

General comments and suggestions:

- The research question addressed by the authors is important due to the lack of knowledge on the topic. Indeed, researchers only recently started to understand the importance of cold soils for the global carbon cycle, and thus global climate. As a consequence, only a few studies related to this topic have been made so far. - The authors did not explicitly state any hypotheses. They described their intent of investigating the effect of climate change on the carbon stabilization in permafrost-affected soils, but they remained vague and did not state any kind of expected results. Therefore, it is difficult to understand to what extent the research contributed to their question. – The study site is situated in the river delta of the River Lena. Chemical composition and structure of the soil could be the result of flooding which is not the case for typical arctic permafrost soils. In general, the isle may be more affected by the Lena itself than by the rising temperature. In addition, the closeness of the Siberian sea will have an influence of the isle too, as the ocean moderates the temperatures. Therefore, the study site on the isle Samoylov maybe not representative for arctic permafrost soils in general. - Do you think is the $\delta^{15}\text{N}$ a suitable method? There are many uncertainties related to it, which could be elaborated upon.

Specific comments and suggestions:

L. 75-78: Here the authors write about their approach and the aims, which are basically to gain better knowledge on the topic. Since this section is at the end of the introduction, we think that this part is the most suited for adding the research questions and hypotheses. We think this is important, especially because the authors took four soil cores in a vast area that might be highly heterogeneous. Therefore, having expectations related to the SOM fractions you expect to find in this area, including also the stratification of the soil layers could help determining how representative the four soil cores are with respect to the whole study area.

L. 94-97: In this part the methods are described. However, the authors then state that “a detailed description of the study area and the sampling of the soil cores can be found in Zubrzycki et al. (2013)”. We advise that the authors include all relevant information also in the presented manuscript. Otherwise, the readers have to go into the literature to find this relevant information.

L. 101-105: Here, the authors describe how samples were collected, but omitted to state how many samples were collected for each SOM fraction and from which soil core they were collected. We advise to provide the number of samples of each SOM fractionation type, because otherwise it might be difficult to interpret the graphs. We also checked the literature but found any information about the number of samples in Zubrzycki et al., 2013.

Fig. 4: The three graphics (figure 4. a, b, c) could be made more similar. Further, for what concerns figure a and b, the authors represented only the two extreme values on the x-axis (10 and 100), which makes it difficult to infer the values of the dots in the middle of the graph. Please include more labels

on the x-axis to make it more continuous and improve readability. We would also advise to put the x-axis on the bottom for both graphs (a and b) and not once on the top and once on the bottom. Further, we noticed a clear positive correlation between the C/N ratio and the $\delta^{13}\text{C}$, however, since the C/N ratio usually decreases during ongoing decomposition, we were expecting the opposite trend. We therefore advise to further explain the meaning of this positive correlation.

Fig.7 & 8: Graphs 7 and 8 are difficult to interpret and would require more information in the captions to make the graphs understandable without the reader having to look up more information in the main text.

Title: Why was the word "fibrous" included in the title? Almost all plants residues are fibrous, except for plant exudates. Do you specifically looked at fibrous plant residues omitting exudates? Further, the fate of organic matter sounds somewhat dramatic.

We think the title could be shortened to, for example: "From plant residues to mineral-associated organic carbon in Arctic permafrost soils".

Author's Response

Response to *Interactive comment on "From fibrous plant residues to mineral-associated organic carbon – the fate of organic matter in Arctic permafrost soils" by Isabel Prater et al. from Marijn Van de Broek and students by the authors*

Dear master students,

Thank you for your helpful comments and for the effort you put into your review. And congratulations that you had the chance to prepare reviews as exercise during a seminar – that is a really useful training. We appreciate your comments; please find our answers below in *italics*:

- The research question addressed by the authors is important due to the lack of knowledge on the topic. Indeed, researchers only recently started to understand the importance of cold soils for the global carbon cycle, and thus global climate. As a consequence, only a few studies related to this topic have been made so far.

We are happy that you share our view on the current rather sparse knowledge on the topic of our research.

- The authors did not explicitly state any hypotheses. They described their intent of investigating the effect of climate change on the carbon stabilization in permafrost-affected soils, but they remained vague and did not state any kind of expected results. Therefore, it is difficult to understand to what extent the research contributed to their question.

We are happy that you raised this issue, we now better clarified our objectives and added our expectations in the final part of the introduction.

– The study site is situated in the river delta of the River Lena. Chemical composition and structure of the soil could be the result of flooding which is not the case for typical arctic permafrost soils. In general, the isle may be more affected by the Lena itself than by the rising temperature. In addition, the closeness of the Siberian sea will have an influence of the isle too, as the ocean moderates the temperatures. Therefore, the study site on the isle Samoylov maybe not representative for arctic permafrost soils in general.

As described in section "2.1 Site characteristics and soil sampling", we took the samples from the Holocene river terrace that is rarely flooded. To avoid the impact of regular flooding, we did not take samples from the active floodplain. Your remark regarding the influence of the river and the Laptev Sea is very important. We address the differences between the climate on the island and on the mainland in our site description. However, the studied Cryosol types can be found throughout the Arctic and thus we assume the demonstrated properties and suggested processes can be applied in a general sense.

- Do you think is the d15N a suitable method? There are many uncertainties related to it, which could be elaborated upon.

The use of d15N as an integrator of the N cycle is a semi-quantitative method, which does not allow to derive quantitative process information. Thus, we agree that this method requires a careful discussion of the various processes/fractionation factors and isotopic signatures of N sources that jointly determine d15N, which is precisely what we do in this manuscript. Taking this into account, it is a very powerful tool to fingerprint dominating N cycle processes and general N cycle patterns (e.g., open N cycle, closed N cycle) without the need for disturbance by e.g., adding enriched isotope tracers. In this context, it is also important to relate d15N to other measured parameters to support its interpretation, which is what we do in this study as well.

Specific comments and suggestions:

L. 75-78: Here the authors write about their approach and the aims, which are basically to gain better knowledge on the topic. Since this section is at the end of the introduction, we think that this part is the most suited for adding the research questions and hypotheses. We think this is important, especially because the authors took four soil cores in a vast area that might be highly heterogeneous. Therefore, having expectations related to the SOM fractions you expect to find in this area, including also the stratification of the soil layers could help determining how representative the four soil cores are with respect to the whole study area.

We agree that hypotheses are one possible way to communicate expectations at the start of a study. We have added a sentence stating what we generally expected to find without naming this explicitly as hypothesis. The cores were taken from a Holocene river terrace and were cryoturbated. We are giving the detailed information on the soil cores in table S1 in the supplement. As we sampled in depth layers, we have no particular information about the stratification, however, there were no indications of flooding events that might have resulted in stratigraphic shifts.

L. 94-97: In this part the methods are described. However, the authors then state that “a detailed description of the study area and the sampling of the soil cores can be found in Zubrzycki et al. (2013)”. We advise that the authors include all relevant information also in the presented manuscript. Otherwise, the readers have to go into the literature to find this relevant information.

Thank you for this remark; we added more information in the text. As we did not want to overload this section, we focused on the information that is relevant for our study. For readers who want to have more information, we added the respective references.

L. 101-105: Here, the authors describe how samples were collected, but omitted to state how many samples were collected for each SOM fraction and from which soil core they were collected. We advise to provide the number of samples of each SOM fractionation type, because otherwise it might be difficult to interpret the graphs. We also checked the literature but found any information about the number of samples in Zubrzycki et al., 2013.

Thank you for this very important remark. A detailed list of the samples and their properties can be found in table S1 and we added this information (l. 113). In addition, we have to excuse that we had a mistake in the reference (it said table 1 instead of table S1 before). We also added the number of selected layers in the same line.

Fig. 4: The three graphics (figure 4. a, b, c) could be made more similar. Further, for what concerns figure a and b, the authors represented only the two extreme values C3 on the x-axis (10 and 100), which makes it difficult to infer the values of the dots in the middle of the graph. Please include more labels on the x-axis to make it more continuous and improve readability. We would also advise to put the x-axis on the bottom for both graphs (a and b) and not once on the top and once on the bottom. Further, we noticed a clear positive correlation between the C/N ratio and the d13C, however, since the C/N ratio usually decreases during ongoing decomposition, we were expecting the opposite trend. We therefore advise to further explain the meaning of this positive correlation.

Thank you for this hint. We reworked all of our figures and made them easier to read and took care of a better quality. We also implemented a color scheme for all figures, changed the values on the x-axis

and put all x-axes to the bottom. Regarding $d^{13}C$ and C/N ratio, we rewrote the respective section (4.3) to better emphasize the differences between the SOM fractions.

Fig.7 & 8: Graphs 7 and 8 are difficult to interpret and would require more information in the captions to make the graphs understandable without the reader having to look up more information in the main text.

We reworked both figures and think they are easier to read and understand now.

Title: Why was the word “fibrous” included in the title? Almost all plants residues are fibrous, except for plant exudates. Do you specifically looked at fibrous plant residues omitting exudates? Further, the fate of organic matter sounds somewhat dramatic. We think the title could be shortened to, for example: “From plant residues to mineral-associated organic carbon in Arctic permafrost soils”.

We use "fibrous" because we see a clear difference between the large, fibrous POM fractions and the oPOMs fraction that is not fibrous anymore – we describe this at length, but the difference becomes especially obvious in figure 9. We want to emphasize the specific macroscopic nature of the cryoturbated materials referring to the occurrence that one is experiencing when sampling. And the fibrous litter residues foster the bulky soil structure that drives the specific oPOMs formation, which is clearly different from less organic temperate soils. We want to use the word “fate” as it summarizes a development from initial litter residues to more transformed soil compartments, which we think best describes the partitioning of compounds to various processes or pools/compounds in biogeochemistry.

Author's changes in manuscript

Please find below our reworked manuscript with changes highlighted in yellow:

From fibrous plant residues to mineral-associated organic carbon – the fate of organic matter in Arctic permafrost soils

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Abstract. Permafrost-affected soils of the Arctic account for 70 % or 727 Pg of the soil organic carbon (C) stored in the Northern circumpolar permafrost region and therefore play a major role in the global C cycle. Most studies on the budgeting of C storage and the quality of soil organic matter (SOM) in the northern circumpolar region focus on bulk soils. Thus, although there is a plethora of assumptions regarding differences in terms of C turnover or stability, only little knowledge is available on the mechanisms stabilizing organic C in Arctic soils besides impaired decomposition due to low temperatures. To gain such knowledge, we investigated soils from Samoylov Island in the Lena River Delta with respect to the composition and distribution of organic C among differently stabilized SOM fractions. The soils were fractionated according to density and particle size to obtain differently stabilized SOM fractions differing in chemical composition and thus bioavailability. To better understand the chemical alterations from plant-derived organic particles in these soils rich in fibrous plant residues to mineral-associated SOM, we analysed the elemental, isotopic and chemical composition of particulate OM (POM) and clay-sized mineral-associated OM (MAOM). We demonstrate that the SOM fractions that contribute with about 17 kg C m⁻³ for more than 60 % of the C stock are highly bioavailable and that most of this labile C can be assumed to be prone to mineralization under warming conditions. Thus, the amount of relatively stable, small occluded POM and clay-sized MAOM that account currently with about 10 kg C m⁻³ for about 40 % of the C stock will most probably be crucial for the quantity of C protected from mineralization in these Arctic soils in a warmer future. Using $\delta^{15}\text{N}$ as proxy for nitrogen (N) balances indicated an important role of N inputs by biological N fixation, while gaseous N losses appeared less important. However, this could change, as with about 0.4 kg N m⁻³ one third of the N is present in bioavailable SOM fractions, which could lead to increases in mineral N cycling and associated N losses under global warming. Our results highlight the vulnerability of SOM in Arctic permafrost-affected soils under rising temperatures, potentially leading to unparalleled greenhouse gas emissions from these soils.

1 Introduction

For several millennia, organic matter (OM) accrued in the remote soils of the Arctic and only recently, researchers started to increasingly understand the importance of these cold soils for the global carbon (C) cycle and, thus, global climate (Ping et al., 2015). Estimates on the northern circumpolar soil organic carbon (SOC) stock within the first meter vary between 445 and 496 Pg (Tarnocai et al., 2009, Hugelius et al., 2014). These C-rich soils are changing from a C sink to a source due to global warming (Oechel et al., 1993; Parmentier et al., 2017). The Arctic is strongly affected by climate change with an increase in

44 surface temperatures during the last two decades that has been more than twice as high as the global average (Meredith et al.,
45 2019). In a warming Arctic, C is lost both via carbon dioxide and methane emissions and by lateral transport with water (Plaza
46 et al., 2019). The C that is released from permafrost-affected soils due to anthropogenically accelerated thawing is assumed to
47 further enhance global warming and thus trigger additional C release from permafrost, a phenomenon known as permafrost C
48 feedback (Davidson and Janssens, 2006; Schuur et al., 2015).

49 An analysis of soils from ten North American ecosystem types reaching from tropical forests to Arctic tundra demonstrated a
50 pronounced longer turnover time for soil organic matter (SOM) in cold regions in comparison to other climate regions as the
51 C stabilization mechanisms clearly differ (Frank et al., 2012). In temperate soils, the main drivers for SOC sequestration are
52 spatial inaccessibility (occlusion in soil aggregates), binding to mineral particles (organo-mineral associated OM), and intrinsic
53 chemical recalcitrance of the OM itself (Six et al., 2002; von Lützow et al., 2006). Besides these specific mechanisms,
54 environmental factors like waterlogging and low temperatures inhibit the turnover of OM in cold regions (Oades, 1988), with
55 cryoturbation additionally supporting the conservation of SOM at greater soil depth and thus in the permafrost (Kaiser et al.,
56 2007). These abiotic mechanisms fail as soon as permafrost collapses, which leads to an increased decomposition of OM
57 (Turetsky, 2004; Plaza et al., 2019).

58 Already in 1982, Post et al. recognized a considerable variability in C stocks in tundra soils, which illustrates that a more
59 detailed knowledge on the biogeochemical cycling of C in permafrost soil needs to involve more analytical approaches that
60 enable to assess possible mechanisms of C stabilization. Thus, besides the quantification of organic C (OC), there is a growing
61 number of studies aiming to elucidate the chemical composition of SOM and the processes and mechanisms involved in C
62 cycling and stabilization in permafrost-affected soils (i.a. Torn et al., 2013; Mueller et al., 2015; Strauss et al., 2017; Jongejans
63 et al., 2018; Kuhry et al., 2019).

64 With ongoing warming, the active layers in cold regions deepen and thus, microbial activity changes and the accessibility and
65 bioavailability of OM in hitherto frozen soil layers increases (Mackelprang et al., 2011; Hultman et al., 2015).
66 Depolymerization and ammonification as well as nitrification of the long sequestered organic nitrogen (N) might also enhance
67 mineral N availability in these permafrost-affected soils, leading to increased emissions of the highly potent greenhouse gas
68 nitrous oxide (Elberling et al., 2010; Wilkerson et al., 2019). The importance of mechanisms restricting SOM decomposition
69 in permafrost soils will possibly shift from climatic stabilization (Schmidt et al., 2011) to spatial inaccessibility and association
70 with minerals (Harden et al., 2012; Mueller et al., 2015) with widely unknown consequences for the C stored in these soils.

71 Several studies estimated the vulnerability of C in permafrost soils to microbial decay from the chemical composition of bulk
72 SOM (i.a. Herndon et al., 2015; Strauss et al., 2017; Tesi et al., 2016; Weiss and Kaal, 2018; Wild et al., 2016; Xue et al.,
73 2016; Zimov et al., 2006). Yet, as SOM represents a continuum of a range of materials of different composition, from fresh
74 plant litter to highly altered compounds (Lehmann and Kleber, 2015) ruled by different stabilization regimes, the investigation
75 of bulk SOM alone is insufficient. The use of more sophisticated approaches, separating SOM into different fractions, allows
76 for a more detailed understanding of the stabilization mechanisms in soil (Golchin et al., 1994). So far, only few studies (i.a.
77 Dao et al., 2018; Diochon et al., 2013; Dutta et al., 2006; Gentsch et al., 2015; Höfle et al., 2013; Mueller et al., 2015; Xu et
78 al., 2009) used fractionation approaches to investigate the distribution and composition of OM pools in permafrost-affected
79 soils (Ping et al., 2015), most of them focusing on the composition of specific fractions or using incubation experiments.

80 The objective of our study is to gain detailed insights into the chemical composition and stabilization mechanisms of SOM in
81 Cryosols from the Siberian Lena River Delta under present conditions; therefore, we used a physical fractionation approach to
82 separate light organic particles and OM associated with minerals, i.e., particulate OM (POM; dominated by bits and pieces of
83 plants and to a lesser extent microbial residues) and mineral-associated OM (MAOM). As it is known from temperate soils
84 that POM and MAOM have different ecological functions and contribute differently to C and N storage and cycling, we expect
85 that also in permafrost-affected soils, these soil C and N pools show marked differences in their chemical composition and
86 thus vulnerability to climate change. Our two major hypotheses for the present work are: 1) SOM in permafrost-affected soils

87 is mainly stored as POM resulting from a restricted decomposition due to climatic stabilization and 2) larger POM is
88 characterized by a high content of rather labile OM that mirrors the plant litter input, whereas smaller POM particles and
89 MAOM resemble microbial transformed OM, independent of the original plant litter.

90 2 Methods

91 2.1 Site characteristics and soil sampling

92 Samoylov Island (72° 22' N, 126° 30' E) is located in one of the main channels of the **Siberian** Lena River Delta, the largest
93 delta of the Arctic. The island developed during the Holocene and belongs to one of three river terraces. While the western
94 third of the island consists of an active floodplain, the eastern part is covered by ice-wedge polygonal tundra that is typical for
95 this terrace (Boike et al., 2013). Located at 10 to 16 m a.s.l., the Holocene river terrace is rarely flooded and its plant cover
96 represents the characteristic wet sedge tundra vegetation (Zubrzycki et al., 2013). **The dominant soils of the terrace are Cryosols**
97 **according to the World Reference Base for Soil Resources (Zubrzycki et al., 2013; IUSS Working Group WRB, 2014; Soil**
98 **Survey Staff, 2014), Orthels and Turbels according to the US Soil Taxonomy.** This terrace has recently been reported to be
99 covered by about 40 % non-degraded polygonal tundra, 40 % collapsed polygons, slopes, and water bodies, and 20 % of
100 polygons that show different stages of degradation (Kartozia, 2019). On the island, active layer thickness varies around 50 cm
101 and the thawing period lasts approximately 129 days (Boike et al., 2013). The climate is Arctic and the 30-year mean (1961-
102 1990) of the closest meteorological station in Tiksi, about 110 km southeast, shows a mean annual air temperature of -13.5° C
103 with a large amplitude between warmest (around 8° C in July and August) and coldest (around -32° C in January) months
104 (Roshydromet, 2019). Precipitation is low on Samoylov Island and, due to the different geographic setting within the river
105 delta, with a mean of 125 mm a⁻¹ markedly lower than the 323 mm a⁻¹ measured in Tiksi (Boike et al., 2013; Roshydromet,
106 2019).

107 We drilled four intact soil cores from ice-wedge polygon centers **on the Holocene river terrace** (Fig. 1; Boike et al., 2012) in
108 April 2011 and May 2013 using a Snow-Ice-Permafrost-Research-Establishment coring auger (Jon's Machine Shop,
109 Fairbanks/USA) with a length of 1 m and a diameter of 76 mm with a STIHL BT 121 engine power head (Andreas Stihl AG
110 & Co. KG, Waiblingen/Germany). A detailed description of the study area and the sampling of the soil cores can be found in
111 Zubrzycki et al. (2013) **and Zubrzycki (2013).**

112 All bulk soil samples were slightly acid with lowest pH values of 4.9 and highest of 6.6, electric conductivity **(EC)** ranged
113 from 66 to 240 µS cm⁻¹ with a mean of 115 µS cm⁻¹ and bulk density varied from 0.2 to 0.9 g cm⁻³ around a mean of 0.5 g cm⁻³.

114 2.2 Geochemical properties of bulk soils, physical soil fractionation and chemical analyses of fractions

115 We separated the drilled cores according to visible mineral soil horizons in frozen condition and subsequently thawed and
116 dried them at 40° C in an oven. Our analyses focused on **23** selected layers only, as shown in Table **S1**.

117 The bulk soils were fractionated according to density and particle-size, following the approach described by Mueller and
118 Koegel-Knabner (2009). Due to the high amount of fibrous material in these Cryosols, some modifications of the procedure
119 were necessary to yield mechanistically different SOM fractions. We unclenched 15 to 20 g – depending on the available
120 amount of sample material – of each soil sample by forceps and gently saturated them with a sodium polytungstate solution
121 with a density of 1.8 g cm⁻³ by slowly adding the salt solution with a pipette. After 12 hours to ensure a complete and gentle
122 saturation, the floating free POM (fPOM, not embedded in stable aggregates, cf. Golchin et al., 1994) was collected using a
123 vacuum system. The removal of the floating fPOM was repeated twice to ensure a high recovery and the obtained fraction was
124 subsequently washed over a sieve of 20 µm mesh size to remove excessive salt. Due to the highly fibrous nature of the fPOM,
125 the washing step also yielded fine mineral particles, which adhered to the fPOM fibers. As the C and N contents and C/N ratios
126 of this mineral material were in the exact same range of the clay-sized MAOM fraction, we added it mathematically to this

127 fraction for the calculation of the C stock. To separate occluded POM fractions (oPOM, incorporated in water-stable
128 aggregates, cf. Golchin et al., 1994) from MAOM, the residual samples were subjected to ultrasonication (Bandelin Sonoplus
129 HD2200, Berlin/Germany) using a calibrated (Graf-Rosenfellner et al., 2018) energy input of 300 J ml⁻¹ after the fPOM
130 removal. On the lines of the fPOM fractions, oPOM was withdrawn using a vacuum system and washed salt-free over a sieve
131 of 20 µm mesh size by repeated washing until the EC dropped below 2 µS cm⁻¹. During the washing of the oPOM through a
132 20 µm sieve, we obtained the small oPOM (oPOMs) fraction representing a fine particulate light OM (Mueller et al., 2015,
133 2017). The remaining heavy residues, constituting the MAOM, were separated by wet sieving and sedimentation to obtain
134 coarse/medium sand (>200 µm), fine sand (63-200 µm), coarse silt (20-63 µm), medium silt (6.3-20 µm) and fine silt/clay
135 (<6.3 µm, further referred to as the clay-sized MAOM fraction). All SOM fractions were analyzed for total C and N contents
136 in duplicate by dry combustion (EuroVector EuroEA3000 Elemental Analyser, Pavia/Italy). After the analyses of each sample,
137 for better clarity for the reader, C and N contents were calculated for the combined sand- and silt-sized fraction per each bulk
138 soil sample. Due to the absence of carbonates (see pH values in Table S1), total C represents OC. Coarse fractions >20 µm
139 were ball milled and homogenized prior to C and N measurements. The bulk soil C and N contents were calculated from the
140 sum of the physical fractions; C and N stocks for the SOM fractions were also calculated and overall C and N stocks projected
141 to 1 m soil depth. The mass recovery rate after fractionation was >90 % in all samples. In addition, to reveal the microscale
142 structure and illustrate possible source materials (microbial vs. plant origin) scanning electron microscope (SEM) images
143 (JSM-7200F, JEOL, Freising/Germany) were obtained for representative POM fractions.

144 2.3 Stable isotope measurements

145 The abundance of ¹⁵N and ¹³C of POM and clay-sized MAOM fractions were determined using an isotope ratio mass
146 spectrometer (Delta V Advantage, Thermo Fisher, Dreieich/Germany) coupled to an elemental analyzer (EuroEA, Eurovector,
147 Pavia/Italy). A lab standard (acetanilide) was used as a standard for every sequence in intervals and different weights as well
148 to quantify isotope linearity of the system. The standard itself was calibrated against several suitable international isotope
149 standards from the International Atomic Energy Agency (IAEA, Vienna/Austria) for both isotopes. Final correction of isotope
150 values was achieved with several international isotope standards and other suitable laboratory standards that cover the range
151 of δ¹⁵N and δ¹³C results. Results are given in delta values relative to air-N₂ for ¹⁵N and relative to Vienna Pee Dee Belemnite
152 (V-PDB) for ¹³C (Werner and Brand, 2001)

153 2.4 ¹³C Nuclear Magnetic Resonance Spectroscopy

154 We subjected all fPOM, oPOM, oPOMs and selected clay-sized MAOM fractions to ¹³C cross-polarization magic angle
155 spinning (CP-MAS) NMR spectroscopy (Bruker DSX 200 spectrometer, Billerica/USA). The ¹³C NMR spectra were recorded
156 at 6,800 Hz with an acquisition time of 0.01024 s. During a contact time of 1 ms, a ramped ¹H pulse was applied to avoid
157 Hartmann-Hahn mismatches. We executed measurements in 7 mm zirconium dioxide rotors with a delay time of 1.0 s for large
158 POM fractions (fPOM and oPOM) and a reduced delay time of 0.4 s for oPOMs and clay-sized MAOM fractions. The acquired
159 number of scans (NS) varied according to the examined fractions and the available sample material. For most of the large
160 POM fractions, a NS between 3,000 and 10,000 provided sufficient signal-to-noise ratios, while most of the oPOMs and clay-
161 sized MAOM fractions required a NS of at least 10,000. Tetramethylsilane was equalized with 0 ppm as reference for the
162 chemical shifts. The spectra were integrated in different chemical shift regions according to Beudert et al. (1989) with slight
163 adjustments according to Mueller and Koegel-Knabner (2009): -10 to 45 ppm (alkyl C), 45 to 110 ppm (O/N alkyl C), 110 to
164 160 ppm (aromatic C) and 160 to 220 ppm (carboxyl C), spinning sidebands were included. Based on these integrated shift
165 regions, we calculated the ratio of alkyl C and O/N alkyl C (a/o-a ratio) as a proxy for the degree of decomposition of plant
166 residues according to Baldock et al. (1997). Furthermore, we calculated the ratio of the integrated ¹³C chemical shift regions 70 to
167 75 ppm (O alkyl C of carbohydrates) and 52 to 57 ppm (methoxyl C of lignin) according to Bonanomi et al. (2013), which

168 provides another proxy for the decomposition stage of plant residues in relation to fresh plant source material (further referred
169 to as 70-75/52-57 ratio). To translate the NMR spectra into OM compound classes (carbohydrate, protein, lignin, lipid,
170 carbonyl) we fitted the NMR data using the molecular mixing model (MMM) developed by Nelson and Baldock (Baldock et
171 al., 2004; Nelson and Baldock, 2005). For the MMM fitting, we utilized the following chemical shift regions: 0 to 45 ppm, 45
172 to 60 ppm, 60 to 95 ppm, 95 to 110 ppm, 110 to 145 ppm, 145 to 165 ppm and 165 to 215 ppm. We applied the five component
173 MMM (without char) with N:C constraint.

174 2.5 Statistics

175 We plotted C/N ratios and C and N concentrations against the N and C stable isotope ratios of SOM fractions using R to
176 identify interrelations. The R software, RStudio and the packages Rcmdr (with the plugin FactoMineR), Hmisc, Factoshiny
177 and corrplot were used for Principal Component Analysis (PCA), correlation matrices and the compilation of plots (Lê et al.,
178 2008; RStudio Team, 2016; R Development Core Team, 2017). We used PCA and correlation matrices to find correlations
179 between the properties of different SOM fractions (fPOM, oPOM, oPOMs, clay-sized MAOM), namely C and N contents,
180 decomposition proxies (C/N ratio of bulk soils and of SOM fractions, a/o-a ratio, 70-75/52-57 ratio), stable isotopes, and the
181 results from the MMM.

182 3 Results

183 3.1 Biogeochemical bulk soil properties and distribution of SOM fractions

184 The bulk soil C contents over all cores and depth layers varied between 31.6 and 144.0 mg g⁻¹. The content of total N ranged
185 from 1.3 to 6.8 mg g⁻¹ for all cores and depth layers. While the C/N ratios ranged between 23 and 38 in three of the four cores,
186 the values of the bulk soils of the fourth core were markedly lower (Table S1). The soil C stocks (projected to 1 m soil depth)
187 ranged between 20.4 and 31.4 kg C m⁻³ with a mean of 27.5±11.8 kg C m⁻³, the N stocks varied between 0.7 and 1.9 kg N m⁻³
188 with a mean of 1.2±0.6 kg N m⁻³ (Table 1).

189 The mass distribution of POM fractions varied throughout all depth layers with proportions between 10.6 and 295.0 mg g⁻¹
190 (fPOM), between 3.0 and 71.7 mg g⁻¹ (oPOM) and between 3.9 and 267.2 mg g⁻¹ (oPOMs). Especially core 3 and 4 showed
191 larger amounts of fPOM and oPOM material at greater depth in between layers dominated by MAOM (Table S1). The MAOM
192 fractions ranged between 37.2 and 244.5 mg g⁻¹ (clay-sized), between 182.4 and 479.3 mg g⁻¹ (silt-sized) and between 79.0
193 and 591.5 mg g⁻¹ (sand-sized).

194 3.2 Elemental composition of SOM fractions

195 The highest C contents were detected in the fPOM and oPOM fractions, with values ranging from 196.3 to 425.5 mg g⁻¹ C for
196 the fPOM and from 368.4 to 449.1 mg g⁻¹ C for the oPOM fractions. Due to the highly fibrous structure of these Cryosols rich
197 in plant residues, fractionation was challenging for some of the samples, leading to one outlier within the fPOM fractions and
198 four outliers within the oPOM fractions. We defined outliers as the measurements laying outside the boxplots' whiskers, thus
199 values lower than 1.5 times the interquartile range below the lower quartile and values higher than 1.5 times the interquartile
200 range above the higher quartile. We excluded these fractions from further calculations as we assume that they point to mineral
201 particles, which we were not able to separate fully from the very fibrous POM structures. The C content of the oPOMs fractions
202 ranged between 61.4 and 344.8 mg g⁻¹ C, the C contents of the clay-sized MAOM fractions between 51.5 and 117.9 mg g⁻¹ C,
203 while silt- and sand-sized MAOM fractions showed the lowest C contents with 2.5 to 11.1 mg g⁻¹ C and 0.7 to 3.1 mg g⁻¹ C,
204 respectively (Fig. 2a).

205 Results for the N content were 5.0 to 19.5 mg g⁻¹ N for fPOM fractions, 3.4 to 23.7 mg g⁻¹ N for oPOM fractions and slightly
206 higher for oPOMs fractions with 4.6 to 26.4 mg g⁻¹ N. The N contents of the clay-sized MAOM fractions ranged between 3.8

207 and 10.1 mg g⁻¹ N, while silt- and sand-sized MAOM fractions contained markedly less N (Fig. 2b). Large POM fractions
208 (fPOM and oPOM) showed a wide variation of C/N ratios with values between 22 and 76 for fPOM and between 18 and 113
209 for oPOM. The values of the oPOMs fractions were clearly lower and had less variability with 13 to 25, while clay-sized
210 MAOM fractions ranged between 11 and 16. Lowest C/N ratios were present in silt- and sand-sized MAOM fractions with 8
211 to 12 and 6 to 19, respectively. Large POM fractions had not only the widest C/N ratios compared to the oPOMs and mineral-
212 associated OM within each soil layer, but also showed the largest variation (Fig. 3, Table S2).

213 The contribution of C and N weighted for the amount of each specific SOM fraction per soil layer showed a great variance in
214 the amount of C and N stored either as POM or MAOM. For C, this ranged between 211.5 and 807.0 mg C per g bulk soil for
215 the large POM fractions (fPOM and oPOM), between 13.7 and 479.7 mg C per g bulk soil for the oPOMs, whereas the clay-
216 sized MAOM ranged between 59.4 and 431.4 mg C per g bulk soil (Table S2).

217 Over all analyzed soil layers, POM fractions accounted for 80 % of the C stock (22.0±9.2 kg C m⁻³), while the MAOM fractions
218 accounted for about 20 % (5.5±2.7 kg C m⁻³). Overall, the fPOM fractions dominated the C stock, with 14.0±4.6 kg C m⁻³
219 representing about half of the total C stock of all analyzed cores and layers. The occluded POM fractions contributed less with
220 2.6±1.1 kg C m⁻³ (oPOM) and 5.4±3.5 kg C m⁻³ (oPOMs). The share of the clay-sized MAOM fractions in the C stock was
221 4.6±2.2 kg C m⁻³, while silt- and sand-sized MAOM fractions played only a subordinate role (Table 1).

222 For the N stock, the contribution of the POM fractions sums up to about 60 % (0.7±0.4 kg N m⁻³) and that of the MAOM
223 fractions to about 40 % (0.5±0.2 kg N m⁻³). The fPOM and oPOM fractions contributed differently to the stock with
224 0.3±0.1 kg N m⁻³ and 0.1±0.1 kg N m⁻³, respectively. The oPOMs and clay-sized MAOM fractions added similarly to the N
225 stock with 0.3±0.2 kg N m⁻³ and 0.4±0.2 kg N m⁻³, but also showed the largest variation. Similar to C stocks, silt- and sand-
226 sized MAOM fractions had a negligible share in the N stocks (Table 1).

227 Although overall the soil C and N storage was dominated by POM, the distribution of POM- vs. MAOM-related C and N
228 varied greatly with depth, with some soil layers showing a dominance of MAOM for C and N storage (Table S2).

229 3.3 Isotopic composition of SOM fractions

230 For POM and clay-sized MAOM fractions, we analyzed the content of stable carbon (¹³C) and nitrogen (¹⁵N) isotopes. With
231 respect to δ¹⁵N, the values differed little between all examined fractions: fPOM (-0.3 to 1.4 ‰), oPOM (0.2 to 2.4 ‰), oPOMs
232 (0.0 to 2.9 ‰) and clay-sized MAOM (-0.4 to 3.4 ‰) fractions with the latter showing the highest values (Table S2). With
233 decreasing C/N ratios, a clear trend towards more negative δ¹³C and lower δ¹⁵N values was demonstrated for all POM fractions
234 (Fig. 4). As shown by PCA (Fig. 5), δ¹⁵N and δ¹³C showed positive dependencies with the C/N ratios. As the deeper soil layers
235 of core 4 were clearly dominated by MAOM with a narrow C/N ratio, the overall δ¹⁵N (0.7 ‰) was lower compared to the
236 other three cores.

237 The δ¹³C values were similar for all fractions and the range of the values and their variability was similar for fPOM (-31.2 to
238 -25.6 ‰), oPOM (-30.6 to -25.3 ‰), oPOMs (-31.5 to -25.0 ‰) and clay-sized MAOM (-31.8 to -24.1 ‰; Table S2). As for
239 δ¹⁵N, also the δ¹³C values of the soil material of core 4 differed from those in the other cores showing clearly lower values.
240 Thus, overall the differences between the cores were larger than the differences between the fractions. Also for the δ¹³C values,
241 a relation to the C/N ratios of all fractions was demonstrated. The C/N ratios of the clay-sized MAOM asymptotically
242 approached a limit when plotted over the δ¹⁵N and δ¹³C, whereas the POM fractions showed a linear increase in the isotope
243 content at higher C/N ratios (Fig. 4).

244 3.4 ¹³C NMR – the molecular level

245 The ¹³C CP-MAS NMR spectra of all examined SOM fractions showed dominant peaks in the O/N alkyl C region. The spectra
246 of both large POM fractions were clearly dominated by the shouldered major peak around 70 ppm and a minor peak around
247 105 ppm. The integration of the spectra fortified the dominance of O/N alkyl C with about 70 % in the fPOM (n=22) and

248 oPOM (n=19) fractions (Table 2). In the regions of carboxyl and alkyl C, small peaks were present, with only a small hump
249 being present in the aromatic C region. The differences between the spectra of the fPOM and oPOM fractions (see Fig. S1)
250 and in their relative composition were only minor, even shoulders and minor side peaks were comparable in the majority of
251 the samples. In contrast, spectra of the oPOMs (n=23) and clay-sized MAOM (n=10) fractions showed pronounced peaks
252 around 30 ppm in the alkyl C region and around 170 to 175 ppm in the carboxyl C region. Throughout all samples, there was
253 a shift from a high percentage of O/N alkyl C in the large POM fractions to a higher percentage of aromatic and alkyl C in
254 oPOMs and clay-sized MAOM fractions (Table 2).

255 To get more differentiated information about the degree of decomposition of the OM, we calculated the a/o-a-ratio for the
256 SOM fractions (Baldock et al., 1997). While fPOM and oPOM fractions revealed identically low values and relatively large
257 standard deviations with 0.2 ± 0.1 , oPOMs and clay-sized MAOM showed clearly higher values with about 0.5. Additional to
258 the a/o-a-ratio, we applied the 70-75/52-57 ratio (Bonanomi et al., 2013) to the SOM fractions and received results consistent
259 with the a/o-a-ratios: fPOM and oPOM showed high values, indicating a low degree of decomposition, while oPOMs and clay-
260 sized MAOM showed very low values. With this ratio, the large POM fractions showed a considerable variance, while the
261 deviation within oPOMs and clay-sized MAOM was marginal (Fig. 6). Figure 7 illustrates the close relation between the C/N
262 ratio of the SOM fractions and the NMR-derived decomposition proxies.

263 By modelling the molecular composition of the SOM fractions using the MMM (Baldock et al., 2004; Nelson and Baldock,
264 2005), we obtained a clear differentiation between the large POM fractions (fPOM, oPOM) and small oPOM and clay-sized
265 OM separates. The composition of the fPOM and oPOM fractions was rather similar: the percentage of carbohydrates (about
266 60 %) was highest and at the same time, the contribution of lipids (about 8 %) was lowest in these fractions (Table 3). Overall,
267 the composition of both large POM fractions was similar with slightly lower amounts of protein and slightly higher amounts
268 of carbonyl in oPOM compared to fPOM. The usage of the MMM revealed once more clear differences between the large
269 POM fractions and oPOMs and clay-sized MAOM. The latter fractions had a lower percentage of carbohydrates (about 40 %),
270 whereas the percentage of protein and lipids was markedly higher. These fractions differed mainly in the proportion of protein
271 and lipids, with clay-sized MAOM containing a larger proportion of protein, but a smaller proportion of lipids (Table 3). The
272 proportion of carbonyl was overall low with high deviations, while the percentage of lignin was rather constant throughout all
273 four examined fractions.

274 The PCA executed on the examined fractions showed slight correlation between the abundance of stable isotopes and NMR-
275 derived decomposition proxies; yet, it confirmed the close relation between fPOM and oPOM and the positioning of oPOMs
276 between large POM and clay-sized MAOM fractions (Fig. 5). The separation of the large POM fractions and oPOMs fractions
277 provided correlation matrices with more details on the correlations (Fig. 8). While the PCA (Fig. 5) already gave a hint, the
278 correlation matrices demonstrated that in the large POM fractions both $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$ were slightly positively correlated with
279 the 70-75/52-57 ratio and negatively correlated with the a/o-a ratio. The positive correlation between $\delta^{13}\text{C}$ and the a/o-a ratio
280 was strong in the oPOMs fractions and the negative correlation between $\delta^{13}\text{C}$ and the 70-75/52-57 ratio was more pronounced,
281 whereas $\delta^{15}\text{N}$ was not correlated with the 70-75/52-57 ratio, but negatively correlated with the a/o-a ratio in the oPOMs
282 fractions.

283 4 Discussion

284 4.1 Cryoturbation determines bulk soil organic matter distribution

285 The projected mean C stock of $27.5 \pm 11.9 \text{ kg C m}^{-3}$ corresponds with those reported in other studies from the Siberian Arctic
286 (cf. Zubrzycki et al., 2014, where the authors demonstrated values between 6.6 and 48.0 kg C m^{-3} in their overview). Besides
287 the large amount of sequestered C, a noteworthy amount of N is stored in permafrost-affected soils. Despite often named as a
288 decisive factor for plant growth in usually N deficient tundra ecosystems (Weintraub and Schimel, 2005), soil N stocks strongly

289 dominated by polymeric organic N might not be related to N availability for plants in the form of amino acids or mineral N.
290 The values for N stocks of permafrost-affected soils reported by other authors (cf. Fuchs et al., 2018; Zubrzycki et al., 2013,
291 demonstrating N stocks ranging between 1.1 and 2.2 kg N m⁻³) are similar to our results of 1.2±0.6 kg N m⁻³.
292 The ample range of the bulk soil C/N ratios points to a wide variance in composition and the degree of decay of the SOM. The
293 C/N ratios notably differed both between the single depth layers and the overall soil cores. The variable bulk soil C/N ratios
294 with depth can be assigned to the translocation of fresh plant-derived OM from top- to subsoils by cryoturbation, leading to
295 specific soil layers which also can contain so called cryoturbated pockets rich in rather less decomposed OM with higher C/N
296 ratios (Kaiser et al., 2007; Krueger et al., 2014). Such an incorporation of OM in the subsoil is also confirmed by the high
297 amounts of POM present in these depth increments dominated by rather fibrous plant residues. Between the analyzed cores,
298 soils from three cores showed wider C/N ratios indicative for the dominance of plant-derived OM, while the fourth core had
299 lower C/N ratios, pointing to a larger amount of microbial-derived OM. Generally, C/N ratios decrease with ongoing
300 decomposition (Kramer et al., 2003) as the proportion of microbial-derived OM with its characteristically low C/N ratio
301 increases after depolymerization of plant-derived organic macromolecules. This goes along with the increased binding of
302 microbial residues to mineral particle surfaces and thus OM becoming less bioavailable (Connin et al., 2001; Vitousek et al.,
303 2002).

304 4.2 POM fractions dominate the C stock stronger than the N stock

305 The large POM fractions (fPOM, oPOM) clearly dominated the C stocks (~ 17 kg m⁻³) in the analyzed Cryosols, whereas small
306 POM (oPOMs) and clay-sized MAOM represented slightly more than one-third of the stored C (~ 10 kg m⁻³). This nicely
307 illustrates that rather large plant-derived fragments (see Fig. 9) dominate the C storage in these OM-rich Cryosols. Especially
308 fPOM, mainly consisting of less decomposed plant material, largely contributes to both C and N stocks. However, in contrast
309 to the C stocks, the oPOMs and clay-sized MAOM fractions act besides fPOM as major contributors to the N stock. A probably
310 accelerated degradation of the fPOM fractions under continued warming could clearly alter the major contribution of the fPOM
311 to the C stock. At the same time, the increased mineralization of fPOM could release vast amounts of N, which are assumed
312 to further foster microbial OM mineralization. This would increase the importance of mineral N cycling such as microbial
313 ammonification-immobilization turnover, compared to organic N cycling. As permafrost-affected soils are often waterlogged
314 during the thawing season with changing oxygen availability and anoxic soil microsites, it can be assumed that in these soils
315 nitrification and denitrification accelerate as well, thereby leading to associated increases in nitrous oxide emissions
316 (Marushchak et al., 2011; Voigt et al., 2017). While a shift from aerobic to anaerobic conditions can hamper the overall
317 decomposition of organic compounds, a shift from anaerobic to aerobic conditions, e.g., when a thawed Arctic soil is exposed
318 to drying conditions, can accelerate decomposition (Keiluweit et al., 2017). With regard to consequences for the role of plants
319 for C and N budgets, some studies point to more plant available N leading to a changing flora and an increasing plant biomass
320 that will possibly be able to counteract the soil C loss caused by thawing (Sistla et al., 2013; Keuper et al., 2017), while others
321 question that gains in biomass will lead to a sufficient compensation of the loss in soil C (Salmon et al., 2016). No matter
322 which of the predictions proves true, as the rather labile fPOM fractions store almost one third of the N in these soils, thawing
323 will lead to a profound change in N budget and N cycling with presumably increasing N bioavailability and increasing
324 importance of mineral N cycling (Voigt et al., 2017; Altshuler et al., 2019).

325 The C and N content, C/N ratio and decomposition proxies based on NMR spectra clearly group the particulate OM fractions
326 into large POM (fPOM and oPOM) and small POM (oPOMs) (Table 1, Fig 3). While the large POM fractions showed rather
327 high C/N ratios, the C/N ratios of oPOMs were considerably lower. This demonstrates that oPOMs represent a discrete type
328 of SOM consisting of smaller, more degraded organic fragments intimately connected with mineral particles, a presumption
329 already made by Wagai et al. (2009) for small particulate OM. We assume that the distinct fibrous structure of the larger POM
330 fractions (see Fig. 9) drives the differentiation into large plant-derived, less decomposed POM and mostly microbial dominated

331 small POM in the studied permafrost-affected soils. The less decomposed fibrous fPOM and oPOM are hot spots for microbial
332 activity and, thus, for the decay of these larger plant structures (Kuzyakov and Blagodatskaya, 2015). These hot spots for the
333 formation of MAOM in Cryosols, plant residues in direct contact with silt- to clay-sized mineral particle surfaces, were already
334 demonstrated on intact Cryosol cross sections using spectromicroscopic imaging (Mueller et al., 2017). The fibrous large
335 POM provides a distinct network that entraps smaller POM and MAOM particles and thereby retains especially the small POM
336 fraction restricting its bioaccessibility. The small OM particles (oPOMs) act as a linking element between the fresh less
337 decomposed plant residues (fPOM, oPOM) and the clay-sized MAOM, to our knowledge a phenomenon not described before
338 in permafrost-affected soils.

339 **4.3 Isotopic composition demonstrates the fate of labile and recalcitrant organic compounds from POM to MAOM and** 340 **the importance of biological N fixation**

341 The $\delta^{13}\text{C}$ values of all samples were well within the range of those obtained for SOM derived from plants with a C_3 metabolism
342 (Sharp, 2007). During the decomposition of plant-derived material, the changes in $\delta^{13}\text{C}$ values are usually subtle and are
343 determined by a variety of factors, especially by the composition of the original plant material (Ågren et al., 1996).
344 Nevertheless, SOM compounds rich in presumably more recalcitrant macromolecules, like lignin or aromatic hydrocarbons,
345 have lower $\delta^{13}\text{C}$ values than labile compounds, like carbohydrates (Schmidt and Gleixner, 1997). Besides the $\delta^{13}\text{C}$ value of
346 the original plant litter input, soil $\delta^{13}\text{C}$ values depend on several factors like climate, soil texture and major soil processes (Nel
347 et al., 2018). We found clear positive correlations between the decomposition stage (a-oa ratio, 70-75/52-57 ratio, C/N ratio)
348 of the large POM fractions and $\delta^{13}\text{C}$ (Fig. 8), which nicely illustrates the initial decomposition stage of the large POM with a
349 relative dominance of labile OM rich in carbohydrates. This is supported by the negative correlation between $\delta^{13}\text{C}$ and the
350 rather recalcitrant lipids (aliphatic C based on NMR spectra) both in the large POM and oPOMs fractions (Fig. 8). This
351 correlation reflects well the relative increase in aliphatic compounds with progressing decomposition (Benner et al., 1987),
352 which corresponds to the fact that aliphatic compounds commonly show a lower ^{13}C abundance (Schmidt and Gleixner, 1997).
353 Thus, although we demonstrate clear mechanistic differences between large (fPOM, oPOM) and small POM (oPOMs) with
354 respect to C sequestration, the decomposition in both OM pools follows the same fundamental principles. The positive
355 correlation of the $\delta^{13}\text{C}$ in large and small POM with the bulk soil C/N ratios demonstrates the dominance of the POM-C pool
356 for the bulk soil C pool. Thus, the positive correlation between ^{13}C and bulk soil C/N reflects the larger amount of
357 undecomposed plant residues in the large POM in some soil horizons, while it demonstrates an increased amount of aliphatic
358 moieties in the small POM for other soils horizons. Thus, the overall elemental composition of the bulk soils can directly be
359 linked to the ^{13}C isotopic composition of the fresh and more decomposed POM fractions.

360 In Arctic ecosystems, N_2 fixation is known as the major N input into ecosystems (Granhall and Selander, 1973; Rousk et al.,
361 2017, 2018) with N fixation rates between 1 and 29 kg N $\text{ha}^{-1} \text{a}^{-1}$, depending on which N_2 fixing species (e.g. free-living or
362 moss-associated cyanobacteria) is dominating (Rousk et al., 2017). Furthermore, Arctic soils are known to be dominated by
363 organic N cycling rather than mineral N cycling (Hobbie and Hobbie, 2008), while atmospheric N deposition is low in this
364 region (Hole et al., 2009). The soil $\delta^{15}\text{N}$ values we found are consistent with $\delta^{15}\text{N}$ values reported for bacterial N_2 fixation as
365 N source (Casciotti, 2009; Hoefs, 2015), but also similar to values reported for plant litter-derived OM (Connin et al., 2001).
366 Other studies reported stable or increasing $\delta^{15}\text{N}$ values with advancing decomposition (e.g. Ågren et al., 1996; Connin et al.,
367 2001). Whether increases in $\delta^{15}\text{N}$ occur with enhanced decomposition and N turnover is largely depending on gaseous N loss
368 processes, such as ammonia volatilization, and nitrous oxide (N_2O) and dinitrogen (N_2) losses through nitrification and
369 denitrification, as highest isotope fractionation factors are reported for these processes, enriching the heavier ^{15}N isotope in
370 soil, while ^{14}N is preferably lost to the atmosphere (Bedard-Haughn et al., 2003; Nel et al., 2018). By illustrating decreasing
371 $\delta^{15}\text{N}$ with increasing OM decomposition, our results seem to contradict this presumption. Therefore, we assume that biological
372 N_2 fixation is a decisive control of $\delta^{15}\text{N}$ in the studied soils, as also recently shown for permafrost-affected soils of Tibet

373 (Chang et al., 2017). Such a dominant role of biological N₂ fixation in regulating δ¹⁵N requires that nitrification/denitrification
374 and associated gaseous N losses as well as atmospheric inputs are not significant for the studied soils, which is in general
375 agreement with the N cycle paradigm for the High Arctic (Schimel and Bennett, 2004).

376 4.4 Distinct differences in chemical composition from large POM to MAOM

377 By using NMR spectroscopy, we were able to differentiate further between large POM fractions (fPOM, oPOM) and oPOMs
378 and clay-sized MAOM, which also allowed a nice clustering of these materials into distinctly different OM pools with respect
379 to assumed bioavailability (see the representative example in Fig. S1). The NMR spectra of both large POM fractions were
380 clearly dominated by a major peak around 70 ppm and a minor peak around 105 ppm, both relating to polysaccharides (Koelbl
381 and Koegel-Knabner, 2004). This was well reflected by the calculated high amounts of carbohydrates, the high 70-75/52-57
382 ratio and low a-oa ratios, which all point to the rather labile undecomposed nature of the larger OM particles.

383 Based on both decomposition proxies pointing in the same direction, we assume a high potential bioavailability for both large
384 POM fractions (fPOM, oPOM). Interestingly, when comparing the decomposition proxies **between** these **two** POM fractions
385 per single soil layer (Table S3), they indicate a less pronounced decomposition for oPOM in most of the samples. These
386 patterns deviate from what is commonly observed in temperate soils, i.e. an increased degree of decomposition with **decreasing**
387 **POM size and advancing** aggregate occlusion (fPOM<oPOM<oPOMs) (Mueller and Koegel-Knabner, 2009). We assume that
388 this demonstrates a reduced bioaccessibility (accessibility of OM by microorganisms and enzymes) of oPOM, which is
389 encrusted by mineral particles, leading to a reduced degree of decomposition of the occluded as compared to the free POM.
390 Thus, the initial microbial decomposition of the surfaces of fresh plant residues (fPOM) driven by microbial decay leads, in
391 part, to the formation of oPOM due to the association with minerals glued to the POM surfaces by microbial residues (see
392 **Mueller et al., 2017**), e.g. extracellular polymeric substances (Tisdall and Oades, 1982; Schimel and Schaeffer, 2012; Costa et
393 al., 2018). We thus demonstrate soil structure formation in Cryosols as driven by microbial activity **via the excretion of**
394 **extracellular polymeric substances** at POM surfaces leading to the stabilization of rather labile POM without necessarily
395 leading to OM with high degrees of decomposition.

396 In contrast to the large POM fractions, the NMR spectra of oPOMs and clay-sized MAOM were dominated by peaks around
397 30 ppm representing long-chain structured aliphatic C derived for example from macromolecules like cutin or suberin (Koegel-
398 Knabner, 2002). **Irrespective of the high amount of alkyl C**, the dominating group of compounds as calculated by the MMM
399 were carbohydrates for both **fractions, oPOMs and clay-sized MAOM** (Table 3). Both fractions also showed distinct peaks
400 around 170 to 175 ppm, representing partly esterified carboxyl groups and amide C that stems predominantly from proteins
401 (Koelbl and Koegel-Knabner, 2004). **Especially the** clay-sized MAOM showed distinctly higher amounts of protein C (Table
402 3) compared to all POM fractions, which corroborates the preferential association of N-rich microbial residues at mineral
403 surfaces (Kleber et al., 2007; Kopittke et al., 2018, 2020). This highlights the fact that the association of OM with mineral
404 surfaces follows the same mechanisms as previously described for temperate soils (Kleber et al., 2007). In the specific context
405 of the studied permafrost-affected soils, the oPOMs represented a kind of passage fraction. Although it clusters with the clay-
406 sized MAOM in the PCA (Fig. 5), the small POM links to the large POM fractions as illustrated in Fig. 7. Thus, in contrast to
407 the larger, relatively undecomposed plant residues, lipids and proteins contribute noteworthy to the oPOMs fractions and the
408 fine MAOM of the clay-sized fraction. This clearly points to the increased amount of microbial-derived compounds in these
409 fractions, as already stated above with respect to the C/N ratio and δ¹⁵N. Thus, the MAOM in these soils is dominated by
410 microbial-derived SOM rich in biologically fixed N. **As demonstrated in the PCA and shown** in Fig. 9, oPOMs is represented
411 by degraded plant residues, fungal hyphae and amorphous material which can be assumed to mainly represent microbial
412 necromass (Miltner et al., 2012). The PCA demonstrated that oPOMs represents a linking fraction between the initial plant
413 residues of the larger POM fractions and the microbial OM dominated clay-sized MAOM (Fig. 5). However, it falls short to

414 assume that all OM in POM fractions is fast-cycling and all MAOM is slow-cycling (Torn et al., 2013). Our results underline
415 that the large and rather undecomposed POM fractions rich in carbohydrates might act as a highly bioavailable substrate in a
416 warmer future. This means, when active layers deepen and the larger POM fractions become accessible to microorganisms,
417 oPOMs and clay-sized MAOM may represent a C pool that is less bioavailable and thus presumably more persistent. Besides
418 the demonstrated occlusion of particulate OM, we were able to show the quantitative importance of MAOM for the C storage
419 in these High Arctic soils. Thus, the oPOMs and clay-sized MAOM representing altered and microbially transformed OM
420 pools, could gain importance regarding C storage under further thawing conditions in soils of the Arctic. Besides the
421 importance for C sequestration, the high amount of biologically fixed N of the MAOM may also be released and foster the
422 microbial decay of the high amounts of C stored in larger POM fractions (Jilling et al., 2018).

423 5 Conclusions

424 Employing physical fractionation and molecular level analyses, we show that the SOM fractions that contribute with about
425 17 kg C m⁻³ for more than 60 % of the C stocks in the investigated Arctic soils are presumably highly labile and vulnerable to
426 environmental changes. In the face of global warming, most of this labile C, currently protected from decomposition by low
427 temperatures, will be prone to mineralization, with severe consequences for the C stocks in Arctic soils. Our results clearly
428 support our hypotheses that the major amount of C and N is stored as POM, with large POM resembling the composition of
429 the initial plant litter. With increasing decomposition and, thus, decreasing size of the OM particles, the material gets microbial
430 transformed, which leads to MAOM dominated by microbial residues, as indicated by C/N ratios, ¹⁵N abundance and chemical
431 composition. Organic C stored in small occluded POM and clay-sized MAOM that accounts with 10 kg C m⁻³ for about 40 %
432 of the C stock currently, will likely dominate the C pools as it is less vulnerable to increased mineralization in Arctic soils in
433 a warmer future. Small occluded POM was found to be acting as a transitional C pool between the larger POM fractions and
434 MAOM, demonstrating the importance of the interfaces between particulate plant residues and the fine mineral fraction as hot
435 spots for microbial activity and thus MAOM formation. Using δ¹⁵N as proxy for N balances, we demonstrate the important
436 role of N inputs by biological N fixation with increasing contribution to organic matter N at higher degree of decomposition,
437 while gaseous N losses appear to be of minor importance. The large soil organic N stocks however might be at risk in future,
438 as with about 0.4 kg N m⁻³ one third of the N is present in presumably bioavailable SOM fractions, which could lead to
439 increases in mineral N cycling and associated N losses under the auspices of global warming.

440
441 *Data availability.* The data that support the findings of this study are available from the corresponding author upon request.

442
443 *Supplement.* Supplementary material is available.

444
445 *Author contributions.* IP conducted analyses in the laboratory (elemental analysis, NMR measurements) and wrote the
446 manuscript. SZ was responsible for the sampling and the selection of the respective cores. LCZF conducted analyses in the
447 laboratory (fractionation, elemental analysis, NMR measurements). FB conducted stable isotope measurements. CWM
448 developed the design of the study. IP, MD, GA and CWM were responsible for data evaluation and the interpretation of results.
449 All authors discussed the data and results and contributed to the final form of the manuscript.

450
451 *Competing interests.* The authors declare that they have no conflict of interest.

452
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Table 1: C and N stocks (projected to 1 m soil depth) and C/N ratios of the SOM fractions. Given are mean values and the standard deviation of free (fPOM), occluded (oPOM) and small occluded (oPOMs) particulate organic matter and of different sized mineral-associated organic matter (MAOM).

SOM fraction	C stock kg C m ⁻³	N stock kg N m ⁻³	C/N ratio
fPOM	14.0±4.6	0.3±0.1	46±16
oPOM	2.6±1.1	0.1±0.1	51±22
oPOMs	5.4±3.5	0.3±0.2	17±3
clay-sized MAOM	4.6±2.2	0.4±0.2	13±6
silt-sized MAOM	0.7±0.4	0.1±0.0	10±1
sand-sized MAOM	0.2±0.1	0.0±0.0	19±0
sum	27.5±11.9	1.2±0.6	721

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Table 2: Relative chemical composition of SOM fractions obtained by ¹³C CP-MAS NMR spectroscopy and decomposition proxies (a/o-a ratio and 70-75/52-57 ratio). Given are mean values and the standard deviation of free (fPOM), occluded (oPOM) and small occluded (oPOMs) particulate organic matter and of clay-sized mineral-associated organic matter (MAOM).

SOM fraction	relative chemical composition ¹				a/o-a ratio ²	70-75/52-57 ratio ³
	alkyl C	O/N alkyl C	aromatic C	carboxyl C		
fPOM	13.3±5.0	70.2±7.6	11.6±2.5	4.9±1.9	0.2±0.1	5.6±2.1
oPOM	12.5±6.0	68.5±8.4	12.2±4.0	6.5±2.6	0.2±0.1	7.4±3.3
oPOMs	25.2±5.9	52.1±6.3	14.0±3.0	8.5±2.2	0.5±0.2	2.6±0.3
clay-sized MAOM	24.0±2.6	49.6±3.4	15.1±1.8	11.2±3.7	0.5±0.1	2.1±0.3

¹ Relative chemical composition determined by integration of the following chemical shift regions: -10 to 45 ppm (alkyl C), 45 to 110 ppm (O/N alkyl C), 110 to 160 ppm (aromatic C) and 160 to 220 ppm (carboxyl C).

² Ratio of alkyl C and O/N alkyl C according to Baldock et al. (1997).

³ Ratio of the chemical shift regions 70 to 75 ppm and 52 to 57 ppm according to Bonanomi et al. (2013).

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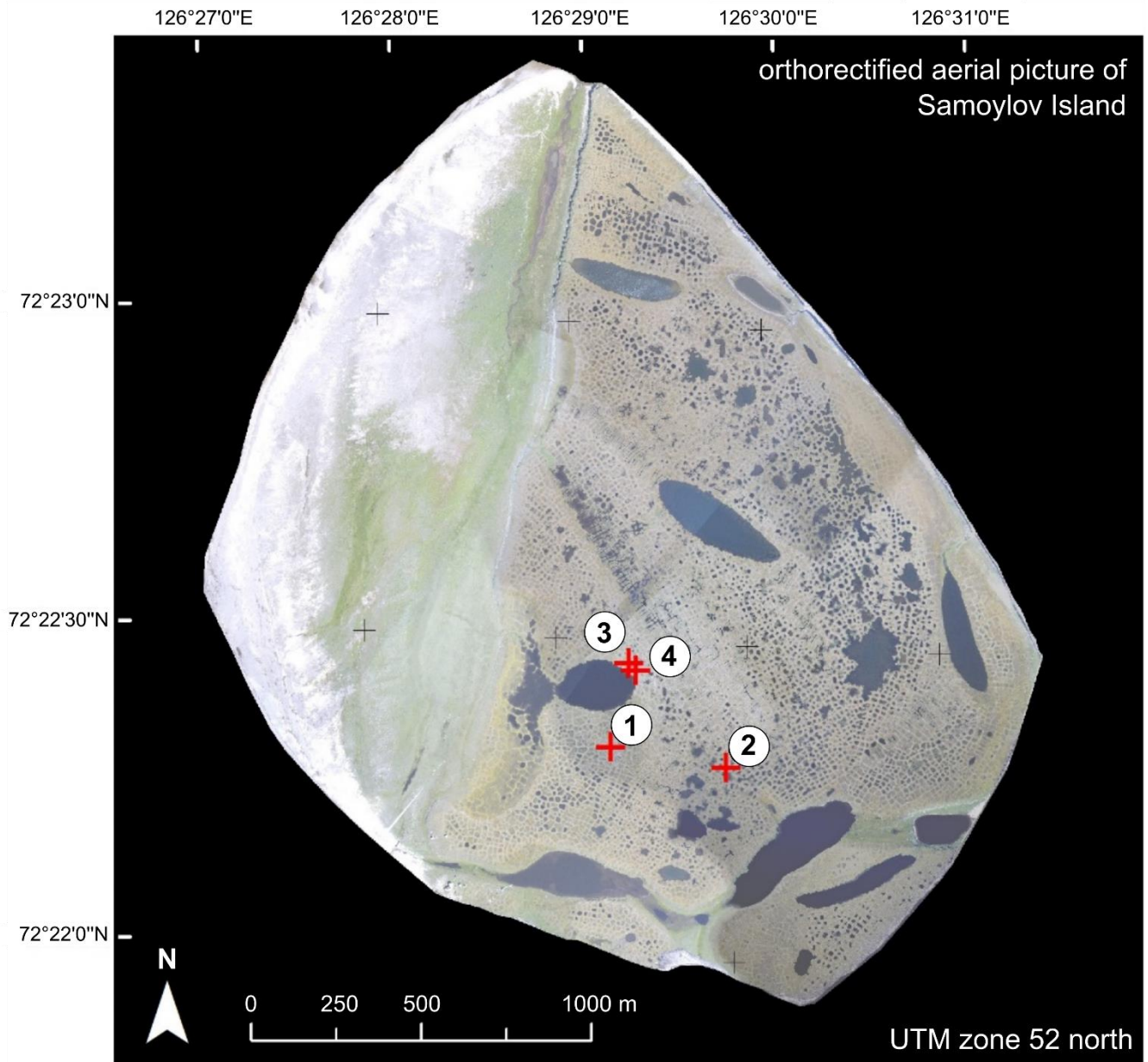
Table 3: Results from the molecular mixing model with data obtained by ¹³C CP-MAS NMR spectroscopy and calculated according to the molecular mixing model by Baldock et al. (2004) and Nelson and Baldock (2005); 5 component model (-char) with N:C constrained. Given are mean values and the standard deviation of free (fPOM), occluded (oPOM) and small occluded (oPOMs) particulate organic matter and of clay-sized mineral-associated organic matter (MAOM).

SOM fraction	molecular mixing model				
	carbohydrate	protein	lignin	lipid	carbonyl
fPOM	61.4±8.0	6.4±2.8	21.7±3.2	8.2±3.9	2.3±1.9
oPOM	60.9±9.9	6.4±3.8	21.2±5.5	7.6±4.8	3.9±4.0
oPOMs	41.9±5.9	17.0±3.2	21.3±4.3	18.9±5.1	0.9±2.0
clay-sized MAOM	41.1±2.9	22.6±2.0	21.2±3.7	13.5±2.8	1.6±2.3

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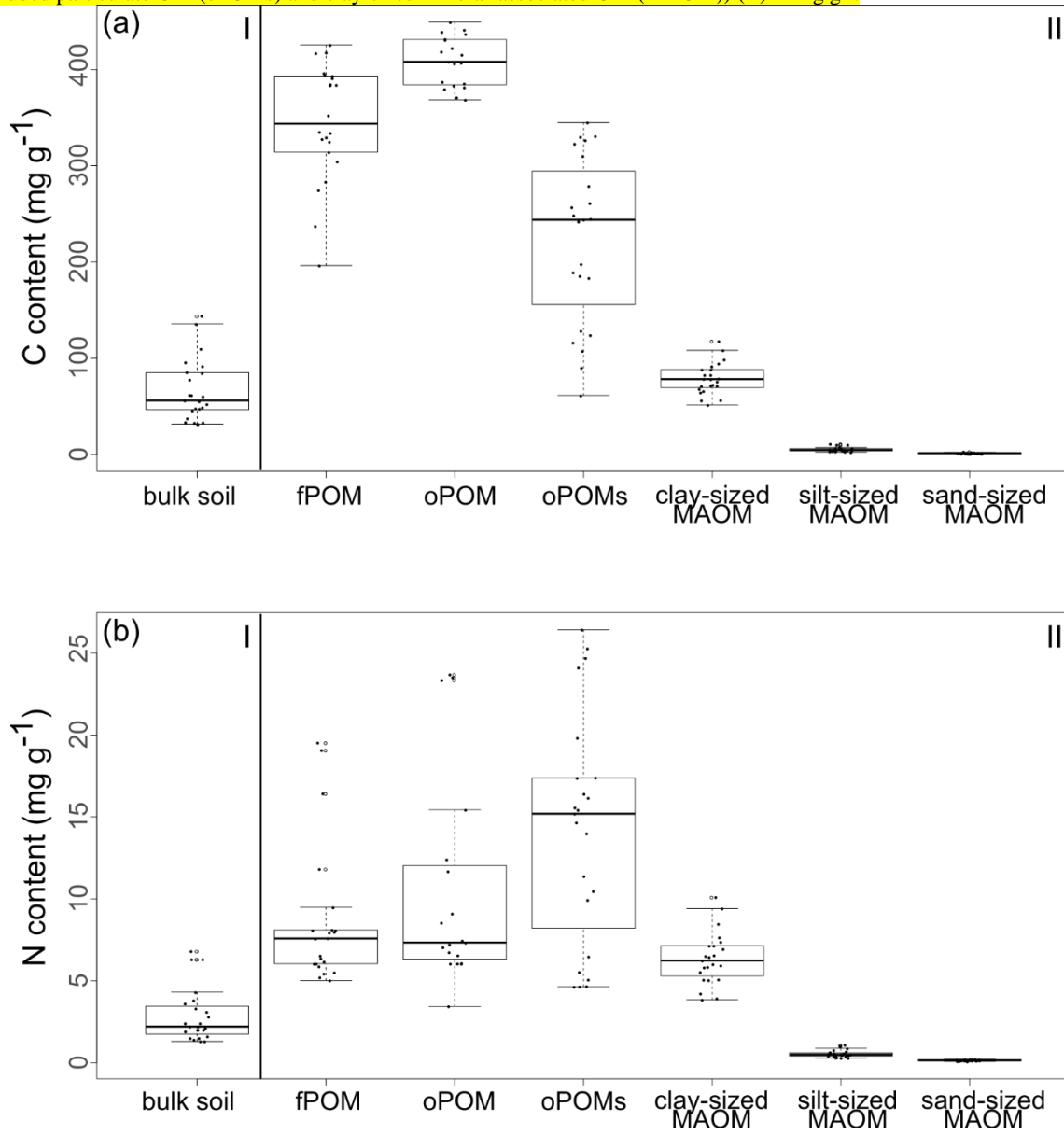
Figure 1: On this aerial image of Samoylov Island, the separation between the floodplain in the west (with the white unvegetated sandy sediment) and the Holocene terrace on the eastern part (with blue-grey spots indicating shallow water and larger water bodies). Red crosses indicate the sampling sites and the identification numbers of the cores are given (Boike et al., 2012).



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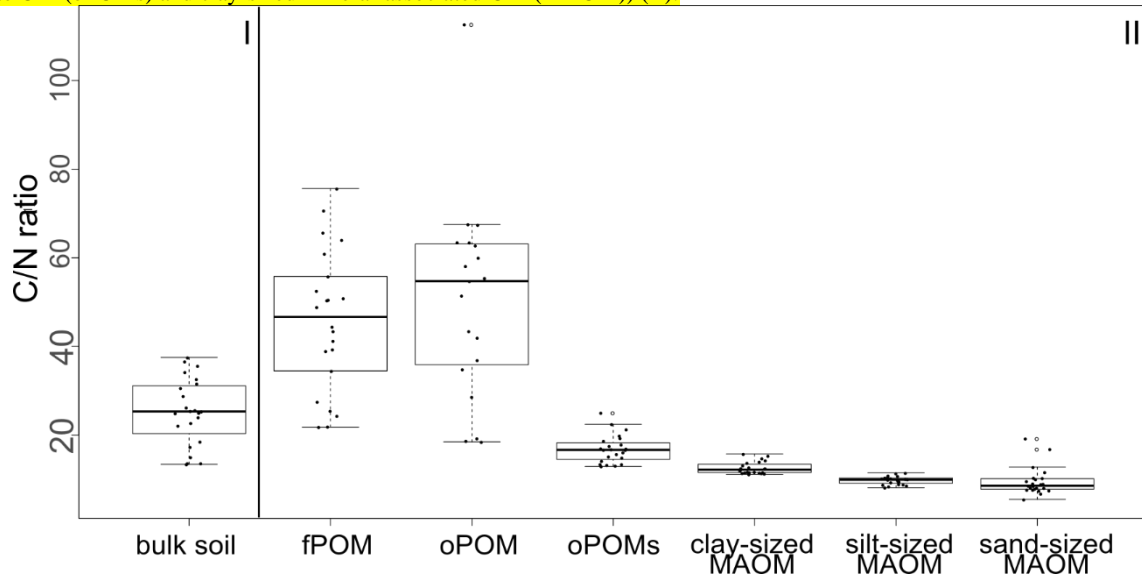
Figure 2: The content of C (a) and N (b) of bulk soils (I) and SOM fractions (free particulate OM (fPOM), occluded particulate OM (oPOM), small occluded particulate OM (oPOMs) and clay-sized mineral-associated OM (MAOM)) (II) in mg g^{-1} .



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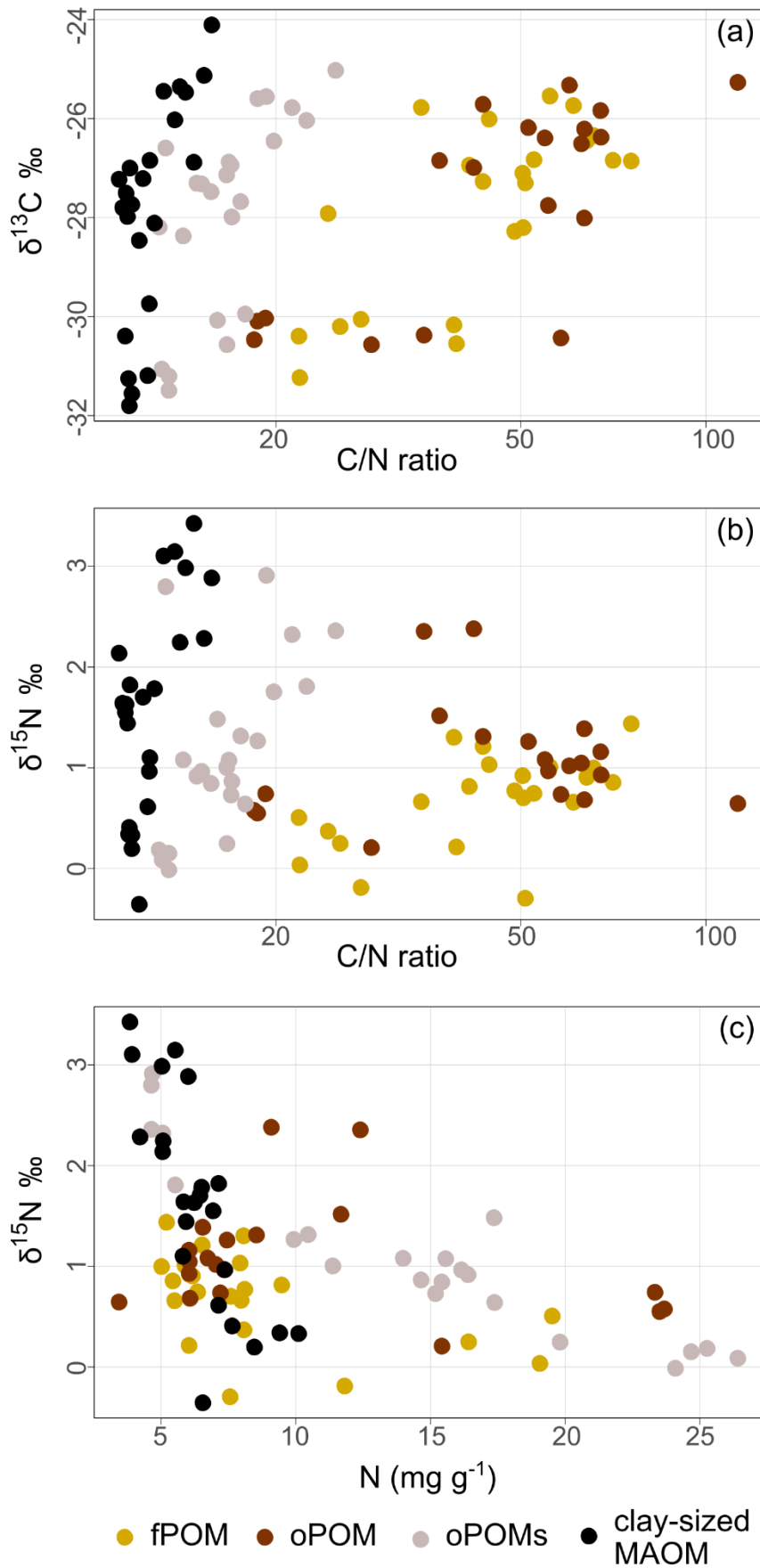
Figure 3: C/N ratios of bulk soils (I) and SOM fractions (free particulate OM (fPOM), occluded particulate OM (oPOM), small occluded particulate OM (oPOMs) and clay-sized mineral-associated OM (MAOM) (II)).



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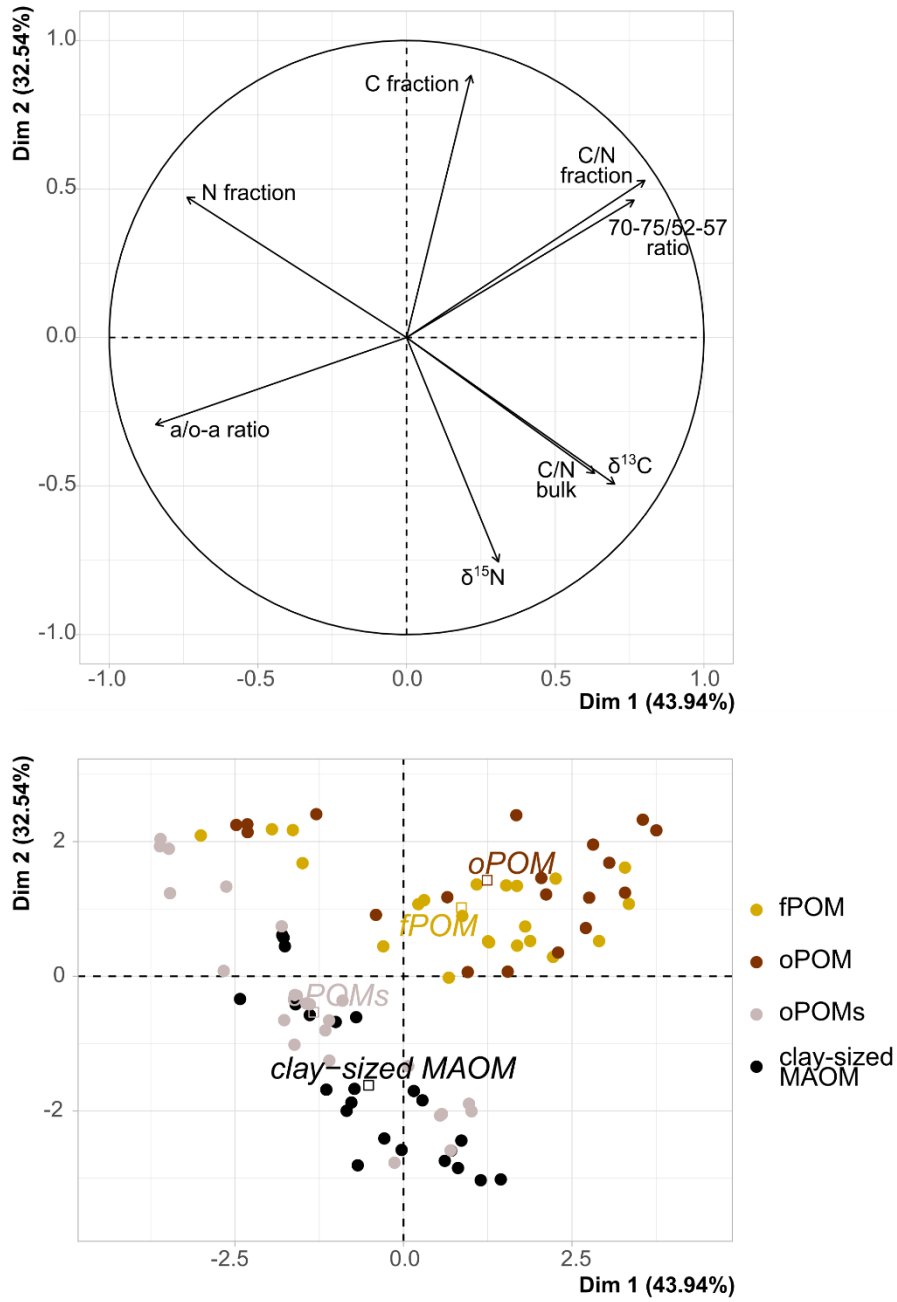
Figure 4: Natural abundance of $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ plotted against the C/N ratios, and the $\delta^{15}\text{N}$ values plotted against the N content of SOM fractions (free particulate OM (fPOM), occluded particulate OM (oPOM), small occluded particulate OM (oPOMs) and clay-sized mineral-associated OM (MAOM)): The values of $\delta^{13}\text{C}$ (‰ relative to V-PDB) (a) and the values of $\delta^{15}\text{N}$ (‰ relative to air N_2) (b) in relation to the C/N ratio (log-converted) of SOM fractions and $\delta^{15}\text{N}$ (‰ relative to air N_2) plotted against N (in mg g^{-1}) content of the SOM fractions (c).



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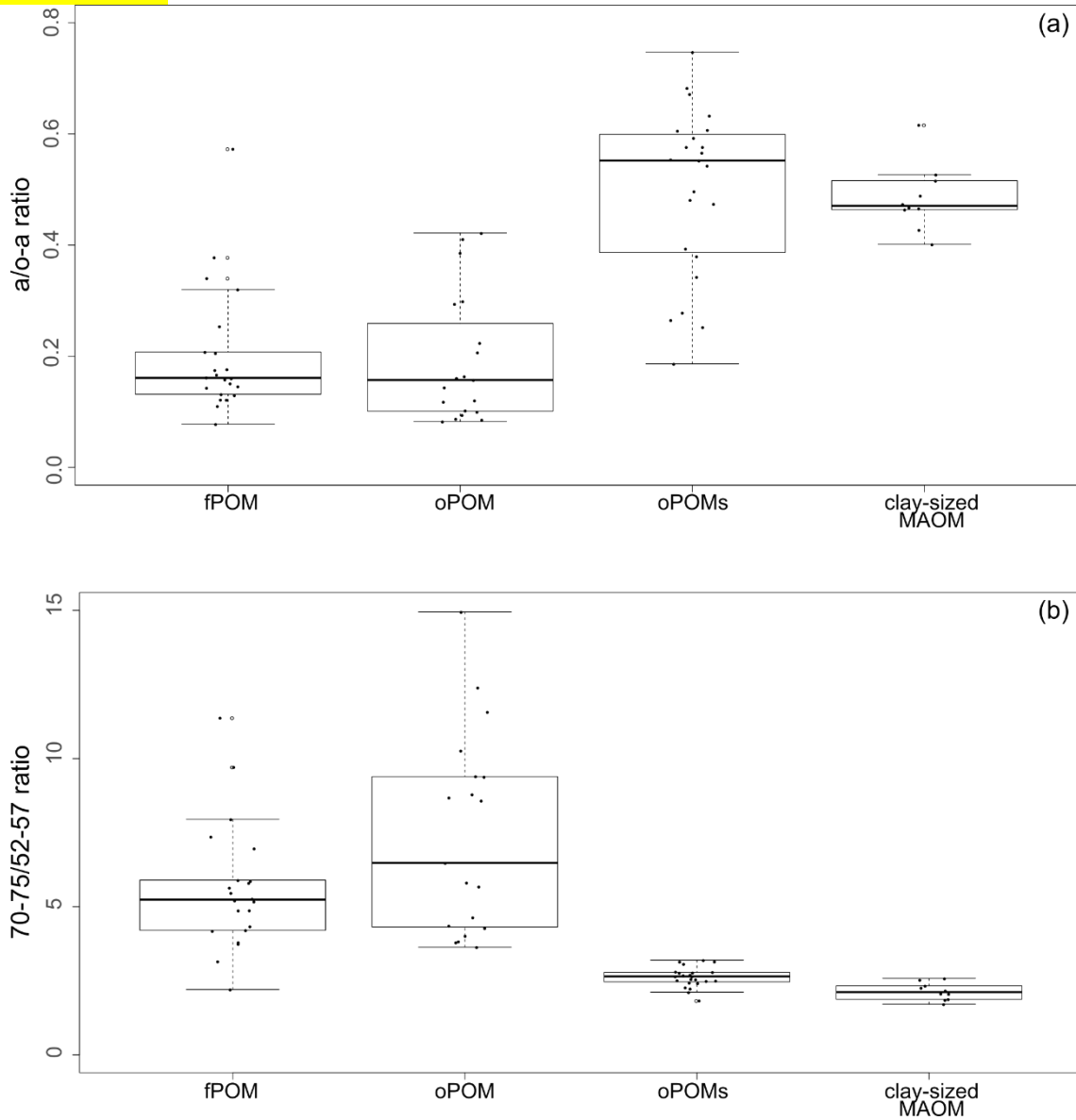
Figure 5: Principal Component Analysis (PCA) of $\delta^{13}\text{C}$ (‰ relative to V-PDB), $\delta^{15}\text{N}$ (‰ relative to air N_2), C and N content of the SOM fractions (free particulate OM (fPOM), occluded particulate OM (oPOM), small occluded particulate OM (oPOMs) and clay-sized mineral-associated OM (MAOM)), C/N ratio of fractions and of bulk soils and ^{13}C CP-MAS NMR-derived decomposition proxies (a/o-a ratio and 70-75/52-57 ratio).



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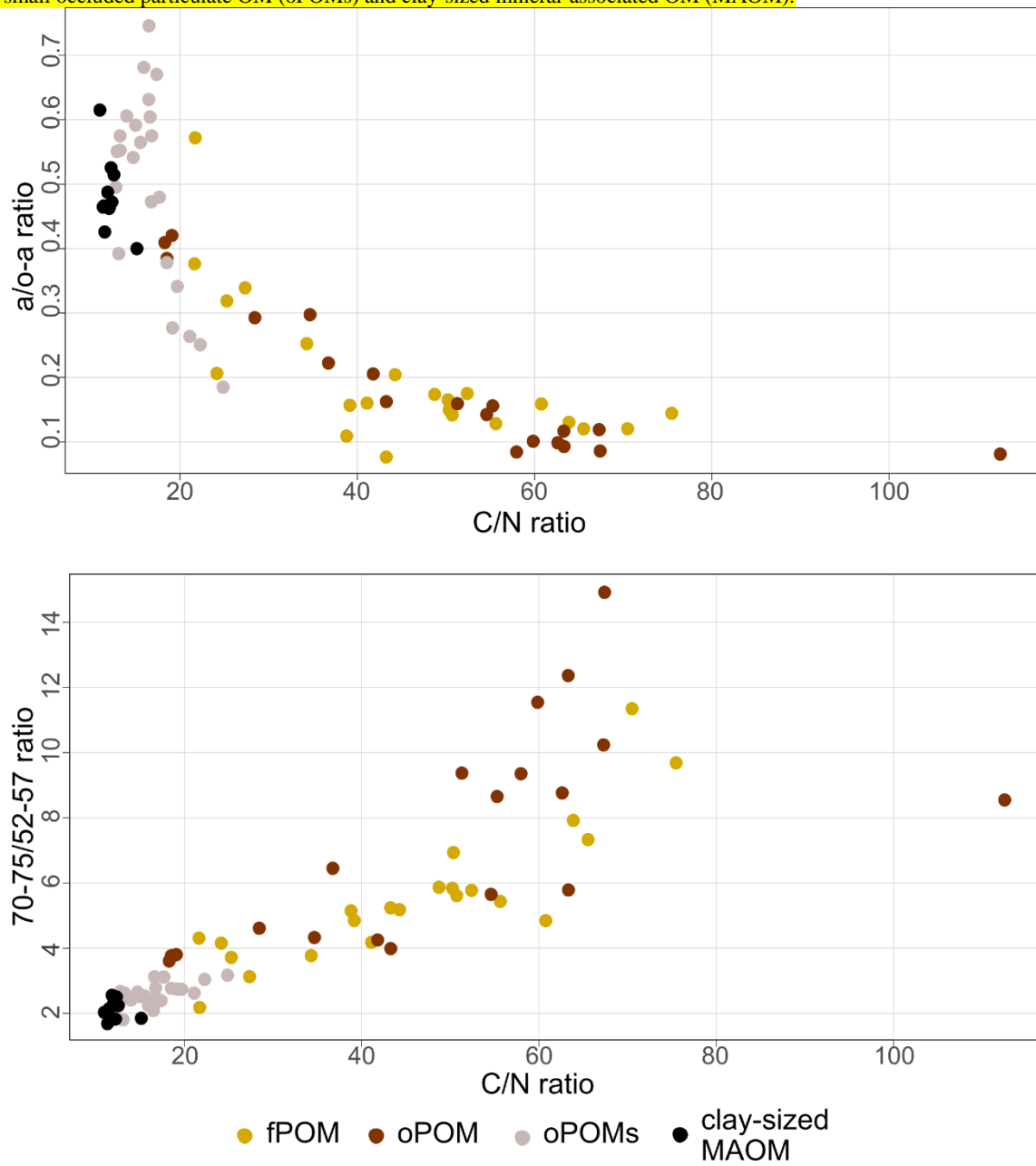
Figure 6: Decomposition proxies obtained by ^{13}C CP-MAS NMR spectroscopy for specific SOM fractions: Both a/o-a ratio (a) and 70-75/52-57 ratio (b) of SOM fractions demonstrate the similarity of large particulate OM fractions – free and occluded POM (fPOM and oPOM) – and the conjunctive characteristics of the small occluded particulate OM (oPOMs) fractions that links large POM fractions and the clay-sized MAOM fraction.



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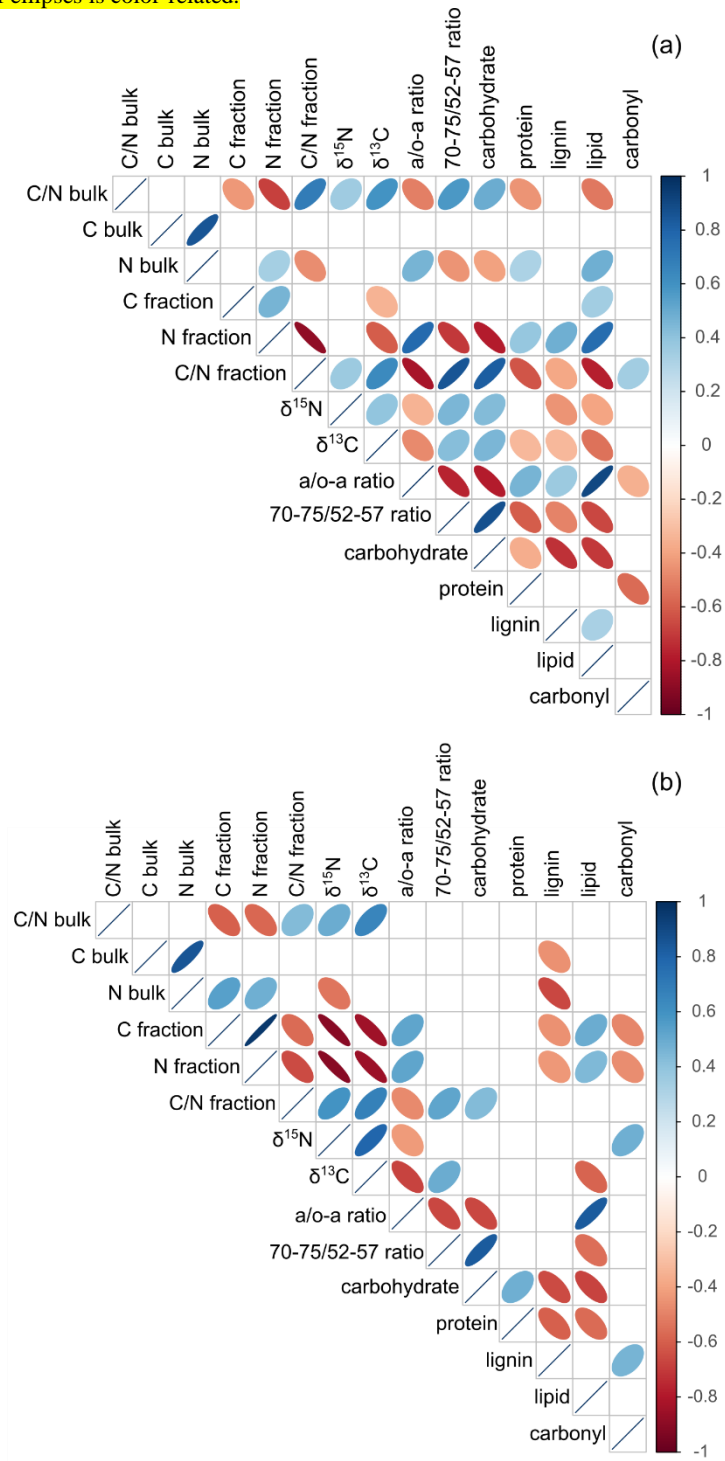
Figure 7: Relation between decomposition proxies and C/N ratio of distinct SOM fractions: ^{13}C CP-MAS NMR spectroscopy-derived decomposition proxies a/o-a ratio (a) and 70-75/52-57 ratio (b) vs. C/N ratio for free particulate OM (fPOM), occluded particulate OM (oPOM), small occluded particulate OM (oPOMs) and clay-sized mineral-associated OM (MAOM).



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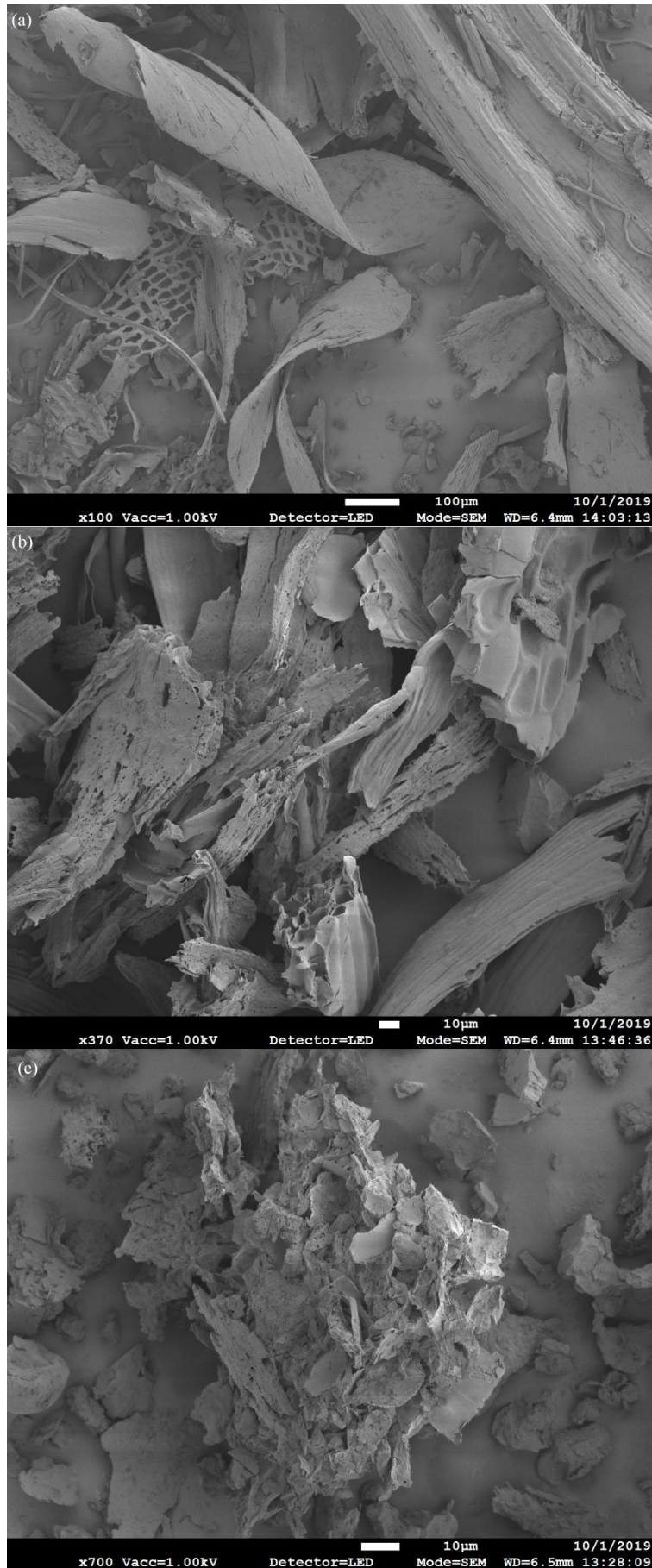
Figure 8: Correlation matrices of POM fractions: The large POM (oPOM and fPOM) fractions (a) show different correlations compared to oPOMs fractions (b). The more intense the color and the smaller the ellipse, the stronger the correlation: blue indicates a positive, red a negative correlation, direction of ellipses is color-related.



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Figure 9: Scanning electron micrographs of particulate organic matter fractions: While fPOM fraction (a) and oPOM fraction (b) consist mainly of larger particles of plant-derived litter with clearly visible cell structures and only minor indications for initial decomposition, the image of the oPOMs fraction (c) of the same sample clearly reveals the intricate association of small organic particles and silt- and clay-sized soil minerals.



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