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Black (pyrogenic) carbon in boreal forests: a synthesis of current knowledge and uncertainties

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Abstract

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The carbon (C) cycle in boreal regions is strongly influenced by fire, which converts biomass and detrital C mainly to gaseous forms (CO_2 and smaller proportions of CO and CH_4), and some 1–7% of mass to pyrogenic C (PyC). PyC is mainly produced as solid charred residues, including visually-defined charcoal, and a black carbon (BC) fraction chemically defined by its resistance to laboratory oxidation, plus much lower proportions of volatile soot and polycyclic aromatic hydrocarbons (PAHs). All PyC is characterized by fused aromatic rings, but varying in cluster sizes, and presence of other elements (N, O) and functional groups. There are several reasons for current interest in defining more precisely the role of PyC in the C cycle of boreal regions. First, PyC is resistant to decomposition, and therefore contributes to very stable C pools in soils and sediments. Second, it influences soil processes, mainly through its sorption properties and cation exchange capacity, and third, soot aerosols absorb solar radiation and may contribute to global warming. However, there are large gaps in the basic information needed to address these topics. While charcoal is commonly defined by visual criteria, analytical methods for BC are mainly based on various measures of oxidation resistance, or on yield of benzenopolycarboxylic acids. These methods are still being developed, and capture different fractions of the PyC “continuum”. There are few quantitative reports of PyC production and stocks in boreal forests (essentially none for boreal peatlands), and results are difficult to compare due to varying experimental goals and methods, as well as inconsistent terminology. There are almost no direct field measurements of BC aerosol production from boreal wildfires, and little direct information on rates and mechanisms for PyC loss. Structural characterization of charred biomass and forest floor from wildfires generally indicates a low level of thermal alteration, with the bulk of the material having H/C ratios still >0.2 , and small aromatic cluster sizes. For the more chemically-recalcitrant BC fraction, a variety of mainly circumstantial evidence suggests very slow decomposition, with turnover on a millennium timescale (5000–10 000 y), depending on environmental conditions and

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PyC properties, but the main limitation to PyC storage in soil is likely consumption by subsequent fires. Degraded, functionalized PyC is also incorporated into humified soil organic matter, and is transported to sediments in dissolved and particulate form. Boreal production is estimated as 7–17 Tg BC y^{-1} of solid residues and 2–2.5 Tg BC y^{-1}

5 as aerosols. Primary research needs include basic field data on PyC production and stocks in boreal forests and peatlands, suitable to support C budget modeling, and development of standardized analytical methods and of improved approaches to assess the chemical recalcitrance of typical chars from boreal wildfires. To accomplish these goals effectively will require much greater emphasis on interdisciplinary cooperation.

10 1 Introduction

Fire is a major driver of ecosystem processes and the carbon (C) cycle in boreal forests and peatlands (Hicke et al., 2003). Globally, it has been estimated that one-third of net primary productivity in boreal forests is consumed by fire (Harden et al., 2000; Wirth et al., 2002), while Bachelet et al. (2005) concluded that 61% of the C gained in Alaska 15 between 1922 to 1996 was lost to fires (1 out of 1.7 Pg C). Conard and Ivanova (1997) estimated that boreal forest fires accounted for about 20% of C emissions from global biomass burning. Amiro et al. (2001) estimated that C emission from Canadian forest fires during 1959–1999 amounted to 18% of emissions from all energy sources in Canada, with a similar magnitude of secondary CO₂ emission due to postfire decomposition. The retrospective analysis by Kurz and Apps (1999) indicated that Canada's 20 forests functioned as a sink for atmospheric C during the period 1920–1990. After 1970 however, increases in fire and insect disturbances have moved the balance closer to neutral or even to C loss. Increased burn areas are predicted for boreal forests of Canada, Russia and Alaska under warmer and drier conditions resulting from increased atmospheric CO₂ (Stocks et al., 1998, 2000; Bachelet et al., 2005; Flannigan et al., 2001, 2005). One obvious byproduct of forest and peatland fires is black, charred 25 residues from incomplete combustion of biomass and forest floor debris. Evidence for

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formation of charred biomass goes back 350 My to the Late Devonian (Schmidt and Noack, 2000). The charcoal record preserved in sediment and peat cores has long been used in studies of ecosystem change and fire frequency during the Holocene (e.g., Clark and Royall, 1994; Laird and Campbell, 2000; Flannigan et al., 2001; Sanborn et al., 2006). In recent years, there has been increasing interest in other aspects of black carbon (BC), or more generally, the whole range of pyrogenic C (PyC) forms (Hedges et al., 2000; Schmidt and Noack, 2000; Schmidt et al., 2001; Masiello, 2004). The PyC continuum (Fig. 1) includes chars that still retain their physical structures, soot and polycyclic aromatic hydrocarbons (PAHs) that condense out of the vapour phase, and highly recalcitrant graphite formed by geological processes. As discussed later, the condensed aromatic structures of PyC, especially of soot and graphite make them highly resistant to decomposition, and therefore they contribute to very stable C pools in soils and sediments. PyC may influence soil processes (microbial activity, nutrient cycling), while soot aerosols have been suggested as a contributor to atmospheric warming. Despite high interest in these aspects of PyC, there are few quantitative reports of PyC production and stocks and little direct information on mechanisms and rates of loss in boreal regions, especially for peatlands. We review the current state of knowledge on PyC, including chemical structures, definition of terms, analytical methods and quantitation, formation of PyC including soot aerosols and PAHs, stocks and characteristics in soil, loss mechanisms and longevity, the role of PyC in boreal forests, (C cycle, effects on soil processes), and identification of knowledge gaps and research needs. We focus on recent and generally available publications providing access to further literature, rather than a comprehensive literature survey.

2 What is PyC?

PyC exists as a continuum from partly charred plant material, through char and charcoal, to soot and graphite particles without distinct boundaries (Seiler and Crutzen, 1980). This is visualized in Fig. 1, which also indicates the terminology, the associated

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chemical and degradation properties, and ranges of operationally defined analytical windows. PyC can form in two fundamentally different ways. The solid residues of plant combustion form char, whereas volatiles formed within (and recondensed from) flames comprise more condensed soot structures, ranging up to highly graphitized organic carbon. The common, defining characteristic of fire-altered biomass is the presence of condensed aromatic rings. As the extent of thermal alteration increases, structures with small cross-linked aromatic clusters (Fig. 2a) give way to larger graphene sheets (Fig. 2b) that tend to form stacks with disordered packing (Fig. 2c). Soot particles that condense from the gas phase typically develop as concentric shells of these graphene stacks (“onion-like” structure, Fig. 2d). In the limit, graphite is a form of pure C, a highly ordered, electrically conducting structure of parallel graphene sheets of indefinite size. Its formation from soot or char precursors requires both heat and pressure.

Functional groups contain mainly oxygen (O) or hydrogen (H), and the degree of functionalization is reflected in the molar ratios O/C and H/C shown as a van Krevelen plot (Fig. 3). The number of functional groups, important for biological degradability, decreases continuously from the initial, thermally unaltered biomass to PyC. Large O/C and H/C ratios indicate the presence of many functional groups, whereas smaller ratios are typical for more condensed structures, with ratios close to 0.0 typical for almost pure carbon. It should be noted that molar ratios are typically used in geochemical studies and in this review, whereas most forestry and soil studies report concentration ratios.

3 Quantitative analysis of PyC

The wide range of PyC structures and properties is reflected in the variety of methods used for analysis, including visual detection of charcoal, measures of resistance to thermal, chemical or photo-oxidation, production of benzenepolycarboxylic acids after nitric acid oxidation (BPCA method), and ^{13}C nuclear magnetic resonance (NMR) spectroscopy to determine the proportion of aromatic C. Figure 1 shows the principal geochemical approaches used to quantify PyC in soils and sediments and the sizes

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and positions of the individual analytical windows. More detailed reviews of the various analytical approaches are given elsewhere (Kuhlbusch, 1998; Schmidt and Noack, 2000; Schmidt et al., 2001; Masiello, 2004; Cornelissen et al., 2005).

Analytical techniques for PyC in sediments and soils attempt to differentiate between three forms of C, i.e. inorganic carbonates, thermally unaltered organic C (such as humic substances and plant material) and PyC. A multitude of methods exists to differentiate PyC from other forms of organic C, but they essentially all fall into one of three categories. First, visual or microscopic techniques rely on visual discrimination of charred (i.e. blackened) vs. uncharred plant particles, followed by particle counting or actual physical separation and weighing. This fraction has been traditionally described as charcoal, although it is unlikely that much of it would meet the criteria of molar O/C and H/C ratios around 0.4, the van Krevelen ratios typically measured in geochemical studies of organic matter (Fig. 3).

Second, resistance to laboratory oxidation (induced through heating, exposure to acids or bases, or ultra-violet light, with or without various pre-extractions) is used to distinguish a BC fraction from other organic matter (Kuhlbusch, 1995; Skjemstad et al., 1996; Gustafsson et al., 1997; Hockaday and Hatcher, 2005). The most vigorous extraction and oxidation treatments leave only graphitic BC (GBC), which requires geochemical processes for its production (heat and pressure) and is not produced by forest fires alone (Gélinas et al., 2001). Third, the release of molecular markers unambiguously related to PyC, such as BPCAs or levoglucosan derived from heating of glucose serves as an indicator for the presence of PyC (Glaser et al., 1998; Elias et al., 2001; Brodowski et al., 2005b). Once the pyrogenic organic matter is separated it can be quantified with elemental analysis (C, N, H, O) and further characterized, e.g. by ^{13}C NMR spectroscopy.

One problem is that terms are not used consistently. BC has commonly been used to describe the whole continuum, but it is now more often used to specify the more resistant PyC fraction determined by resistance to laboratory oxidation or by production of BPCAs. The term elemental C (EC) has become more widespread to define the

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oxidation-resistant fraction in analysis of atmospheric aerosols and soot.

In this paper, we use PyC for the whole continuum of fire-altered biomass and organic matter (sometimes called pyromorphic materials), and char for the visually-detected fraction, reserving BC and GBC for the fractions defined chemically by resistance to oxidation, or by production of BPCAs. We use charcoal only where it has been reported as such, based on visual assessment, and similarly, use EC only where it has been reported as such for atmospheric studies. As methods for characterization and analysis of PyC are still under development, and definitions are not standardized, readers should check how terms are used in different publications.

Again, PyC is a continuum, whereas all analysis relies on clear-cut boundaries to separate the different forms of carbon – an obvious contradiction. The sizes and positions of analytical windows may vary greatly, capturing different parts of the combustion continuum. A recent comparative analysis of soils (Schmidt et al., 2001) showed that BC concentrations obtained by different methods can vary by two orders of magnitude, depending on the type of separation technique used (e.g. Currie et al., 2002). Briefly, particle size, morphology, and the presence or absence of a mineral matrix may lead to an over- or underestimation of the content of pyrogenic organic matter (Masiello, 2004). A new systematic comparative analysis was started in 2003 (Schmidt et al., 2003), and further details and continuous updates can be found at web site of the BC Ring Trial (<http://www.geo.unizh.ch/phys/bc/>).

4 Production of PyC

4.1 Production of PyC from forest biomass burning

4.1.1 Production determined by visually-detected charcoal

As shown in Table 1, few studies are available, and none with a complete accounting of BC or charcoal produced in a boreal wildfire. Three studies determined charcoal

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production of 235–932 kg ha⁻¹ from experimental fires with crowning or partial crowning (Clark et al., 1998; Ohlson and Tryterud, 2000; Lynch et al., 2004). These were designed specifically to provide information on how charcoal is transported to, and deposited in nearby sediments, with production determined from recovery of charcoal particles in surface traps. Higher yields have been found in other studies, including several of slash-burning after forest clearing: i.e., 7400 kg ha⁻¹ of forest floor charcoal after cutting and slash-burning of a forest site in Virginia (Schiffman and Johnson, 1989), and 1140–4540 kg ha⁻¹ char in four studies of forest clearing and burning in Amazonia (Fearnside et al., 1993, 1999, 2001; Graça et al., 1999). Conversion of 4.8% of initial mass to charred particles was found in a slash-pile-and-burn experiment designed to emulate possible Neolithic practices in temperate deciduous forest (Eckmeier et al., 2005). Much greater increases in site charcoal (<2 mm, to 30 cm depth), of 20 000 and 170 000 kg ha⁻¹, after clearfelling and burning of two eucalypt sites were reported by Hopmans et al. (2005).

In the only study following wildfire rather than experimental burns, Tinker and Knight (2000) determined 6400 kg ha⁻¹ of char production on coarse woody debris after an intense crown fire in Yellowstone National Park. For the boreal and Amazonian fires, factors for conversion of fuel (including living biomass, slash and coarse woody debris) to visually-determined char were 0.9–2.2% (mass basis), with a higher conversion factor (8%) for coarse woody debris. Using average C concentrations of 500 g kg⁻¹ for biomass and 700 g kg⁻¹ for char, these correspond to approximately 1.3–3.1% conversion on a C basis, or 11% for coarse woody debris.

Few other data are available, and comparison is problematic. In addition to these studies from specific fire sites, Clark and Royall (1994) estimated charcoal deposition rates from analysis of particles >10 µm in lake sediments. For pre-settlement North America, the highest rates (5000 kg ha⁻¹ y⁻¹) were found in prairie/forest border areas, followed by conifer, largely boreal forest, with rates of 500–5000 kg ha⁻¹ y⁻¹. Rates appeared to decline sharply after 1900, which was assumed to be due to fire suppression. Few studies of charcoal in sediments, peats, or forest floor measure down to

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$10\text{ }\mu\text{m}$ particle size, which may account in part for the high values.

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4.1.2 Production determined by chemically-defined BC

In the single study located, Czimczik et al. (2003) used the BPCA method to determine that a surface fire in a Scots pine (*Pinus sylvestris*) stand in Siberia generated

- 5 61 kg ha^{-1} of BC in forest floor (conversion of 0.7% of prefire forest floor C), adding to the prefire stock of 141 kg BC ha^{-1} . The BPCA method detects C only in larger aromatic clusters and thus gives lower results than visual detection of charcoal. Carbon-13 NMR analysis of the same samples showed that estimation of BC by total aromatic C was approximately tenfold higher than the BPCA value.

10 The conversion rates for woody biomass burning may be compared to those from savannah fires in South Africa, for which BC was quantified by a method based on resistance to thermal oxidation (Kuhlbusch et al., 1996). On average, pre-fire vegetation and litter was approximately 2700 kg ha^{-1} from which 90% of C was volatilized. Conversion to BC was 0.5 to 2% (mean 1%) based on prefire C stocks (defined as the
15 ratio BC/CE, where CE is carbon exposed to fire), or 1.3%, based on BC/ $\text{CO}_2\text{-C}$. From laboratory burns of 20 non-woody biomass substrates, Kuhlbusch and Crutzen (1995) found that BC/CE was 0.14–2.2%, or 0.15–2.9% as BC/ $\text{CO}_2\text{-C}$. For deciduous wood (mean for two substrates), the values were higher, with BC/CE 3.1%, BC/ $\text{CO}_2\text{-C}$ 4%, and BC constituting 58% of the charred residues. Despite the differences in methodology, burning conditions, and biomass substrates, these results for BC yields based
20 on chemical oxidation are in a similar range to those in Table 1 based on visual char assessment. However, it seems clear that the limited data on char or BC production from forest fires are an inadequate basis for modelling their role in boreal C budgets.

25 At the other extreme, two studies provide essentially background values for BC deposition, away from the influence of forest fires. Muri et al. (2002) analysed BC in sediments of remote Alpine lakes in Slovenia, above and below the treeline. The purpose was to track the effects of atmospheric deposition from fossil fuel burning, and forest fires do not seem to have been common. For all remote lakes, the BC/OC ratio

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was less than 0.1, within the range found for marine sediments, and the pre-industrial BC sedimentation rate was estimated to be $<5\text{ kg C ha}^{-1}\text{ y}^{-1}$. This is similar to estimates of Wang et al. (2005) of BC deposition for the Chinese Loess Plateau over the last two glacial cycles ($>200\text{ ky}$).

5 4.2 Aerosol BC emissions from forest fires

As this review focuses on the solid charred residues produced by fire, we present a less detailed overview of BC as a component of volatile emissions. Techniques used to quantify atmospheric BC (often called EC in this context) generally differ from those used for solid soot and char, and will not be discussed here (e.g., Schmid et al., 2001; Conny and Slater, 2002). In addition to soot, volatile emissions from forest fires include a complex mixture of organic compounds and poorly-characterized high-molecular-weight substances (Andreae and Merlet, 2001; Friedli et al., 2001; Hays et al., 2002; Duarte et al., 2005), collectively referred to as organic C (OC) or particulate organic matter (POM). Smoke aerosols affect air quality locally, such as demonstrated for the Missoula Valley, Montana (Ward and Smith, 2005), but can also be transported over long distances. For example, smoke plumes from Siberian forest fires in 2003 affected air quality over Korea (Lee et al., 2005), and Canadian forest fires have been associated with events of elevated CO, BC and OC over the USA (Wotawa and Trainer, 2000; Park et al., 2003). Boreal forest fires contribute substantially to atmospheric BC in the Arctic (Cooke and Wilson, 1996), and soot deposition reduces the albedo of Arctic sea ice (Kim et al., 2005); even Antarctica receives BC from biomass burning in the tropics (Wolff and Cachier, 1998).

In addition to impacts on visibility and health, atmospheric BC produced from fossil fuel and biomass combustion has been identified as a contributor to global warming due to its direct absorption of solar radiation (Jacobson, 2002), and there have been calls for its control to mitigate this effect (Hansen et al., 2000; Bond and Sun, 2005). This is opposite to the effect of sulphate aerosols that tend to mitigate the greenhouse effect by reflecting solar radiation; secondary effects of atmospheric BC

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and other aerosols are discussed elsewhere (Jacobson, 2002; Menon et al., 2002).

Liousse et al. (1996) and Cooke and Wilson (1996) first modeled global atmospheric BC concentrations (typically measured in ng m^{-3} or $\mu\text{g m}^{-3}$). The latter study estimated BC emission as 5.98 Tg y^{-1} from biomass combustion plus 7.96 Tg y^{-1} from fossil fuels.
5 It estimated a lifetime of 7.85 days for BC aerosol particles, during which they increase in hydrophilic character, and are then removed in precipitation with the same efficiency as sulphate. Liousse et al. (1996) estimated $6.64 \text{ Tg BC y}^{-1}$ from fossil fuel, and $5.63 \text{ Tg BC y}^{-1}$ from biomass combustion, but did not include BC production from natural fires of temperate and boreal regions. They also concluded that wet deposition
10 was at least 4 times larger than dry deposition in most locations, but both studies noted the scarcity of precipitation data.

Other studies have yielded similar estimates: Andreae and Merlet (2001) evaluated data on biomass combustion and emission factors (EF, expressed in g BC per kg of biomass burned), yielding an estimate of 4.8 Tg BC y^{-1} for the late 1990s. Streets et
15 al. (2003) used these EFs to estimate BC emissions in Asia of 0.45 Tg y^{-1} from 730 Tg of biomass burned (a mean EF of $0.62 \text{ mg BC kg}^{-1}$ biomass, in the lower end of the range). Note that these four studies included biomass fuel use: Bond and Sun (2005) recently estimated global production of 3.3 Tg BC y^{-1} specifically from open vegetation burning.
20 The most recent study of global BC aerosol emissions from all sources shows 2 to 2.5 Tg BC y^{-1} for northern hemisphere open vegetation burning for the period 1960–2000 (Ito and Penner, 2005).

A detailed spatial and monthly inventory of carbonaceous particle emission from boreal and temperate wildfires in forests, shrublands and grasslands for 1960–1997 was produced by Lavoué et al. (2000). One key result was the very high interannual variability:
25 fires in boreal and temperate regions accounted for 4%, on average, of global biomass burned, but ranged up to 11.5% of biomass burned, with emissions up to 8.8% of BC and 19.4% of POM, due to less complete combustion than in other ecosystems. They noted that particle emissions from boreal fires may have especially high impact in Arctic regions that are highly sensitive to climate change. The BC emissions from

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Canadian fires were mainly from sparsely populated regions (Northwest Territories, and northern Saskatchewan, Manitoba and northwestern Ontario). Total BC emission from Canadian forest fires has been increasing, with estimates (Tg y^{-1}) of 0.016 in 1960–1969, 0.019 in 1970–1979, 0.038 in 1980–1989, and 0.041 in 1990–1997, consistent with the total C trends reported by Kurz and Apps (1999). The increase in fire disturbance has been mainly attributed to warmer and drier conditions, especially during the 1980s.

Park et al. (2003) estimated 0.84 Tg y^{-1} of OC emission from Canadian fire emissions (climatological mean 1997–2000). Using their emission ratio of OC/EC of 7, this amounts to 0.12 Tg EC . The discrepancy between this and the lower values in Lavoué et al. (2000) for 1980–1997 may be largely due to the different EFs used. Park et al. (2003) used $2 \text{ g EC per kg dry mass burned}$, whereas Lavoué et al. (2000) used 0.75, assuming 50% biomass consumption during flaming and 50% during smoldering.

These analyses of global and regional BC emissions and transport rest on many assumptions and uncertain estimates, highlighting the need for more direct measurements of BC and OC emissions from boreal forest fires, and atmospheric BC concentrations, and BC deposition rates, both wet and dry. Estimated or measured EFs for BC production are generally within the range of $0.5\text{--}2 \text{ g BC kg}^{-1}$ biomass. Hays et al. (2002) recently determined EFs for EC and many elements and organic compounds for six foliar fuels under open burning to simulate natural events. The EFs for needles of loblolly pine (*Pinus taeda* L.), western hemlock (*Tsuga heterophylla*), and ponderosa pine (*Pinus ponderosa*) were (g EC kg^{-1}) 1.3, 0.4 and 0.4, respectively, consistent with this range.

Until recently, there was little direct information on aerosols and gases produced by boreal wildfires. As part of the International Crown Fire Modelling Experiment (ICFME), three studies were carried out near Fort Providence, NWT, Canada, in jack pine (*Pinus banksiana*) stands of 12 m height. Particle production and size distribution in smoke plumes was measured in 2000 (Payne et al., 2004), gases and total nonmethane hy-

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drocarbons in 1997 (Cofer et al., 1998), and BC and OC in aerosol particles in 1998 (Conny and Slater, 2002). In the latter study, which used both ground-based and aerial sampling of particles up to $2.5\text{ }\mu\text{m}$ diameter, the ratio of BC/TC was found to be 0.085 for the flaming stage and 0.0087 for the smouldering stage, but EFs were not determined.

More information on aerosol emissions is now available from experimental fires in three Scots pine sites of central Siberia (Samsonov et al., 2005). On average, total organic substances (assuming 50% C content) constituted 50–70% of total aerosol mass, while EC amounted to 7–15%. Aerosol emissions corresponded to 1%–7% of the mass of fire-consumed biomass, scaled up to an estimated 3–10 Tg aerosol mass per year for boreal Russia from 12–14 million ha burned. This corresponds to 0.2–1.5 Tg of EC, in line with previous estimates.

4.3 PAHs from forest fires

The products from incomplete combustion of carbon-based fuels include polycyclic aromatic hydrocarbons (PAHs), having structures of 2–7 fused aromatic rings. The natural background of PAH deposition from wildfires is preserved in sediments, which also record the recent increases from fossil fuel combustion, industrial pulping and direct contamination with oil and coal (Yunker and Macdonald, 2003). In urban areas, atmospheric PAHs originate largely from fossil fuel combustion, but extensive use of wood for home heating was responsible for half the atmospheric PAHs at a semirural forested Swedish site (Mandalakis et al., 2005). PAHs can be transported long distances, especially the lighter ones (Bucheli et al., 2004), and they are deposited in Arctic regions far from their sources (Yunker et al., 2002; Hung et al., 2005).

PAHs are produced in the gas phase during combustion, but at lower temperatures they rapidly condense onto ash particles (Kim et al., 2003). Therefore PAHs formed in forest fires may be emitted as volatiles or deposited on the soil surface with the ash. We did not locate any direct measurements of PAH emission from forest fires, but the previously-noted study of Hays et al. (2002) on open burning of foliar fuels

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found that PAHs constituted 0.2 to 2 g kg⁻¹ of PM_{2.5} (fine particulate matter with aerodynamic diameter <2.5 µm) which itself was formed with an of EF of 15–35 g kg⁻¹. Kim et al. (2003) measured changes in PAH concentrations in 0–5 cm soil after forest fires at three sites in Korea. On average, soil PAH concentrations (sum of the 16 US Environmental Protection Agency (EPA) priority PAHs, denoted Σ16PAH) were 1200, 200 and 300 µg kg⁻¹ at one, five, and nine months after fire, compared to 50 µg kg⁻¹ in control sites. The decline in the first few months was attributed to loss of ash by wind and rain erosion. The same 16 PAHs were determined in 0–10 cm soils of the Swiss soil monitoring network (Bucheli et al., 2004), with median Σ16PAH values increasing in the order of arable soils (66 µg kg⁻¹), grassland and pasture (142 µg kg⁻¹), forests (164 µg kg⁻¹) and urban soils of (451 µg kg⁻¹). Wilke and Amelung (2000) found similar values for the sum of 20 PAHs in North American native grassland sites (63–321 µg kg⁻¹). No data were located in a literature search for information on PAHs in boreal forest soils.

Compared to non-pyrogenic OM in soils and sediments, BC has a much higher ability to adsorb PAHs and other hydrophobic organic compounds. This has been the subject of much recent research, especially concerning the fate and bioavailability of these compounds in BC-containing harbour sediments (e.g., Accardi-Dey and Geschwend, 2003; Cornelissen et al., 2005; Jonker et al., 2005; Lohmann et al., 2005). Laboratory-produced biomass chars (Chun et al., 2004; Zhu and Pignatello, 2005) also showed high affinity for aromatic compounds, depending on the degree of carbonization and oxygen loss. The influence of soil and sediment BC on the fate of organic pollutants is not likely of immediate concern for boreal regions which are mostly affected by natural fires. Studies of PAH deposition rates in peatlands of eastern Canada (Dreyer et al., 2005), and of PAHs in precipitation in Altantic Canada (Brun et al., 2004) indicated that emissions were predominantly from coal and fossil fuel combustion rather than forest fires.

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5 Characteristics and stocks of soil PyC

5.1 General effects of fire on SOM properties

As recently reviewed (Certini, 2005; González-Pérez et al., 2004), the effects of fire on soil organic matter (SOM) include a relative decrease in the proportion of humic acid (the fraction soluble in NaOH), with increase in the insoluble humin fraction, due to production of BC and melanoidins. The latter are hypothesized to form by condensation of reducing sugars with amine groups upon heating. There is a loss of carbohydrates and oxygen-containing functional groups. The proportion of alkyl C may increase, and this contributes to soil erosion, landslides and flooding in mountainous territory (Neary et al., 1999; DeBano, 2000). The loss of carbohydrates (indicated by reduction of O-alkyl C in the ^{13}C NMR spectra) also contributes to loss of aggregate stability (Kavdir et al., 2005). Thus, fire generally causes the remaining organic matter to become less soluble and more resistant to oxidation and biological decomposition. It has also been suggested that podzolization processes are inhibited by frequent fires that destroy the forest floor and C available for translocation (Mokma and Vance, 1989; Czimczik et al., 2005).

The increasing proportions of aromatic C and loss of oxygen-containing functional groups with charring can be observed directly by ^{13}C NMR. Spectra of charred biomass and SOM are characterized by a broad aromatic signal with maximum around 126 ppm, which can account for up to 100% of the organic C (Baldock and Smernik, 2002; Czimczik et al., 2002, 2003; González-Pérez et al., 2004; Simpson and Hatcher, 2004a, b; Knicker et al., 2005a, b). Condensed aromatic structures are poorly detected by routine solid-state NMR with cross-polarization (CP), so that a more time-consuming quantitative technique (Bloch decay, BD) must be used. The ^{13}C NMR spectra in Fig. 4 illustrate the effects of charring on Scots pine wood from the wildfire site in Czimczik et al. (2003). The CP spectrum of the fresh wood (Fig. 4a) is dominated by the O-alkyl peak of cellulose at 73 ppm. The CP spectrum of the charred wood (Fig. 4b) is dom-

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inated by aromatic C at 125 ppm, with small peaks due to residual cellulose, and an increase in alkyl C (37 ppm). The corresponding quantitative BD spectrum (Fig. 4c) has a higher proportion of aromatic carbon than the CP spectrum.

High-resolution transmission electron microscopy actually reveals the condensed aromatic clusters, in both disordered and concentric onion-like structures (Poirier et al., 2000; Schmidt et al., 2002). Other novel approaches to BC characterization include ultra-high-resolution mass spectrometry (Kim et al., 2004; Kramer et al., 2004), and various forms of X-ray spectroscopy (Solomon et al., 2005), which have been coupled with microscopic techniques to produce spatial distribution of BC forms (Brodowski et al., 2005a; Lehmann et al., 2005).

Charring often results in only small changes (Czimczik et al., 2003; Knicker et al., 2005a) or a decrease (Baldock and Smernik, 2002; Harden et al., 2004; Neff et al., 2005) in the C/N ratio, and an increase in more recalcitrant organic N forms (González-Pérez et al., 2004; Castro et al., 2006). In SOM unaffected by fire, ^{15}N NMR shows that amide is the predominant form of soil N, whereas a small proportion of pyrrole-type N is found in fire-affected soils, and can be considered a molecular marker of fire (Knicker et al., 2005a).

Effects of charring on $\delta^{13}\text{C}$ are generally small and somewhat erratic (Bird and Gröcke, 1997; Czimczik et al., 2002; Rumpel et al., 2006). With increasing thermal alteration, biomass should become isotopically lighter with preferential loss of cellulose, and greater contribution of more-depleted lignin to formation of condensed aromatic structures. However, this would be counteracted by the kinetic effect, so that ^{12}C should be lost slightly faster than ^{13}C , leading to isotopic enrichment. In general, char retains the signature of its C3 or C4 plant origin (Hiradate et al., 2004), although much greater depletion has been observed for char derived from some C4 grasses, especially for natural rather than oxygen-limited laboratory burns (Turekian et al., 1998; Krull et al., 2003). Measurement of changes in $\delta^{13}\text{C}$ of atmospheric CO_2 is one of the approaches used to quantify CO_2 emissions from forest fires (Langenfelds et al., 2002; Schuur et al., 2003; Randerson et al., 2005).

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5.2.1 Qualitative studies

There is increasing evidence that pyrogenic C can make a substantial contribution to SOM in fire-prone ecosystems, and can influence pedogenesis, although studies from boreal regions are lacking. The isolation of highly aromatic humic acids (the fraction obtained by extraction with base and subsequent precipitation with acid) from SOM has long been considered an indicator of BC inputs (Haumaier and Zech, 1995; Golchin et al. 1997; Zech et al., 1997; Almendros et al., 2003; Benites et al., 2005), as is development of stable humus forms highly resistant to decomposition (Almendros et al., 2003; Benites et al., 2005). Highly aromatic humic acids (Shindo et al., 1986; Hiradate et al., 2004) and charred plant fragments constituting 3–33% of SOC (Shindo et al., 2004, 2005) were found in volcanic ash soils from Japanese grasslands subject to frequent burning.

The development of dark Chernozemic soils in the Canadian prairies has been attributed to frequent fire (Ponomarenko and Anderson, 2001). Charcoal inputs have also been proposed to account for the patchy occurrence of relict dark Chernozems in Europe, especially where there were Neolithic settlements (Schmid et al., 2002; Schmidt et al., 2002; Kleber et al., 2003). Black carbon is a defining property of the South American anthropogenic soils (*Terra Preta*) developed by native populations before European contact. They are dark in colour and still high in fertility compared to the surrounding highly weathered and infertile Oxisols (Glaser et al., 2000, 2001, 2002). In the deep A horizons (40–80 cm), BC (BPCA method) typically constitutes 20% or more of SOC.

5.2.2 Quantitative analysis

Table 2a summarizes literature data for soil BC from mainly non-forest sites, as percentage of total SOC if available. The values obtained from photo-oxidation and ^{13}C NMR

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of the <53 µm fraction (Skjemstad et al., 1999a, 2002, 2004), up to 35% of SOC, are in general higher than those from other thermal and chemical measures of oxidation resistance (Schmidt et al., 2001). Some higher values were found by Ponomarenko and Anderson (2001), who determined BC in soils and size fractions by mass remaining after photo-oxidation. These higher values are likely because the residues from photooxidation still contain other resistant or protected C structures that are detected by NMR. The highest values came from a pyrogenic morphon; i.e., a small component in the soil profile with large amounts of charred particles that were visually obvious. Lower results were found using the BPCA method (Glaser and Amelung, 2003; Dai et al., 2005), that only detects BCPAs released from polycyclic aromatic structures, and by thermal oxidation with some pre-extraction (Bucheli et al., 2004).

5.3 PyC in forest soils

5.3.1 Qualitative characterization

Few studies have characterized or quantified the BC component of forest soils, especially for boreal regions. Solid-state ^{13}C NMR was used to elucidate changes in organic matter composition for wildfires in Spain (Knicker et al., 2005a) and Turkey (Kavdir et al., 2005), and for prescribed fire in eucalypt plantations in Australia (Guinto et al., 1999). The general results are similar to trends reported in recent reviews (González-Pérez et al., 2004; Certini, 2005) and for a moorland site (Haslam et al., 1998), mainly 15 a relative decrease in O-alkyl C and increase in alkyl and aromatic C. While individual charred particles may be very high in aromatic C, there may actually be little change in aromatic C in bulk forest floor or surface mineral horizons of fire-affected sites. This can 20 be the case where organic matter is consumed by fire, leaving ash rather than char, with little alteration of underlying material. In addition, within a few years, new organic matter is built up from the pulse of detritus from fire-killed biomass and litter from new vegetation.

Preston et al. (2002b) found evidence for BC in coastal temperate forests of southern

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Vancouver Island, British Columbia, in a solid-state ^{13}C NMR of five organic matter pools: fine woody debris, forest floor, roots from the forest floor, and two water-floatable fractions (2–8 mm and <2 mm) from 10–30 cm mineral soil. Several samples of the 2–8 mm floatables were high in aromatic C characteristic of BC, all from the drier east side of Vancouver Island, where fire was historically more prevalent.

Although sparse, information on boreal forests includes two studies on the Boreal Forest Transect Case Study (BFTCS) in northern Saskatchewan and Manitoba. Preston et al. (2002a) separated mineral soil from two jack pine and one black spruce (*Picea mariana*) sites into water-floatables and three size fractions. Solid-state ^{13}C NMR spectra suggested a small BC contribution to the <63 μm fraction of the A horizon of the jack pine sites, but not to the B horizon samples. For the black spruce site, the broad aromatic peak in the 250–63 μm fraction of the A+B horizon also indicated possible incorporation of BC into mineral soil OM. In a survey of forest floor chemistry along the BFTCS transect, Preston et al. (2006) found high aromatic intensity from charcoal in some of the samples from jack pine sites, but not from black spruce sites. No attempt was made to quantify BC, as the studies were not designed with this objective.

Also using ^{13}C NMR, Czimczik et al. (2003) characterized the changes in organic composition of forest floor, by sampling burned and unburned patches shortly after a wildfire in Scots pine in central Siberia. The highly aromatic structures produced from fire were underrepresented by routine CP, and quantitative spectra required the much slower and less sensitive Bloch decay (BD) method. As discussed later, this study also determined stocks of forest floor BC before and after fire using the BPCA method, which accounted for approximately one-tenth of the aromatic C in NMR spectra.

Harden et al. (2004) used the approach of thermogravimetry in O_2 to characterize fire effects on organic horizons as part of the FROSTFIRE experimental burn study in black spruce in Alaska. With burning, the proportion of mass loss at the first peak (280°C) decreased and that at the second peak (400°C) increased, and there was lower total mass loss up to 1000°C. These changes indicate loss of labile C, mainly carbohydrates,

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and increase in lignin and/or black C. Similar changes in organic composition were found following wildfire in Alaska (Neff et al., 2005). Although peatlands comprise large areas of the boreal forest, and are also vulnerable to fire (Zoltai et al., 1998; Benscoter and Wieder, 2003; Turetsky et al., 2004), we found no information on BC characteristics or stocks. Analyses of char in peat profiles to investigate paleobotany and peat development (e.g., Kuhry, 1994; Laird and Campbell, 2000; Flannigan et al., 2001) are not generally designed for this purpose.

As part of their studies of C cycling in the BOREAS Old Black Spruce study area near Thompson Manitoba, Harden et al. (1997) and Trumbore and Harden (1997) concluded that charred material was an important input to deep organic layers in black spruce-feathermoss. In a later study, Manies et al. (2005) estimated that 10 to 60% of C in deep organic horizons is derived from wood biomass, specifically from fire-killed trees (i.e., data were only used from buried wood with evidence of charring). However, charcoal was not specifically quantified.

5.3.2 BC and charcoal in forest sites – quantitative analyses

For organic horizons, only two studies were found, based on visual assessment of charcoal (Table 2b). Higher amounts ($984\text{--}2074\text{ kg ha}^{-1}$) were found by Zackrisson et al. (1996) in Sweden than in the Quebec study ($343\text{--}1295\text{ kg ha}^{-1}$) by Bélanger et al. (2004 and personal communication). This is probably due to several factors, including low intensity of the fire, low biomass of the stands, and no quantification of charcoal fragments smaller than 2 mm.

Carcaillet and Talon (2001) determined charcoal stocks in soil profiles along altitudinal transects in dry valleys in the French Alps. In the current conifer-dominated forest belt (1700 to 2200 m a.s.l.) charcoal C accounted for 0.1 to 20% of SOC, corresponding to $10\text{ to }30\,000\text{ kg ha}^{-1}$ of charcoal. The high variability likely resulted from both local spatial variation in fuel type (e.g., log v.s. twigs or shrubs), and variation in fire frequency throughout the Holocene.

Two studies of Scots pine stands in Siberia produced widely differing results. For a

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215-y-old stand (Schulze et al., 1999), BC determined by a thermal method amounted to 20–24% of total C in forest floor plus 0–75 cm mineral soil, and 13% for 75–300 cm. Using the BPCA method, however, Czimczik et al. (2005) found much lower soil BC stocks for a fire chronosequence in the same study area, with 99% of the BC found in the organic layer (Fig. 5). This study also indicated that BC could be lost in subsequent fires: for sites with low fire frequency, BC stocks in the organic layer were up to 720 kg ha^{-1} , but much lower values ($<50 \text{ kg ha}^{-1}$) were found for stands subject to intense fires that had previously consumed the whole organic layer. Since the BPCA method detects only more highly condensed aromatic structures, these BC stocks are likely much lower than what would be found by visual detection of charcoal.

A recent Australian study produced high estimates of both charcoal generated in slashburning (Sect. 4.1.1, Table 1) and soil charcoal C stocks ($>2 \text{ mm}$, to 30 cm depth). Charcoal accounted for 15–32% of total soil C, or $15\,000\text{--}140\,000 \text{ kg ha}^{-1}$, and would have been higher if smaller particles could have been isolated. A similar result was found for Corsican forests, with 9800 to $14\,800 \text{ kg ha}^{-1}$ of soil charcoal (Carcaillet and Talon, 2001). In general, estimates of both production and stocks of PyC as BC or charcoal are very limited, especially for boreal regions, and very difficult to compare, due to the variations of sampling and analysis. However, estimates of forest floor charcoal stocks in the order of $1000\text{--}2000 \text{ kg ha}^{-1}$ for boreal forest sites may well underestimate total site charcoal, including that in mineral soil, coarse woody debris, and very fine particles.

5.4 BC in marine sediments

Atmospheric deposition, and dissolved, colloidal and particulate organic matter of rivers deliver BC to marine sediments, where it constitutes up to one-third of organic C (Gustafsson and Gschwend, 1998; Masiello and Druffel, 1998, 2001; Middelburg et al., 1999; Mitra et al., 2002; Golding et al., 2004). Only recently has analytical evidence for the presence of BC in the dissolved or colloidal fractions of soil been obtained. In acid extracts from soils with frequent vegetation fires, Kramer et al. (2004) detected heavily

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carboxylated, condensed aromatic structures, characteristic of BC-like material. Similar structures have been detected in the dissolved fractions of water samples from the Delaware and Amazon River systems (Kim et al., 2004).

Pyrogenic organic matter and its breakdown products have been suspected to be exported from boreal forests to rivers and ultimately to the Arctic Ocean (Schmidt, 2004). There are no published analyses of the dissolved fraction of Arctic rivers draining boreal forests. However, in sediments of Siberian rivers Guo et al. (2004) found that the proportion of BC increased consistently (1–15% of the sedimentary organic matter) from east to west. With increasing proportions of BC the apparent ^{14}C ages also increased, supporting the idea that BC is more resistant than other soil organic carbon. Göni et al. (2005) found that ancient organic C (radiocarbon age exceeding 7000 y) accounted for ~70% of the particulate organic matter delivered by the Mackenzie River to the Mackenzie Delta/Beaufort Shelf region. It was suggested to originate from a combination of fossil C (bitumen and/or kerogen) eroding from sedimentary rocks, and old soil C released from permafrost deposits. It is possible that the former category could include graphite, and the latter BC from ancient fires. Degradation products of BC have also been found to contribute to the old, pre-aged organic matter found in shelf sediments originating from temperate regions (Masiello and Druffel, 2003; Dickens et al., 2004a, b).

20 6 Mechanisms of PyC loss

6.1 Background

Pyrogenic C is produced in enormous quantities both from wildfires and from burning fossil fuels (Kuhlbusch, 1998) and is ubiquitous in the terrestrial environment (Schmidt and Noack, 2000). Although PyC is generally assumed to be relatively inert to degradation (especially the BC fraction that is oxidation-resistant), it must degrade eventually (Schmidt and Noack, 2000). High-latitude coniferous forests are large fire-prone

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ecosystems, with presumed high production of BC, and yet the amounts of BC found on the forest floor can be surprisingly small (Czimczik et al., 2003), emphasizing how poorly the fate of BC in soil is understood.

There is ample evidence that char, or highly aromatic carbon structures are decomposed more slowly than other plant residues. Studies of C-depleted plots from long-term bare fallows showed that the remaining soil C was enriched in aromatic structures (Kiem et al., 2000). Similar results were found in two Australian studies of agricultural soils (Skjemstad et al., 1999b, 2004). The Terra Preta anthropogenic soils of South America have retained their higher C and BC contents centuries after being abandoned (Glaser et al., 2001).

There is increasing evidence, however, that BC is susceptible to losses, especially over longer time scales. One likely mechanism for BC loss is oxidation by subsequent fires. Based on measurements of charcoal production, stocks in forest floor, and fire return intervals, Ohlson and Tryterud (2000) concluded that charcoal stocks in boreal forests should be approximately ten times higher than actually observed. To explain this discrepancy, they suggested that some charcoal is consumed by subsequent fires, a conclusion also reached by Czimczik et al. (2005). Slow chemical oxidation also likely plays a role; there is evidence that atmospheric BC can be oxidized, carboxylated and eventually become water soluble (Decessari et al., 2002). The surfaces of BC particles aged in soils for decades to millennia become increasingly oxidized (Schmidt et al., 2002; Brodowski et al., 2005b), characteristically exhibiting more carboxylic and phenolic groups (Lehmann et al., 2005).

Pyrogenic carbon is not immune to biological degradation. Even graphite, which can be weathered out of rock, transported and reburied in ocean sediments (Dickens et al., 2004a, b) is not immune; 2% oxidation was reported by Shneour (1966) for a 96-day incubation of artificial graphite in soil. Compared to the PyC formed from fossil fuel combustion, however, PyC formed from biomass burning has a relatively low degree of thermal transformation (Fernandes et al., 2003; Knicker et al., 2005b), with higher polarity, and small and less-ordered aromatic regions. Similar to laboratory-produced

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chars from wood (Baldock and Smernik, 2002; Czimczik et al., 2002), BC found in boreal forest floor after wildfire had small cluster size with many functional groups (Czimczik et al., 2003). Such PyC would probably fall in the intermediate region termed charcoal in Fig. 1. Small cluster size combined with a relatively high degree of functionalization would probably make these particles more susceptible to chemical degradation than more highly condensed structures. By comparing burned and protected savanna plots, Bird et al. (1999) concluded that the half-life of BC was <100 years at 0–5 cm. He also summarized literature from archaeological studies consistent with charcoal loss, and noted that charcoal fragments are well-preserved in peat or sediment cores where decomposition is generally hindered by lack of oxygen.

6.2 Incubation studies

The few incubation studies show very low rates of decomposition in a laboratory setting. Shindo (1991) used charred plant residues from an experimental fire in a grassland (mainly *Miscanthus sinensis* A.), and found “hardly any” decomposition after 40 weeks incubation in volcanic ash soil. Baldock and Smernik (2002) used red pine (*Pinus resinosa*) wood charred at different temperatures. After 120 days incubation in sand, 20% of C was mineralized from wood heated at 70°C (essentially unaltered). Carbon mineralization decreased to 13% for wood heated to 150°C, and to less than 2% for chars produced at 200–350°C, with increasing proportions of aromatic C. Incubation of wheat and rice chars (produced at 350°C for 2 h) with soil and quartz showed a rapid initial loss of 5–50% of the initial BC (determined by the BPCA method) within the first 6 months, but this levelled off in the following 1.5 years (Brodowski, 2004). No changes in BC content were found in the sterile controls, consistent with microbial rather than abiotic processes.

The importance of an additional source of more readily-available C (co-metabolism) for the breakdown of PyC was shown by Hamer et al. (2004), in a study of charred maize (*Zea mays* L.), rye (*Secale cereale* L.) and oak wood incubated in sand for 60 days. For the charred substrates, carbon mineralization of 0.3%–0.8% was increased

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to 0.6%–1.2% by glucose additions at 0 and 26 days. However, glucose mineralization was also enhanced by addition of charcoal, suggesting that its high surface area and porous structure may promote growth of microorganisms. Similar effects have also been found in field studies, as discussed in Sect. 7.3. Hamer et al. (2004) suggested that small molecules may not be stabilized by sorption into charcoal pores, but could actually become even more susceptible to microbial activity.

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The possibilities for microbial degradation of charred biomass are likely to be similar to those occurring in forest sites that have shown microbial utilization of C from lignite combustion ash (Machulla et al., 2004), and from lignite in soils developed after open-pit coal mining (Rumpel and Kögel-Knabner, 2002, 2004). The latter studies also showed increasing transformation of lignite into humic acids and the importance of co-metabolism for decomposition of lignite.

6.3 Movement and fragmentation of charcoal

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Charcoal production mainly occurs within organic horizons and detritus, but there is much evidence for its fragmentation and movement down into mineral horizons. Movement of fine char particles down through mineral soil was demonstrated for sugar cane fields in Australia (Skjemstad et al., 1999b), native grassland in Texas (Dai et al., 2005) and slash-and-burn soils in Thailand (Rumpel et al., 2006). As noted previously, Ponamarenko and Anderson (2001) found high proportions of charcoal C in aspen parkland sites. They also used scanning electron microscopy to show the importance of pedoturbation and mechanical processes that cause downward movement, and fragmentation and rounding of charcoal particles, especially the more fragile combustion products of herbaceous plants compared to those of woody origin.

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Little information is available for forested regions, although some studies have been undertaken to deal with the effects of charcoal movement on reconstruction of Holocene vegetation and disturbance regimes. For an altitudinal transect in the French Alps, Carcaillet (2001a, b) showed that fragmentation by freeze-thaw processes, bioturbation by soil fauna, and profile disturbance by tree uprooting and surface erosion

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following fires all contribute to downward migration of charcoal. This study concluded that “the minimum time for a charcoal particle to migrate down the soil profile to a depth of 100 cm is less than 500 y at 2000 m a.s.l., but most of the charcoal production of the last two millennia remains in the upper 30 cm. After 2500–6000 y, the major part of the charcoal mass was buried at a depth of 40–70 cm for elevations below 2000 m.”

Tree uprooting was considered to be the main cause of charcoal movement for a coastal temperate rainforest of Vancouver Island (Clayoquot River watershed), where cryoturbation, bioturbation and landslides were of minor importance (Gavin, 2003; Gavin et al., 2003). Figure 6 from Gavin (2003) shows how charcoal could be transported from organic to mineral horizons from fires at 2500 y and 1500 y before present. The importance of charcoal burial by tree uprooting also shown by Talon et al. (2005), who used the charcoal fragments in podzolic soils to reconstruct fire history in an old-growth deciduous forest in southern Québec. On steeper slopes of British Columbia interior rainforest, charcoal is transported and buried mainly by shallow earth landslides resulting from loss of root strength after wildfires (Sanborn et al., 2006). A recent study of Alaskan soils (Ping et al., 2005) noted that charcoal particles typically accumulated at the base of the O horizon, and throughout the A and upper B horizons, and that profile development can be influenced by gelifluction and cyroturbation. Therefore, charcoal movement and fragmentation in boreal regions may occur by several mechanisms, including tree uprooting, mass wasting, gelifluction and cryoturbation.

6.4 Degradation of geological and sedimentary substrates

Studies using a variety of techniques indicate that the resistance of BC and char to mineralization is relative, and while the rates may be slow, they appear to be affected by similar factors that inhibit or enhance loss of other organic substrates. The results of short-term incubations would mainly represent the less-condensed fraction, so rates would be expected to decline with time. However, decomposition in field settings could also be influenced by other factors such as chemical and photooxidation, leaching, and availability of nutrients and more readily-available C. While these have not been

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investigated for charcoal and BC in forest or peatland settings, some information can be gathered from studies of other recalcitrant organic substrates such as coal, shale and kerogen.

At geological timescales, chemical weathering of ancient organic matter plays a role in the global C cycle. These slow processes consume oxygen, release CO₂, and generate dissolved and solid oxidation products (Mackenzie et al., 2004a). Recent studies of coal (Chang and Berner, 1998, 1999) and black shale (Petch et al., 2000, 2001) have highlighted the importance of contact with oxygen-rich surface waters. The laboratory coal oxidation studies of Chang and Berner (1998, 1999) show that while chemical oxidation rates are low, they are significant on a geological time scale. Chang and Berner (1999) estimated lifetimes of 8000 and 80 000 y for coal with grain sizes of 10 µm and 100 µm diameter, respectively. The coal and shale studies also show that CO₂ production accounts for only 30–50% of oxygen consumption, with the rest producing partially oxidized solids and a small fraction of DOC. Thus similar to aging of PyC in soil, the residues become enriched in oxygen-containing functional groups such as carboxylic acid, esters, aldehydes and ketones, enhancing their water-solubility and initiating their transformation into humic fractions of SOM. Keller and Bacon (1998) determined that Cretaceous-age till kerogen in the vadose zone would be consumed in 10 000 y or less with oxygen exposure.

Results in the same range have been found for loss of BC in ocean sediments. Middelburg et al. (1999) estimated 64% BC degradation in marine sediments with 10–20 ky of oxygen exposure. By comparing radiocarbon ages of BC and concurrently deposited non-BC organic matter Masiello and Druffel (1998, 2003) estimated BC residence times of 2400 to 13 900 y with oxygen exposure. Recalcitrant substrates such as coal, shale, kerogen and BC in sediments can be oxidized in the order of 20 000 y, and similar results might be expected for PyC from forest fires, depending on structure and oxygen exposure.

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7 The PyC cycle in boreal forest regions

7.1 How much BC is produced in boreal regions?

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Kuhlbusch and Crutzen (1995) estimated annual release of 90–180 Tg C as CO₂-C from boreal and temperate fires. Using their BC/CO₂-C ratio of 5–7% for boreal and temperate forest fires gives a range of 4.6–12.6 Tg BC y⁻¹. Lynch et al. (2004) calculated fuel consumption and charcoal production for the boreal regions of Canada and Alaska for 1959–1995, based on 2% conversion of fuel mass to charcoal mass (Sect. 4.1.1). For Canada, fuel consumption was 46.86 Tg y⁻¹, resulting in 0.94 Tg charcoal annually. For Alaska, fuel consumption was 8.51 Tg y⁻¹, corresponding to 0.17 Tg charcoal. Assuming charcoal C concentration of 700 g C kg⁻¹ gives 0.66 Tg charcoal C for Canada and 0.12 Tg charcoal C for Alaska, totaling production of 0.78 Tg C y⁻¹ as charcoal particles that were airborne at some point during the fire.

Comparison is difficult because of the different methods and approaches. Kuhlbusch and Crutzen (1995) define BC as C remaining after pre-extraction and chemical oxidation. It would be expected that production of this oxidation-resistant BC fraction would be lower than conversion to C as visually-determined char. However, the 5–7% ratio of BC/CO₂-C corresponds to the upper range of char C yield (Table 1). On the other hand, the 2% mass conversion reported by Lynch et al. (2004) is probably low, as it is based on collection of charcoal particles in surface traps. It does not include char produced on CWD and within the forest floor, without becoming airborne.

Kasischke et al. (2005) recently modeled boreal fire emissions for 1992, and 1995–2003, including detailed consideration of flaming vs. smouldering combustion stages, and extent of forest floor consumption. For the whole high Northern Hemisphere (HNH), emissions were 162.9, 225.8 and 265.5 Tg C y⁻¹ for low-, moderate- and high-severity fire scenarios, respectively, based on depth of forest floor consumption. For Canada, the corresponding emissions were 35.3, 58.2 and 64.1 Tg C y⁻¹. Combining the emissions range from Kasischke et al. (2005), with a general estimate of 90%

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conversion of C to CO_2 and 5–7% BC/ CO_2 -C (Kuhlbusch and Crutzen, 1995), results in 7.3–16.7 Tg BC y^{-1} from fires in the HNH, and 1.6 to 4.0 Tg C y^{-1} for Canada. Using the more conservative 46.86 Tg estimate of annual boreal fuel consumption in Canada from Lynch et al. (2004), and assuming 500 g C kg $^{-1}$ in fuel, 90% conversion of C to CO_2 , and 5–7% BC/ CO_2 -C results in 1.2 to 1.6 Tg BC y^{-1} . This is in the low range predicted by Kasischke et al. (2005), but higher than the 0.66 Tg charcoal C calculated above, using 2% conversion of mass to charcoal. This is mainly due to higher consumption of forest floor in the work of Kasischke et al. (2005), especially by smouldering combustion.

Further refinements of these estimates are really beyond the scope of this review, and our expertise. These preliminary estimates, however, provide a starting point to consider the significance of BC as a component of the very stable soil C pool. Recently modeled estimates of soil C for north of 50 degrees (White et al., 2005) were 514 Pg C for forests and peatlands and 26 Pg C for grasslands, plus another 230 Pg C for tundra, totaling 870 Pg C. Bhatti et al. (2003) report 207 Pg C in boreal forest soil and detritus plus 415 Pg C for peat, totaling 622 Pg C. For Canada, the amounts were 65 Pg C in forest soil and detritus, and 113 Pg C in peat. Boreal production of the order of 7 to 17 Tg BC y^{-1} is therefore a small input to some global 600 Pg C in boreal forest, peatland and grassland soils. However, much of the soil C in boreal regions is in poorly-decomposed forest floor and peat, protected from rapid decomposition mainly by environmental conditions of cold, waterlogging or permafrost (e.g., Preston et al., 2006; Harden et al., 1997). Future predictions of warmer temperatures, lower precipitation, loss of permafrost, and increased fire could enhance soil and detrital C loss, and increase the relative importance of the BC sink. It would also be useful to consider annual boreal BC production in relation to detrital inputs, in particular, that proportion of detrital C transferred to more stable soil pools.

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7.2 How long does BC last in boreal soils?

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Field data on the fate of PyC hardly exist, but circumstantial evidence from other experiments designed for different purposes may help to elucidate the long-term fate of BC under field conditions. For BC that is not consumed in a subsequent fire, the previously-
5 noted studies of coal, shale, kerogen and BC in marine sediments suggest a half-life for BC perhaps up to 10 000 y under oxic conditions. For soils, there is little information, especially for boreal regions. As noted previously, Bird et al. (1999) postulated a half-life of <100 years for BC at 0–5 cm depth in a tropical savannah. By contrast,
10 soils of temperate rainforests in coastal British Columbia contain charcoal fragments dating back to deglaciation, approximately 13 000 y (Lertzman et al., 2002). For one site, Gavin et al. (2003) plotted charcoal mass vs. time since last fire determined by
15 ^{14}C dating of individual charred particles, and found that charcoal mass decreased exponentially with increasing time since the last fire. From their data we calculate an average half-life of 6623 years, assuming that all fires initially produced similar amounts of BC. This is similar to the time-scale for BC loss in sediments, and for geological substrates, and also to results for charcoal buried by landslides in interior British Columbia
20 (Sanborn et al., 2006). Soil charcoal radiocarbon ages ranged up to nearly 10 000 y BP, but only a few samples were older than 6000 y.

The rapid decomposition found in Bird et al. (1999) could be due to higher temperatures, good aeration and stronger photooxidation, and also because char produced
25 from savanna vegetation is finer than from wood. By contrast, in the British Columbia studies (Gavin et al., 2003; Sanborn et al., 2006), decomposition would be much slower due to colder temperatures, larger size of char fragments produced from wood, burial in soil and moss layers, and especially in coastal sites, wet conditions in winter. Even slower decomposition may be expected in colder boreal forests, especially for poorly-drained black spruce sites. Char is of course well-preserved in peat bogs, due to lack of oxygen, and has been used to reconstruct vegetation history through the Holocene.
We could not, however, locate any studies on the rate of BC loss in peat.

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7.3 Does charcoal influence boreal forest soil function?

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- In some boreal ecosystems, stands that have not experienced fire for many years develop thick forest floor derived from feathermoss (mainly *Pleurozium schreberi*); over time there is increasing cover of ericaceous plants, higher concentrations of forest floor phenolics and tannins, and lower N mineralization and nutrient availability, with N increasingly sequestered in poorly-available organic forms. Ericaceous species are better able to utilize organic N (Read et al., 2004; Treseder et al., 2004), and late-successional increase in N-fixation by feathermoss in association with cyanobacteria may mainly benefit ground vegetation rather than trees (Zackrisson et al., 2004).
- Decomposition is further impeded by lower growing-season temperatures in the thick feathermoss layer, and sometimes by excessive moisture, with development of impermeable layers or sphagnum growth and incipient peat profile development. Fire consumes the insulating moss layer, producing warmer surface soil temperatures, and increasing the depth of the active layer in permafrost-affected soils. The ash from forest floor and biomass provides nutrients and increases pH. Reduction of competition and allelopathy from ericaceous plants improves germination and growth of tree seedlings.

Nitrogen losses from fire are highly variable and soil N may even increase from inputs of canopy necromass (Dryness et al., 1989; Harden et al., 2004). As discussed earlier, fire generally reduces the biological availability of organic N. However, thermal ammonification of organic N, increased microbial activity after reduction of thick forest floor, and inputs of freshly killed litter may result in increased availability of NH_4^+ for 2–5 y (Choromanska and DeLuca, 2002). These general concepts of the role of fire in ecosystem function have been developed through many studies, notably of black spruce in North America (Richter et al., 2000), and Norway spruce (*Picea abies* (L.) Karst.) and Scots pine in Scandinavia (DeLuca et al., 2002; Zackrisson et al., 1996, 2004). A similar trajectory of ecosystem development and forest floor characteristics with time from fire was found for mixed forests of ponderosa pine (*Pinus ponderosa* Doug. Ex. laws) and Douglas-fir (*Pseudotsuga menszii* (Mirb.) Franco) in Montana

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(MacKenzie et al., 2004b).

In addition to direct effects of fire, such as changes in nutrient cycling, and temperature and depth of forest floor, Zackrisson et al. (1996) recently proposed that charcoal in boreal forests may directly influence microbial activity and nutrient cycling in soils. This mainly derives from its porous structure that can sorb organic molecules and provide microbial habitat (Harvey et al., 1976; Pietikäinen et al., 2000; Hamer et al., 2004), and from its contribution to cation exchange capacity (CEC). Charcoal produced from laboratory heating of twigs and stems adsorbed phenolics and reduced their phytotoxic effects on germination (Zackrisson et al., 1996; Keech et al., 2005), and increased growth of birch (*Betula pendula*) seedlings in pots (Wardle et al., 1998). For pitch pine (*Pinus rigida* Mill.), addition of commercial activated charcoal to leaf litter extracts did not affect germination, but improved seedling elongation (Garnett et al., 2004). Somewhat contradictory results were reported by Wallstedt et al. (2002) from a pot trial, in which activated charcoal reduced the concentration of water-soluble phenolics in soil, but also reduced seedling growth. Increased nitrification was found by DeLuca et al. (2002) and Berglund et al. (2004) with activated carbon amendments, although results for field studies were weaker than for laboratory incubations. This is likely because addition of activated charcoal is not a complete surrogate for the effects of fire and its properties are not identical to charcoal from wildfire (Hille and den Ouden, 2005).

Apart from these specific studies of boreal and temperate forest soils, charcoal amendment has been associated with general improvement and sustainability of soil fertility. Glaser and Amelung (2003) suggest that BC has contributed to the fertility of fire-prone native grassland sites in the North American Great Plains. The combination of long-lasting charcoal plus more quickly-decomposed plant residue is an effective soil amendment to promote long-term fertility in the tropics (Topoliantz et al., 2005), including the anthropogenic South American Terra Preta soils (Glaser et al., 2000, 2001, 2002).

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8 Boreal BC knowledge gaps

Figure 7 shows our initial view of the BC cycle, on a C basis. Fuel conversion to solid char in forest fires is probably in the range of 1–3% on a C basis, although higher values may be found where there are large stocks of coarse woody debris. Conversion to volatile soot is much lower, around 0.1–0.4% of fuel C, and deposition data are lacking. The solid char may be consumed in a subsequent fire, or slowly lost by photochemical, chemical and biological oxidation, whose rates and mechanisms are poorly understood. Partially degraded and modified BC structures are likely to contribute to DOC, and to C deposited in ocean sediments. In the absence of fire, char and BC persistence is affected by the same factors as other forms of SOM. First, its recalcitrance to oxidative degradation increases with increasingly condensed polycyclic aromatic structures with lower H/C and O/C ratios. Second, it appears that BC can become stabilized by association with mineral particles, and third, persistence is enhanced by cold and anoxic conditions, so that BC is well-preserved in peat bogs and sediments. We estimate a half-life of 5000–10 000 y under these conditions, but loss rates may be higher in warmer, drier, surface environments.

While analytical problems persist, and methodologies for BC are still under intense development and discussion, this should not be an impediment to acquiring some basic information on PyC in boreal regions, especially as required for C budget modelling.

Techniques for visual assessment of char are well-established, but there appear to be no studies with a complete accounting of char in any boreal forest site, and we found no quantitative information on BC storage in boreal peatlands. Interdisciplinary approaches are required, so that, for example, charcoal or BC measurements could be integrated into experimental fire studies such as the ICFME (Stocks et al., 2004), and the painstaking efforts to assess char abundance in peat cores could also be used to develop estimates of area-based char stocks. Char height has been used to predict tree mortality after fire (Hély et al., 2003), and there are assessments of stocks of downed coarse woody debris and snags in Canadian boreal regions (Lee, 1998; Greif

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and Archibald, 2000; Pedlar et al., 2002), but only one study (Manies et al., 2005) has linked fire-killed wood (but not wood-associated char) to soil C storage.

Analysis of BC by thermal or chemical oxidation resistance, BPCA production or photochemical oxidation plus NMR is expensive and time-consuming, and it is unlikely that any one method will prove suitable for all samples. Valuable information is coming from the BC comparative analysis ring trial, but these results need to be expanded to the types of char and BC produced in boreal fires. It would also be useful to develop more capacity for routine analysis of H concentration as well as C, as the H/C ratio is a good indicator of aromatic condensation. Radiocarbon dating of char fragments and soil fractions is essential to understanding the long-term sink potential of BC, including its contribution to marine sediments.

Efforts should be made to acquire more field data on production and stocks of visually-defined char in boreal regions, while analysing a subset of samples for BC by several methods. This approach should allow development of a two-component model for PyC, and a conversion factor to estimate what fraction of visually-determined char has the more recalcitrant structures that will contribute to a long-lasting BC sink. For mineral soils, char may be less visible and chemical methods essential to assess adequately the BC contribution to SOM. Information on the atmospheric component is also inadequate, and requires measurement and tracking of volatile C emission from fires, and monitoring of atmospheric BC concentrations and deposition rates.

Despite the impact of wildfire in boreal regions, there has been little study of stocks, production, characteristics or longevity of BC in boreal forests and even less for peatlands. We still lack much basic information required to assess the role of BC as a C store in boreal forest soils, or whether it substantively influences soil processes by sorption of phenolics and provision of microbial habitat. It seems equally urgent to quantify soot production, transport, and potential for atmospheric warming.

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Table 1. Production of charcoal and BC from forest biomass burning, on a mass^a or carbon^b basis. Based on 500 g C kg⁻¹ for biomass and 700 g C kg⁻¹ for char, conversion of 1% of biomass to char on a mass basis is equivalent to 1.4% conversion on a C basis. Conversely, 1% conversion on a C basis corresponds to 0.71% on a mass basis.

Region/site/cause of fire	Method	Char or BC production	Reference
(a) Boreal and Temperate Forests			
Bor Island, Siberia, experimental crown fire in Scots pine (<i>Pinus sylvestris</i>)	Visual char collected in surface traps	^a 729 kg ha ⁻¹ (2% fuel conversion) ^b 235 kg ha ⁻¹	Clark et al. (1998) Ohlson and Tryterud (2000)
Norway and Sweden, 3 boreal sites, mostly coniferous, experimental fires with partial crowning	Visual char collected in surface traps	^a 932 kg ha ⁻¹ (2.2% fuel conversion)	Lynch et al. (2004)
Northwest Territories, Canada, experimental crown fire (ICFME), jack pine (<i>Pinus banksiana</i>)	Visual char collected in surface traps	^a 7400 kg ha ⁻¹	Schiffman and Johnson (1989)
Virginia, USA after cutting and slash-burning loblolly pine (<i>Pinus taeda</i> L.)	Visual char in forest floor	^a 5.3%	Eckmeier et al. (2005)
Germany, to imitate Neolithic agricultural slash-and-burn in deciduous forest, one 8×12 m ² plot	Visual char >2 mm	^a 6400 kg ha ⁻¹ (8% CWD conversion)	Tinker and Knight (2000)
Yellowstone National Park, Wyoming, USA, after intense crowning wildfire	Visual char on CWD	^b 61 kg C ha ⁻¹ (0.7% forest floor C conversion)	Czimczik et al. (2003)
Siberia, Yenisey R., Scots pine, mainly surface fire	BPCA in forest floor		
(b) Tropical and Subtropical			
Amazon; Forest clearing; cut, dried 2–3 months, burned, as per local practice.	Visual char on soil or partially burned biomass	^b 3500 kg C ha ⁻¹ (2.5% fuel C conversion) ^b 1600 kg C ha ⁻¹ (1.3% fuel C conversion) ^b 3200 kg C ha ⁻¹ (1.8% fuel C conversion) ^b 6400 kg C ha ⁻¹ (2.9% fuel C conversion)	Fearnside et al. (1993) Fearnside et al. (1999) Fearnside et al. (2001) Graça et al. (1999)
Australia, slashburning after clearfelling eucalypt stands	Visual char >2 mm, to 30 cm depth	^b 20 000 and 170 000 kg C ha ⁻¹	Hopmans et al. (2005)

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Table 2. Soil BC or charcoal in soil for (a) non-forest and (b) forest sites. Unless otherwise stated, results given as percentage of total soil organic carbon (SOC). Abbreviations: NMR (nuclear magnetic resonance), OX (oxidation), BPCA (benzenepolycarboxylic acids).

Region/site	Method	BC or Char quantities	Reference
(a) Non-forest sites			
Australia, 8 surface soils	Photo OX+NMR of <53 µm	<1 to 30%	Skjemstad et al. (1999a)
Australia, 2 long-term agriculture sites, 0–30 cm	Photo OX+NMR of <53 µm	10 to 27% (4–15 Mg C ha ⁻¹)	Skjemstad et al. (2004)
USA, surface soils (4 cultivated, 1 prairie)	Photo OX+NMR of <53 µm	10–35%	Skjemstad et al. (2002)
Canada, Black Chernozem, aspen parkland native and cultivated	Photo OX	20–60% (whole soils), 10–80% (size fractions)	Ponomarenko and Anderson (2001)
North American great plains, 18 native grass-land sites, 0–10 cm	BPCA	4–18%, average 9% (1–15 Mg C ha ⁻¹)	Glaser and Amelung (2003)
Texas grassland fire study, 0–20 cm	BPCA	5–13%	Dai et al. (2005)
Switzerland, 23 monitoring sites including forests, 0–10 cm	Thermal OX	1–6%, median 2%	Bucheli et al. (2004)
Thailand, slash-and-burn agriculture	Chemical OX	3–7%	Rumpel et al. (2006)
China, Loess Plateau, very deep profiles	Chemical OX	0.41–5.7 g C kg ⁻¹ soil	Wang et al. (2005)
(b) Forest sites			
Forest floor/humus only			
Sweden, 12 lake islands, 1–350 y after fire	Visual Char	984–2074 kg ha ⁻¹	Zackrisson et al. (1996)
Canada, 6 stands, southern Quebec, 75 y after fire,	Visual Char >2 mm	343–1295 kg ha ⁻¹	Bélanger et al. (2004) and personal communication
Whole soil profile			
France, Alpine conifer forest	Visual Char >0.4 mm	0.1–20% (10–30 000 kg ha ⁻¹)	Carcailliet and Talon (2001)
Siberia, Yenisey R., 215-y <i>Pinus sylvestris</i>	Thermal OX	20–24% in forest floor +0–75 cm; 13% for 75–300 cm	Schulze et al. (1999)
Siberia, Yenisey R., <i>Pinus sylvestris</i> fire chronosequence study	BPCA	≤3.5% in organic layer (up to 720 kg C ha ⁻¹) 0.01–0.02% (0.16 to 0.25 g m ⁻²) for 0–100 cm mineral soil	Czimczik et al. (2005) More detail in Fig. 5
Australia, 3 eucalypt forest management sites, to 30 cm depth	Visual char >2 mm	15–32% (15 000–140 000 kg C ha ⁻¹)	Hopmans et al. (2005)
Corsica (France)		9800–148 000 kg ha ⁻¹	Carcailliet and Talon (2001)

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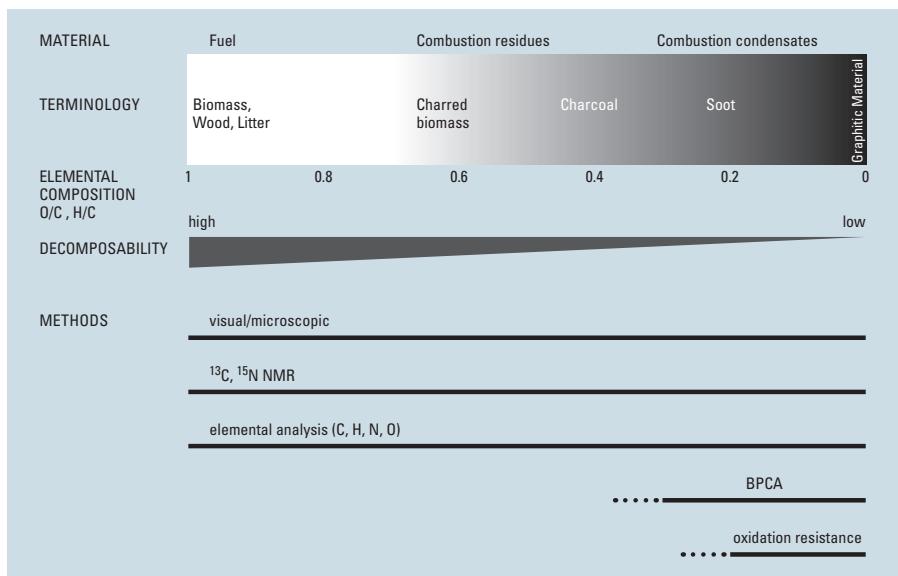


Fig. 1. Definitions and analytical windows for PyC.

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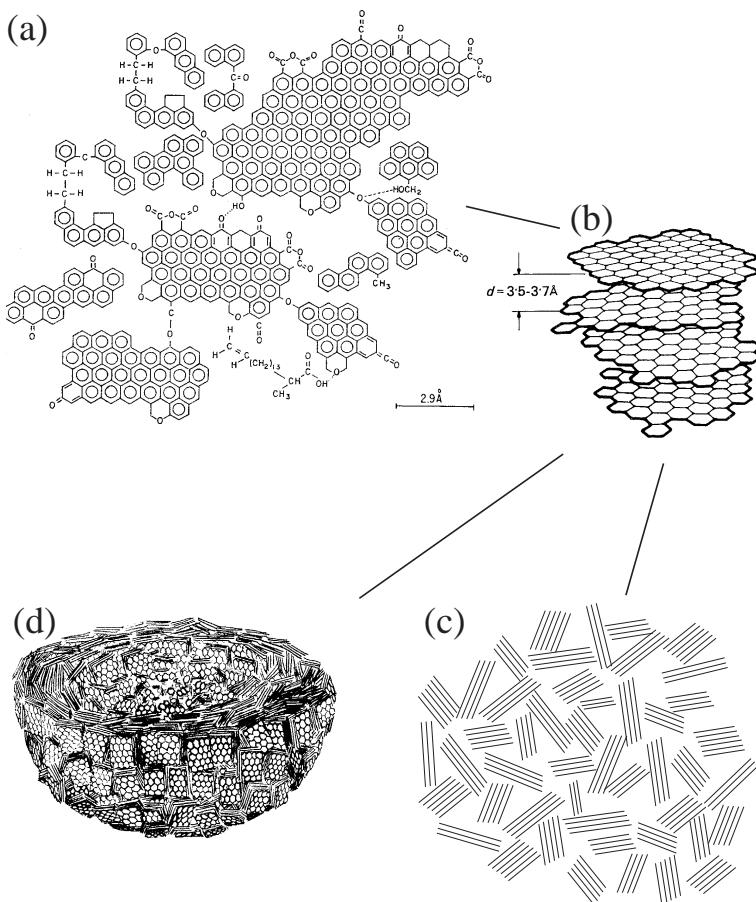
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Fig. 2. Black carbon and soot molecular structures, reproduced with permission from Schmidt and Noack (2000).

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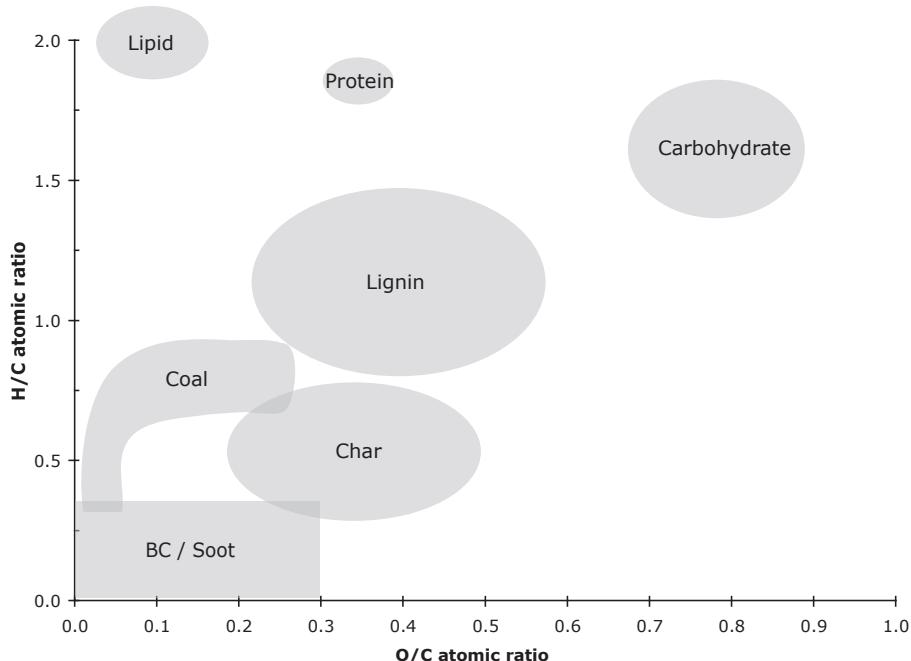


Fig. 3. van Krevelen plot of molar ratios for typical biopolymers, biomass and pyrogenic materials.

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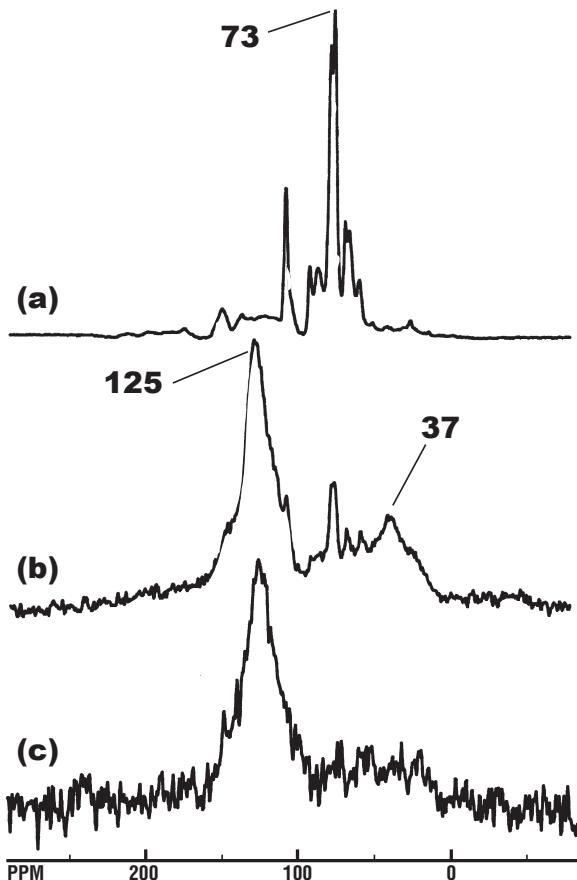


Fig. 4. Solid-state ^{13}C NMR spectra of Scots pine wood **(a)** fresh wood with standard CP acquisition and freshly charred outer layer with **(b)** CP and **(c)** quantitative BD acquisition. Sampling site and NMR methods described in Czimczik et al. (2003). Magic-angle spinning rate was 4700 Hz for fresh wood and 12 000 Hz for charred wood.

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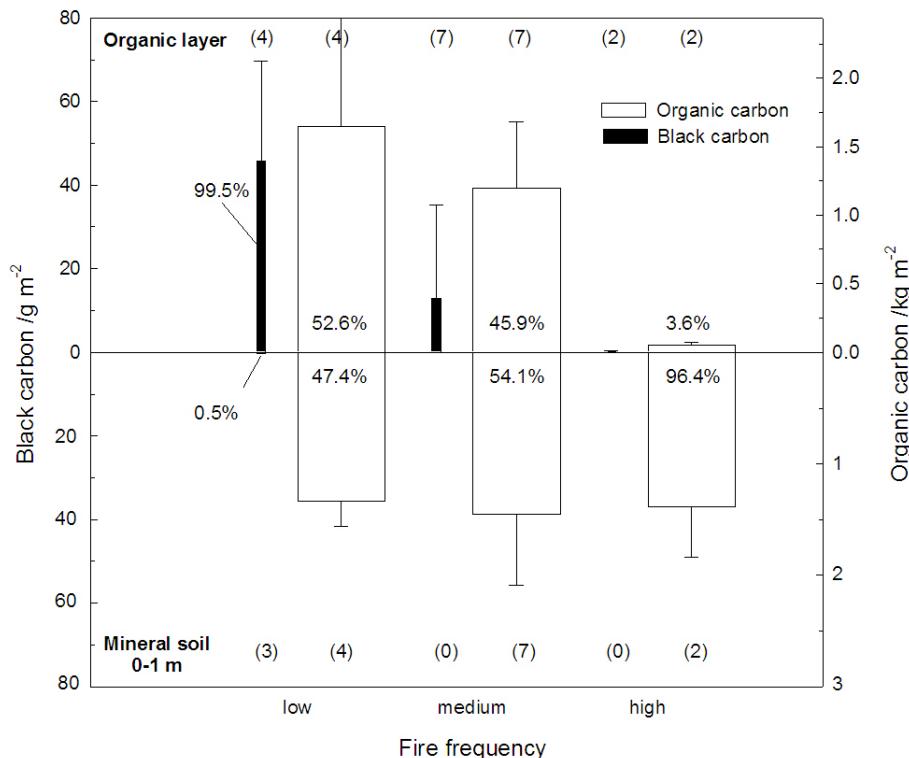


Fig. 5. Distribution of PyC above and below ground in a Siberian Scots pine site, reproduced as permitted from Czimczik et al. (2005).

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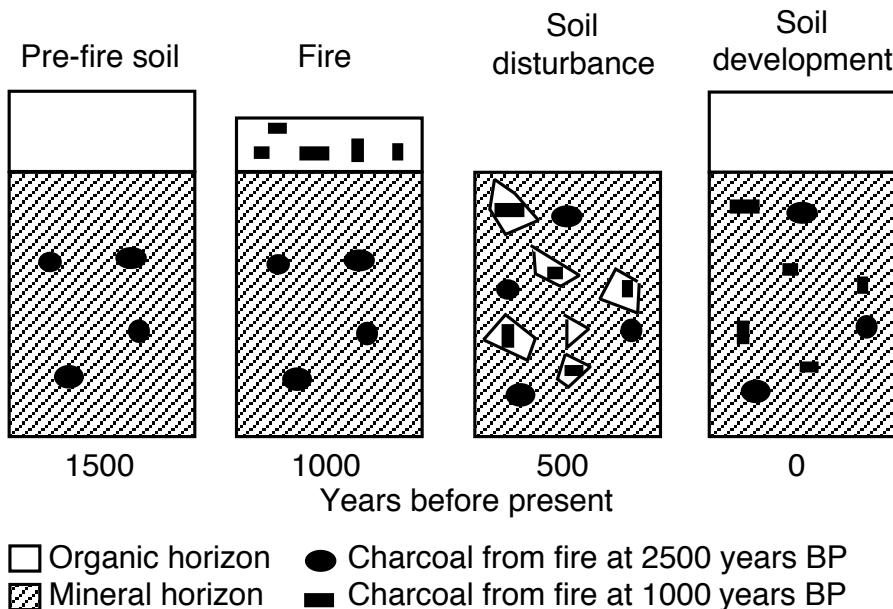


Fig. 6. Schematic showing how charcoal could be transported from organic to mineral horizons in Podzols (Vancouver Island, British Columbia). Charcoal from a fire 1000 years before present (BP) is deposited in the organic horizon and mixed into the mineral soil at 500 y BP. A new charcoal-free organic horizon has developed by 0 y BP. Reproduced with permission from Gavin (2003).

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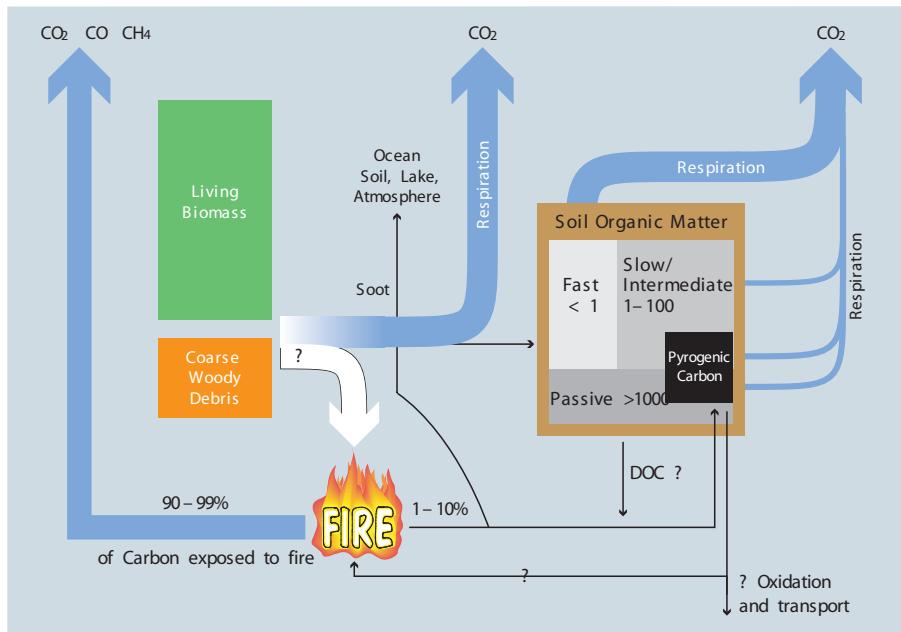


Fig. 7. A preliminary concept for pools and fluxes of PyC in boreal forests.

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