LETTERS

Australian climate–carbon cycle feedback reduced by soil black carbon

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Annual emissions of carbon dioxide from soil organic carbon are an order of magnitude greater than all anthropogenic carbon dioxide emissions taken together[1]. Global warming is likely to increase the decomposition of soil organic carbon, and thus the release of carbon dioxide from soils[2–5], creating a positive feedback[6–9]. Current models of global climate change that recognize this soil carbon feedback are inaccurate if a larger fraction of soil organic carbon than postulated has a very slow decomposition rate. Here we show that by including realistic stocks of black carbon in prediction models, carbon dioxide emissions are reduced by 18.3 and 24.4% in two Australian savannah regions in response to a warming of 3 °C over 100 years[1]. This reduction in temperature sensitivity, and thus the magnitude of the positive feedback, results from the long mean residence time of black carbon, which we estimate to be approximately 1,300 and 2,600 years, respectively. The inclusion of black carbon in climate models is likely to require spatially explicit information about its distribution, given that the black carbon content of soils ranged from 0 to 82% of soil organic carbon in a continental-scale analysis of Australia. We conclude that accurate information about the distribution of black carbon in soils is important for projections of future climate change.

For predictions of climate change to be accurate, they must include key processes controlling responses of C pools to global warming. Recent improvements in general circulation models include the coupling of terrestrial biogeochemical processes with atmospheric and oceanic processes[1]. Soil C cycling has an important role in such coupling because soil organic C (SOC) stocks are almost four times greater than C in the atmosphere, and annual emissions of CO_2 from soil are one order of magnitude greater than all anthropogenic CO_2 emissions together[1]. Therefore, small uncertainties in soil processes may have large effects on climate-change predictions for those general circulation models that incorporate terrestrial biogeochemical cycles[9].

Mineralization of organic C by microorganisms is generally found to increase with warming[5]. Thus, emission of CO_2 from soils will probably increase with increasing temperature[2–5], creating a positive climate-change feedback[6]. SOC consists of a complex mixture of compounds interacting with soil minerals and aggregates leading to the recognition of multiple pools of SOC with different turnover times[8]. Such variation causes biogeochemical simulation models that include multiple soil C pools[8] to predict a slower response to temperature change than do models with a single pool[7]. However, for models that will fully incorporate soil C stocks[9], failure to identify significant but highly stable fractions could render predictions of global climate change, even by simulation models with multiple soil C pools, inaccurate.

Black C, the carbonaceous residue of incomplete combustion of biomass and fossil fuels[10], has recently been detected in several soils worldwide, and is generally considered to be a highly stable form of SOC[10–12]. Charring typically increases the mean residence time (MRT) of biomass by one order of magnitude in short-term decomposition experiments[13]. The ^{14}C ages of black C were found to lie between 1,160 and 5,040 years BP (ref. 14) and are greater than the most stable non-black C pools[15]. However, reliable information about the MRT of black C in soils is hampered by a lack of longitudinal mass balance data[16] and by the heterogeneity of black C (ref. 10), which poses a challenge to its quantification (see the Methods section).

We hypothesize that neglecting or underestimating the size and stability of black C significantly inflates the mineralization predicted for total SOC under warming and thus future CO_2 emission from soils. In turn, recognizing black C as a soil C pool with a long MRT decreases estimates of the temperature sensitivity of SOC, with important implications for predictions of global warming. Using longitudinal field data of black C cycling and stocks, we carried out a modelling exercise to estimate MRT using contrasting assumptions about formation and loss. Assuming uniform sensitivity of these MRTs to warming, we assess the likely significance of the existence of black C to the climate-change feedback from total soil C using a multi-pool SOC model, RothC[2]. The soil samples analysed in the study were the representative soil profiles of the Australian National Soil Archive (NSA), and two landscape transects (Queensland QLD; and Darwin DWN) of approximately 3,000 km, totalling about 1,900 black C data from 452 soil profiles.

An analysis of the black C pool size suggested, at a continental scale, a wide variation in black C proportions, ranging from

Table 1 Proportion (%) of black C or the IOM pool as a fraction of total organic C in topsoils of Australia. A horizons (to depths of 0.1–0.9 m) for sites from the Australian National Soil Archive (NSA), 0–0.3 m depth for sites from the Queensland (QLD) and Darwin (DWN) transects.
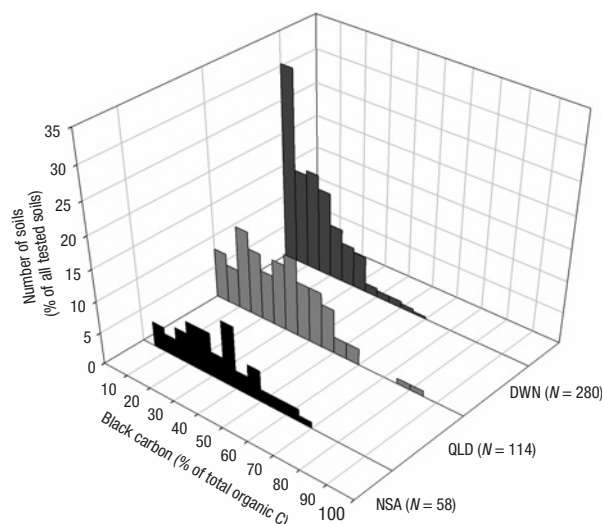
	NSA	QLD	DWN
Mean	32.9	31.5	13.6
Median	32.7	27.4	11.9
Coefficient of variation of mean (%)	47.8	52.2	87.8
Minimum	7.0	0	0
Maximum	67.0	81.7	58.6
Number of profiles	58	114	280
Predicted IOM*	5.9	4.8	7.4

*Calculated according to Falloon et al.[17], organic C in IOM is computed using the following equation: $IOM = 0.049 \times SOC^{1.139}$

Figure 1 Proportion (% of SOC) of black C in selected Australian soils. Black C in A horizons (0–0.1 to 0.9 m deep) of 58 profiles from the Australian NSA, and to a standardized depth of 0.3 m in 114 profiles from a transect in Queensland (QLD) and in 280 profiles from a transect south of Darwin (DWN) in the Northern Territory of Australia.

an absence of measurable black C contents to 82% of SOC (Table 1; Fig. 1). Representative soil profiles for Australia obtained from the NSA had an average black C content of 33% of SOC (Table 1), with half of the values lying between 20 and 40% (Fig. 1). This distribution was very similar for the QLD transect, presumably because it was entirely located inland (see Supplementary Information, Fig. S5). The DWN transect encompassed soils that lie close to the coast, including sandy soils with relatively steep relief and high rainfall intensities, and contained distinctly lower proportions of black C (Fig. 1). The average proportion for all 452 profiles was 20.4%. In comparison, the proportion of the most stable soil fraction that is used in RothC for the prediction of SOC turnover[2] (C in the so-called inert organic matter (IOM), which is assumed by RothC to not mineralize over the modelled period of time) and which should include black C, was calculated to be 6.6%. In the absence of radiocarbon data, this IOM pool is computed as a function of total SOC[17]. For our data set, SOC is poorly correlated to black C, as demonstrated by the large coefficient of variation (Table 1), and we propose that such estimation of stable organic C on the basis of total SOC may therefore not be applicable in situations where black C is a significant component. On the basis of our large Australian data set, the portion of SOC that is stable with respect to scenarios of climate change may be incorrect at the scale of relevance, if black C was ignored.

Measured black C and SOC stocks in two Australian savannah regions of the DWN transect (similar in climate, differing in soil clay content) could be predicted by RothC and an externally computed model on black C formation and disappearance only if all of the following three processes were recognized (Fig. 2): (1) regular burning of the above-ground biomass; (2) formation of black C; and (3) decomposition of black C. Disregarding burning (with oxidation of above-ground biomass to CO_2), total SOC was overestimated by 18% (Fig. 2). Accounting for 80% biomass burning every two years, but disregarding consequent formation of black C, SOC was underestimated by 17%. Modelling the formation of black C (2.5% of the C present in burnt biomass) without decomposition or other disappearance resulted in a steep increase in SOC without reaching an equilibrium (Fig. 2). Only by assuming burning, formation of black C and applying a MRT of 1,300 years could SOC and black C at modelled equilibrium be matched to the experimental observations (Fig. 2). The resulting black C stocks at equilibrium were significantly greater than a calculated IOM content of 7% using the conventional equation based on SOC[17].

Testing a range of scenarios with 60–90% biomass burned, and 1–4.5% conversion of burnt biomass to black C, resulted in calculated MRTs from 718 to 9,259 years (see Supplementary Information, Table S1). In addition to microbial decomposition,
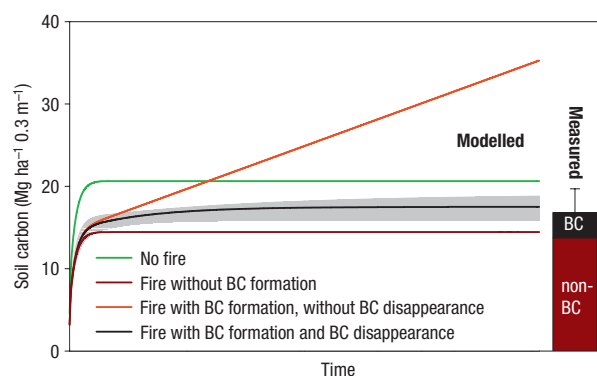

Figure 2 Convergence of model equilibrium conditions (lines) to measured soil stocks (bars). Average of 13 light-textured Inceptisols (Katherine region, DWN). Total measured SOC (0–0.3 m) is the sum of non-black carbon (non-BC) and black carbon (mean and standard error). Errors for model equilibrium conditions (grey-shaded area) were calculated using a range from 90% burnt every 1.5 years to 70% every 3 years (for clarity shown only for fire with black carbon formation and black carbon disappearance).

the calculated MRT is potentially a result of several other processes of black C disappearance such as abiotic oxidation, reburning by subsequent fires, leaching or erosion[11,12,18]. Underestimating the quantitative importance of these chemical and physical losses in the modelling studies means that mineralization rates are likely to be even lower than shown here. For example, assuming that 30% of the produced black C is eroded or reburnt during the next fire-increases the estimated MRT based on microbial mineralization by 41%, from 1,300 to 1,837 years calculated for the light-textured Inceptisols. The calculated MRTs are therefore conservative estimates for microbial decay that will most likely be slower than suggested by the MRT shown here.
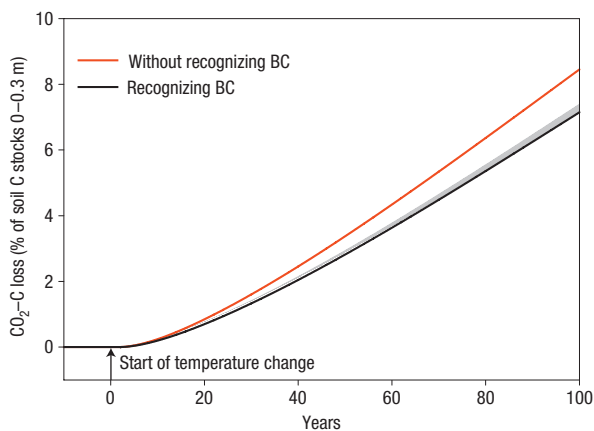
Figure 3 **Effect of ignoring black carbon as IOM on SOC losses by mineralization in response to warming by 3 °C over 100 years.** Response to different warming shown in Supplementary Information, Fig. S3. Average of 13 light-textured Inceptisols (Katherine region, DWN). The shaded area shows responses with mean residence times of black C ranging from 718 to 4,292 years around the most likely scenario of 1,300 years (corresponding to measured 80% biomass burnt and 2.5% black C formation every second year; Supplementary Information, Table S1).

Owing to measured proportions of black C that were greater than predicted IOM, we tested predictions of soil warming using a version of the RothC model (Version 26.3, ref. 19) conservatively modified to account for black C as IOM (ref. 19). In contrast to RothC, we recognize that this IOM mineralizes (albeit slowly), and applied the MRT determined above. Without black C as IOM, C losses on warming from the two test regions increase by 7–8.5% of initial SOC over 100 years (Fig. 3; Supplementary Information, Fig. S2). If the existence of black C is not considered, the three-pool model predicts an 18% greater increase in CO_2 production compared with a scenario that recognizes black C (Fig. 3; 24% for the example in Supplementary Information, Fig. S2). The demonstrated overestimation of the temperature sensitivity of SOC is a significant error and increases by 21% with each degree Celsius of predicted temperature increase over 100 years (see Supplementary Information, Fig. S3) and is nearly proportional to the fraction of black C (see Supplementary Information, Fig. S4).

For Australia, a proportion of 20% black C would lead to a 135 Tg overestimation of CO_2–C losses from soil on a continental scale, using net SOC decreases from topsoils (0–0.1 m) calculated for a 4.5 °C increase over 100 years[20]. Overestimation may even be greater for the total soil profile, because topsoil SOC values to 0.2 m are typically only 41% of total C stores to 1 m (ref. 21). On an annual basis, an inflated prediction from topsoils alone equates to 13% of CO_2 emissions from land use, land use change and forestry, or 84% of CO_2 emissions associated with aviation for Australia using values obtained for 2006 (ref. 22).

An extrapolation of these results to the global scale is currently not possible, because very few data are available that report black C contents in soils outside Australia[23,24]. However, vegetation fires as the source of black C are a global-scale phenomenon[25] and fire frequency and size may regionally even increase in the future[1,26]. Therefore, investigation of black C in soils on a global scale is required to capture what are probably significant regional differences depending on black C production and disappearance as evident in the studied Australian soils.

A limitation to rigorous quantification of black C in soils has been the dependence of its determination on the analytical method used[27]. This dependence is due in part to the broad range of compounds that may be categorized as black C in soils[10]. The base method that we used here (ultraviolet oxidation followed by NMR, see the Methods section) encompasses the widest range of such compounds[27]. However, recovery may still be less than 100% (see the Methods section). As we made conservative assumptions, resolving such analytical issues or issues of modelling MRT of black C for the studied sites (as pointed out above) would increase rather than decrease the effects demonstrated herein.

Several potential feedbacks in response to global warming were not considered, such as changes in fire frequencies, soil water contents and biomass production, which could each influence black C production and disappearance. Regardless, the significant effects of including black C stocks on the projection of the temperature sensitivity of CO_2 release from soil highlight the need for improved assessment of global and regional black C stocks and fluxes.

METHODS

Three archived collections of soil profiles were used in this study: (1) 58 representative soil profiles of the CSIRO Australian National Soil Archive (NSA); (2) 114 profiles from a 900 km transect in Queensland (QLD), Australia; and (3) 280 profiles from an approximately 2,100 km transect near Darwin in the Northern Territory (DWN), Australia (see Supplementary Information, Methods).

This provided a total of 1,877 soil samples analysed for black C and SOC. SOC was determined by dry combustion. The base method for quantification of black C was ultraviolet oxidation followed by NMR spectroscopy[28]. The NSA sample set was fully analysed by the ultraviolet–NMR method. Black C in the transect (QLD and DWN) samples was quantified by a combination of the ultraviolet–NMR method and a new mid-infrared method[29] that enables rapid analysis of a much larger number of samples (see Supplementary Information, Methods for details).

An established multi-pool SOC model was used to assess the response of CO_2 emissions to soil warming, in combination with a separately modelled black C pool. The dynamics of non-black C were modelled using RothC (Version 26.3; modified by reducing the decomposition rate of the resistant plant material pool from 0.3 to 0.15 years^{-1} according to Skjemstad et al.[19]). Black C mineralization was calculated with a first-order decay to CO_2. Simulations were carried out for two subsets of the DWN transect selected for their similar soil type and climate, but contrasting soil textures: a set of 13 light-textured Inceptisols (12.5% clay, Katherine) and of 15 heavier-textured Inceptisols (21.0% clay, Daly Waters).

First, equilibrium conditions were established for black C and non-black C pools for an average of each soil set. The balance between black and non-black C input was then calculated for different combinations of burning rates and black C conversion factors (details in Supplementary Information, Methods and Supplementary Information, Table S1). To initialize SOC pools for light- and heavy-textured Inceptisols, model runs were carried out to equilibrium (shown to 4,000 years in Fig. 2) after setting all initial pool sizes to zero, under the soil and climate scenarios of the two regions. Four scenarios for calculating the annual input of black C and non-black C to soil were then compared: (1) no fires occur and the entire above-ground biomass production is an input to soil (no fire); (2) biomass consumption by fire is considered but no black C is formed—residual non-black C after burning is input to soil (fire without BC formation); (3) both biomass consumption by fire and the formation of black C is considered but black C does not mineralize or disappear and accumulates in soil (fire with BC formation, without BC disappearance); (4) both biomass consumption by fire and the formation of black C is considered and black C disappears over time (fire with BC formation and BC disappearance). Modelled equilibrium stocks of C obtained by applying externally modelled biomass production for the C input to RothC matched analytical observations of non-black C very closely, and did not require further modifications. For scenario 4 above, the mean residence time of black C was independently calculated with a single exponential function, which was adjusted to fit the observed black C stock at equilibrium.

The feedback of total SOC stock changes to soil warming was then computed in response to a temperature increase of 1, 3 and 5 °C over 100 years. The response of non-black C to warming was modelled with RothC using pool distributions[19] established for equilibrium computations, whereas the response

to warming of black C was modelled externally using the MRT quantified (see Supplementary Information, Table S1). Two scenarios, representing different assumptions about initial pool size conditions were computed (Fig. 3). (1) The IOM pool in RothC is zero to simulate soils that do not contain any black C (and no other IOM) and the humified, resistant, decomposable and microbial biomass pools are initialized with values obtained from the equilibrium calculations to match vegetation input to measured SOC. (2) The humified, resistant, decomposable and microbial biomass pools are initialized with equilibrium values (Fig. 2); the IOM pool is assumed equal to measured black C (ref. 19; which is a conservative assumption as other very stable organic matter fractions may fall in this pool, as well; at minimum, IOM should include black C); MRTs calculated above are applied and mineralization of black C is set to increase with soil warming using the default rate modifier for temperature used for all other pools for the appropriate mean annual temperature determined at each site (see Supplementary Information, Methods). Sensitivity analysis was carried out for different MRTs of black C using varying assumptions about burning severity and formation (from Supplementary Information, Table S1, shown in Fig. 3).

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Author contributions

J.L., S.S., P.F., P.W. and K.C. planned the temperature response modelling. J.S. and M.B. planned and conducted the soil assessments, J.S. and E.K. conducted the soil analyses. J.C. contributed the vegetation model. J.L. and J.S. provided the modelling framework and analysed the data. J.L. developed the manuscript and J.S., S.S., P.F., K.C., P.W., K.C. and E.K. provided input to the manuscript.