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Fossil charcoal in Late Devonian marine shales: An indicator of expanding terrestrial ecosystems, fire, and atmospheric change

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ABSTRACT

Fossil charcoal provides direct evidence for fire events that, in turn, have implications for the evolution of both terrestrial ecosystems and the atmosphere. Most of the charcoal record is known from terrestrial or near-shore environments and indicates the earliest occurrences of fire in the Late Silurian. However, despite the rise in available fuel through the Devonian as vascular land plants became larger and trees and forests evolved, charcoal occurrences are very sparse until the Early Mississippian where extensive charcoal suggests well established fire systems. We present data from the latest Devonian of North America from terrestrial and marine rocks indicating that fire became more widespread and significant at this time. This may be a function of rising O_2 levels and the occurrence of fire itself may have contributed to this rise through positive feedback. Recent atmospheric modelling suggests an O_2 low during the Middle Devonian (around 13%), with O_2 rising steadily through the Late Devonian and Early Carboniferous (Mississippian) (from 17-19%). In Devonian-Carboniferous marine black shales, fossil charcoal (inertinite) steadily increases up-section suggesting the rise of widespread fire systems. Scanning electron and reflectance microscopy of charcoal from Late Devonian sites indicate that the fires were moderately hot (around 550°C) and burnt mainly surface vegetation dominated by zygopterid ferns and lycopsids, rather than being produced by forest crown fires.

Keywords: Wildfire, charcoal, black shale, Devonian, Kentucky.

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INTRODUCTION

Fire plays a significant role not only in the regulation of plant biome distribution (Bond and Keely, 2005) but also in the regulation of atmospheric O_2 (Lenton and Watson, 2000; Berner et al., 2003). The origin of widespread fire systems is significant, therefore, in our understanding of the evolution of both terrestrial ecosystems and the atmosphere. Berner et al. (2003) suggested an important positive feedback loop in the relationship between fire and O_2: as O_2 rises, more fires occur, leading to increased charcoal production and burial, thus giving rise to further increases in O_2. Fossil charcoal (a pyrolysis residue of wildfire) provides evidence for the earliest fires in the latest Silurian (Glasspool et al., 2004), yet records of charcoal through the Devonian are rare (Rowe and Jones, 2000; Scott and Glasspool, 2006), possibly reflecting an insufficient fuel supply together with low levels of atmospheric O_2 (Scott and Glasspool, 2006).

The occurrence and spread of wildfire in recent and ancient vegetation has been discussed widely (Pyne et al., 1996; Scott, 2000; Scott and Glasspool, 2006). Under modern atmospheric O_2 conditions (Present Atmospheric Level (PAL) = 21%), a low plant-moisture content is necessary for fire to spread (Wildman et al., 2004) as is a sufficient fuel build-up and an ignition source (usually lightning) (Pyne et al., 1996). Atmospheric O_2 levels are crucial: below 13% fires will not ignite and spread (Chaloner, 1989) whereas at high O_2 levels (above 35%) plants may burn irrespective of fuel moisture, resulting in an upper limit for atmospheric O_2 concentration as above this level no fire could be extinguished (Watson et al., 1978; Lenton and Watson, 2000; Lenton, 2001). This has led to the concept of the fire window (Jones and Chaloner, 1991).

The Devonian saw a rapid rise and spread of vegetation into a wide range of habitats (Kenrick and Crane, 1997; Edwards and Wellman, 2001; Hotton et al., 2001). Plant size rose dramatically, resulting in increased fuel loads. By the Middle Devonian (Givetian) there is evidence for woody shrubs and small tree-sized plants (Algeo et al., 2001), and by the Mid Frasnian there were lowland forests dominated by the progymnosperm tree *Archaeopteris* (Meyer-Berthaud et al., 1999; Scheckler, 2001). However, only scattered terrestrial records of Late Devonian charcoal exist and it is not until the Carboniferous that extensive evidence of widespread fire systems is seen (Scott and Glasspool, 2006). Thus, there appears to be a significant ‘charcoal gap’ in the latest Middle Devonian and earliest Upper Devonian coinciding with a suggested O_2 low of 13% (Berner, in press; Scott and Glasspool, 2006). However, we demonstrate here that the marine fossil record may provide a more continuous record of Late Devonian-Early Carboniferous fire systems that may have played a significant role in the rise in atmospheric O_2 levels.

STUDY SITES AND METHODS

Samples were collected from the New Albany Shale (Famennian) and the Sunbury Shale (Tournaisian) from core D6 (east-central Kentucky) (see Rimmer et al., 2004). In addition, samples were obtained from two well-documented terrestrial environments, Red Hill (Famennian, Duncannon Member, Catskill Fm., PA) (Cressler, 2001, 2006) and Elkins (Famennian, Hampshire Fm., WV) (Gillespie et al., 1981; Scheckler, 1986). These terrestrial sites represent floodplain and deltaic deposits on the Catskill Delta, including some of the earliest know coal swamps (Hampshire Fm.) (Scheckler, 1986). They are time-correlative (Famennian 2c) (Cressler, 2001; Gillespie et al., 1981) with the upper parts of the section of the core from central Kentucky (Cleveland Shale Member of the New Albany Shale), and thus represent areas from which terrestrial OM likely originated during accumulation of the marine black shales.

For organic petrographic analysis, shale kerogen concentrates (HF-H_3BO_3 digestion) were embedded in epoxy resin and polished. A combined white-light, blue-light (450-490 nm excitation) quantitative point-count analysis (300 points) was performed on each kerogen concentrate ($n = 63$) using a Zeiss Universal microscope (640x magnification) (see Rimmer et al., 2004, for details of analytical techniques). As inertinite reflectance is indicative of formation temperature (Scott, 2000; Scott and Glasspool, in press), reflectance analyses (min. 100 readings) were performed on three terrestrial samples and a subset ($n = 6$) of the marine shale samples using a Zeiss Universal scope fitted with a photomultiplier tube; data collection was controlled by custom computer hardware and software. SEM analysis provided additional support for a charcoal origin of the inertinites; homogenization of cell walls and loss of the middle lamella (300-325°C) are indicative of charcoal (Scott, 2000).

RESULTS AND DISCUSSION

Variations in OM Type in the Devonian Black Shales

The Devonian marine black shales include marine liptinitic organic matter (OM) (bituminite and alginite), as well as terrestrially derived OM including vitrinite and inertinite. The base of the Huron contains as much as 95% marine liptinitic OM (Fig. 1), but inertinite rises to over 20% towards the top of the section at the expense of marine OM; vitrinite content generally remains fairly constant and low. This inertinite consists of small fragments (2-50 µm) that show discernable cell structure (Fig. 2) and is considered to be fossil charcoal (after Scott and Glasspool, in press). Transport of wildfire-derived inertinite into the marine basin may have been by both water and wind (Scott, 2000; Rimmer et al., 2004). Studies of modern wind-blown charcoal suggest aeolian transport of fragments ranging from 1-150 µm (Patterson, 1987; Clark, 1988), encompassing the range of inertinite fragments observed in this study. Aeolian transport of charcoal into the basin from the Catskill Delta would be consistent with paleogeography and prevailing winds (trade winds) during the Late Devonian (Ettensohn, 1985), and such long-distance transport of charcoal has been demonstrated in other marine basins (Smith et al., 1973; Herring, 1985). Charcoal also would have been introduced into the marine basin associated with increased runoff and sediment load following fire events.

Reflectance and SEM analysis of inertinites

Inertinite reflectances of 2% correspond to combustion temperatures ~ 400°C, 5% to temperatures of 600°C or higher, and 6% to 850°C or higher (Scott and Jones, 1994; Guo and Bustin, 1998; Scott, 2000). Regardless of the duration of heating, reflectances will not exceed 2% at temperatures < 400°C (Guo and Bustin, 1998; Scott and Glasspool, in press). Mean random reflectance provides a minimum temperature if the duration of charring is unknown (McParland et al., in press), thus the data given here are for minimum fire temperatures. Reflectance levels for inertinites from both the Red Hill (mean R_o = 4.4%; mode = 4.75%) and Elkins (mean R_o = 3.2%; mode = 3.25%) sites confirm that these materials are fossil charcoal and were produced at high temperatures, predominantly 575°C at Red Hill and 500°C at Elkins (Table 1). Note that burial maturation produced vitrinite reflectances of 1.5% and 1.1% at the Red Hill and Elkins sites, respectively. The absence of lower reflectance inertinite from the Red Hill site is probably due to the higher level of maturation.

Morphological evidence (from SEM and organic petrography) suggests that these materials are derived mostly from herbaceous and shrubby plants rather than from large pieces of wood from trees (Fig. 2), possibly from the fern *Rhacophyton* that has been identified as a major component at both sites (Scheckler, 1986; Cressler, 2001). In modern forests, ground fires smoulder at low temperatures, surface fires burn litter at temperatures <350°C, whereas intense crown fires may exceed 600°C (Scott and Jones, 1994; Scott, 2000). Possibly, the moderately high temperatures indicated for this herbaceous and shrub-derived charcoal arose because of excessive build-up of litter.

Within the marine black shales, inertinite reflectance levels also indicate a wildfire origin (Table 1). In all three units studied, reflectance distributions show a bimodal distribution, with a mode around 2% and a second one between 4-5%. Similar reflectance distributions were obtained for inertinites from three cores from the Illinois and Appalachian basins (Hawkins, 2006). Mean values (3.25-4.15%) correspond to temperatures around 500-560°C, comparable to those suggested for the terrestrial sites at Red Hill and Elkins (Table 1). Thus, reflectance data are consistent with a terrestrial wildfire origin for the inertinites seen in the marine black shales.

DISCUSSION

We know of no terrestrial charcoal records from the latest Frasnian or early Famennian (Fig. 3); this represents the 'charcoal gap' (Scott and Glasspool, 2006). Macroscopic charcoal records from both Europe and North America appear to be restricted to the latest Famennian (Fa2c). Most of the North American charcoal occurrences are not of the tree *Archaeopteris*/*Callixylon*, but from herbaceous plants or shrubs such as the zygopterid fern *Rhacophyton*. Similarly, studies of Famennian sediments in Europe have indicated charcoal assemblages dominated by a range of herbaceous plants, with *Callixylon* wood being rare (Fairon-Demaret and Hartkopf-Fröder, 2004), a pattern that appears to continue through the Mississippian (Scott and Glasspool, 2006). This suggests extensive surface fires. If there were extensive forests then at least some crown fires may be expected, yet there are no confirmed occurrences of *Archeopteris* leaf charcoal. If surface fires spread through the forest, then fallen branches and leaves in the litter would have been burnt. Possibly forest litter remained moist but open 'scrub-like' vegetation dried out occasionally and burnt after a significant fuel build-up.

The rise in fire activity, as indicated by the black shale inertinite record, appears to have been gradual but sustained, and fits well with the suggested rise in atmospheric O_2 at this time (Berner, in press; Scott and Glasspool, 2006). The occurrence of charcoal in marine black shales may not only be significant in the Devonian but also during other periods in Earth history such as the Triassic where there are widespread black shales above the Permian-Triassic boundary (Wignall and Twitchett, 2002). It has been suggested that O_2 levels were around 24% (Berner, in press), a level at which plant material with relatively high moisture contents should burn, but there are no terrestrial records of charcoal at this time. Using the microscopic marine charcoal record may offer a window on to the nature of fire systems at this time and contribute towards our understanding of terrestrial ecosystem recovery.

CONCLUSIONS

Despite the occurrence of scattered records of charcoal in the Late Silurian and Early Devonian, evidence of fire through most of the Devonian is rare despite the diversification of land vegetation and the potential build-up of fuel. There is a significant ‘charcoal gap’ in the latest Middle Devonian and earliest Late Devonian, followed by increasing evidence of fire in the latest Famennian (Fa2c). Scanning electron microscopy of macroscopic charcoals from the Late Famennian indicates that herbaceous plants and shrubs were being burnt and, thus suggests the occurrence of surface fires, rather than forest crown fires, a pattern that was maintained at least through the Early to Middle Mississippian. The marine record of inertinite (fossil charcoal) in black shales of Kentucky, which cross the Devonian-Carboniferous boundary, indicates increasing fire activity though this time. Reflectance data on both terrestrial and marine charcoal indicates minimum fire temperatures of 500-550°C. Atmospheric modelling suggests a rise in O_2 levels from 13-19% though the Late Devonian and the charcoal record is consistent with this prediction.

The occurrence of charcoal in marine black shales is significant in yielding a more continuous record of fire than is possible in many terrestrial systems. It is probable that petrographic study of black shales from other ages may provide a similar wildfire record.

ACKNOWLEDGMENTS

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Figure Captions

Figure 1. Stratigraphic variability in organic petrographic composition for core D6, central Kentucky (vol. %, mineral-free basis) showing vitrinite (woody tissue), inertinite (charcoal), and alginite plus bituminite (marine OM) (data from Rimmer et al., 2004).

Figure 2. Photomicrographs of inertinite in (a) Red Hill (reflected-light microscopy); (b) Red Hill (SEM); (c) Elkins siltstone (reflected-light microscopy); and (d) Cleveland Shale (reflected-light microscopy).

Figure 3. Atmospheric O_2 levels from the Silurian through the Mississippian), based on Berner, in press (solid circles, margin of error), Berner et al., 2003 (open circles), and known occurrences of fossil charcoal. * = present study, Red Hill site, Catskill Fm., PA, USA; Elkins site, Hampshire Fm., WV, USA; New Albany Shale and Sunbury Shale, KY, USA. (1) Platyschima Shale Member, Dowton Fm., Ludlow, England (Glasspool et al., 2004); (2) Ditton Fm., Shropshire, England (Edwards and Axe, 2004); (3) Hecla Bay

Fm., Melville Island, Canada (Goodarzi and Goodbody, 1990); (4) Red Hill site, Duncannon Fm., PA, USA (Cressler, 2001); (5) Ardennes-Rhenish Massif, Germany (Fairon-Demaret and Hartkopf-Fröder, 2004), (6) Horton Grp., Nova Scotia, Canada (Falcon-Long, 2000); (7) Upper Shalwy Beds, Donegal, Ireland (Nichols and Jones, 1992; Scott and Jones, 1994); (8) Moyny Limestone, Creevagh Head, County Mayo, Ireland (Falcon-Long, 1998); (9) Strathclyde Group, Scotland (Falcon-Long, 2000); also see Scott and Glasspool (2006) for additional Mississippian occurrences).

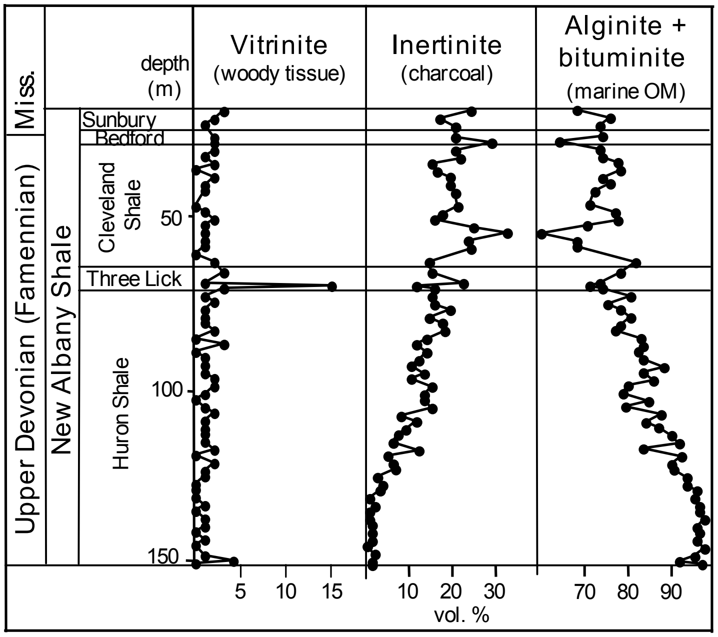

Miss.
Upper Devonian (Famennian)
New Albany Shale
depth (m)
Sunbury
Bedford
Cleveland Shale
Three Lick
Huron Shale
50
100
150
Vitrinite (woody tissue)
Inertinite (charcoal)
Alginite + bituminite (marine OM)
5
10
15
10
20
30
70
80
90
vol. %

Figure 1

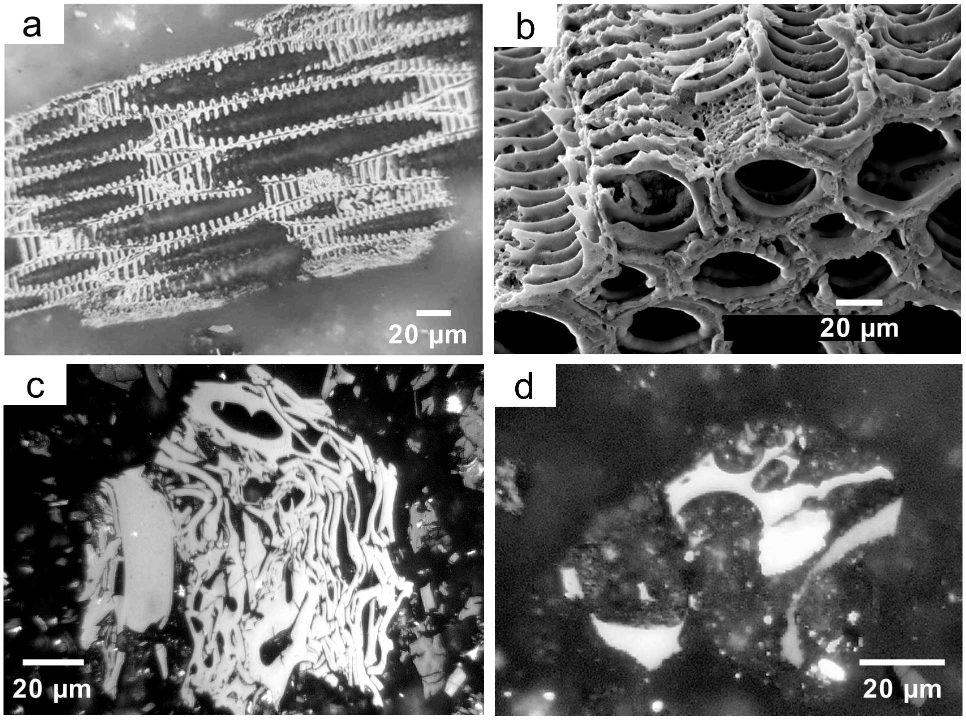
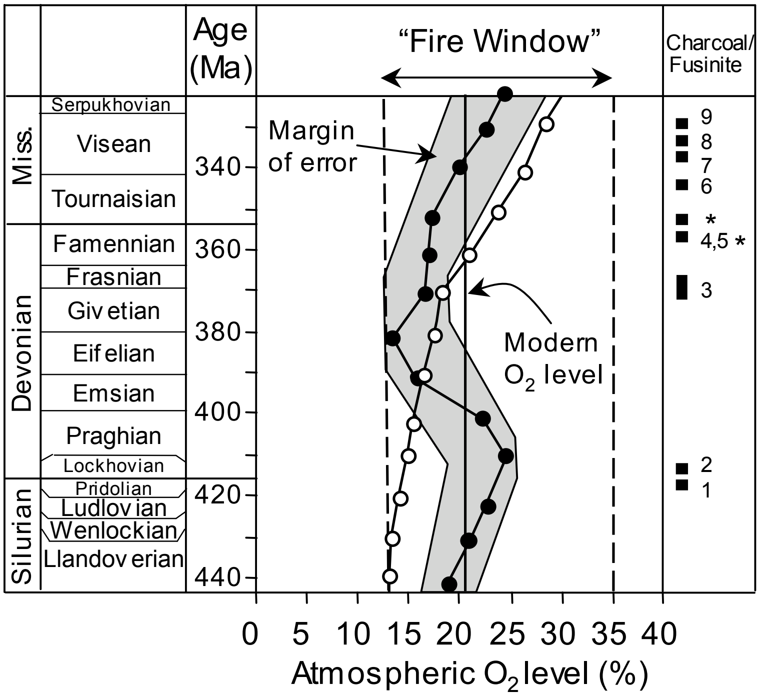


Figure 2



Age (Ma)
"Fire Window"
Charcoal/ Fusinite
Miss.
Devonian
Silurian
Serpukhovian
Visean
Tournaisian
Famennian
Frasnian
Givetian
Eifelian
Emsian
Praghian
Lockhovian
Pridolian
Ludlovian
Wenlockian
Llandoverian
340
360
380
400
420
440
Margin of error
Modern O$_2$ level
9
8
7
6
*
4,5 *
3
2
1
0 5 10 15 20 25 30 35 40
Atmospheric O$_2$ level (%)

Figure 3

Table 1. Inertinite reflectance (R_o) data for terrestrial and marine samples. Calculated minimum fire temperatures based on means using data for *Sequoia* in Scott and Glasspool, in press).

Sample	Unit	Age*	Inertinite R_o (%)			Calc. Min. Temp °C
			Min	Mean	Max	
Terrestrial samples:						
Red Hill siltstone	Catskill Fm.	Fam	2.32	4.40	5.51	575
Elkins coal	Hampshire Fm.	Fam	1.48	3.14	5.06	500
Elkins siltstone	Hampshire Fm.	Fam	1.05	3.19	4.48	500
Marine samples:						
DN6-2	Sunbury Sh.	Tour	1.39	3.95	5.62	550
DN6-4	Sunbury Sh.	Tour	1.44	3.99	6.27	550
DN6-9	Cleveland Sh.	Fam	1.47	3.44	5.51	525
DN6-16	Cleveland Sh.	Fam	0.98	3.26	5.37	500
DN6-22	Cleveland Sh.	Fam	1.25	4.15	6.11	550
D6R-7	Huron Sh.	Fam	1.33	4.11	5.64	550

* Fam = Famennian; Tour = Tournaisian