

Answers to referee 3:

We thank the referee for the comments, which helped to improve the understandability and clarity of the manuscript.

#### General comment

1) Lack of specific objectives and hypothesis that guided this work

The introduction is restructured and specific objectives are added to the revised version of the manuscripts

2) Lack of justification for why they modified existing methods

Justification of the modification of methods are given and were added were necessary to the methods part

3) Excessive reliance on linear correlations

see comments below

#### Major comments:

Page 13086, Line 19 and discussion the authors state that they believe 'geogenic' parent material affected the  $^{14}\text{C}$  values of the fLF – both from rock derived C and fossil C. If there is believed to be significant contamination from rock derived C, it is likely that they could have had major contamination of the fLF with minerals. The procedure must have picked a considerable amount of mineral material during the first mixing step that is meant to extract fLF OM. This is serious issue that the authors have to explain. When reading the methods section, the description of the density fractionation procedure doesn't indicate why  $1.6\text{ g/cm}^3$  was chosen as a density of SPT solution. Where there any preliminary tests done to determine that that density level is in fact appropriate for the range of soils that were used in this study? This could explain why they have high amount of mineral matter being picked up during the FL extraction steps. If the density is too low for the soils they are studying, you can float considerable amount of mineral matter with the fLF organic matter.

The density of  $1.6\text{ g cm}^{-3}$  was selected since based on proper theoretical consideration (see Christensen 1992) and is in line with the original density fractionation procedure by Golchin et al. (and accordingly no modification). Testing the optimal density cutoff to separate LF-OC from HF-OC, for example Cerli et al (2012) re-confirmed the applicability of the  $1.6\text{ g cm}^{-3}$  cutoff as best suited for testing soils. The references will be added with a comment to the methods part.

Nevertheless, it is well known that fLF-OC can hold small amounts of minerals. Since obtained without sonication treatment, some minerals can still adhere, e.g., to small roots without preventing them from floating. Accordingly, OC concentrations of the separated oLF are typically slightly larger than those of fLF-OC, and so minor contributions of HF-OC to fLF-OC seem possible. These contaminations cannot be responsible for the extremely small  $\Delta^{14}\text{C}$  values of fLF-OC at Grignon, Carlow, and Easter Bush, because the  $\Delta^{14}\text{C}$  values are much lower than HF-OC. So, contaminations of the LF with HF-OC cannot be the reason for small  $\Delta^{14}\text{C}$  values of LF-OC.

Instead we think rather lignite/coal, which typically have densities of between 0.8 and 1.5 g cm<sup>-3</sup>, released from the parent material, was responsible for the old average fLF-OC. As the density range of coal overlaps with that of pure organic matter, it is not possible to separate out coal by density. At Grignon and EasterBush (Easter Bush is close to places where surface mining of coal and lignite occurs in the surroundings of Edinburgh), coal particles were clearly visible and larger particles could be manually picked out from the samples, which was not possible for smaller particles. As we were not able to quantify the contribution of fossil carbon, these sites were excluded for a number of analyses. As minor contributions of coal at these study sites would still have a strong effect on estimated turnover times based on <sup>14</sup>C values alone, we believe that the risk of biased results due to fossil C inherited from the parent material needs to be considered.

Starting in Page 13089, Line 1 and many other places in the manuscript the authors state that there are no studies that employed the approach they used here. This is not true, although there are not a lot of studies that consider these two analyses, there are some. For example Berhe and co-workers did just that in work that was published in two papers in 2008 and 2012, both in JGR-Biogeosciences.

Berhe and co-workers related soil OC stability with landscape position, by analyzing total soil profiles, trying to elucidate the impact of erosion on OC in soil. Since erosion is no likely factor at the study sites, the articles have little meaning to the discussion and interpretation of our results.

Page 13092, Line 21 – Here, the authors are doing at least two things different than the Golchin and others technique. First, they are reusing SPT without any cleaning steps (which basically means they are shaking the samples after the fLF extraction step in water/SPT extractable organic matter solution). Almost all of the studies using this method, do not reuse SPT solution from the fLF steps to oLF and HF extraction steps, even for the same sample in the procedure, at least not without cleaning it first according to the methods of Six and others. The cleaning of the SPT after the end of each sample's extraction is completed is described in the methods section, but not the reusing of the SPT used for

extraction of fLF for OLF and HF steps that they describe in the method description. Second, they are incorporating a sequential sonication step instead of the onetime ultrasonication step that is recommended in the procedure. In the procedure, typically, one does a preliminary test to determine an appropriate energy level for the specific samples that you are using and you stick with that for the rest of your samples. Although, I don't necessarily disagree with the need for these steps (I actually think the second one especially is a very useful modification of the technique), I think the authors should properly justify why these modifications need to be included in their method. In addition, the authors should go back to this in the discussion and discuss whether it had any influence on their results and how they interpreted them. While doing so, the authors should also indicate whether they meant to say they stopped the sonication when there was no further release of particulate OM or any and all OM (including water extractable, dissolved OM). For the second point, refer to the recently published work of Kaiser et al 2012 (Journal of Soil Science and Plant Nutrition) in regards to maximum energy levels for disruption of aggregates in soils that vary in mineralogy and carbon content.

Yes, it means that after separating the fLF, the remaining sample was extracted in the same SPT solution (after checking the density), which probably contained some dissolved OC from the first extraction step. We assume that the referee is afraid that this could lead to contamination of the oLF/HF fraction with OC dissolved during the first step? However, why should there be re-adsorption of material previously released from the soil under otherwise identical conditions? Release of OC into the SPT solution is based on its absence in the solution and the desorptive action of tungstate. Thus, using new SPT solution might even cause increased loss of OC during fractionation instead of improving the results.

Sequential sonication was only applied to determine the energy level required to achieve total aggregate disruption. The actual density fractionation was carried out by one single sonication, at the energy input level identified in the previous test. This is explicitly stated in the revised version. We are aware of the article by Kaiser et al. (2012), however, the soils we studied did not contain extraordinarily stable aggregates. Also, the release of dissolved OC following upon different levels of sonication as studied by Kaiser et al. (2012) is an unsuitable criteria to test for the complete liberation of particulate organic matter from inside aggregates. Instead, we selected the sonication level when there was no further release of particulate organic matter, similar to the procedure used by Cerli et al. (2012, see reference list), which showed that for different soil types also different sonication energies are required to achieve full release of oLF from inside aggregates.

Page 13094, Lines 3-4 – is there any evidence that addition of H<sub>2</sub>SO<sub>4</sub> directly into tin capsules doesn't have any effects on the results presented in this study? The most common method used for this application is addition of HCl to silver capsules. The authors did not provide a reference for this method. We did not use sulfuric but sulfurous acid and apologize for that mistake. Sulfurous acid is strong enough to remove carbonates from carbonate-poor soils, as the ones studied, but less aggressive than HCl and it is volatile at 60°C. We did not use HCl as it also attacks organic carbon (see e.g., Bisutti et al. 2004, Trends in Analytical Chemistry) and was shown to cause shifts in the isotopic composition (see e.g., Fernandes and Krull 2008, Environmental Chemistry). Also, it tends to interfere with the mass spectrometer. Unless oxygen isotopes are studied, tin capsules are standard.

Mineralization experiment – the authors do not state if during the time they stopped the mineralization experiment they had achieved near constant flux of CO<sub>2</sub> from the incubation jars (the relatively flat part of the curve of CO<sub>2</sub> flux over time, following the rapid, typically exponential decline). If they did not, then what is the justification for stopping the incubation experiment on the 20th day?

The CO<sub>2</sub> evolution rate in incubation jars normally declines with time, because no fresh substrate is added during incubation (in contrast to the field situation). Typical curves are given in Kutsch et al. (2010, Biogeochemistry, see reference list). During the first 20-30 days, the materials often show similar rates; after that period rates starts to decline. We aimed at identifying and comparing CO<sub>2</sub>-release rates just after sampling, before substantial decomposition of labile compounds occurred. Therefore the initial 20-day period should be the most appropriate estimate of labile material. We will add a respective comment to the revised version.

Page 13095-Line 28-19, check the first figure a) in figure 1. How is it possible that you have higher concentration of C in the HF compared to the fLF and oLF? Are you reporting

Values reported in Figure 1 A are not the concentration of OC in the HF (that is g OC per kg fraction), which would indeed be much smaller than in the two LF. Instead we report g OC in each fraction per kg of soil. As the total soil is dominated by HF-OC, larger amounts of OC are with the HF than with the LF, despite smaller OC concentrations in the HF.

Page 13101, Lines 6-15 and section 5.1 – the reasoning in this whole paragraph is not well supported by the data. You can have a linear correlation between root biomass and C in fLF, but that doesn't necessarily mean roots contributed more to fLF than above-ground biomass. It could be, but it is in no way certain based on this correlation. Reword this paragraph to restate that this could only be one of the possible (not the one) explanations.

Above and belowground litter are two possible sources for fLF-OC. We agree that the mentioned correlation would also be obtained for example if aboveground litter contributed with equal proportions to fLF-OC than roots across study sites so that roots are not the only possible source based on this relation. Therefore we find the relation between the depth distribution of roots and fLF-OC the more convincing argument for our statement that roots are more important than aboveground litter. Contribution of aboveground litter to fLF-OC is probably most relevant in topsoil layers where bioturbation enables the mixing of aboveground litter into the mineral soil. However, we did not find the topsoils having larger LF-OC accumulations than roots, which points at little incorporation of aboveground litter, i.e., by bioturbation. However, since we aimed at tracking that the effects of roots shaping belowground LF-OC profiles and not so at judging the relevance of aboveground and belowground sources, we decided to omit that part as it seems to be rather confusing.

Figure 16 and Summary – the schematic and discussion ignores at least two important factors for mineralization of C and distribution of C in different soil organic matter fractions – bioturbation and lateral distribution by soil erosion. Most of the earths land surface experiences if not both, at least one of these processes on a regular basis. The effect of both on total C in soil, distribution of C in different soil fractions, loss of C through gaseous and/or dissolved fluxes is extensively covered in soil science and biogeochemistry literature. The authors are advised to consider the contribution of these two important processes on their conceptual model and interpretation of their results overall.

We agree that both processes are important for soil OC transport and stability within the soil profile and in the landscape. But with our dataset we are not able to discuss the relevance of these processes at our study sites apart from acknowledging their existence. We explicitly add bioturbation and advection as transport processes to the former Figure 16.

Supporting information files:

S, Figure 1 – the bar graphs (the left most charts in each line) of figure 1 in the supporting informing information file is confusing. As far as I can tell it looks like you are presenting C concentration of each fraction in these figures. I think these bar graphs are good if you present data on C distribution between the different fractions, but if all you want to present is

Similar to the former Figure 2, the bars show average total OC concentrations for each soil layer. The bars are divided with three different colors into segments showing the relative contribution of each fraction to the total OC concentration.

S, Table 1 – Mass losses of 20-38.

The average mass recovery across all samples was 95%. Smallest mass losses occurred for the sandy soils of Le Bray (1%), largest for the Hainich soils (6–15%), which had the largest clay contents. The mass recoveries are in the range of values reported for example by Chenu & Plante (2006, >97%) and Grünwald *et al.* (2006, 96–104%). We add a paragraph comparing the mass and OC losses of our study to others to the supplement.

S, Table A2, what bulk density values were used to convert the C in fLF, oLF, and HF values to kg OC/m<sup>2</sup> values? This information should be included at least in the supplementary information file.

Bulk densities were determined for each soil core and are already published in Schrumpf *et al.* (2011), as stated in the methods part. We add a hint to the calculation to the methods section.

Minor comments:

Page 13086, Line 15 – revise the sentence for clarity.

Re-worded.

Page 13086, Line 18 – you can't say 'easily available' without testing the decomposability of the FL fraction :: the best you can say is 'potentially available'

Sorry, I do not understand the problem here.

Page 13086, Line 19 – consider replacing 'geogenic'

we replaced geogenic with "coal and lignite, probably inherited from the parent material".