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Fire-derived organic carbon turnover in soils on a centennial scale

N. Singh¹, S. Abiven¹, M. S. Torn², and M. W. I. Schmidt¹

¹University of Zurich, Department of Geography, Winterthurerstrasse 190, Zurich 8057, Switzerland

²Lawrence Berkeley National Laboratory, Berkeley, CA 94720, USA

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Correspondence to: N. Singh (nimisha.nimisha@geo.uzh.ch)

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Abstract

Pyrogenic Carbon (PyC), the residue of an incomplete combustion of plant biomass, is considered as a carbon (C) sink due to its assumed stability in soil. Our meta-analysis of studies on PyC degradation challenges the assumption that PyC persist in soil for several thousand years. The turnover time for PyC estimated here ranges from decadal to centennial time scales, and is not slower than decomposition of bulk Soil Organic Matter (SOM) and differs with initial biomass, pyrolysis temperature and climate. Thus, using PyC as a strategy for offsetting carbon emissions requires caution and further research.

1 Introduction

Wildfires transfer approximately 1.7 to 4.1 Pg C yr^{-1} ($1 \text{ Pg} = 10^{15} \text{ g}$) to the atmosphere as carbon dioxide (CO_2) (Lavorel et al., 2007) and 0.05 to 0.2 Pg C yr^{-1} to the soil (Kuhlbusch, 1998; Seiler and Crutzen, 1980), as incomplete combustion residue of plant biomass known as pyrogenic carbon (PyC) (Goldberg, 1985). Moreover, climate change is projected to increase wildfire frequency in many parts of the world (Flannigan, et al., 2006), which could modify the input of PyC and consequently the terrestrial carbon cycle considerably (Westerling et al., 2006). PyC is ubiquitous in the environment and ranges from 2% to 45% of the total soil organic carbon (SOC) in various terrestrial systems (Bird et al., 1999; Skjemstad et al., 2002; Schmidt et al., 1999). PyC contributes to stable C pools in the soil (Preston and Schmidt, 2006; Marschner, 2008) and is assumed to transfer fast cycling biomass-C to much slower-cycling C in soils (Ohlson et al., 2009) and is therefore expected to act as a carbon sink (Marris, 2006; Seifritz, 1993). In the last decade, PyC has gained significant interest as a strategy for sequestering atmospheric CO_2 to partly offset carbon emissions (Lehmann et al., 2006).

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The ability of PyC to act as a carbon sink depends on its persistence in the soil. PyC is widely considered to be relatively “inert” (Forbes et al., 2006) because PyC has been preserved in geological records (Forbes et al., 2006), archaeological sites (Schmid et al., 2002) and old anthropogenic soil (Glaser et al., 2000; Knicker, 2011), giving an impression that it persist for a long time in terrestrial systems. Moreover, experimental evidence suggests that PyC is resistant to chemical oxidants (Skjemstad et al., 1996) and contributes to the oldest soil organic carbon (SOC) pool (Krull et al., 2006). Based on the ^{14}C age of PyC macro-pieces/charcoal (Pessenda et al., 2001; Schmidt et al., 2002) and budget calculations (Forbes et al., 2006), PyC age in soil has been estimated to be on the scale of hundreds to ten thousand years (Liang et al., 2008). The limitation of using a radiocarbon age to estimate turnover time is that we rarely have knowledge of the input rate (or, for isolated systems, initial stock when the radiocarbon “clock” started) and hence is not possible to estimate turnover times.

However, recent experimental studies observe transformation and mineralization of PyC over weeks to year time (Kuzyakov et al., 2009; Hilscher et al., 2009; Hilscher and Knicker, 2011; Hamer et al., 2004; Bruun et al., 2008) and significant losses of PyC from the soil profile in the long-term field studies (Bird et al., 1999; Hammes et al., 2008a; Nguyen et al., 2008). PyC is presumed to degrade physically (Carcaillet, 2001; Carcaillet and Talon, 1996; Lehmann et al., 2003; Hammes and Schmidt, 2009) and chemically by abiotic (Cheng et al., 2006; Hockaday et al., 2006; Lehmann et al., 2005) and/or microbial agents (Goldberg, 1985; Potter, 1908; Shneour, 1966). Incubation identified abiotic (Cheng et al., 2006) and/or biotic oxidation processes (Zimmerman, 2010; Potter, 1908; Hamer et al., 2004; Kuzyakov et al., 2009), as an important mechanism leading to PyC degradation. Turnover time of PyC as reported in these experimental studies ranged between a hundred and a thousand years.

These recent observations contradict the perception that PyC persists in soil for many millenia. The uncertainty in PyC persistence is accompanied by a basic lack of understanding about PyC dynamics in soil. What is the turnover time of PyC in different soils? To reconcile the apparent discrepancies between assumed persistence of

PyC based on radiocarbon age and degradation of PyC as observed in experimental studies, we assembled data from published studies on PyC losses from soil and for the first time, calculated turnover times within and across all studies with one consistent approach.

2 What are PyC turnover times in soil?

We compiled data from published studies ($n = 15$, comprising 47 data sets, Supplement Table 1) on PyC degradation. In our approach, PyC is modelled as a single homogeneous C pool and assumed to follow first order kinetics. We further assumed that there were no new PyC inputs between time = 0 and time = t (yr). This yields an equation for the decay rate k based on the loss of PyC over time (i.e., as a function of the mass remaining (C_t/C_0) and time) as follows:

$$C_t = C_0 \cdot \exp(-k \cdot t) \quad (1)$$

where C_t is the remaining stock after time t (yr), C_0 is the initial stock of PyC ($t = 0$), k is the decay rate (yr^{-1}) and turnover time τ (yr) = $1/k$.

We adopted the simplifications of one homogeneous pool and first order decay kinetics because that allowed us to compute and compare turnover time of PyC despite the differences in type of PyC, experimental set up, or analytical method employed in various studies. Moreover, first order kinetics, meaning the rate of mass loss is a constant proportion of mass, is a simple and robust formulation that is commonly used to describe the turnover of SOM (Six and Jastrow, 2002). We justify the assumption of no new PyC inputs because the incubation studies were set in that way and the field experiments were deliberately chosen where inputs had been low (although quantified poorly or not at all) after the initial sample collection.

Therefore, the data set includes those studies where (1) the initial inputs and stock were known or could be estimated; (2) the initial stock decreased or remained constant with time; and (3) the experimental set up included terrestrial systems.

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We calculated turnover time for each study using Eq. (1), finding they range from < 10–600 yr (Supplement Fig. 1). Critically, the turnover times calculated are much shorter than earlier estimates that used the radiocarbon age of PyC in soil as a proxy for turnover time. The PyC degradation studies were done for few months to a year and thereby potentially biased towards shorter turnover times (Derrien and Amelung, 2011). Also, we see a high scatter in the turnover time between different studies. For instance, Brodowski (Brodowski, 2005) observed 16–22 % PyC degradation in 104 weeks of incubation study (yielding a turnover time of 8 yr), while, Shindo (Shindo, 1991) observed no decomposition of grassland plant PyC in volcanic ash soil for 40 weeks of incubation indicating that the duration of the experiment is not the only factor controlling the turnover time.

For the meta-analysis, we pooled all the data and expressed them as percentage of the initial PyC and fit the first order kinetic model, Eq. (1), to compute a single decay rate. The overall turnover time of PyC computed was 291 yr (r^2 value of regression is 0.41, root mean square error = 9.735). Although having much faster decay than previously thought, PyC remains more stable than all known plant-derived organic compound classes in soil based on ^{13}C natural abundance labelling experiments (Amelung et al., 2008) and is comparable to the turnover time of bulk SOM.

Turnover of SOM is sensitive to methodological differences due to edaphic factors (Trumbore, 2000) and correspondingly could also influence PyC turnover. Additionally, initial biomass (Franklin, 1951), pyrolysis temperature (Schneider et al., 2010) and environmental conditions (Cheng et al., 2008) may affect PyC turnover in soil and could bias the turnover times calculations. Therefore, we grouped the data to see whether these factors had an influence on turnover time when all other factors were allowed to vary, namely (1) incubation vs. field studies; (2) initial biomass (grass vs. wood); (3) pyrolysis temperature (< 400 °C and \geq 400 °C) and (4) quartz sand vs. soil medium (Fig. 1). Data were not grouped by other factors that control SOM decomposition, like climate, degree of soil development, soil types, topography, and biota because they were either not available or kept constant in most studies. We computed individual

turnover time for each study within above-mentioned grouped data set using Eq. (1) to get the range of turnover times, which was averaged to get turnover time. The turnover times of grouped factor were compared using a non-parametric Wilcoxon rank sum test. Interactions between factors on the compiled data were evaluated by multi way ANOVA using R software (Supplement Table 2). Our analysis shows that we do not have any significant interactions between the factors. Therefore, the imbalanced design of the grouped data does not introduce any significant error in the interpretation.

2.1 Incubation vs. field study

Incubation studies have significantly ($p < 0.001$) shorter turnover times (average 60 yr; range 2–180 yr) than field-based studies (average 272 yr; range 90–600 yr). It could be that short-term decay studies capture mainly the rapid initial loss of more labile or/and non charred components of pyrolysed material before it is physically stabilized by association with mineral or aggregates, and therefore are not a good indication of the long-term degradation rates. These possible explanations for the short turnover time in incubations also pertain to short-term field experiments (Major et al., 2010). Incubation studies were conducted with fine-sized PyC (Hamer et al., 2004; Major et al., 2010; Nocentini et al., 2010b) at optimum temperature and field capacity moisture content (Baldock and Smernik, 2002; Hilscher et al., 2009; Zimmerman, 2010), which partly explains the accelerated degradation rates and the faster turnover time (Nocentini et al., 2010a).

Long-term field studies provide more realistic estimation of turnover time of PyC, which not only includes the rapid initial phase but also the phase when mineralization rate decreases with time (Kuzyakov et al., 2009). We took advantage of pre existing long-term field experiments, which were designed with scientific objectives other than PyC loss and still tried to estimate the turnover time for PyC in situ. It was, however, not possible to conduct a straightforward mass balance for the multi-year field studies because there was uncertainty regarding PyC initial input/stock and atmospheric deposition of PyC from other sources. Our analysis shows that long-term field based

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studies indicate a turnover time on a centennial scale which is still shorter than previously assumed. The shorter turnover time may be attributed to mechanisms like frequent freeze-thaw cycles (Carcaillet, 2001), root growth (Carcaillet and Talon, 1996; Lehmann, et al., 2003), fungal hyphae (Hammes and Schmidt, 2009), soil fauna (Ponge et al., 2006) and erosion that exposes PyC to biological and chemical degradation. However, at present our knowledge of the underlying mechanisms of PyC degradation on a longer time scale remains scarce.

2.2 Initial biomass

We identified two types of initial biomass reported in the studies here that were representative for grassland and forest ecosystems, namely grass and wood (Fig. 1). Grass PyC turned over (average 45.8 yr, range 2–170 yr) significantly ($p < 0.05$) faster than wood PyC (average turnover time = 79.3 yr, ranges from 2 to 181 yr) in the incubation studies. This is consistent with a previous study that observed higher oxidation of grass PyC as compared to wood reflecting differences in their chemical structure (Nguyen and Lehmann, 2009). Fourier Transformed Infra Red (FTIR) spectra of grass and wood PyC produced at the same temperature (Keiluweit et al., 2010) show differences in the type of PyC produced. Knicker et al. (Knicker et al., 2008) proposed that a significant amount of grass derived PyC consists of N-heteroaromatic carbon with the average cluster size of the aromatic units smaller than six rings. If this holds true, grass PyC would probably be easier to degrade. On the other hand, lignocellulose rich substrate like wood is transformed by charring into a more aromatic structure (Czimczik et al., 2002). However, the influence of initial biomass on chemical structure diminishes with increasing pyrolysis temperature of PyC (Schneider et al., 2011). Therefore, the persistence of PyC largely depends on the chemical and physical structure of PyC.

2.3 Pyrolysis temperature

We compared degradation rates of PyC based on the pyrolysis temperature. We chose 400 °C as temperature threshold based on PyC thermosequence studies (Keiluweit

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et al., 2010; Schneider et al., 2010), which showed maximum modification in PyC structure around 400 °C. Pyrolysis temperature data were only available for incubation-based studies. Turnover time was significantly shorter ($p < 0.05$) for < 400 °C (average turnover time = 25 yr, range 2–82 yr) than PyC formed at ≥ 400 °C (average turnover time = 98.9 yr, range 10–520 yr). Lower temperature PyC retains uncharred initial biomass (Zimmerman, 2010), which are comparatively more labile than charred biomass and thus results in a faster turnover time. Moreover, PyC formed at lower temperature had a greater internal microporosity (Hammes et al., 2008b), allowing easier access to oxidizing agents like water or microbes that facilitates degradation. The degree of condensation increases with higher pyrolysis temperature (Schneider et al., 2010; McBeath and Smernik, 2009; Nishimiya et al., 1998; Baldock and Smernik, 2002) and thus could explain slow turnover for high temperature PyC.

2.4 Quartz sand vs. soil medium

Most incubation studies used either quartz sand with microbial inoculum or fresh soil, while all field studies took place in soil. To compare turnover times by medium, we used only incubation studies because the large differences between field and incubation, including duration of study, would confound the analysis. In soil, PyC has a shorter turnover time (average = 24 yr, range 4–109 yr) than in quartz sand (average = 65 yr, range 2–181 yr). The use of quartz sand probably increases the permeability and the oxygen accessibility for PyC (Zimmerman, 2010) whereas soil as a medium would yield more realistic values because it holds a larger range of microbial populations than a microbial inoculum. The faster turnover time in soil may reflect the role of microbial community in PyC degradation. However, the influence of the microbial community dynamics for PyC degradation is poorly understood (Pietikainen et al., 2000; Czimczik and Masiello, 2007) and needs further exploration.

PyC is known to interact with soil minerals (Glaser et al., 2000; Piccolo et al., 1997; Brodowski et al., 2005; Liang et al., 2008), in some cases resulting in aggregation (Brodowski et al., 2006; Vasilyeva et al., 2011) and stabilization in the soil system.

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However, short-term incubation studies might not capture the stabilizing effects of organo-mineral interaction. Therefore, we did not attempt to examine the stability mechanisms like aggregation or organo-mineral interaction that might influence the turnover times for PyC.

2.5 Climate

The effect of climate, including temperature and moisture, which is one of the factors controlling SOM and PyC decomposition, has not been covered in our discussion because of insufficient data. A few studies observed a positive correlation between mean annual temperature of the field site and PyC degradation (Cheng et al., 2008; Glaser and Amelung, 2003) but not with mean annual precipitation (Cheng et al., 2008). Further, Nguyen et al. (Nguyen et al., 2010) also showed that degradation rates of PyC is accelerated with increasing temperature. Therefore, faster turnover of < 100 yr observed in tropical (Nguyen et al., 2008) and sub-tropical climate (Bird et al., 1999) could be attributed to the more favourable climate. On the contrary, slower degradation rates could be expected in boreal forest (Preston and Schmidt, 2006). However, in a boreal forest (Ohlson et al., 2009) PyC content decreased to the concentration of the surrounding organic soil matrix in nearly 100 yr only. Therefore, we need to directly examine a range of climatic conditions to understand the influence of environment on PyC turnover time.

3 Conclusion and future research

Based on this analysis of the published data, the turnover time of PyC is much shorter than widely assumed. PyC is degrading in the soil, whatever the PyC properties and the edaphic conditions are. PyC comprises an array of compounds and is present in different environmental matrices, thus, a single true rate of decomposition cannot exist. We used a simple but robust model that may be subject to quantitative uncertainty, but

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the general trend shows that PyC degrade on a centennial scale. This implies that PyC may not be a long-term storage of carbon in soil.

The questions that remain unanswered are: what are the factors that make PyC susceptible to degradation? Does rapid turnover of PyC result from microbial activity and changes thereof? Details of the experimental and modelling approaches aside, the use of PyC as a strategy for C sequestration on a regional scale requires further research and we recommend to

- start long term PyC degradation field experiments in different climate and soil types
- identify underlying mechanisms of PyC degradation and factors controlling stability, and
- address decomposition dynamics of ageing PyC.

4 Author contributions

N.S. assembled the data, N.S. and S.A. analyzed the data, and all authors contributed to the research design and the text.

5 Additional information

The authors declare no competing financial interests. Supplementary information accompanies this paper. Correspondence and requests for materials should be addressed to NS.

Supplementary material related to this article is available online at:
[http://www.biogeosciences-discuss.net/8/12179/2011/
bgd-8-12179-2011-supplement.pdf](http://www.biogeosciences-discuss.net/8/12179/2011/bgd-8-12179-2011-supplement.pdf).

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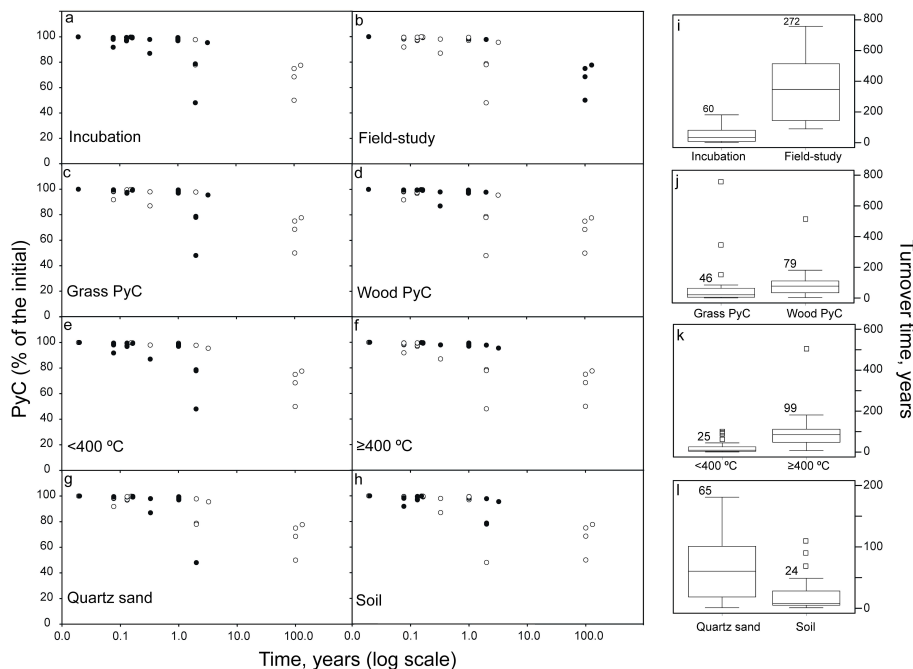


Fig. 1. Pyrogenic stock expressed as percentage of the initial from the data reported in literatures. Circles represent all data points and solid circles represent the grouped data set namely, **(a)** incubation study, **(b)** field study, **(c)** grass PyC, **(d)** wood PyC, **(e)** pyrolysis temperature < 400 °C, **(f)** pyrolysis temperature ≥ 400 °C, **(g)** sand medium and **(h)** soil medium. The time is expressed in years (log scale). The box-plot (right) of turnover time of each variable **(i)–(l)**, where square is outliers beyond 10th or 90th percentiles and numbers denote average turnover time. The calculated turnover time varies from decades to century.

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