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Evaluating the two-pool decay model for biochar carbon permanence

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Abstract

Accurate estimation of biochar carbon permanence is essential for assessing its effectiveness as a carbon dioxide removal (CDR) strategy. The widely adopted framework, based on the two-pool carbon exponential decay model, forms the basis of policy guidelines and national CDR accounting. However, our re-analysis of the meta-data used in this model reveals significant deficiencies in its parameterization, leading to two critical issues. First, the current parameterization assigns a disproportionally low percentage of the labile carbon fraction (C1) relative to the recalcitrant fraction (C2), effectively reducing the model to a single-pool approach. Due to the limited duration of incubation experiments, the decay constant of the labile fraction is incorrectly applied to the entire biochar mass, resulting in a considerable overestimation of the biochar decay rate. Second, our analysis reveals a lack of causal correlation between the assigned proportions of C1 and C2 and key carbonization parameters such as production temperature and hydrogen-to-carbon (H/C) ratios, suggesting that the model does not accurately represent the underlying chemistry. This misalignment contradicts the established relationship between increased biochar stability and a higher degree of carbonization. Consequently, the the parameterization of current model may not adequately reflect the carbon sequestration potential of biochar. While a multi-pool decay model is suitable for predicting the permanence of biochar, the primary issue with the current model lies in its parameterization rather than its structure. To address these limitations, we recommend that future research prioritize the development of a revised multi-pool decay model with improved parameterization, supported by empirical decomposition data from a variety of experimental methods, including incubation studies, accelerated aging experiments, and comprehensive physicochemical characterization. This refined approach will improve the accuracy of biochar permanence estimations, strengthening its role in global carbon management strategies.

Highlights

- Reassessment of biochar decay model reveals key parameterization deficiencies.
- Current model substantially overestimates biochar decay rates.
- Model misaligns with the chemistry underlying the decay behavior of biochar.
- New decay models for biochar permanence need improved parametrization.
- Enhanced model aims to strengthen biochar's role in carbon management strategies.

Keywords Biochar permanence, Carbon sequestration, Two-pool model, Decay rates, Parameterization

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1 Introduction

The permanence of biochar carbon in soil is a crucial factor in its efficacy as a climate change mitigation strategy (Woolf et al. 2021; Lehmann et al. 2015, 2021; Lefebvre et al. 2023). Over the past two decades, extensive research has been conducted on biochar carbon permanence, primarily through laboratory incubation experiments (Major et al. 2010; Zimmerman 2010; Singh et al. 2012; Zimmerman and Gao 2013; Fang et al. 2014, 2019; Herath et al. 2015; Kuzyakov et al. 2014; Dharmakeerthi et al. 2015; Wu et al. 2016; Budai et al. 2016). These controlled studies minimize the risk of biochar loss through erosion, enabling precise measurements of biochar degradation rates over time (Wang et al. 2016).

The standard methodology involves mixing biochar with soil in an incubation chamber at a constant temperature, capturing the CO_2 released, and using carbon isotope analysis to distinguish between carbon released from the biochar and the soil (IBI 2013; Camps-Arbestein et al. 2015; Lehmann et al. 2015; Whitman et al. 2015). Empirical data from these studies provide cumulative carbon loss of biochar over various time intervals, which are then used to fit mathematical functions, most commonly exponential decay functions (Woolf et al. 2021; Lehmann et al. 2015, 2021; Wang et al. 2016; Ogle et al. 2006; Azzi et al. 2024). These functions project the ongoing decay rate to predict biochar carbon loss over specific future periods. To ensure comparability across different experiments, the decay constant from each study is adjusted to the average global soil temperature of 14.9 °C (Woolf et al. 2021; Lehmann et al. 2015, 2021).

The results from these incubation experiments, alongside associated mathematical modeling processes, are extensively reviewed in academic literature. These studies form the basis for methodologies used by the IPCC (Ogle et al. 2006) to integrate biochar into national emission inventories. Woolf et al. (2021) aimed to refine the IPCC biochar methodology by incorporating additional analyses from existing scientific literature and introducing more parameterization options for estimating biochar permanence, henceforth the 'Woolf's model'. Their approach involved a meta-analysis and curve fitting based on incubation study results, providing estimates of the fraction of biochar carbon remaining in soil (F_{perm}) after 100, 500, and 1000 years. This methodology is also utilized by several biochar certification programs, including Puro.earth, Verra VCS, and Riverse (Puro.earth 2024; Verra 2023; Riverse 2013).

In this study, we critically re-examined the metadata and methods presented by Woolf et al. (2021) to assess the validity of the prevalent certification approach for biochar as a CDR strategy. We discussed the limitations of this method and underscored the necessity for refining the Woolf's model to enhance the accuracy of biochar permanence estimation, thereby strengthening the role of biochar in global carbon management strategies.

2 Examining carbon pool parameterization in the 'two-pool' model

The rationale for using the double exponential model, or "two-pool model," in Woolf's meta-analysis, as well as in studies by Lehmann et al. (2015, 2021), Ogle et al. (2006), and Azzi et al. (2024), is grounded in the widely accepted understanding that biochar is composed of a mixture of both aliphatic and aromatic organic compounds. The larger aromatic structures are generally more persistent than the aliphatic ones. As a result, it is advocated that biochar decomposition is more accurately described using a multi-pool decay function rather than a single-pool model, to better reflect the heterogeneous compositional chemistry of biochar.

The Woolf et al. (2021) meta-analysis of incubation results explicitly recommends a minimum of a two-pool exponential model, consistent with other works preceding and following theirs (Singh et al. 2012, 2015; Rodrigues et al. 2023). In Woolf's model, the first carbon fraction (C1) corresponds to the labile/aliphatic fraction and exhibits a higher decay rate (K1 decay constant), while the second carbon pool (C2) corresponds to the recalcitrant/aromatic fraction and demonstrates a lower decay rate (K2 decay constant).

Woolf's meta-analysis also includes a small subset of six decay series modeled with a three-pool exponential model (Herath et al. 2015; Kuzyakov et al. 2014), which is outside the scope of our discussion.

An examination of Woolf's meta-analysis (presented in Woolf et al. (2021) supplementary information) reveals that the carbon content in biochar is divided such that the relative proportion of C1 to C2 is minimal, effectively rendering the impact of C1 in their "two-pool" model insignificant. Across the dataset, the median value of C1 is 0.6%, while the median C2 fraction is 99.4%.

Figure 1 compares the estimated F_{perm} (fraction of carbon remaining after 100 years) based on the incubation metadata reported by Woolf et al. (2021) using their two-pool model, with F_{perm} calculations in which the C1 carbon pool is intentionally set to zero. This comparison highlights the effect of minimizing the C1 fraction within the two-pool model. The results show that both sets of estimates align closely with the parity line, indicating that, although Woolf's model claims to represent a 'two-pool' exponential decay, it is effectively governed by the decay of a single carbon pool within the biochar.

The division of biochar carbon into C1 and C2 pools in Woolf's model was primarily undertaken to facilitate curve fitting, with the objective of achieving the closest

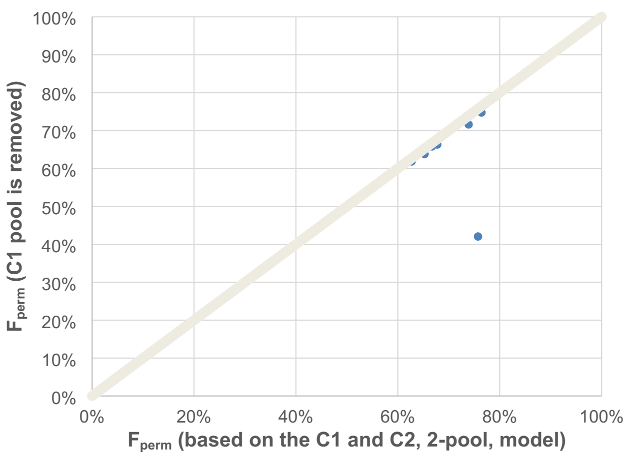

Fig. 1 Comparison of the estimated F_{perm} (fraction of carbon remaining after 100 years) from Woolf et al. (2021) using their two-pool model and the F_{perm} calculations with the C1 carbon pool intentionally set to zero. This comparison demonstrates almost perfect alignment with the parity line, indicating that the C1 pool is irrelevant in the model. The results suggest that the F_{perm} estimates are effectively governed by a single carbon pool decay, despite the nominal use of a two-pool model

fit to the empirical data for carbon decay observed over time during the incubation experiments. However, the crucial question remains: to what extent does this division reflect the actual chemistry of biochar? Specifically, what proportion of the observed carbon loss during each incubation experiment can be attributed to the labile fraction of biochar, and what proportion can be attributed to the recalcitrant fraction of biochar?

The most recent published metadata compilation of incubation studies by Azzi et al. (2024), which also includes those in Woolf et al. (2021), shows that the total carbon loss during the incubation of biochar produced at temperatures between 350 °C and 800 °C ranges from 15% to 1%, respectively. Additionally, recent metadata of 75 biochar samples published by Sanei et al. (2024) and its update (unpublished) for biochar produced at the same temperature range of 350 °C to 800 °C indicates that their measured labile carbon fraction varies from 16% to 1%, respectively. The total carbon loss during incubation experiments is therefore likely to be always within or less than the actual labile carbon fraction of biochar samples. Therefore, the incubation experiments to date have measured either entirely or predominantly the degradation rates of the labile fraction of the biochar.

This has significant implications for interpreting the incubation results. Given that the total carbon loss during multi-year incubation experiments is attributed entirely to the decay of the labile fraction of biochar, the decay rates of the recalcitrant fraction remain empirically unknown due to the relatively short duration of the reported experiments thus far. Consequently, the projected F_{perm} data by Woolf's model and later compilations (Azzi et al. 2024; Rodrigues et al. 2023) reflect the permanence of only the labile fraction of biochar. The estimates of permanence based on such models significantly underestimate the true permanence of biochar. This finding aligns with and explains the discrepancies and problems outlined by Sanei et al. (2024) regarding the Woolf's model.

Sanei et al. (2024) argued that biochar, thermodynamically considered on par with some of the most stable forms of organic carbon, should exhibit much greater permanence. If the performance of biochar was as short as suggested by current models, it would imply that surface processes are so strongly oxidizing that no organic matter could persist long enough to transition to geological stages. This contradicts the abundant presence of natural fossil charcoal in geological rocks found in various depositional environments (Scott 1989, 2000), particularly in shallow and surface outcrops of sedimentary rocks that contain significant amounts of preserved charcoal (ICCP 2001; Sanei et al. 2024 and references therein).

3 The disconnect between the current model and biochar chemistry

As mentioned earlier, there is a general consensus that biochar contains roughly two fractions: labile and recalcitrant carbon (Lehmann et al. 2015; Woolf et al. 2012). These fractions are quantified through analytical techniques, such as proximate analysis (volatile matter ≈ labile fraction; fixed carbon ≈ recalcitrant) or other thermal analyses (labile versus refractory; Petersen et al. 2023; Sanei et al. 2024), or by direct spatial measurement of the percent inertinite in a biochar sample using reflected light microscopy (Sanei et al. 2024). It is also generally accepted that the relative proportion of labile to recalcitrant fractions decreases with increasing degrees of carbonization, aromatization, and condensation of organic molecules, resulting from higher production temperatures and/or longer heating residence times (Carr and Williamson 1990; Morga 2011; Wiedemeier et al. 2015; Budai et al. 2017; Zhang et al. 2017; Liu et al. 2020; Howell et al. 2022; Sanei et al. 2024). The fractionation of the C1 and C2 proportions in Woolf 's model should follow the same general trend rooted in the chemistry of biochar that defines its carbon stability.

A critical question regarding Woolf's model is whether the estimated decay of biochar aligns with the fundamental chemistry of the biochar used in the incubation studies. One way to investigate this is by examining how the relative proportions of the two carbon fractions (C1, C2) vary in biochar samples produced at different pyrolysis temperatures. Biochar produced at higher temperatures is expected to have lower H/C ratios and a higher proportion of the C2 fraction, reflecting an increased degree of carbonization.

Figure 2 illustrates the relationship between the C2 fraction and both production temperature and molar H/C ratio for all modeled biochar samples from the Woolf et al. (2021) meta-analysis. These plots aim to investigate how production temperature and carbonization level influence the distribution of recalcitrant, C2 carbon fraction in biochar.

Contrary to expectations, the results show no correlation between the C2 fraction and either production temperature or H/C ratio. Across a wide range of production conditions, the C2 fraction fluctuates randomly between 95% and 100%, regardless of production temperature or H/C ratio. Notably, there is no observable distinction in the C2 fraction between biochars produced at lower temperatures (< 400 °C; H/C > 0.8) and those produced at higher temperatures (> 600 °C; H/C < 0.4).

The absence of a clear relationship between the assigned C2 fractions and the degree of carbonization in Woolf's model indicates a misalignment with the established chemistry of biochar. This discrepancy undermines the validity of the model, as proper parameterization of a decay model must be rooted in a robust understanding of the chemical processes governing carbon degradation and stability. Specifically, biochar produced at higher pyrolysis temperatures should exhibit greater

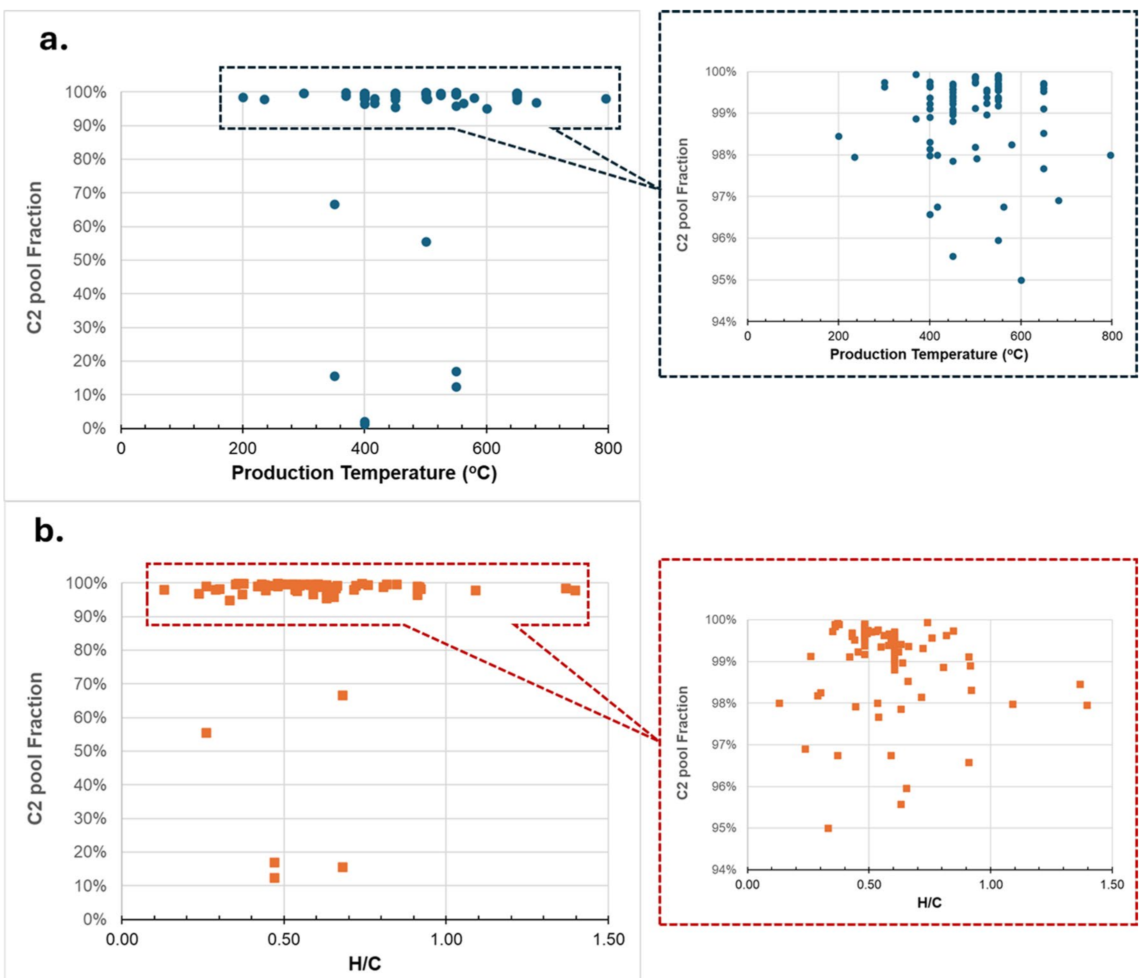

Fig. 2 **a** Relationship between the recalcitrant, C2 fraction and production temperature (the pyrolysis temperature at which biochar is produced). **b** Relationship between the C2 fraction and the H/C molar ratio across the entire dataset from the Woolf et al. (2021) meta-analysis of modeled biochar samples. These plots indicate that neither production temperature nor the atomic H/C molar ratio has a significant influence on the degree of carbonization in biochar, as reflected by the size of the recalcitrant (C2) carbon fraction in the Woolf model. This observation contradicts the chemical expectation that increased carbonization, driven by higher pyrolysis temperatures, should systematically decrease the H/C molar ratio and increase the recalcitrant carbon fraction size in biochar

carbonization, characterized by a smaller proportion of labile carbon and a larger recalcitrant fraction (C2).

4 Recommendations for improving biochar permanence models

To address challenges in biochar carbon permanence models, we proposed several recommendations to improve their accuracy and reliability. A revised multi-pool decay model remains a suitable framework for predicting biochar permanence, provided it is parameterized to reflect the chemical properties of biochar accurately. Key considerations for this revised model include the need for accurate parameterization, where the model incorporates precise proportions of labile (C1) and recalcitrant (C2) carbon fractions based on empirical data. This requires comprehensive chemical analysis of biochar samples to determine their exact composition. Additionally, the model should be calibrated using decomposition data that capture both the rapid degradation of labile fractions and the slower degradation of recalcitrant fractions, ensuring a more accurate representation of biochar long-term stability.

The model should also clearly differentiate between various carbon pools within biochar, potentially expanding beyond the simple two-pool model to include multiple fractions that reflect the complex nature of biochar degradation. Moreover, the model should account for production variables such as production temperature and H/C ratios, which significantly influence the composition and stability of biochar. This allows the model to be tailored to specific types of biochar, thereby improving its predictive accuracy.

In addition to revising the multi-pool model, alternative approaches should be employed to complement and validate the model, especially in the absence of long-term incubation data. These approaches include accelerated aging studies, which simulate long-term environmental conditions in a shorter timeframe and can provide valuable data on the long-term stability of both labile and particularly recalcitrant carbon fractions.

Detailed physicochemical characterization, such as spectroscopy, chromatography, and microscopy, should be conducted to understand the structural and compositional characteristics of biochar. This information can refine model parameters and enhance the accuracy of carbon stability predictions. Furthermore, molecular models that simulate the molecular structure and degradation pathways of biochar can offer insights into the long-term behavior of different carbon fractions, helping to predict how biochar interacts with soil components and environmental factors over extended periods.

Finally, integrating data from incubation experiments, accelerated aging studies, physicochemical analyses, and molecular models can create a comprehensive dataset. This integrated approach ensures that the model reflects the complex interactions and long-term stability of biochar in various environmental contexts.

By adopting these recommendations, future models can more accurately predict the long-term permanence of biochar carbon in soil, providing reliable estimates for its role in carbon management and climate mitigation strategies.

5 Conclusion

Our critical review of the commonly used two-pool exponential decay methodology, based on a meta-analysis of over 100 incubation experiments compiled by Woolf et al. (2021) and subsequent studies, reveals significant shortcomings in current biochar permanence models.

This model theoretically divides total carbon loss during incubation into labile carbon (C1) and recalcitrant carbon (C2) fractions, with C1 assigned a higher decay rate (K1) than C2 (K2) to account for the heterogeneous composition of biochar. However, in the current implementation, the proportion of C1 relative to C2 is so low that the model effectively functions as a single-pool model. Empirical carbon loss observed during incubation primarily reflects the degradation of the labile fraction, as the degradation of permanent fraction cannot be distinguished during the experiments. Consequently, the model inaccurately assumes biochar is predominantly composed of the labile fraction, applying an incorrect, higher decay constant to almost the entire biochar mass. This results in a gross overestimation of the decay rate for the largely inert recalcitrant fraction.

Furthermore, the proportion of the recalcitrant C2 fraction assigned to biochar samples in Woolf's model exhibits no correlation with production temperature or H/C ratio. This observation challenges the well-established principle that higher production temperatures and increased carbonization typically lead to a greater relative proportion of the recalcitrant fraction. The disparity between the meta-data and the expected chemical behavior of biochar's labile and recalcitrant fractions underscores a critical limitation in current models, highlighting their inability to accurately represent the long-term carbon sequestration potential of biochar. This has significant implications for biochar certification and climate policy (European Commission 2022, 2024), highlighting the need for more robust models that account for the complex nature of biochar degradation, and presence of a fraction with decomposition rate so low that it cannot be reliably measured in incubation studies lasting only a few years.

Addressing these issues involves using a revised multi-pool decay model that is appropriately parameterized to reflect the chemical properties of biochar and calibrated with decomposition data that capture both labile and stable fractions. In the absence of long-term incubation data, this approach should be complemented by combining incubation data with accelerated aging studies, physicochemical characterization of biochar, and molecular models to ensure accurate predictions of biochar permanence.

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Author contributions
All authors contributed to the literature analysis, conceptual development, and writing of this manuscript. Each author played an active role in reviewing and refining the analysis and interpretation presented.

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Availability of data and materials
This perspective paper is based exclusively on data available in the literature. No new data were generated or analyzed during this study. All sources are appropriately cited, and the information discussed is fully documented within the manuscript. No additional data or materials will be made available beyond what is presented here.

Declarations

Competing interests
Ondrej Masek is an EBM of the journal Biochar, and he was not involved in the peer-review or handling of the manuscript. The authors have no other competing interests to disclose.

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