

Answers to referee 1:

First, we like to thank the reviewer for thoughtful comments, which helped much to improve the structure and clarity of the manuscript.

General comments:

The manuscript would be easier to understand when:

- Objectives of the study in the “Introduction” are stated more clearly
- The methods section is elucidated for the data used to make the figures
- The results section present in figures are reconstructed to make the whole story easy to catch
- The summary section is stated more explicitly and concisely

We agree with these points and incorporated them into the revised version of the manuscript.

Specific comments:

Abstract and Introduction:

No clear and concise statements of the objectives from the beginning.

We added a one-sentence statement of objectives to the abstracts. However, in the Introduction, we prefer to state objectives at the end; we think that to be the more logic position.

Lines 30-31: this manuscript does not fully discuss how soil type and parent material affect SOC stock and stabilization. As far as I understand it discussed how SOC stocks and stabilization differed among soil horizons and among soil with different contents of Fe/Al oxides.

We did not mean to say that the study aimed at elucidating the effect of soil types and parent material on SOC. Instead, we focused on the distribution of OC among density fractions as related to soil properties (texture, Fe/Al oxides, pH, OC-to-TN ratio) and how the overall stability of soil OC relates to its distribution over density fractions. We explain that better in the revised version.

Lines 36-37: what is the exact meaning of lines 36-37, especially “reflect plant functional types in soil profiles not subject to plowing”?

It means that the depth profiles of LF-OC followed that of roots for forest and grassland sites unless the soil profiles was disturbed by plowing. We explain that better in the revised version.

Line 39: how did authors draw the conclusion that “fossil C derived from the geogenic...”? What is the evidence/data/citation of fossil C in this study?

The  $\Delta^{14}\text{C}$  of LF-OC indicated that OC was very old, i.e., from a time before onset of the recent soil formation. The  $\Delta^{14}\text{C}$  of the LF being less than that of the HF suggests a source of old carbon of low density. Also, we found coal particles in the samples, and so, we assume coal caused the small  $\Delta^{14}\text{C}$  values. Contamination by dust or other depositions would concentrate at or near the surface, but the  $\Delta^{14}\text{C}$  of the LF decreased with soil depth, pointing at the contribution of old carbon to increase with depth. Carbonic sedimentary parent material and coal mining in the surrounding of one of the sites led to the conclusion that the fossil C was inherited from the parent material. This has been stated more clearly also in the abstract now.

In lines 57-59, what are the references for this statement?

The review paper by Peltoniemi et al. (2007, *silva fennica*) gives a good summary of the features of common soil carbon models (Roth C, Century, Yasso, ROMUL, SOILN, and Forest DNDC). It shows that the separation of bulk OC into pools of different turnover times is mainly based on differences in OM quality. Also, the review of Gärdenäs et al (2011) gives a nice overview to the history of soil carbon modeling, and includes the statement that most models still rely on the idea that decomposition products become progressively more recalcitrant. We incorporated these references into the revised manuscript.

Lines 102-103: I do not quite understand this sentence

Actually the sentence is explained in the next one. We re-worded the sentences.

Line 118: I suggest to delete “in summary” in this line:

Omitted.

Materials and methods:

Lines 154-158: what is the measurement of root biomass along the soil profile

It is the dry mass of roots sorted out during sample preparation. We now explain in greater detail the determination of root distributions.

Line 161: please state which soil fraction was used to determine mineralization rates of soil OC, bulk soil OC, or fLF, or oLF, or HF

Mineralization rates were determined on bulk soil samples. We state that explicitly in the revised version.

Are 20 days long enough for the majority of the soil labile OC respired? Therefore, how can normalized CO<sub>2</sub> respiration rates over 20 days be used to indicate stability of OC?

We do not think that 20 days of incubation are enough to respire most labile OC. Nevertheless, the rate at which CO<sub>2</sub> is released during that period of time relates to the overall availability of labile material. That means, CO<sub>2</sub> rates normalized to total soil OC is an indicator of the share of labile matter in relation to total organic matter, thus, an indicator of stability. An OC-rich soil giving a small OC-normalized CO<sub>2</sub> respiration rate has little labile but much stable organic matter.

Line 173-174, Please specify how the calculation of “mass of C evolved per container and hour was calculated by taking the pH-dependent solubility of CO<sub>2</sub> in the soil water into account”

Since CO<sub>2</sub> evolution was calculated as the difference between two CO<sub>2</sub> concentrations, the accumulation of CO<sub>2</sub> caused increased dissolution of CO<sub>2</sub> in the water phase of the sample. The solubility of CO<sub>2</sub> is pH-dependent (solubility increased with low pH). Therefore we had to include pH in the calculations as shown below. The mass of C evolved per container and hour was calculated (from Persson et al. 1989, given in the reference list) as:

$$R_C = (12 \times 10^6 (C - C_0) p (V_g/RT + A \times V_{aq})) / t$$

where R<sub>C</sub> =  $\mu\text{g CO}_2\text{-C container}^{-1}$ , C = sample concentration of CO<sub>2</sub> (mL mL<sup>-1</sup>), C<sub>0</sub> = CO<sub>2</sub> concentration (mL mL<sup>-1</sup>) in the container directly after closure, t = time between closure and gas sampling (hours), V<sub>g</sub> = gas volume (mL) in the container, V<sub>aq</sub> = water volume (L) in the container, A = pH-dependent CO<sub>2</sub>-absorption factor, where:

$$A = K_1 + (K_1 K_2 / [H^+]) + (K_1 K_2 K_3 / [H^+]^2)$$

where K<sub>1</sub> (Henry's law constant) =  $4.79 \times 10^{-2}$  (mol L<sup>-1</sup> atm<sup>-1</sup>), K<sub>2</sub> =  $3.81 \times 10^{-7}$  (mol L<sup>-1</sup>), K<sub>3</sub> =  $3.71 \times 10^{-11}$  (mol L<sup>-1</sup>) (all values given for 288.15°K) and [H<sup>+</sup>] =  $10^{-\text{pH}}$  (mol L<sup>-1</sup>), p = atmospheric pressure (atm) when the container was closed, R = the gas constant (82.05) (mL atm °K<sup>-1</sup> mol<sup>-1</sup>) and T = temperature during incubation (288.15°K). The A factors were calculated from the final pH values at day 20.

These calculations were added to the revised manuscript.

Lines 220-222: ... is that the dry combustion of samples at 450°C for 16 hours can remove carbonate. While, is 450°C high enough to remove carbonate efficiently? What are the citations to support this method?

No, dry combustion was used to remove OC from the samples, not inorganic C. The remaining inorganic C was then analyzed with the elemental analyzer and OC calculated as the difference between total and inorganic C. We use a certified reference soil material (HEKATech GmbH, Wegberg, Germany) to assure quality control. Details on accuracy of the method have been published before (Steinbeiss et al 2008 GCB). We now refer to that work in the revised manuscript.

Line 234: where are soil samples from 5-10 cm? are they missing?

Due to limited budgets and instrument capacities, we analyzed only every second sample of the depth increments. However, we included instead more profiles to gain information on the spatial variation of  $\Delta^{14}\text{C}$  and more reliable averages for the sites.

Line 235: what do "these" samples refer to? Please clarify which samples were missing and which samples were analyzed.

Only the density fractions of the site Le Bray were lost before  $^{14}\text{C}$  analyses. Therefore, we only have bulk  $^{14}\text{C}$  data for that site. For all other sites, density fractions from the 0-5, 10-20, and 30-40 cm layers were analyzed. We added a more detailed explanation to the revised version.

Lines 234-235: please specify how we can know that density fractionation showed no contribution of fossil C.

We knew that only and made our decision that after we saw the  $^{14}\text{C}$  results of the density fractions. As it is not possible to interpret  $^{14}\text{C}$  results of soils containing fossil C ecologically, we decided to analyze only bulk soil samples of sites where reasonable interpretation of results seem possible. We added a detailed explanation to the revised version.

Results:

I suggest to restructure this section...:

We re-structured that section.

Definition of terms and their units in the methods section and consequent application in the rest of the manuscript:

Done.

Lines 271-272: why is only the data from cropland sites and the ploughed grassland site selected in Figure 2

At all these sites the soil profile was disturbed by ploughing and most of them contained coal in the LF, thus, hampering the interpretation of LF-OC values as quality indices for litter input of the current vegetation.

Lines 361-362: how were the contributions of organic layers to the total CO<sub>2</sub> evolved calculated? No description can be found in the methods section.

The CO<sub>2</sub> evolved per g soil/litter layer was determined in the incubations. Using the bulk densities or soil masses per area determined with the soil cores, CO<sub>2</sub> evolution can be calculated for each layer and then summed up for the total profile. The contribution of the litter layer is then the sum of CO<sub>2</sub> evolution from each litter layer divided by the CO<sub>2</sub> evolution from the profile. A description has been added to the methods section.

Lines 372-374: should be moved to the methods section

Done.

## Discussion

Lines 402-403: just results, better remove

Sentence omitted.

Lines 405-412: it is confusing that which OC is more important contribution to LF-OC, fossil C or geogenic C. In addition, what are the evidence/data of the contribution of fossil C to LF-OC in 3/12 sites?

The <sup>14</sup>C values of the three sites show that oldest fLF-samples had an average C age of about 11 150 - 11 300 years (97.7%, OxCal version 4.1.7) at Carlow, of 18 950 - 19 650 years at Easter Bush, and of 38 500 - 40 500 years at Grignon. As it can be assumed that also more recent plant material contributes to the LF at these sites, the old average ages justify the assumption of fossil C contained. We added age estimates a reference to the OxCal program to the manuscript. We did not aim to distinguish between geogenic and fossil C. We assume that fossil C can be inherited by the parent material at certain sites and we suggest considering that possibility when interpreting <sup>14</sup>C data of subsoils.

Lines 418-420: why can the authors state that roots are more stable and more important for soil OC formation? The results of this study show the relations between root mass and fLF-OC and oLF-OC rather than HF-OC, which do not support the statement above.

The results show that root amounts are linked to LF-OC and depth distributions of roots mirror those of LF-OC. A larger contribution of aboveground litter to LF-OC should have resulted in increased proportions of LF-OC located in topsoil layers versus subsoils, where aboveground litter likely will not be transported onto. Therefore we assume that roots form much of the LF-OC.

Lines 440-441: what is the evidence for the statement “there seems to be a similar situation at our topsoils”(Tables, Figures)

We removed that part from the revised version as the discussion does not really contribute to the main aims of the manuscript.

Line 450-453: sentence too long.

The sentence has been revised.

Lines 453-454: it would be better to say that “little or no relation between mineral indicators and OC can be expected when available binding sites at mineral surfaces are not limited” because limited factors are always the driving factors and can cause changes of dependent variables. However, the availability of binding sites at mineral surfaces does not indicate that mineral indicators are limited factors.

We agree and changed the sentence accordingly.

Lines 494-495: I did not see the reasons of put this sentence here

Sentence omitted.

Line 51-503: it does not make sense that “correlations between CO<sub>2</sub>-rease rates and OC in all three density fractions suggest that OC in all three fractions turns over fast enough to...”. First, what is this “CO<sub>2</sub>-release rate”, absolute or relative rates? I did not see the relations between fast turnover of OC and significant contribution to mineralization rates. The CO<sub>2</sub> evolved might be due to large amounts of labile OC in soil fractions rather than fast turnover of small amounts of soil OC.

The CO<sub>2</sub> release per g of soil was used for these correlations. As we are referring to the first two weeks of incubations, we cannot separate between fast turnover of a small amount of soil C or a large amount of labile OC in the fractions. It seems as if each fraction has a large enough labile component to contribute significantly to CO<sub>2</sub> release.

Line 503: what is the respiration rate: absolute or relative rate?

Again, this refers to CO<sub>2</sub> release per g of soil. We specify the used terminology in the revised version.

Lines 507-508: the statement is not new and commonly accepted, why repeat here?

Because it seems that the statement is true irrespective of the assumed stability of OC in the HF as also indicated by the link between the age of OC in the density fractions across sites in the topsoil layer: while OC stability in the HF is assumed, faster turnover of the LF seems to go along with faster turnover in the HF irrespective of soil mineralogy etc.

Line 519: it does not make sense that "... which is the prerequisite for degradation of mineral bound...".

What is the exact meaning here?

If sorption is assumed to be the reason for OC stabilization, desorption is necessary for the further decomposition or mineralization. Stronger sorption of OC at depth, i.e., reduced desorption will increase soil OC stability.

Line 520: which type of correlation is between OC-normalized CO<sub>2</sub>-release rates and bulk sample Δ<sup>14</sup>C, positive or negative? Moreover, how can the negative correlations for samples from 10-20 cm be used to draw the conclusion for samples at larger scale? Deeper soils may refer to soils in horizons deeper than 20 cm.

We added "positive" to the correlation. The positive correlation between Δ<sup>14</sup>C and specific mineralization rates shows that mineralization is linked to the mean residence time of OC. Specific mineralization rates depending on the OC in density fractions were not only found at 10-20 but also at 20-30 cm depth (for which we have no <sup>14</sup>C data). Except for the croplands, that depth increment is already within the B horizon, thus in subsoil. As we have no data for deeper soil layers, our statements are only backed up by the data of the 20-30 cm layer.

Line 542-545: What is the exact meaning of that sentence?

Older <sup>14</sup>C ages of OC in the oLF than of the fLF indicate that LF-OC is stabilized by occlusion within aggregates as. This is a confirmation of results of previous studies.

Summary

Line 561: it is strange to say "the distribution of OC over density fractions"

Since grammatically correct, we prefer to keep that expression.

Lines 562-567: long sentence

Re-worded.

Lines 571-573: what is the evidence for transfer of fLF-or oLF-OC to HF-OC?

Litter decomposition studies cited in the discussion showed that fLF-OC is transferred to oLF-OC and HF-OC. Our results show that  $^{14}\text{C}$  values of fractions are linked, showing that at sites with a short mean residence time of fLF-OC also oLF-OC and HF-OC have faster mean residence times.

Lines 573-574. The statement does not make sense. Please clarify.

This statement offers an alternative explanation for the differences in isotopic composition and OC-to-TN ratios between density fractions in the subsoil. In the subsoil, the transfer of LF material into the HF cannot explain the composition of the HF organic matter but movement of aged and processes dissolved OC can.

Title for the third point in summary section:

Occlusion within aggregates increases the longevity of LF-OC throughout the soil profile

Lines 582-584: please explain how HF-associated OM in topsoils is prone to be replaced by freshly produced reactive compounds

Organic matter sorption to fresh minerals shows comparably little fractionation. With the organic loading of the minerals increasing, less strongly bound compounds become replaced by more strongly sorbing compounds produced in the overlying layers. Evidence for that comes from the preferential removal of certain organic matter fractions (fractionation) and the depth increase in  $^{14}\text{C}$  age of dissolved OC in soil water with depth. This fits well to our observations that the age difference between fractions is little in topsoils and widens with depth.

Lines 592-593: is it necessary to say that OC in the LF represents fresh, unprotected OC? It is widely accepted.

We remove the second part of the sentence.

In lines 595-596: there should be no further discussion on unsolved problems in the summary section.

We agree and remove that sentence.

Line 602-604: results show weak correlations between mineral properties and OC. So how was the statement developed?

That statement refers actually not to correlation but simply to the fact that the utmost part of OC in the study soils was recovered with the HF.

Figures:

Figure 1: The last sentence of the figure capture belongs to the results section

The sentence was transferred.

Figure 2 Rescale x-axis of OC graph, OC-to-TN ratio graph, and d13C and make graphs easier to read.

The Figure is replaced by a figure showing just OC-to-TN ratios and  $^{14}\text{C}$  values of the sites Grignon, Easter Bush, and Carlow, which contained very old OC.

Please clarify the unit of OC g kg<sup>-1</sup>. Does it refer to g kg (bulk soil)<sup>-1</sup> or g kg (soil fraction)<sup>-1</sup>?

g kg (bulk soil)<sup>-1</sup>

Is the large value in the graph of OC-to-TN in Carlow outlier or not? If the point is outlier, why plot the point in the graph?

There was an error in the dataset which is now corrected

Figure 3 Why put these two graphs into one group? How was total soil respiration calculated? Does it make sense to use the word “total” for CO<sub>2</sub>-release per day?

They were placed together to show the potential CO<sub>2</sub> release from the profiles as related to OC stocks in the litter layer, OC stocks in the fLF, and total OC stocks. We replace the y-axis title by “potential CO<sub>2</sub> release of soil profiles” as values were obtained under optimal temperature and moisture conditions.

Figure 4 Are data in the two graphs average values or not? Please clarify

Yes, they are averages, standard errors are added.

Figure 5: Please specify the calculation of relative contribution per layer. What is the denominator?

The denominator is the total stock of LF-OC or roots in 0-60 cm. The contribution was calculated by dividing the LF-OC stock of each 10 cm depth increment by the total stock (times 100 to get %). The same was done for root masses. Calculation is explained in the methods part now.

Figure 6: Are the values in the graphs averages? If so, where are the standard errors? Perhaps Figure 6 can be replaced by a table or text to demonstrate the main point that root biomass was correlated with fLF-OC and oLF-OC at the top 0-10 cm considering the many figures of the manuscript.

The values are averages for each site. We used site averages of extracted roots as indicators for the root distributions at each site (100 cores per site) as single cores might not be representative. The figure is omitted.

Figure 8: "0-10cm" in the Figure caption should be deleted

Deleted and labels of x-axis were corrected.

Figure 9: "0-10cm" in the Figure caption should be deleted. The values of x-axis are too high, are they correct? Are the values in the graphs the average? If so, please add standard errors.

Deleted, values of the x-axis are correct, they were unfortunately wrong in Figure 8 (% instead of  $\text{g kg}^{-1}$ ). So, Figure 9 was corrected. The standard errors are given, but are about the size of the symbols or smaller.

Figure 11: The values in the right graph are too high, is the unit of x-axis of the right graph correct?

Yes, it is  $\mu\text{g CO}_2\text{-C g OC}^{-1}$ , 1000 times the values used for Figure 14, where  $\text{mg CO}_2\text{-C g OC}^{-1}$  is plotted to save space.

Figure 12: CO<sub>2</sub>-evolved during incubations have been found to be positively correlated with initial soil OC in many studies. Can be removed.

We agree and removed the figure.

Figure 13: This graph shows the relation between CO<sub>2</sub> evolved and OC of different soil fractions. The results suggest that each fraction has easily degraded OC. What will be the multi-regression between CO<sub>2</sub> evolved and OC of fLF, oLF, and HF? The multi-regression results might tell the different contributions of OC of each soil fraction to CO<sub>2</sub> evolved.

We tried multiple regressions, but it did not help to separate contributions of each fraction. Due to the small number of sites, the degrees of freedom are too small to address interactions and as variables are collinear, only the first variable in the model is significant. The result on different contributions depends largely on the treatment of the sandy sites Bugac and Le Bray with large amounts of OC in the fLF but small respiration rates. Including them, HF-OC would be more important, but fLF-OC when they are excluded. Therefore we rather present the data as it is.

Figure 15: “10-20 cm” in the Figure caption should be deleted. In addition, why were only the data of the samples from 10-20 cm reported here?

Values at 0-5 cm depth were not related, as stated in the text. For other soil depths, either data on mineralization rates (only 0-30 cm) or  $^{14}\text{C}$  (only 0-5 cm, 10-20 cm, and 30-40 cm) are not available.

Figure 16: What do the arrows from fLF to oLF and from oLF to HF mean? There are no straightforward hierarchical relationships among OC of the three soil fractions. SO, the arrows between them would be misleading.

The arrows are dotted and no straight line to indicate that the relative importance is unsure. The arrows pointing at the HF come from fLF, oLF, and dissolved OC. The figure shows no hierarchy among arrows but points at possible sources of OC in each fraction. Leaving the boxes unconnected would imply however, that they are independent of each other, which is likely not the case.

#### Technical corrections

We agree with the technical corrections and implemented them in the revised version.