

Answer to Dr Ding comments:

We wish to thank Dr Ding for his positive feedbacks and comments on our preprint. His major concern is about the discussion on the role of physical and physico-chemical protection in soil organic carbon persistence and the role of chemical recalcitrance, considered to be limited to pyrogenic organic carbon. We agree that this point could be improved. In order to emphasize this, we modified the discussion as follows: (i) The distinction between the three mechanisms leading to soil organic carbon persistence is made in the introduction (p.2, ll.7–32). We refer to broad and highly-cited papers on these topics to support our introduction (*e.g.* Sollins et al. 1996, Balesdent et al., 2000, von Lützow et al., 2006, Angst et al., 2016). In order to emphasize that pyrogenic organic carbon is a specific form of soil organic carbon for which chemical recalcitrance may be a determinant mechanism explaining its persistence, we added this sentence : “Even if chemical recalcitrance is regarded as a secondary parameter to explain bulk SOC persistence (Amelung et al., 2008), it could be a relevant parameter for a specific form of SOC: pyrogenic organic carbon (PyOC; Schmidt et al., 2011).” (ii) A specific section (4.3.1) of the discussion is dedicated to the discussion on the major role of physical and physico-chemical protection in soil organic carbon persistence in the fine fractions in light of the major works which studied and explained these processes. (iii) We discuss the specificities of pyrogenic organic persistence and the likely role of chemical recalcitrance in a separate section (4.3.2) to distinguish properly this mechanism, specific to pyrogenic organic carbon, from the two other mechanisms, more widely acknowledged. As detailed in this discussion, we must exclude physical and physico-chemical protection from the dominant mechanisms explaining pyrogenic organic carbon persistence considering the size of the fraction.

Other minor comments, detailed in the attachment, are addressed as follows:

p.8, l.20: The reviewer is right, the increase of the OC content is not so clear at first sight. We will modify the sentence accordingly: “Inputs of OC in the fine clay subfraction is evidenced by the absolute increase of OC content after 10 years of experiment (Fig. 1b). Compared to the decreasing trend, such increase is also observed in the clay fraction and subfractions.”

p.8, l.25: The table numbering was wrong, Table 2 will be modified to Table 3.

p.18, l.17: We acknowledge that the sentence highlighted by the reviewer was not clear and we will modify it as follows: “Breakdown of coarse OM would lead to inputs of new OC with variable chemistry in the finest fractions, particularly in the clay subfractions. The scattering of the HI and OI values in the clay subfractions during the intermediate years of LTBF experiment strengthen this idea (Figs. 3b and 3d).”

Answer to Dr Van Zwieten comments:

We would like to thank Dr Van Zwieten for his careful review and feedbacks on our manuscript. Three major concerns arise from his comments.

Dr Van Zwieten suggests to reinforce the discussion on pyrogenic organic carbon persistence and support it with studies on the residence time of natural sources of pyrogenic organic carbon. To the best of our knowledge, only few studies exist calculating natural pyrogenic organic carbon residence time over several decades (Hammes et al., 2008, Nguyen et al., 2008, Cheng et al., 2008, Lehman et al., 2008) reporting residence time gives values of the same order of magnitude than our findings (data reviewed in Singh et al., 2012, average fast pool 91 years and slow pool 1034 years). As two pools of PyOC with long (>100 years MRT) and short (40 years MRT) residence times are found, we expect that bulk PyOC pool would exhibit a centennial turnover, as shown by Lutfalla et al., 2017. In addition, it is increasingly acknowledged that pyrogenic organic carbon *in situ* residence time (where the PyC was produced, so accounting for lateral and horizontal losses) in soil is shorter than previously thought, with possible fast turnover controlled by environmental conditions (Abney & Berhe, 2018). On this basis, we will reinforce the discussion as follows: “The equivalent persistence of total OC and PyOC in the clay fraction detected in this study contradicts most literature stating that PyOC is more persistent than soil OC (Forbes et al., 2006). However, analyses of published data show that PyOC persistence is less than previously expected (Abney and Berhe, 2018). Long-term field-based studies show mean residence times ranging from 90 years to 1034 years (Singh et al., 2012). The average MRT for the intermediate OC pool in the clay fraction was estimated at 40 years (SD = 15 years) from this study and PyOC decrease was following the same trend. Therefore, this study suggests that MRT of the finest PyOC particles may even be shorter than lowest bulk estimates. These results agree with incubation experiments which demonstrated that pyrogenic material can decrease at similar rates than total OC in the first phases of biodegradation (Hilscher et al., 2009). The balance between centennially persistent coarse PyOC particles and more labile fine PyOC fraction would lead to centennial residence time, consistent with residence times found for natural pyrogenic carbon in field experiments (182 years to 541 years, Hammes et al., 2008). This is also consistent with previous studies of PyOC persistence in bulk samples from LTBF showing limited MRT differences with total OC, of the order of the century (Lutfalla et al., 2017).”

Vertical or horizontal transfer of PyOC (as soluble or particulate) may also be an important factor on the long term (Abney & Berhe, 2018). Two recent studies showed that the importance of the transfer is mainly a function of the soil properties (Schiedlung et al., 2020 ; Bellè et al., 2020). These studies show that soil with a large porosity or low C content will favour the transfer of soluble and particulate PyOC in soil depths. While these mechanisms are not a loss of PyOC from the soil *per se*, it is still a loss from our study perspective, since it quitted our study boundaries. So the *in situ* MRT of PyOC can decrease because of this. In term of quantities, the soluble fraction of PyOC is small, less than 0.1 % (Abiven et al., 2011) and is thus negligible in the calculated MRT. Vertical transfer of PyOC particles compared to average soil particles results from its low density. Fine PyOC particles are more likely to interact with mineral phases than coarse PyOC particles, and are therefore more likely included in aggregates with higher density compared to free coarse PyOC particles. The leaching processes should thus be more important for coarse than fine grained PyOC. The longer residence time of coarse PyOC in this study

indicates that vertical or horizontal transfer is expected to play only a minor role in PyOC persistence in this work. It will be discussed in more details as follows: “Different mechanisms can explain the decrease of PyOC content in the clay fraction: biotic degradation, photo-oxidation and vertical and horizontal transport or erosion. Due to twice-yearly tillage, photo-oxidation, impacting PyOC material situated at the soil surface, can occur despite the sampling depth (25 cm) of this study. All plots from the experimental setup of the LTBF experiment were fenced in the late seventies, limiting horizontal losses of OC and more specifically of PyOC by erosion from this date. Earlier horizontal losses can account for PyOC decrease. Vertical transport cannot be excluded either from this setup, but it should affect both coarse and fine PyOC particles. The content in sand-sized PyOC particles remaining stable over the duration of the experiment, erosion and transport of PyOC must be limited. In addition, potential losses of PyOC by solubilisation is minor compared to biological degradation (Abiven et al., 2011; Maestrini et al., 2014, Schiedung et al., 2020). In general, biological or biotic degradation is demonstrated to be the predominant degradation pathway for PyOC (Santos et al., 2012). The observed loss of PyOC must thus result mainly from biotic processes.”

The third major comment relates to the development of comprehensive hypotheses in the introduction. We will detail the introduction to develop our working hypotheses as follows: “The objectives and hypotheses of this work are threefold. (i) We aim to identify SOC pools with distinct dynamics (*i.e.* storage and movement of OC in the soil and between soil compartments) as the heterogeneous properties of the studied fractions should lead to varied SOC residence times giving the opportunity to separate SOC in distinct pools. (ii) We want to determine the thermo-chemical characteristics of each pool as their dynamics is expected to depend on their stoichiometry (*e.g.* H and O content), chemistry (*e.g.* aromaticity for PyOC) and thermal stability. (iii) We wish to deduce the mechanistic origin of their dynamics (*i.e.* the physical, chemical and biological mechanisms at stake). The correlations between SOC dynamics and thermo-chemical properties in a given pool will enlighten the nature of the underlying mechanisms explaining SOC persistence.” Each hypothesis is then addressed in a specific section of the discussion (4.1, 4.2, 4.3).

More specific minor comments are addressed as follows:

p. 2, l. 28: We agree with Dr Van Zwieten, what is behind the term dynamics can be unclear to some readers. However, this term, widely used in the literature, describes properly the purpose of this part of our work. For this reason, we decided to keep it but give a clear definition of what we mean: “(*i.e.* persistence and decline of OC in the soil and between soil compartments due to mineralization, leaching or transfer towards other soil fractions)”.

p. 2, l. 29: Mechanistic may also seem to be a loose term and will be clarified: “(*i.e.* the physical, chemical and biological mechanisms at stake)”. Nonetheless, the term will be kept as it is a convenient word to cover the variety of mechanisms controlling soil organic carbon persistence in soil without prior assumption on their nature or relative importance.

Fig. 4: Dr Van Zwieten wonder if these data would be better presented as a table. Even if the number of data points is limited, we believe that a graph is more appropriate as it gives the opportunity for the reader to see the trend in the data and the standard deviation at first sight. The precise value of each data point is also given in Supplementary Information.

p. 19, l. 3: Following Dr Van Zwieten advice, we will modify the title of the section 4.1.3 to “Pyrogenic organic carbon exhibits contrasting dynamics between coarse and fine fractions”, in order to explain with respect to what (granulometry) the term dynamics was used.

The other minor comments will be modified according to Dr Van Zwieten suggestions.