Reply to Reviewer#3

While the concept presented in this manuscript is highly compelling and represents a potentially novel insight when viewed in broad strokes, there are several shortcomings to both the experimental approach and its interpretations as well as to the manuscript itself.

We thank the reviewer for pointing out that the concept presented in this manuscript is highly compelling, for acknowledging the novelty of the study, and for providing constructive comments on how to improve the manuscript. The reviewer's comments helped us to see that more explanations of the results and the approach are needed given that this is a novel approach.

The manuscript:

The word "very" in the title should be omitted. Adverbs are challenging in a quantitative domain. that is, what does "very" mean in terms of N concentration? The word is ambiguous and doesn't provide additional information. While mineral associated organic matter tends to have a low C:N ratio, the organic matter in the fractions studied here has an even lower C:N ratio, which is in the range of the C:N ratios of proteins. Therefore, we used the adverb "very". However, we understand that this is not clear if the if the title is considered in isolation. Therefore, we will remove the word "very" from the title of the revised manuscript, as suggested by the reviewer.

This is a short communication, that is actually shorter than it appears because a great deal of its content is repeated several times. When the repetition is removed, there is not much left. This is unfortunate because there are many angles that need to be unpacked and elaborated in the interpretation of the results before the reader might have much confidence in the conclusions. We will extend the Discussion of the manuscript, following the suggestions made below. The reviewer's comments helped us to see that more explanations are needed given that this is a novel approach. Specifically, we will extend the discussion about the coupling and decoupling of carbon and nitrogen in soil organic matter (as detailed below). In addition, we will also explain explicitly that our conclusions about soil organic nitrogen being on average old are not based on the oxidation rate of the organic matter (see below). We will also carefully check that there are no repetitions in the manuscript.

While the manuscript is readable, it could use some fine-tuning to make the English more natural and fluid. We will fine-tune the language of the manuscript, as suggested.

No paragraph should be a single sentence. Too many sections are broken up into unnecessarily into multiple too-short paragraphs. We will remove some of the line breaks to increase readability, as suggested.

The authors frequently use a sentence structure that requires the use of "respectively" throughout the manuscript. The structure "A, B, C are related to X,Y,Z, respectively" requires the reader to perform mental matching and disrupts the fluidity of reading. Instead, the sentence could be structured as "A is related to X, B to Y, and C to Z." This improves the reading experience. We will carefully check all sentences that contain the word respectively, and we will improve the readability of the manuscript.

The authors need to be careful about distinct diction for "fraction" and "pool". Generally, "pool" is used to refer to a non-tangible, non-measured, modeled collection, while "fraction" is a tangible"/"real", measured, physical entity. We agree with this comment. The word pool is used four times in the manuscript. We will replace the word pool by the word fraction throughout.

The methods section could be re-structured to distinguish between characterization and analysis. That is, measurements of total soil organic C and N and soil texture might come first in the order because hey are basic characterization measurements. These would then be followed the thermal fractionation procedure (perhaps as a separate subsection) and the isotopic analysis. The descriptions of the C and N analysis and the isotope analysis follow the description of the fractionation in the Methods section since the analyses are also conducted on the fractions. If we adopted the order suggested by the reviewer, we would have to repeat the description of the analytical methods twice.

The section on statistical analysis needs elaboration. What variables are being examined? Which are dependent and which are independent? In the Method section, we will add the information that C and N are dependent variables and clay content and pH are independent variables in the regression analysis, as suggested.

Conceptual/Scientific/Experimental:

The authors cite Sanderman & Grandy (2020) and Stoner et al (2023) in the background on thermal analysis coupled with isotopic analysis, but while the method might be slightly different, these studies are pre-dated by those of Lopez-Capel et al. (2005, 2006) that coupled ramped combustion with 13C and Plante et al. (2013) that coupled ramped combustion to 14C. We thank the reviewer for pointing out these previous studies. We will add these older references to the manuscript.

If I understand the approach correctly, samples are subjected to ramped combustion to a fixed temperature and the residue is used for subsequent analyses. But what happens to the samples once the threshold temperature is reached? Are they removed from the instrument immediately? Whta is the cooling rate? How long are they exposed to elevated temperatures? The temperature was increased at a rate of 5°C per minute until either 325°C or 400°C was reached (as stated in lines 85 and 86). We will add the information about cooling to room temperature (after reaching the target temperature) at room temperature, as suggested.

The authors seem to implicitly treat C and N are independent pools in some arguments, or implicitly link the behaviour of the two in other arguments. Which is it? That is, just because the C is old (indicated by 14C) what makes the N old (indicated by ?). Alternative, are compounds rich in N-containing known to be more thermostable? The interpretation as written seems to confound biochemical-thermal stability with mineral-association-stability, (both) in relation to thermal stability. There is a lot that needs to be unpacked here before there can be any confidence in the conclusions. The two soil fractions obtained in this study have a molar total organic carbon-to total organic nitrogen (TOC:TON) ratio of 7.5 and 4.8. Given the very low TOC:TON ratios, it seems highly likely that the age of the carbon in these fractions equals the age of the nitrogen in this fraction because the low TOC:TON ratio indicates that all carbon in this fraction is covalently bound to nitrogen. We agree with the reviewer that this needs to be explained in more detail in the manuscript, and we will revise the manuscript accordingly.

Our conclusion about soil organic nitrogen being on average old is not based on the thermostability of the soil organic matter, but on the radiocarbon data. We discuss the reason why old soil organic matter is thermostable, but out conclusions are not based on the thermostability soil organic matter. In the present version of the manuscript, we refer to the organic matter fractions as the thermostable organic matter fractions. We understand that this might be misleading, and we will change this in the revised manuscript to avoid misunderstandings. In the revised version of the manuscript, we will call the two fractions old

fraction and intermediate fraction, similarly as in other studies about soil organic matter fractions.

We found that a larger percentage of the soil total organic nitrogen than of the soil total organic carbon is found in the two fraction that have a low $\Delta^{14}C$. This indicates that a larger part of the soil total organic nitrogen than of the soil total organic carbon is old. In this sense soil organic N and organic C are to some extent decoupled. We will explain this in more detail in the revised version of the manuscript.

I'm not sure there is sufficient understand of the behavior of N-containing compounds to assume that they oxidize at the same rate/temperature as non-N containing compounds. While this may be an underlying assumption or requirement, it is not adequately elaborated. There may be something to be learned from examining the pyrogenesis literature (e.g., Knicker (2010, Org Geochem); DeLaRose (2011, SBB). We do NOT assume that N-containing compounds oxidize at the same rate as non-N-containing compounds. Furthermore, our conclusions are NOT based on the thermostability of the fractions. Instead, our conclusions are based on the finding that the fractions gained by thermal fractionation have a low $\Delta^{14}C$ and a low TOC:TON ratio. We discuss the reason why old soil organic matter is thermostable, but our conclusions are not based on the thermostability of soil organic matter. We will explain this in more detail in the revised version of the manuscript.

The discussion is centered on two rather strong assertions that are written in a highly speculative tone. How can these assertions be supported? Are they known to be true? Can they be tested? There is much elaboration needed here. As pointed out above, we do NOT assume that N-containing compounds oxidize at the same rate as non-N-containing compounds. In fact, our conclusions are not based on any assumption about the oxidation rate but on the radiocarbon data. Furthermore, our conclusions are NOT based on the thermostability of the fractions. The reviewer's comments helped us to see that this is not clear for readers, and we will explain this in the revised version of the manuscript.

I found the correlations with clay content compelling, but it was unclear why pH was included in the statistical models. pH as a control on microbial activity is a much different mechanisms that pH as a driver for mineral-association. There may be some confounding of cause and effect here. pH affects the protonation and charge of organic matter and is therefore key for the formation of organo-mineral association. We will point this out in the revised version of the manuscript.

In terms of these statistical analysis, I believe the authors may be using "correlation" and "regression" - but should not. Theauthors should consult papers by Webster on the distinction (1997, EJSS and 1989, SUM). In addition, the improvement of model performance with the addition of pH to clay should be tested using model selection methods (e.g., AIC). We will add the AIC of the models, as suggested. The first part of the comment is not entirely clear to us.

Overall, the outcomes of the study seem intuitive and compelling, but as the saying goes "the devil is in the details", and this manuscript lack a great deal of details that are needed for the reader to have sufficient confidence in the conclusions reached based on the collected evidence. We thank the reviewer for pointing out that the outcomes of our study are compelling and for making us aware that more explanations are needed. We will add the requested details and explanations to the manuscript, as explained above.