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Can inert pool models improve predictions of biochar long-term persistence in soils?

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ABSTRACT

The long-term persistence of biochar in soil is often predicted by extrapolating mineralization data from short-term laboratory incubations. Single first-order, double first-order, triple first-order and power models have been employed for this purpose, all of which have an inherent assumption that biochar is biodegradable. However, recent insights challenge this assumption by suggesting that a large fraction of biochar is inert. If so, it would make sense to reflect this in the models used, by incorporating an inert carbon (C) pool. We hypothesized that such inert pool models would fit better to incubation data than existing models and give more reliable long-term predictions. We evaluated this by fitting the models to data from a recently compiled extensive dataset of biochar incubations. The inclusion of an inert pool enhanced the model fits over first-order models in most cases. However, inert pool models overestimated biochar persistence compared to the measured outcomes. By contrast, the double first-order model, which has been the most widely used to date, underestimated biochar persistence even in the short term. The power model in general outperformed all other models and gave the most reliable predictions, although it was sensitive to increasing or fluctuating mineralization rates in the datasets.

1. Introduction

Following the pioneering work by Wim Sombroek in Amazon soils (Sombroek, 1966), biochar garnered attention due to its beneficial impacts on soil fertility (Glaser et al., 2001; Glaser et al., 2002). In recent decades, emphasis has been placed on biochar's potential to store carbon (C) in soils, owing to its resistant nature (Lehmann, 2007; Smith, 2016; Wu et al., 2019). Compared to raw biomass, biochars are one to two orders of magnitude more persistent (Lehmann et al., 2021; Azzi et al., 2024), which can be attributed to the formation of fused aromatic structures during biomass pyrolysis (Howell et al., 2022). The inclusion of methods for estimating carbon stock change as a result of biochar application in the *2019 Refinement to the 2006 IPCC Guidelines for National Greenhouse Gas Inventories* is a recognition of biochar's potential as a carbon sequestration method (IPCC, 2019). While biochar is gaining recognition as a method for CO_2 removal, prediction of its persistence in soil is difficult.

Such predictions rely on extrapolation from short-term mineralization data from laboratory incubations. Single first-order (SFO) (Rasse et al., 2017), double first-order (DFO) (Major et al., 2010; Singh et al. 2012a; Singh et al. 2012b; Fang et al., 2014), triple first-order (TFO) models and power model (Zimmerman, 2010; Liu et al., 2020) have all been used to fit mineralization datasets and predict biochar persistence. These models differ in their assumptions about the degradation dynamics of biochar components. The SFO model assumes that biochar is homogenous and that the decay follows first-order kinetics, while the more commonly used DFO model assumes the existence of one rapidly degrading, labile (months-years), and one more recalcitrant, stable (decades-centuries) C pool. The TFO model further expands to three pools: a labile, a semi-labile and a recalcitrant pool. By contrast, the power model assumes an infinite number of degrading pools, with a linear relationship between the logarithm of decay rate and the logarithm of time, implying that biochar consists of a continuum from more labile to more refractory C compounds (Zimmerman, 2010). Current models are based on the assumption that all biochar C pools are biodegradable, albeit some of the C pools degrade slowly. This assumption has been challenged by a perspective that condensed aromatic structures within biochars are not only stable and capable of

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persisting for centuries in the environment but completely resistant to biological transformation (Schmidt et al., 2022; Sanei et al., 2024). For instance, Sanei et al. (2024) concluded that the majority of commercial biochars consist entirely of inertinite, and suggested a half-life of approximately 100 million years for such biochars, even in an oxidizing abiotic environment. This view is in line with geological evidence that black C can be preserved for millions of years and be partially inert (Cope and Chaloner, 1980; Forbes et al., 2006).

From this, it seems logic to incorporate an inert C pool (i.e. a constant that does not degrade over time) into existing models used to fit laboratory incubation data. Such inert pool models could be used as a way to assess if results from biochar incubations are consistent with the view that biochars contain an inert fraction. If it works, it would have the added benefit of allowing us to estimate the size of this inert pool for different biochars from the mineralization data and compare it with estimates from other sources, such as chemical characterization. The concept of using inert-pool models is not new; it has been applied to simulate soil organic C (SOC) changes in fallow soils (Barré et al., 2010) and it was employed to estimate the inert-pool sizes of biochar based on short incubations (only 74 days, Farrell et al., 2013) and to predict the persistent aromatic C pool (Schmidt et al., 2022). However, to the best of our knowledge, no systematic evaluation of using inert-pool models for biochar incubation data has been performed.

Recently, we compiled biochar decomposition data from 134 decomposition time series (Azzi et al., 2024). Here, we explore the potential of using inert pool models for estimating biochar persistence based on this dataset. Our first objective (i) was to assess whether inert pool models (SFO + I) and (DFO + I) would improve the fits to incubation data compared to existing models (SFO, DFO, TFO and power model). Our second objective (ii) was to evaluate the ability of the models to predict biochar persistence, based on the incubation data from Kuzyakov et al. (2014) and similar studies spanning at least 2 years. Our hypothesis was that inert pool models would fit better to biochar mineralization data than existing models and provide more reliable long-term predictions.

2. Materials and methods

2.1. Data sources

We used the database compiled during our previous synthesis study (Azzi et al., 2024), but we made a selection of data based on the following inclusion criteria: (1) biochar incubated under aerobic conditions (i.e. excluding flooded conditions as in Wu et al., 2016); (2) duration exceeding 365 days (i.e. excluding the studies from Rasse et al., 2017; Zhu et al., 2019); (3) incubation temperature maintained below 40°C (excluding some data from Fang et al., 2014); (4) soil incubation under constant conditions, i.e., without changes in moisture content, temperature or nutrients (excluding the field studies from Major et al., 2010; Ventura et al., 2015, Ventura et al., 2019); (5) biochar incubated in soil (excluding Aubertin et al., 2021). Additionally, we digitized three observations from Zimmerman (2010) instead of using the reconstructed data that was present in the original database. Based on these criteria, a total of 9 articles containing 72 datasets (observations) of biochar mineralization were retained (Supplementary material 1). For comparative purposes, the analysis incorporated 4 observations derived from non-pyrolyzed raw biomass decomposition from Budai et al. (2016) and Santos et al. (2012).

2.2. Data quality selection

The models used in this study assume a pattern of faster biochar decomposition during the initial stages of incubation, followed by a gradual slowing down over time, reaching a plateau in the case of the inert pool models, and are sensitive to deviations from this pattern. Such deviations can arise due to fluctuations in soil temperature, other unidentified factors or measurement errors can affect model fits, the accuracy and reliability of model predictions. To avoid such problems, datasets were visually inspected by plotting the biochar C remaining over time in scatter plots, and the decomposition datasets that contained obvious increases or fluctuations in mineralization rate were excluded (31 observations). The reasons for data exclusion are further detailed in Table S1. In total, 41 observations were selected for the subsequent curve fitting. An attempt was also made to fit the excluded datasets in order to provide additional rationale for their exclusion from further discussion (Fig. S1).

2.3. Biochar C modeling

The selected biochar C mineralization observations were fitted using Sigmaplot 14.0 through non-linear regression, employing the Levenberg–Marquardt algorithm. This algorithm estimates the parameter values that minimize the sum of squares of variances between observed and predicted values, and it is commonly used in biochar mineralization curve fitting (Fang et al., 2014; Budai et al., 2016; Santos et al., 2021). All C pools and decay rates were constrained to be larger than zero except for the inert C pools where the decay rate was zero by default. Initial parameter values were estimated based on the provided data using the built-in equations in Sigmaplot. The detailed curve fitting results are shown in Supplementary Material 1. The following models were evaluated:

SFO model:

$$C_r = C_0 * \exp(-k*t) \tag{1}$$

where C_r is biochar C remaining at time t, C_0 is initial biochar C and k is the first order mineralization rate, respectively.

DFO model:

$$C_r = C_1 * \exp(-k_1 * t) + C_2 * \exp(-k_2 * t) \tag{2}$$

where C_r biochar C remaining at time t and C_1, C_2, k_1, k_2 represent a labile biochar C pool, a stable biochar C pool, and the first order rate constants for the labile and stable pools, respectively.

TFO model:

$$C_r = C_1 * \exp(-k_1 * t) + C_2 * \exp(-k_2 * t) C_3 * \exp(-k_3 * t) \tag{3}$$

where C_r biochar C remaining at time t and C_1, C_2, C_3, k_1, k_2, k_3 represent a labile biochar C pool, semi-labile biochar C pool, a stable biochar C pool, and the first order rate constants for the labile, semi-labile and stable pools, respectively.

Power model:

$$C_r = C_0 - \left(\frac{C_0 * e^b}{m+1}\right) * t^{m+1} \tag{4}$$

The log transformed decay rate k and log transformed incubation time (t) follows a linear relationship as Eq. (5), where m and b are the slope and intercept, respectively:

$$\ln(-k) = m * \ln(t) + b \tag{5}$$

For simplicity, Eq. (4) can also be written as

$$C_r = C_0 - c * t^d \tag{6}$$

where c and d are positive constants.

Based on Eq. (1) and (2), we parametrized two inert pool models, denoted as the SFO + I and DFO + I models, under the assumption that biochar encompasses an inert C pool, as described by the following equations.

SFO + I model:

$$C_r = C_1 * \exp(-k_1 * t) + C_i \tag{7}$$

where C_r is biochar C remaining at time t, C_1 is the fraction of labile biochar C pool and k_1 is the first order mineralization rate, respectively. C_i represents the size of the inert biochar C pool.

DFO + I model:

$$C_r = C_1 * \exp(-k_1 * t) + C_2 * \exp(-k_2 * t) + C_i \tag{8}$$

where C_r is biochar C remaining at time t, C_1, C_2, C_i k_1, k_2 represent the fraction of labile biochar C pool, semi-labile biochar C pool, the inert biochar pool, and the first order rate constants for the labile, semi-labile pools, respectively.

The fits were considered acceptable if the estimated parameters had smaller standard errors (SE) than the parameter values *per se*. The goodness of the fit of the models was evaluated by the Akaike information criterion (AIC), which serves as a metric for model selection by considering both the goodness of fit and the number of parameters. The best-fit models for each observation were identified based on the lowest AIC values. Adjusted R^2 values were also included to assess the overall fitting.

2.4. Biochar C remaining after 100 years

To evaluate biochar C remaining after 100 years (BC100) predictions, we calculated BC100 for the 31 observations that could be satisfactorily fitted with the SFO, SFO + I, DFO, DFO + I and power models (i.e., the estimated SEs < the parameters). The TFO model was excluded from predicting BC100 because a large portion of the model fits had unrealistically high SEs.

2.5. Assessing the predictive performance of the models

In order to evaluate the predictive ability of the models, we utilized the longest biochar decomposition dataset generated to date from Kuzyakov et al. (2014). The models were fitted to biochar decomposition data from progressively shortened incubation periods (i.e. 1480/1475, 734/732, 380/378, 213/211, 54/51 days) and the predicted mineralization was compared against the measured data on day 3102, according to the method proposed by Sleutel et al. (2005). Moreover, to assess the overall predictive performance of the selected models, we extended our analysis to include data (21 observations) from incubations spanning at least two years, by fitting models using biochar decomposition data from 3-month and 1-year of incubation and comparing the extrapolated results with the 2-year measured data.

2.6. Non-parametric test

The Kruskal–Wallis test with multiple non-parametric comparisons was used to detect differences in BC100 predicted by five of the different models (SFO, SFO + I, DFO, DFO + I, and power) since the condition of normality was not fulfilled.

3. Results and discussion

3.1. Inert pool models

Inert pool models generated the best fits in about 25 % of the cases, as measured by AIC, with 9 for the DFO + I model but only 2 for the SFO + I model when comparing all the models (Fig. 1). However, when comparing the performance only to the first order models without inert pools, the addition of an inert pool improved fits in most cases (84 % for the SFO vs SFO + I comparison and 63 % for the DFO vs DFO + I one as evaluated by AIC, Supplementary material 1). The SFO + I model performed less well in fitting data and had lower adjusted R^2 values compared to the DFO + I model (Fig. 1).

For biochar persistence estimations based on biochar decomposition data, the key criterion for assessing a model's performance lies in its capacity to extrapolate beyond the observed period. Even if a model fits the data well, it may still have poor predictability (Sleutel et al., 2005). Notably, the predictions of biochar C remaining (BC100) was highest for

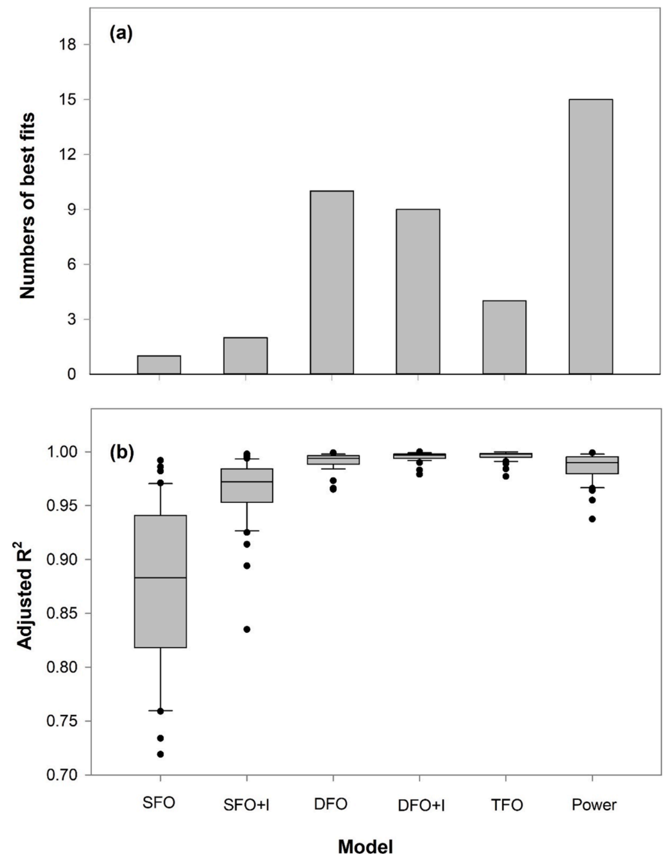

Fig. 1. (a) Numbers of best fits as evaluated by comparing AIC (Akaike information criterion) values fitted by 6 different models (SFO, single-first order; SFO + I, single-first order + an inert pool; DFO, double-first order model; DFO + I, double-first order model + an inert pool; TFO, triple-first order model; power model); (b) Adjusted R^2 values for the fits generated by abovementioned models, n = 41.

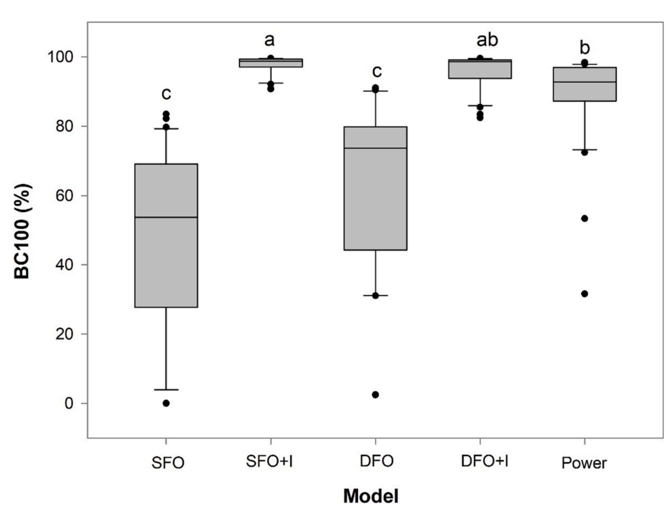

Fig. 2. Predictions of biochar C remaining after 100 years (BC100) using five different models (SFO, single-first order; SFO + I, single-first order + an inert pool; DFO, double-first order model; DFO + I, double-first order model + an inert pool). Different letters indicate significant differences in BC100 predictions (n = 31, p < 0.05).

the inert pool models among the five models assessed (Fig. 2). The inert pool models (SFO + I and DFO + I) also overestimated biochar persistence compared to measured outcomes (Figs. 3 and 4, Fig. S2). When evaluating the predictive power of inert models using a larger dataset (21 observations), they consistently overestimated biochar persistence in all cases (Fig. 4).

Moreover, the models estimated relatively large inert C pool sizes (41 % to 58 %) even when fitted to mineralization data from highly degradable raw biomass. While these estimates are much lower than the estimated inert pool sizes for the biochars (on average 96 %), they are considerably higher than what would be expected for raw biomass and the inert C pool size appears to essentially be determined by the amount of C remaining by the end of the incubation. This is problematic since most incubations are very short in comparison with the timescale that biochars persist in soil. However, dismissing the idea of using the inert pool model (DFO + I) entirely may not be necessary. Some researchers

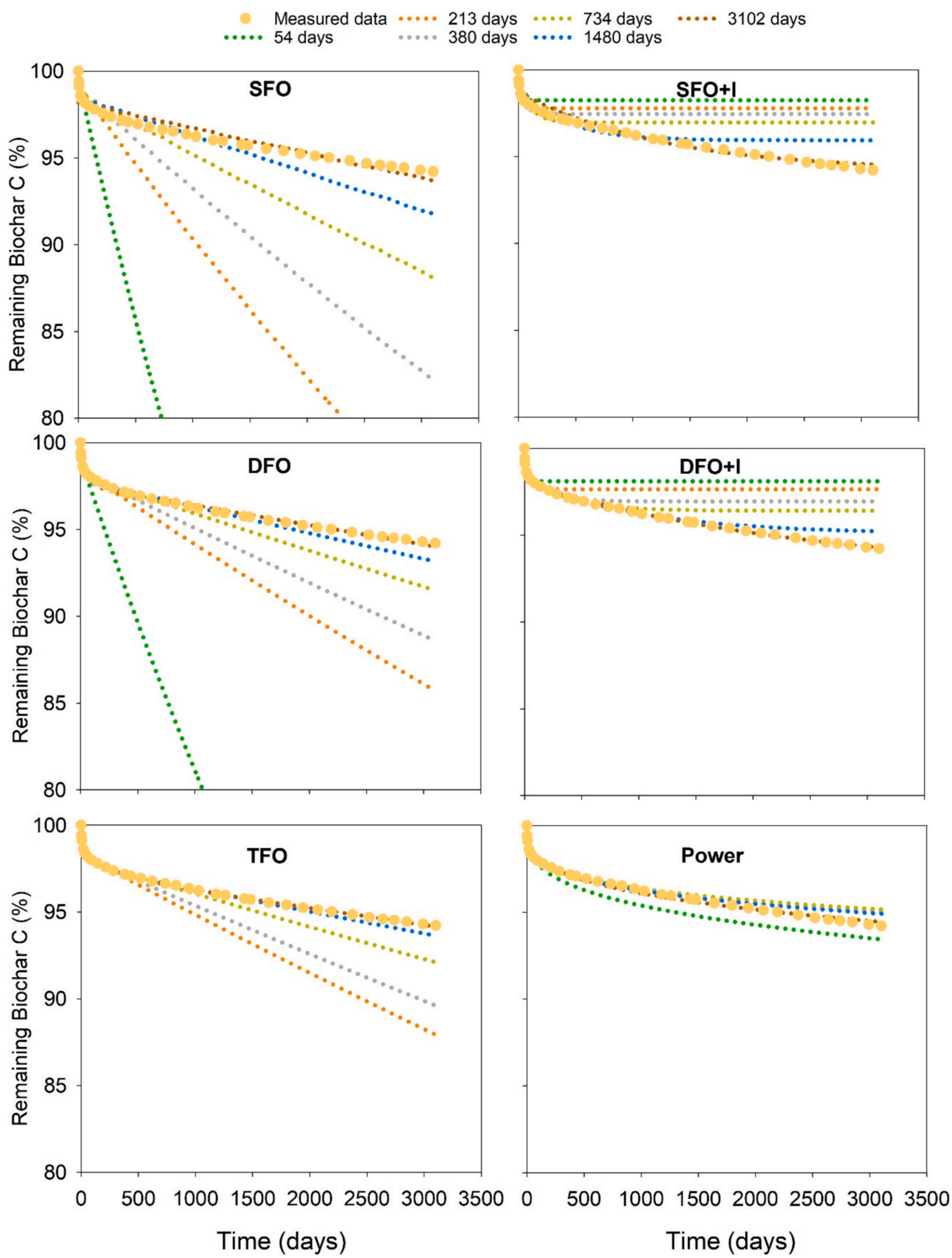

Fig. 3. Predictions from six models (SFO, single-first order; SFO + I, single-first order + an inert pool; DFO, double-first order model; DFO + I, double-first order model + an inert pool; TFO, triple-first order and power models) when fitting to gradually reduced incubation lengths from the longest available biochar incubation in a loess soil (Kuzyakov et al., 2014, observation 37) compared to the measured outcome. Note: the TFO model did not fit to incubation datasets shorter than 51 days.

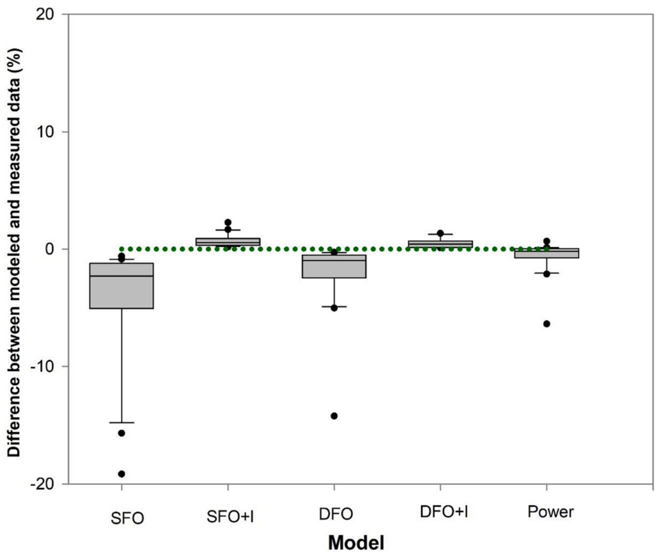

Fig. 4. Difference between the measured and the extrapolated biochar C remaining for five different models (SFO, single-first order; SFO + I, single-first order + an inert pool; DFO, double-first order model; DFO + I, double-first order model + an inert pool) when fitting to 3-month biochar incubation data and subsequent comparison with the measured data after 2 years (n = 21).

have argued that biochar becomes biologically inert during the process of biochar production, as indicated by high random reflectance values (Petersen et al., 2023; Sanei et al., et al., 2024). Formation of such inertinite fractions occur particularly in biochars produced at temperatures exceeding 600 °C. Thus, it is possible that inert pool models would be more useful for incubations of commercial biochars produced at higher temperatures with lower H/C ratios (<0.3) than studied here (Fig. S3). More research is needed in order to validate the efficacy of the inert pool models for such biochars and to refine the conditions under which they could be correctly applied, e.g. by comparing inert pool size estimates with measures of degrees of aromatic condensation or inertinite content in biochar.

3.2. First order kinetic models

The SFO model only had one out of 41 best fits for the selected observations and the adjusted R^2 values were lower than for all other models (Fig. 1). Upon visual inspection, the goodness of fits generated by the SFO model were poor in most cases. By fitting the exponential models to data from a 380-day incubation from Kuzyakov et al. (2014), the estimates of biochar C remaining after 8.5 years were underestimated to greater extent than the predictions from other models (Fig. 3 and Fig. S2). Similar results were obtained when comparing 2-year measured outcomes with the predictions using 3-month and 1-year incubation data (21 observations, Fig. 4, Fig. S4). As the fits for the SFO model are in general poor, the model is not recommended for predicting biochar persistence.

The DFO model performed better and had 10 best fits (24 %) and higher adjusted R^2 values than the SFO model (Fig. 1). In contrast to inert pool models, the DFO model is prone to underestimation of biochar persistence, even within the relatively short time-span that we could evaluate. In our fitting extrapolations from shortened datasets, the DFO model underestimated biochar C persistence, even within a timescale of less than 10 years (Figs. 3 and 4, Fig. S4). The incubation duration had a significant impact on the accuracy of biochar persistence prediction. Within a timeframe of 8.5 years, the prediction accuracy decreased markedly with shortened incubation times, particularly for the predictions from the DFO model when the incubation were shorter than 400 days (Fig. 3 and Fig. S2). This suggests that the incubation time should be at least one year when employing the DFO model, which aligns with the recommendation in Lehmann et al. (2021). Nevertheless, the discrepancy between estimating BC100 from one-year vs. from 8.5-year of incubation data would be as large as 35–46 % based on the longest incubation study by Kuzyakov et al. (2014), which is larger than for the inert pool models (only 3.4–4.1 % for the DFO + I model). When comparing the predictions using 1-year incubation data with 2-year measured data (21 observations, Fig. S4), DFO model again underestimated biochar persistence.

The DFO model has been the most commonly used model for estimation of biochar persistence from both shorter and longer (> 1 year) incubation studies and it was employed in 18 out of the 24 studies listed in Table 1. Despite the underestimation in predicting biochar

Table 1
Overview of the publications from which biochar incubation datasets were extracted for this paper (No. 1–9), other studies containing shorter (<1 year) datasets (No. 10–17), synthesis studies (No. 18–24), and the models used to fit the data in the respective studies.

No.	Publications	Feedstock	Pyrolysis temperature (°C)	Duration (days)	Model (s)
1	Santos et al. 2021	wood	300, 450	745	DFO
2	Zimmerman 2010	wood	250, 400, 525	379	Power
3	Singh et al. 2012a	wood, biosoilds, manure	400, 550	1829	DFO
4	Fang et al. 2014	wood	450, 550	730	DFO
5	Kuzyakov et al. 2014	grass	400	3102	NA
6	Herath et al. 2015	crop	350, 550	510	TFO
7	Liu et al. 2020	crop	200, 300, 500	368	Power
8	Budai et al. 2016	crop, grass	230–796	364	DFO
9	Fang et al. 2019	wood	450, 550	758	DFO
10	Keith et al. 2011	crop	450, 550	120	DFO
11	Maestrini et al. 2014	grass	450	158	DFO
12	Nguyen et al. 2014	grass	375–475	189	DFO
13	Santos et al. 2012	wood	450	180	DFO
14	Yang et al. 2022	crop	300, 450, 600	180	DFO
15	Rasse et al. 2017	grass	500–750	90	SFO
16	Farrell et al. 2013	Crop, wood	450	74	DFO + I
17	Bai et al. 2013	grass	200, 475	200	SFO, DFO, Power
18	Singh et al. 2012b	Various	150–800, including wild fire	–	SFO, DFO
19	Budai et al. 2013	Various	250–650	365, 1829	DFO
20	Wang et al. 2016	Various	200–1200	>57	DFO
21	Lehmann et al. 2021	Various	200–800	>365	DFO
22	Woolf et al. 2021	Various	350–800	>365	DFO, TFO
23	Rodrigues et al. 2023	Various	200–796	>365	DFO
24	Azzi et al. 2024	Various	200–1200	>349	SFO, DFO, TFO, Power

persistence, the DFO model has remained widely used in synthesis studies (Singh et al. 2012b; Budai et al., 2013; Wang et al., 2016; Lehmann et al., 2021; Woolf et al., 2021; Rodrigues et al., 2023). Taking a more cautious stance, biochar persistence as predicted by the DFO model offers a conservative estimate of the C sequestration potential (Budai et al., 2013). However, there is a risk that these underestimations may mislead policymakers and stakeholders, so care should be taken when interpreting the biochar persistence estimates predicted by the DFO model.

The TFO model provided more accurate predictions compared with the DFO model but only had four of the best fits (10 %). Further, only 41 % of the selected datasets could be fitted with the TFO model without large SEs (Fig. S1). The SEs of the estimates by the TFO model tended to be larger than the parameter estimates. Such large SEs may indicate over-parameterization making the results unreliable and the model may not suitable for biochar persistence estimation in most cases.

3.3. Power model

The power model, which has been employed in a limited number of biochar persistence studies (Zimmerman, 2010; Bai et al., 2013; Liu et al., 2020, Azzi et al., 2024), fit all the selected data without large SEs, with an average adjusted R^2 of 0.99 and with 15 best fits (37 %), the highest number of all models. Moreover, when extrapolating from the progressively shorter biochar incubation datasets, the power model gave the smallest differences between predictions and measured outcomes, and the predictions were not as sensitive to shortening the dataset as the other models (Fig. 3 and Fig. S2). These results are in line with experimental findings indicating a diverse range of chemical structures and arrangements within biochar (Brewer et al., 2009; Keiluweit et al., 2010). However, the extrapolations made here were based on a "high-quality" subset of observations, i.e. without sudden increases or fluctuations in decay rates. When evaluating the power model on some of the datasets that we excluded, there were several cases where the power model underestimated biochar persistence even to a larger extent than the SFO model. For instance, the power model predicted that a wood-based biochar produced at 550 °C, using the data from Fang et al. (2014) (observation 27), would be fully mineralized already after 39 years, while the estimates from the SFO model suggested that more than 30 % biochar C would remain after 100 years (Fig. S5). Although complete decay within only 39 years is not biologically impossible, it is not in line with current literature estimates. In general, caution is warranted in interpreting curve fitting if the underlying data is not deemed reliable or suitable (see Table S1 for the criteria we used).

3.4. Challenges with extrapolating from laboratory incubations to field conditions

The biochar decomposition data in this study were obtained under well-controlled laboratory conditions. Under real field conditions, biochar is subjected to drying-wetting and freeze–thaw cycles, dramatic temperature fluctuations and other environmental variations. Moreover, fresh organic matter is continuously added, maintaining the activity of microorganisms at a higher level. All these factors can potentially speed up degradation of biochar compared to lab incubations, which likely explains, at least in part, the significant decrease in biochar stocks after 11 years in a recent field study (Gross et al., 2024). The unpredictable conditions in the field might lead to erratic patterns in biochar decomposition rates, posing challenges for data fitting using the models studied here. Various sophisticated SOC models have been developed to estimate SOC changes over time under field conditions, with decomposition rate modifiers to account for the effects of key factors such as soil temperature, soil moisture and clay content on SOC dynamics (Le Noë et al., 2023). Recently, Pulcher et al. (2021) attempted to incorporate biochar in the RothC model to assess and predict how biochar influences soil C dynamics under Italian climate conditions. However, further studies are still needed to parameterize existing SOC models for biochar in order to predict biochar C dynamics under long-term field conditions.

4. Conclusion

The incorporation of an inert pool often improve model fits to biochar mineralization data compared to first order models. However, the inert pool models tended to overestimate biochar persistence and gave unexpectedly high estimates of the inert pool size even when fitted to mineralization data from biodegradable materials. The most commonly used models for biochar stability predictions, the first order exponential models, underestimated biochar C persistence compared with the measured outcomes. This suggests that most current predictions of biochar long-term persistence from incubation studies, especially the ones stemming from short (< 1 year) incubations, are likely underestimated. The power model yielded the highest number of best fits and gave the most reliable predictions. It appears to be a better option for estimating biochar persistence compared to the other models. However, careful data quality assessment is important since the power model is sensitive to fluctuations in mineralization rate.

CRediT authorship contribution statement

Haichao Li: Writing – review & editing, Writing – original draft, Visualization, Methodology, Formal analysis, Conceptualization. **Elias S. Azzi:** Writing – review & editing, Funding acquisition, Data curation, Methodology. **Cecilia Sundberg:** Writing – review & editing, Funding acquisition, Methodology. **Erik Karltun:** Writing – review & editing, Funding acquisition, Methodology. **Harald Cederlund:** Writing – review & editing, Supervision, Methodology, Funding acquisition, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.geoderma.2024.117093.

Data availability

All data will be published.

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