

## Review Discussion

The comments of both reviewers indicate that our manuscript was flawed due to poor definition and inconsistent use of a number of terms in the paper. For example, we clearly were not consistent or even always correct in our use of the 'clay' as a particle-size designator on the one hand and as a descriptor of crystalline layer silicates on the other. Given the topic of the paper the confusion created by our admittedly sloppy use of terms created understandable frustration for the reviewers. We recognize that a substantial revision of the manuscript is required before the validity of our interpretations can be fairly judged.

Reviewer 1 pointed out numerous instances of confusing terminology and further suggests that our confusion led to misinterpretations of the results. Reviewer 2 echoed many of those concerns but was a bit more open to following our line of argument even when our terms were imprecise. An example might be our use of the phrase "crystalline Fe and Al" which reviewer 1 rightly points out makes no sense. What we should have said was "DCB extracted Fe and Al oxide or hydroxide compounds from which oxalate extracted Fe and Al oxide or hydroxide compounds had been subtracted." Obviously the latter is too cumbersome but the former was too truncated. One can probably get the sense of what we meant from the former. However, such a lack of precision is inappropriate and annoying in a scientific paper.

We hope the editor will allow us to revise our manuscript. Among the things we will change will be to give clear definitions of all of the various terms used in the manuscript, as given below.

One of the major problems we have is that there is no universally agreed upon method to quantify the mineral components in soil. We quantified the crystalline aluminosilicates such as kaolinite and smectite, but also Fe oxihydroxide minerals like hematite and goethite using XRD on clay-sized material isolated from the bulk soil. Separately, we quantified the Fe oxihydroxides and SRO minerals using standard Dithionite citrate and oxalate extractions followed by measurement of Fe in the dithionite citrate extract and Fe and Al in the oxalate extract. A major problem with our originally submitted manuscript was that we did not clearly state in all cases which of the methods was used when we discussed results.

Here we define some of the "mineral" terms we use in this ms and note that we will use these definitions consistently in the revision:

Clay (<2- $\mu$ m size fraction)

Clay Minerals (all the identifiable minerals that reside in the <2- $\mu$ m fraction – this includes layer silicate minerals, Fe oxihydroxides, Al oxide, carbonates)

Layer Silicate Minerals (crystalline layered aluminosilicate minerals such as kaolinite and smectite)

Fe oxihydroxides (Fe compounds that can be dissolved using a standard dithionite citrate extraction but which are not dissolved by a standard oxalate extraction – these compounds are assumed to be pedogenic Fe molecules although it is possible that some geogenic compounds are also dissolved by the dithionite extraction). This fraction also includes coatings on minerals with size  $>2 \mu\text{m}$  but  $<2 \text{mm}$ .

XRD-measured Fe oxihydroxides in the clay fraction. These are the Fe oxidhydroxides measured by XRD in the  $<2\text{-}\mu\text{m}$  fraction, as goethite, hematite, goethite, magnetite, maghemite and ilmenite. We normally will not refer to this fraction, as the standard dithionite citrate and oxalate extractions were performed on more soils.

SRO minerals (aluminosilicate or Fe oxihydroxides that are minimally polymerized and tend to be linked to organic compounds or water – effectively these compounds are extracted using a standard oxalate extraction and quantified by measurement of Fe and Al in solution after extraction). In SRO minerals the crystallites are so small that they do not provide a coherent XRD signal

Al oxide or gibbsite is another component of clay minerals but one that we are not explicitly quantifying in this analysis – as noted below we used Al as an internal standard for the XRD quantification of the Layer Silicate Minerals.

A second issue was the nomenclature used to define the different organic matter fractions that were measured for C and  $^{14}\text{C}$  content.

We define our use of organic matter as follows:

Light Fraction carbon: The organic carbon in material that floats in a solution with density  $1.7 \text{g cm}^{-3}$ . (Questions by reviewer 2 about details of the density fractionation procedures are given below).

Root-free light fraction carbon: The organic carbon remaining once visible roots have been picked out of the light fraction (please see Castanha et al. and other comments below).

Heavy Fraction carbon: The organic carbon in material that sinks in a solution with density  $1.7 \text{g cm}^{-3}$ .

Carbon strongly associated with XRD-clay. The carbon in the same clay fraction measured by XRD for mineralogy. This is ‘strongly bound’ because the material measured for XRD was treated with hydrogen peroxide and presumably only material that has a strong association with minerals or aggregates survives this treatment. Carbon strongly bounded to clay is a subset of the Heavy Fraction carbon (which can include C associated with Fe oxihydroxides coating sand grains)

Carbon not strongly associated with XRD-clay. This is determined by mass balance between the Carbon strongly bound to XRD-clay and the Bulk soil C. It includes a heterogeneous mix of materials, from very fine roots to C associated with Fe oxihydroxides coating sand grains.

## Research Design

We know from a couple of decades of work that SRO minerals store a lot of carbon and hold that carbon for long periods of time. As noted by Reviewer 2 there has been far less work done evaluating carbon storage in soils where SRO minerals make up a very small proportion of the clay mineral fraction of the soil. Our goal in this paper was to evaluate carbon storage in soils with low concentrations of SRO minerals where we would expect the heavy carbon fraction to be associated with other clay mineral components.

We agree with both reviewers that overlaps among the different mineral and C fractions we analyzed can be confusing, and that a more streamlined procedure is advisable in the future. However, we also think that in reporting the data we have, we are able to draw robust conclusions about the role of smectite clays influencing the age of carbon in soils with few SRO minerals.

Detailed responses are given below. To ease reading, we have put our responses in red below the referee's comment.

### Anonymous Referee #1

Received and published: 11 June 2016

#### General comments

The idea of the present work was to test the organic carbon accumulation and turnover as related to crystalline mineral phases. To do so, soils low in short range order (SRO) mineral phases were studied. The authors report differential effects of certain mineral phases on accumulation and turnover. I am not convinced that their assumptions are valid. Major misconceptions as well as serious methodological flaws question the entire study. I therefore cannot recommend publication.

We understand the reviewer's frustration with the originally submitted manuscript and hope to convince them that the major errors were in communication rather than interpretation. We thank the reviewer for their patience in giving such a thorough review.

#### Major concerns:

Throughout the manuscript there is a tendency to mix up "accumulation" and "stabilisation" of organic matter. These are no synonyms. Please try to be specific. There is also an overall tendency of imprecise expressions and mixing up of concepts. The result is a partly confused text hard to read and understand. The definition of SRO phases used by authors is rather vague and changes throughout the text. Sometimes it is based in oxalate-extractable Fe and Al, sometimes it seems also the dithionite-citrate-bicarbonate (DCB)-extractable Fe and Al is included.

We will revise the text with these concerns in mind, and will provide clear definitions and stick to our terminology.

Analyses of the clay fraction were carried out after treatment with H<sub>2</sub>O<sub>2</sub>. This will remove part of the light fraction-type material but also part of the mineral-associated material. Since the exact share of mineral-associated organic carbon removed by the H<sub>2</sub>O<sub>2</sub> may vary, all data organic carbon data obtained on the clay fraction are biased, thus, need to be dropped. Since much of the conclusions are based on the clay fraction-related data this questions the overall meaning of the work. Justifying the analyses of carbon in H<sub>2</sub>O<sub>2</sub>-treated material by claiming that some carbon survived the treatment is not valid.

We agree that the H<sub>2</sub>O<sub>2</sub> treatment removes organic C from the clay fraction. However, our goal was to measure the radiocarbon in C that was in exactly the same fraction as was measured for mineralogy using XRD. As long as the H<sub>2</sub>O<sub>2</sub> treatment was performed consistently, we do not agree that biases associated with differential efficiencies of removal of organic matter by H<sub>2</sub>O<sub>2</sub> (e.g. surface Fe oxihydroxide coatings versus layered silicate minerals) make the measurements meaningless. In a revision, we will take care to point out that there **are** potential biases, but feel that the comparison of the minerals and age of carbon strongly associated with XRD-clay from the same fraction is useful information.

Also, analysis of the mineral assemblage of the clay fractions is based entirely on X-ray diffraction with Cu K $\alpha$  radiation. Thus, there is no proper estimate of possible contents of SRO phases and oxides of the clay fractions. Several sections of the manuscripts left me under the impression the authors are not aware that the clay fraction is not composed entirely of clay minerals but also contains other phases capable to interact with organic matter.

We do understand the point and will clarify throughout the text.

The density separation used is also rather strange. The cut-off density of 1.7 g/cm<sup>3</sup> is not justified, at least not by the reference given. In addition, the sonication energy used was little, thus, total dispersion of samples with stable aggregates has to be questioned. Especially, the more oxide and kaolinite-rich samples may not disperse completely, thus, the heavy fraction likely still may contain light fraction-type material. In turn, undispersed aggregates may still contain enough light material to make them float.

We decided to measure only the so-called 'free' light fraction, i.e. the material that floats but not the material that requires strong sonication to destroy aggregates. The cutoff density of 1.7 g cm<sup>-3</sup> is one that means most SRO minerals (i.e. lowest density mineral phases) will not float. Density separation is a technique that is adapted to the soils used and there is not really a standard protocol. It is very clear that the procedure used definitely influences the results (please see Castanha et. al, 2013 who discuss this in detail). As we used a common procedure for all samples, we assume that results can be compared within our

study, though care must be taken when comparing with other studies that may have used other methods. .

The rather variable and often very small contents of organic carbon in the light fractions point at major problems with the separation. In summary, the density separation has been carried out in a hardly reproducible manner.

Often in clay-rich soils, there are some mineral phases that are attached to low density material, or that remain floating in the sodium polytungstate solution even after a very long time of centrifugation. Some of these can be siphoned on to the filter when removing the floating organic matter. Including the total weight of these phases and the C content is important for determining the yield of the procedure, and reported low C contents are not uncommon, especially in B horizons and (in our data set) in clay-rich soils. The presence of small amounts of mineral materials on the filter can dilute the C content overall but have a negligible effect on the C isotope signature. For example if 50% of the weight of isolated material is mineral-dominated with a concentration of 0.5%C, and the other 50% of the weight is free organic C with 40%C. then the overall %C of the mixture on the filter would be 20.25%, a large dilution. However if we combust and analyze the isotopic signature of the mixture, the part of the mixture with 0.5%C would contribute  $0.5/25.25$ , or about 2%, of the total C in the sample. In the case of the basalt soils (which averaged about 10%C, the contribution from the mineral-associated C could be higher (in our example,  $.5/10$ , or 5%). Using our own data as an example, assuming  $^{14}\text{C}$  signatures of free organic matter of 1.100 fraction modern and 0.8 fraction modern for the mineral-C, the total fraction modern we measure on the mixture would be 1.096 (instead of 1.100), and reduces the presumed TT by 1-2 years (either from 10 to 9 years, or 65 to 63 years).

For this reason, we are not concerned by the degree of dilution in reporting our isotopic signatures for the LF fraction. In a revision, we will point out that %C results are subject to uncertainties in the LF fraction due to the potential inclusion of mineral material on the filters.

Minor concerns:

Title: As written, the title suggests a study of only local importance. The authors may consider changing it into "Timescales of C turnover in soils with mixed crystalline mineralogies".

p.1, l. 17-18: The conclusion that the enrichment of OC in the clay fraction is due to stabilisation by clays is either trivial (in case of the authors refer to clay as size fraction) or disputable (in case of referring to clay minerals; see above and below).

We choose the disputable and will clear up confusion throughout the text as detailed below.

p. 1, l. 23: What made the authors believe that crystalline Al oxyhydroxides contributed to the accumulation of faster turning over fraction? Is that based on DCB-extractable Al? If

yes, please note that the extractant is not capable of extracting much Al from crystalline Al phases such as gibbsite.

We recognize the confusion created here and note for the record that we do not believe that the DCB extractable Al is a useful indicator of mineral composition. We will remove Al from that sentence.

p. 1, l. 26: What does SRO refer to? Seemingly it refers to the observed relationship between DCB-extractable Fe and organic carbon. Dithionite-citrate-bicarbonate-extractable Fe includes Fe from crystalline Fe oxides, such as goethite and haematite.

As this reviewer points out the definition of SRO minerals is vague. Here we use the standard approach of evaluating the oxalate extract for the amounts of Fe and Al released during extraction. We assume that Fe release is due to decomposition of ferrihydrite or perhaps nano-crystalline goethite and that the Al release is due to decomposition of nano-crystalline aluminosilicates such as allophane and imogolite. We recognize that we may not have been clear about this operational definition and will clarify throughout the text.

p.1, l. 29-30 (and elsewhere in the manuscript): Expressions such as “crystalline Fe” are nonsense and misleading. Iron is an element. Please refer to the correct mineral phase, e.g., Fe oxides.

The reviewer is correct. We regret the sloppy short hand that crept into our text.

p. 2, l. 3-5: Some references given do not refer to physical protection or recalcitrance. Please be re-consider.

We intend to simplify the text in the introduction based on this and several other comments by the reviewer below.

p. 2, l. 7-16: Somehow, I am under the impression the authors have some problems with mechanisms involved in organic-mineral interactions. For example, what is “de- hydration bonding”? Also, the choice of references is a bit strange. For instance, two references are on methods for estimating mineral phases but do not address binding mechanisms. Did Masiello et al. (2004) really refer to crystalline Al and Fe sesquioxides?

The reviewer is justified in not understanding our highly compressed text which tried to cover too much ground in a short space – something we think is not really important for the paper anyway, and therefore will be rewritten to achieve greater simplicity and clarity.

p. 2, l. 10: Replace “metallic” by “metal”.

Yes this will be done.

p. 3, l. 13-16: Note, the studies of Wattel-Koekoek et al. (2003, 2004) include no proper estimates of possible contents of oxides, thus, do not allow for distinguishing of effects by clay minerals and other phases, including SRO phases.

The reviewer is strictly correct in this point although those authors selected samples to analyze that ensured a dominance of crystalline aluminosilicate clays with or without crystalline Fe oxides.

p. 3, l. 17-23: I suggest adding proper research questions and/or hypotheses.

We will re-write this section to make the questions more explicit.

p. 3, l. 27-28: What is “soil residence time”? Please explain.

The Kruger sampling sites offer a unique landscape for soil sampling. All the streams that cross the park from west to east are maintained at the same erosional base level by a strata of rhyolite that is much more resistant to erosion than the granites and other volcanics that are upstream of it. Furthermore we are able to establish erosion rates on the granites using  $^{10}\text{Be}$  accumulated in quartz sampled in this case from stream channels sands. As we state in the referenced citation (Chadwick et al., 2013): “Using average regolith depth and catchment-averaged erosion rate estimates, we infer long hillcrest regolith residence times of 0.11, 0.15, and 0.57 m.y. for the dry, intermediate, and wet sites, respectively.” These data are corroborated by measured soil production rates (Heimsath et al. in prep.). The importance of the stream channel base level control is that it means that all landscapes regardless of whether they are underlain by granite or volcanic rocks are eroding at the same overall rate. This gives us confidence that the soil landscape is highly stable as one would expect for a craton in a non-glaciated environment and as a consequence mineral transformations can be expected to have moved past the meta-stable SRO stage toward a stable end product (given a specific climate condition). This approach to sample selection was also used by Wattel-Koekoek et al. (2003, 2004), except they used it a more global context without a specific local landscape context.

p. 4, l. 24-25: Please give more information on the determination of the clay contents. Did the procedure involve pre-treatments such as destruction of organic matter and oxides?

As mentioned in the text, we used  $\text{H}_2\text{O}_2$  that destroyed part of the organic matter in the clay fraction that was measured by XRD.

p. 4, l. 32-33: The location of the CN analyser is probably of no importance. Omit.

Yes will do that.

p. 5, l. 3: As stated, it seems only the characterisation of clay minerals was attempted.

The reviewer is correct that “as stated” it appears that only characterization of clay minerals was attempted. There are several parts to that statement. First we specifically did not characterize sand and silt size mineralogies. We did use a separate approach to characterize the SRO minerals and Fe oxyhydroxide minerals. For these we conducted oxalate and DCB extractions on the <2-mm fine earth fraction. The reason for using the fine earth was that we were concerned that some of these minerals would be coating the sands and silts in ways that would be missed if we only conducted those extractions on the <2- $\mu\text{m}$  (clay size) fraction. In the methods section we covered these extractions in the previous section on soil characterization which led to an artificial separation of the extraction quantification of the clay minerals from the XRD characterization. We will move the extraction material to the section on quantification of clay minerals to make it clear that we are relying on both the extractions and the XRD approaches to develop the quantitative understanding of the clay mineral composition. It should be noted that we recognize that mixing these approaches is not the best way to get a soil mineral compositions, but we also argue that there is no readily accepted single approach to full quantitative mineral characterization of soils. As a consequence we are fully aware that our development of graphical relationships among mineral compositions and carbon turnover is flawed by our acceptance of specific operational approaches toward mineral quantification.

p. 5, l. 14-16: What was the idea behind adding Al oxide to the clay fractions before X-ray diffractometry? Why not quartz? The addition of Al oxide limits estimation of Al oxide phases. Was the quantification of X-ray diffraction data supplemented by data on the elemental composition? What software was used?

The manuscript has been clarified to state that "corundum" was used as the XRD standard. Corundum has sharp peaks in XRD spectra that overlap with relatively few phases common in soil (including gibbsite) and these peaks degrade minimally during the grinding process used to mix sample and standard. Preliminary processing of the XRD spectra did not suggest gibbsite was an important constituent of the clay mineral fraction and gibbsite is not considered to be a major sorber of organic matter in soils. The word "software" has been added to clarify that the Rockjock software was used to for the quantification of minerals from XRD spectra.

p. 5, l. 24: Why a density-off of 1.7 g/cm<sup>3</sup> was used? The reference given refers to 1.6 g/cm<sup>3</sup>.

The density of 1.6 g/cm<sup>3</sup> is typically below those of all SRO minerals; so is 1.7 g/cm<sup>3</sup>. We consulted with the author of the reference (Marion Schrumppf) about which density to use, and this was her suggestion. There is no general agreement on methods to use for density separations and many different density cut-offs can be found in the literature.

p. 5, l. 25: What was the reason of using varying amounts of sample for the density separation? Why no standard protocol was used? Did the authors attempt estimating the carbon recovery?



The amount used was 10-15 grams, we did not feel the need to control the amount of sample extracted to better than within a few grams as the yield was determined based on the measured initial weight for each sample. We did estimate C recovered in each fraction (these data are given in Supplementary Table 1, we are most confident of the % of total C in the HF-fraction as there are issues with weight change in filters and low masses with the quantification of the low-density fraction). An additional amount of C is dissolved and not recovered in the dense liquid. We admit that our mass balance (as occurs in many density separation procedures) was not perfect. However, as outlined above, we do not think this affected isotopic results – or at least it affected them in the same systematic ways. Please see Castanha et al. (2008) for a detailed discussion of the various ways density fractions are affected by the procedures used.

\*Castanha, C, S Trumbore, R Amundson (2008) [Methods of separating soil carbon pools affect the chemistry and turnover time of isolated fractions](#) Radiocarbon, 50, 83-97.

p. 5, l. 26: What is meant with “ultrasonicated at 60 J mL<sup>-1</sup> for 2.5. min”? Is 60 J/ml the total energy input? Did the authors control for proper dispersion, i.e., disaggregation? And if yes, how this was done?

60 J/mL is an estimate of the energy input, determined after calorimetric calibration of the sonicator. This is a relatively low energy and not likely to disrupt strong aggregates. Schrumpf et al. 2013 used stepwise increases in energy input to determine the level at which all aggregates were dispersed (we are using the identical system that she used). They found that “Energy input of 100 J mL<sup>-1</sup> was sufficient” (to destroy all aggregates) “for sandy soils (Bugac, Bordeaux), and between 300 and 450 J mL<sup>-1</sup> for most other soils. For the clay-rich Hainich soil, the energy input had to be raised to up to 900 J mL<sup>-1</sup>). Clearly we did not destroy all aggregates with this procedure, and this was not our intent. Thus our mineral fraction may include low density material that was protected in aggregates. This is part of the general problem in such operationally defined fractionation methods, and one of the points of the paper is to explain the common observation that the heavy fraction is a mix of materials with different <sup>14</sup>C signatures.

p. 5, l. 29-30: What is the idea behind removing visible roots from the light fraction? Actually, dead roots are part of the debris material that makes up the light fraction. Why the authors did not remove living roots from fresh samples?

Castanha et al. (2008, reference above) demonstrated that the radiocarbon signature of the low density fraction is strongly affected by the presence of fine roots. Normally these are picked from samples as part of the sieving to <2mm; however, different people pick fine roots more or less diligently. Castanha et al. (2008) showed that picking the fine roots out of the low density fraction minimized variability among ‘operators’. Also, we know (because we measured them) that the fine roots have mostly contemporary C, and wanted to know what the rest of the C in the low density fraction contained.

p. 6, l. 1-4: Due to the bias in the clay and density separation I do not think the data set to be solid and comprehensive. I am wondering why the authors did not try a more logic approach, such as first separating all light material, then, separating the clay fraction from the heavy fraction.

Figure 4 was intended to be transparent about the overlaps between isolated fractions. While we agree with the reviewer that it might be more satisfying to have all fractions isolated sequentially so that there is no such overlap, this is not what we did. One reason for this is that the density separation is expensive, and did not always yield enough clay for the mineralogy step. We would reconsider this in the future, but cannot change the past.

p. 6, l 13-14: Is acidification to pH 6 really enough remove all carbonate, including that bonded to other mineral phases? I doubt.

Actually carbonates are remarkably non-bonded to other mineral phases and tend to reside in soil as their own unique bodies (K fabric concepts). We do expect that acidification to pH 6 will remove the carbonates although there is the possibility that some carbonates could avoid decomposition if protected within aggregates. As pointed out in the text the carbonate in the horizons sampled was primarily in relatively large aggregates (sand and pebble size) whereas the bulk of the fine-earth fraction was non-calcareous (did not react to acid in the field).

p. 7, l. 13: No, DCB does not extract only crystalline Fe oxide phases but all Fe oxide phases, including ferrihydrite. In turn, oxalate extracts only the poorly crystalline portion of DCB-extractable Fe.

Yes the reviewer is correct: to get at the crystalline Fe oxide phase we subtract the oxalate extracted Fe from the DCB extracted Fe. We will insert an equation along the lines of  $Fe(\text{oxides}) = Fe(\text{d}) - Fe(\text{o})$  and create a new column in Supplementary Table 1 with output of that calculation.

p. 7, l. 14: Note, DCB-extractable Al does not represent Al oxide phases.

Right the DCB-extracted Al is meaningless in this context – we will remove that sentence.

p. 7, l. 18: Is there are reason why the clay contents increased with depth?

Increasing clay with depth through the solum is quite typical for soils due to hydrological transfer of colloids. Typically the downward transfer of colloids is countered by bioturbation which mixes profiles, but our observation is that more often than not soils have a subsurface accumulation of clay-size materials, often skewed to the small particle sizes.

p. 8, l. 27-32: Is it correct, the soils containing pedogenic carbonates are those rich in smectite? So, could it be that their radiocarbon signature was affected by carbonate? By the

way, the picrite (back basalt)-derived soil is classified as Calciustert but not listed as containing carbonate? Isn't that illogic?

There was no evidence from the XRD data to suggest that the clay fraction harbored calcite. If the picrite soil was carbonate free or mostly so then the classification should be Typic Haplustert

p. 9, l. 8-9. Due to the H<sub>2</sub>O<sub>2</sub> treatment of the clay fraction, I doubt that proper estimates on non-clay carbon are possible.

The problem here is with the definition of non-clay C. We meant this to mean all C (including that oxidized by H<sub>2</sub>O<sub>2</sub>) that was not in the clay fraction measured for XRD. We will be more careful with this definition in the future. The mass balance stands – the C removed included all non-clay sized material and all material removed by H<sub>2</sub>O<sub>2</sub> from clay sized material.

p. 10, l. 9-15: These correlations need to be re-considered bearing in mind that the clay fraction contains most of the oxides.

We will make clear in the text that subtracting the Fe<sub>o</sub> from the Fe<sub>d</sub> prior to determining the crystalline Fe oxide concentrations.

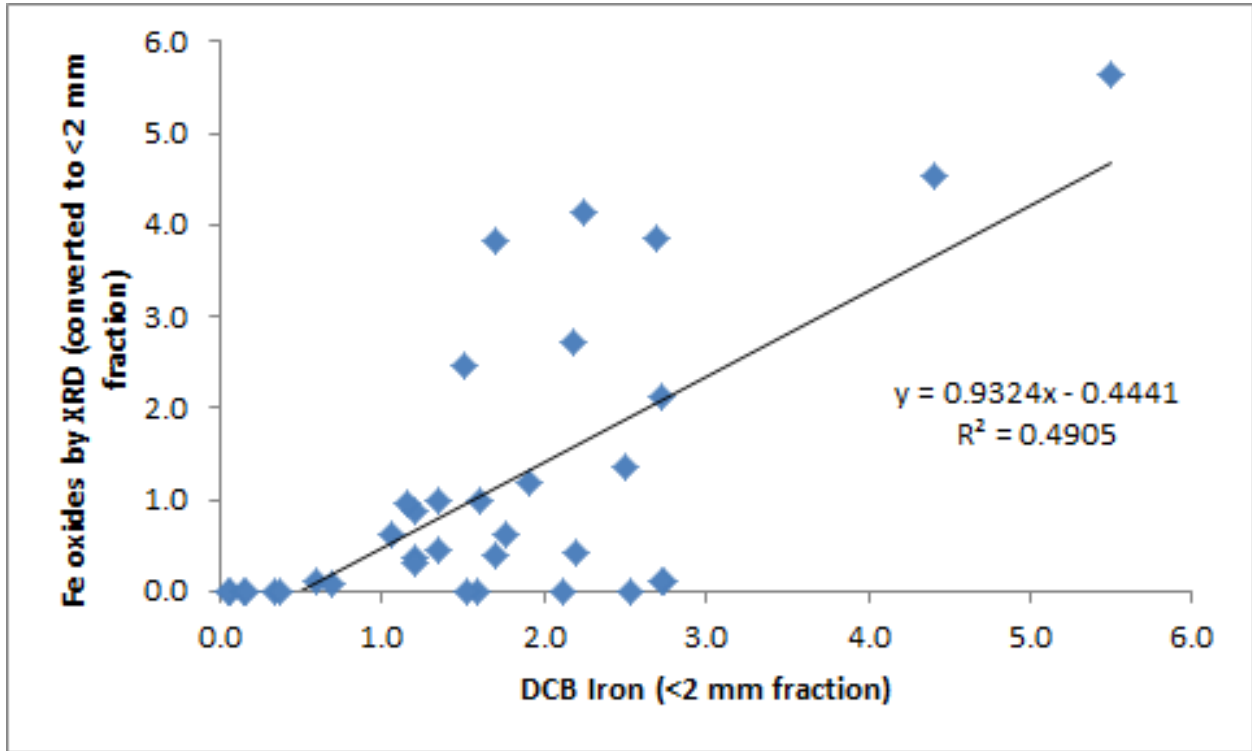
p. 11, l. 9-17: The discussion here is rather speculative since composition of organic matter was not addressed.

We agree, but also feel that we did point out in the text that we were speculating on this – effectively connecting the dots from pieces of the literature and our measurements.

p. 11, l. 18-29: Here, it becomes clear the authors partly equal clay-sized particles and clay minerals. They relate the estimated non-clay fraction carbon to “other, non clay mineral stabilization mechanisms”. This is simply wrong, since the clay fraction holds also most if not all oxide phases (even the Cu K $\alpha$  X-ray diffraction, despite of being rather insensitive to Fe oxides, indicated their presence). Also, I have problems with the authors' logic. The organic carbon of all study soils relates well to the DCB-extractable Fe. Seemingly, the clay mineral type does not matter much to the accumulation of organic carbon. So, there is no reason for all the clay mineral discussion. I am also wondering, why the authors did not comment on the rather small contents of organic carbon in the smectite-rich soils. There is much writing on organic matter stabilisation by smectites. The results presented, however, suggest that smectites are rather poor organic carbon accumulators.

The only oxides quantified by XRD were Fe-bearing and included hematite, goethite, magnetite, maghemite and ilmenite. We agree that we create confusion when we also report data on crystalline Fe oxihydroxides based on the Fe(d) – Fe(o) for the bulk soil. In the plot below, we compare the DCB-oxalate Fe phases for the bulk soil (x-axis) to an upscaling of the Fe oxihydroxides measured with XRD corrected for the % of clay-sized

material (y-axis). There is general correspondence, although we agree that the bulk extracts are a better measure since they also include things like coatings on sand or silt-sized materials. Also, we have far more data for the bulk extracts.



The clay minerals are important for the radiocarbon, where a small amount of old material has influence. The bulk of the C is stabilized by mechanisms that have timescales that yield similar  $^{14}\text{C}$  signatures. Something similar was found by Lawrence et al. (2015), so we thought it important to point this out.

By the way, why do the authors consider “crystalline Fe and Al (oxyhydr)oxides” as controlling carbon accumulation? The close relationship is for DCB-extractable Fe and not for any Al. Also remember, DCB extracts all non-crystalline Fe oxides (see above).

We understand the problem and will increase clarity throughout the text.

Assuming that the relationship between organic carbon and Fe oxides is also valid for the smectite-rich soils, i.e., Fe oxides do the accumulation; it is absurd to assume that the smectites make the organic carbon turning over slowly. As already pointed out, I assume an issue with incomplete removal of carbonates to be the reason of the radiocarbon signature of the smectite-rich soils.

We disagree that carbonates can be responsible for the old ages in smectite clays. To obtain an age of 2000 radiocarbon years, roughly 20% of the carbon in the sample would have to be radiocarbon-free. As the calcites we measured were not radiocarbon free, they would have to make up an even larger portion of the total C measured for isotopes. Of the the smectite-rich clays in Table 2, all but one had <1% Carbonate. It is highly unlikely that inclusion of carbonates can be possible for the low  $^{14}\text{C}$  values we measured. We will measure carbonate contents of this material separately (using  $\text{CO}_2$  evolution on acidification) to address this in the revision.

Tables and Figures (general): Please give proper dimensions. Percentages of smectite, organic carbon, smectite likely refer to weight-%. Consider giving instead g/kg. What is the dimension of the cation exchange capacity (CEC)?  
None of the tables and figures is truly self-explanatory.

We will revise accordingly.

Table 2: I am wondering if the “oxides” included also Al phases. The little clay content of the non-mafic soils is no good reason for not analysing the composition of their clay fractions.

The oxides presented here are for information only. We used the Fed-Feo data from the extractions to develop the Fe-oxide – carbon relationships because of concerns about loss of oxides (as coatings) during particle size separation.

Table 3: The rather low organic carbon contents of some of the light fractions support my concerns on issues with the density separation. The light fractions with little carbon may contain a good portion of mineral-bound (older) carbon, thus, suggesting a slow turnover.

Please see the answer to this issue above. While we agree that there are likely mineral-bound (older) C diluting the low density C, (a) we can not absolutely rule out that the C contents of this fraction can be as low as 10%C and (b) dilution with mineral phases would contribute only a few per cent of the total C measured for isotopes, that would not have really big effects. We have not made much of an effort to interpret the LF radiocarbon data in this paper, and present them mostly for completeness.

Figure 5: Giving an enrichment factor for clay fractions treated with  $\text{H}_2\text{O}_2$  is not valid.

We do not completely understand this comment. There is no ‘enrichment factor’ in the sense you would use for  $^{13}\text{C}$  isotopes. Radiocarbon data are all corrected for the  $^{13}\text{C}$  in the sample, and any such enrichment factors are corrected for. What is left is the mean age information that is given in the figure.