



## Review article

## On the fundamental difference between coal rank and coal type



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## ABSTRACT

This article addresses the fundamental difference between coal rank and coal type. While theoretically settled long ago as being different aspects of coal systems science, the two concepts are still often confounded. In recent years, this has resulted in the publication of several works stating that coal type changes with coal rank. Coal type refers solely to coals' depositional origin and the maceral–mineral admixture resulting from that origin. Coal types typically fall in to two categories: humic coals, developed from peat, and sapropelic coals, developed from organic mud. Either type may be allocthonous or autochthonous, and within types, further refinement of depositional environment can be made. Coal rank refers to the changes in geochemistry and resultant changes in reflectance caused by increasing thermal maturity of the coal. Thus, it provides an overprint of maturity on existing coal types. With proper techniques, such as use of crossed polars and etching, maceral forms can be differentiated even at high ranks, and the original coal type determined.

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## Contents

1.	Introduction . . . . .	59
1.1.	What is coal? . . . . .	59
1.2.	Coal forming environments: humic and sapropelic coals . . . . .	59
1.2.1.	Humic coals . . . . .	60
1.2.2.	Sapropelic coals . . . . .	60
1.2.3.	Co-occurrence of sapropelic and humic coals and the fate of liptinite macerals at high ranks . . . . .	60
1.3.	Coal diagenesis and metamorphism . . . . .	61
1.4.	Aims of the present work . . . . .	63
2.	Environments of peat formation . . . . .	63
2.1.	Environments of autochthonous peat formation . . . . .	63
2.1.1.	Controls on the locus of peat formation . . . . .	63
2.1.2.	Organic production . . . . .	63
2.1.3.	Sediment restriction . . . . .	64
2.1.4.	Pennsylvanian coal and sequence stratigraphy . . . . .	65

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2.2.	Macerals forming in the mire . . . . .	65
2.2.1.	Huminite/vitrinite . . . . .	65
2.2.2.	Pseudovitrinite . . . . .	66
2.2.3.	Inertinite . . . . .	68
2.2.4.	Liptinite macerals . . . . .	73
3.	Coal rank . . . . .	74
3.1.	Mechanisms of coal rank . . . . .	74
3.2.	General physical changes to coal with rank . . . . .	74
3.3.	Maceral changes with rank . . . . .	74
4.	Organic geochemical changes with rank . . . . .	75
5.	The impact of type and rank on coal as a source rock and reservoir rock . . . . .	78
6.	Conclusions . . . . .	79
	References . . . . .	79

## 1. Introduction

### 1.1. What is coal?

Coal is a complex combustible sedimentary rock, composed largely, but not exclusively, of helophytic ( $\pm$  aquatic) plant debris and plant derivatives. Originally deposited primarily as peat, secondarily as mud, to be discussed in more detail below, it transitions to coal through physical and chemical processes brought about by compaction and heat with prolonged burial at depths of up to several kilometers and over periods of up to several hundred million years. Thermal metamorphism from igneous intrusions, while important in some settings, will not be discussed in detail here.

At the most basic level, coal properties are a function of three fundamental parameters of coal composition, each of which is determined by some aspect of the coal's origin and evolution (after Diessel, 1992; Suárez-Ruiz et al., 2012; Taylor et al., 1998; Ward, 1984; among others):

- *organic petrological/geochemical*, including the nature of the organic constituents (macerals), but also with consideration of the organic geochemistry of the macerals and non-maceral organic compounds incorporated in the coal structure;
- *inorganic petrological/geochemical*, including the minerals (crystalline and amorphous inorganic components) and any inorganic entities associated with the organic structure of the maceral components; and
- *coal rank*, the extent of diagenetic/metamorphic transformation in the macerals and minerals, reflecting the maximum temperature to which the coal has been exposed and the time it was held at that temperature and, to a lesser degree, the pressure regime through the latter time and temperature. For most coals, this indirectly reflects the depth of burial and geothermal gradient prevailing at the time of coalification, although heat from igneous intrusions and hydrothermal fluids can also be an influence. The expression as parameters such as huminite/vitrinite reflectance and geochemistry is a function of the irreversible chemical changes, such as increased aromatization, undergone during metamorphism. With a few notable exceptions, to be discussed, while macerals will undergo changes in composition with metamorphism, they do not transform into different macerals. *Provenance prevails through the rank series.*

Ideally, each of the fundamental components is independent of the other two. In reality, this is not the case. Organic and inorganic compositions are, at the onset of coalification, inextricably linked as they are both the product of the environment of peat accumulation. As coalification progresses, both the nature of the macerals and the minerals may vary with coal rank. Maceral chemistry can also influence coal rank; for example, perhydrous vitrinite will have a lower reflectance than 'normal' vitrinite of the same rank (Gurba and Ward, 1998; Hutton and Cook, 1980; Kalkreuth, 1982; Li et al., 2010; Petersen and Vosgerau, 1999). So, while coal petrology can be expressed by a number

of fundamental parameters, each largely independent of each other, we need to be cognizant of the inter-relationships among the parameters.

*Coal type* and *grade* are related but more genetic concepts. Type reflects the nature of the plant debris from which the original organic matter was derived, including the mixture of plant and non-plant components involved (wood, leaves, algae, fungi, etc.). Coal type reflects the depositional environments at the time of peat accumulation, and the amount of biogeochemical degradation experienced by organic components prior to burial. Coal type is expressed as the maceral composition of the coal and is independent of coal rank. Humic and sapropelic coal types, to be discussed in this paper, are the fundamental end members of coal type. Within the humic coals there is also a range from bright (vitrinite-rich) to dull (liptinite- and inertinite-rich) materials (lithotypes). Cady (1942), building on discussions of White and Thiessen (1913), Stopes (1919, 1935), Thiessen (1920a,b,c, 1921, 1926, 1930), Hickling (1932), Thiessen and Sprunk (1936), and Sprunk et al. (1940), defined the term 'phyteral' to refer to the fossil plants in coals. Timofeev et al. (1962) and Timofeev and Bogoliubova (1964) developed a similar approach. Phyterals are composed of macerals, but are themselves useful descriptors of coal type in that they can potentially be recognized throughout the rank series, even in cases where the constituent macerals may become difficult to differentiate, such as in anthracite and meta-anthracite.

Grade reflects the extent to which the accumulation of plant debris has been kept free of contamination by inorganic material (mineral matter), including the periods before burial (i.e., during peat accumulation; syngenetically), after burial (epigenetically), and during rank advance. Regardless of its type or rank, a high-grade coal has a low overall proportion of mineral matter, and, hence, a high organic-matter content.

### 1.2. Coal forming environments: humic and sapropelic coals

The distinction between humic and sapropelic coals dates at least to Potonié (1893) and has been well established in coal geology since that time. Both coal types are functions of the depositional environments and consequently of the botanical constituents that form coal deposits.

Confusion between the two major coal types is not new. Potonié (1908) temporarily confused matters, placing all anthracite at the high-rank end of the gytja-sapropel type series, rather than also at the high-rank end of the peat-lignite-bright coal series. While this mistake was rectified in later works (e.g., Potonié, 1912), the misinterpretation has persisted. More recently, workers such as Sen (1999), Sahay (2006, 2008, 2010a,b), and Jones (2009) have confounded the inter-relationships between humic vs. sapropelic coals and coal rank vs. coal type. Sen (1999), building on 30 years of observations in coal geology, points out that Indian scientists consider Gondwanan coals in the Indian subcontinent to be allochthonous, and argues persuasively that non-bright bands are sapropelic, while bright bands represent floated and concentrated log deposits. However, while non-bright

bands may derive from sapropelic material, not all non-bright bands represent sapropelic material; materials such as bright clarain (detrovitrinite/detrohuminite), clarain (mixed macerals), or fusain (inertinite) may also produce non-bright bands. Sahay (2006, 2008, 2010a) in his study of Cenozoic lignite from the Panandrho Mine, Kutch Basin, western India, not fully appreciating the rank-related changes in coal macerals, much less the complex botanical origins of macerals, stated that coals are universally sapropelic until the onset of medium volatile bituminous rank. O'Keefe et al. (2008) and Hower et al. (2010b) addressed this, and other, issues raised by Sahay (2006, 2008, 2010a,b) in significant detail.

### 1.2.1. Humic coals

Humic coals are those that developed from largely woody and/or reed/sedge remains through the processes of peatification, primarily those that occur in the relatively moist aerobic zone of a mire (i.e., humification). Thus, they typically form primarily from wood or reed/sedge-rich peat and secondarily from organic mud in thin layers. Macroscopically, humic coals may be vitrains or bright clarains, or banded, with vitrain layers alternating with other lithotypes. By microlithotype, they consist of vitrite, clarite, vitrinertite, duroclarite, or inertite. In terms of maceral percentage composition, vitrinite or inertinite (especially fusinite and semifusinite) predominate. Humic coals tend to be dark brown to black and fracture along both cleat and banding planes.

### 1.2.2. Sapropelic coals

Sapropelic coals, on the other hand, developed from a largely non-woody source (algae, spores, pollen, cuticles, other leaf waxes, and macerated herbaceous plant remains) through decay in stagnant or standing water (i.e., putrefaction), primarily in the anaerobic zone. Macroscopically, they are dull clarains and durains, with a tendency to be matte black and fracture conchoidally, rather than along cleats or bands. In terms of microlithotypes, sapropelic coals are liptite, vitrinertoliptite and durite. In terms of maceral composition, they are dominated by liptinite, especially alginite (in the case of boghead coal or torbanite) or sporinite (in the case of cannel coal). In the earliest stages of coalification, the precursors to sapropelic coals are organic muds (gyttja in periglacial environments) or organic-rich oozes that form in primarily freshwater environments.

Sapropelic coals, in general, have distinctly different origins than humic coals. Extreme degradation of humic material can result in a lithology, on the macroscopic level, which is essentially indistinguishable from a sapropelic coal. Microscopically, they are fundamentally different. As discussed by Hutton and Hower (1999), following discussions by Moore (1968) and Taylor et al. (1998), sapropelic coals have two end members:

- Cannel coals with fine humic and liptinite macerals, mainly sporinite, but containing minor alginite; and
- Bogheads or torbanites,<sup>1</sup> which are rich in alginite (although Jeffrey, 1910, argued against an algal origin).

Based on the relative abundances of macerals in the coals, Hutton and Hower (1999) further divided sapropelic coals into the following assemblages:

- Sporinite with abundant medium-grained vitrinite and inertinite;
- Sporinite with fine-grained vitrinite and inertinite (detrovitrinite and inertodetrinite) and abundant micrinite (the Cannel City coal, Morgan County, Kentucky, falls in this category);
- Telalginite with abundant fine-grained vitrinite and inertinite; and
- Bituminite (or lamalginite) and telalginite with minor vitrinite and inertinite (see Fig. 1).

The latter category includes the Pennsylvanian Scottish torbanites and the Breckenridge coal, Western Kentucky (Hower et al., 1987); the Cretaceous King cannel, Utah (Given et al., 1984, 1985); the Permian Joadja torbanite, New South Wales (Hutton and Cook, 1980); and, by extrapolation from its retorting properties, the Pennsylvanian Bell County, Kentucky, cannel described by Ashley (1918).

Han et al. (1999) identified seven categories of sapropelic coals:

- (1) torbanite and (2) boghead, which by definition, are synonymous;
- (3) cannel boghead (Puxiang, China);
- (4) boghead cannel (Breckenridge, Kentucky, which petrographically belongs with the torbanite/boghead category);
- (5) cannel (including many eastern US cannels);
- (6) spore cannel (Melville Island, Canada); and
- (7) canneloid (Indiana paper coal and Texas Eocene cannels).

This classification, while overlapping the Hutton and Hower (1999) classification, differentiates sapropelic coals that Hutton and Hower (1999) did not (see Fig. 1). While both classification schemes were developed for terrestrial deposits, they expand the Hutton (1987) classification for source rocks (see Figures 1 & 2 in Hutton, 1987), providing a continuum from humic coals to oil shales.

The origin of torbanite has been considered to be analogous to modern coorongite, balkhashite, and n'hangellite (Boodle, 1907; Cane, 1976; Thiessen, 1925a,b). Coorongite, from southeastern Australia, is largely derived from the Chlorophyceae *Botryococcus* (telalginite maceral) (David, 1889; Thiessen, 1925a,b; Zalesky, 1914, 1926), with evidence of bacterial alteration (Glikson, 1984). Hutton and Hower (1999), however, disputed the validity of coorongite as a torbanite precursor because the Coorong deposits are highly ephemeral, the last period of algal growth (at the time of their paper) having been in 1962 (and, based on Thiessen (1925b), the previous deposit dated to 1865). In addition, unlike the Coorong deposits, most torbanites are associated with humic coals, rather than siliciclastic sediments.

Another potential modern analog to boghead coals is found in Mud Lake, Florida. The organic matter, devoid of humic contributions, consists of non-decaying blue-green algae which, in part, is eaten and excreted by chironomid larvae, cladocera, ostracods, and copepods (Bradley, 1966, 1970, 1973; Bradley and Beard, 1969). About 20% of the algae are excreted in a viable state (Bradley, 1966). Fecal pellets with blue-green algae growing both within and between the pellets, all suspended with no exact interface between the water and the ooze, comprise the substrate of the lake's organic sediments. At about 0.3-m depth within the ooze, the algae undergo a change in metabolism, producing oils, fats, and pigments instead of the proteins, carbohydrates, and minor lipids produced by the younger, shallower algae. Slow anaerobic decomposition takes place below 0.3 m, and 'copropellic structure persists and the cell walls of the blue-green algae remain sharply defined, but the ooze has a more gelatinous consistency' (Bradley, 1966). Lamalginite, the finer, less-structured algal material in torbanites, may be derived from Cyanophyceae (Hutton et al., 1980), with the mix eloquently described by Alpern (1980) as *gels algaires et bacteriens* (algal and bacterial gels).

### 1.2.3. Co-occurrence of sapropelic and humic coals and the fate of liptinite macerals at high ranks

At low ranks, humic and sapropelic coals are readily distinguished by their maceral composition and chemistry. In general, humic coals are vitrinite- and inertinite-rich, while sapropelic coals are liptinite-rich. This liptinite-enrichment makes sapropelic coals much higher in hydrogen than humic coals, and prone to elute hydrocarbons when under even moderate pressure. For example, the sapropelic portions of the Eocene Carlisle lignite from Western Kentucky will begin to elute hydrocarbons under low vacuum (O'Keefe, personal observation), while the humic portions do not. At higher ranks (low volatile bituminous and above), differentiating the two types can be difficult, as the lipids that constitute most sapropelic coals break down during coalification

<sup>1</sup> Named after the coal mined at the Boghead Estate, Torbane Hill, Bathgate, Scotland.

Type	Sub-type	Source material(s)	Depositional process	Transformation process	Sediment	Dominant Macerals	Example		
Humic Coal		Helophytic (woody and herbaceous) plant remains, locally minor aquatic/subaquatic components	sedentary	peatification	peat	Huminite/Vitrinite and/or Inertinite	Fire Clay Coal (Hazard No. 4), Kentucky (Greb et al. 1999)		
							Carlisle Lignite Deposit, Kentucky, USA (O'Keefe, 2008)		
							Pitt Meadows Bog, Fraser River Delta, British Columbia, Canada (Styan and Bustin, 1983)		
Sapropelic Coal	Canneloid	Aquatic/subaquatic plant remains with	sedimentary	putrefaction	organic mud (gyttja)	Cutinite and Sporinite	Indiana Paper Coal, Indiana, USA (Han et al., 1999)		
	Spore Cannel						spores/pollen and minor woody material	Sporinite and Vitrinite	Melville Island, Canada (Han et al., 1999)
	Cannel						spores/pollen	Sporinite	Cannel City Coal, Kentucky, USA (Han et al., 1999)
	Cannel Boghead						algae and decay products of algae	Near-equal amounts of alginite and bituminite	Puxiang County, Shanxi Province, China (Han et al., 1999)
	Boghead Cannel						algae and minor woody material	Alginite and Vitrinite	Breckenridge, Kentucky, USA (Han et al., 1999)
	Boghead/Torbanite						algae with rare decay products of algae	Alginite, sometimes Bituminite	Torban Hill, Scotland, UK (Han et al., 1999)

Fig. 1. Examples of how humic and sapropelic coals differ in terms of origin and petrography. Classification is after Hutton (1987), Hutton and Hower (1999), and Han et al. (1999).

to produce mobile hydrocarbons, leaving behind materials that greatly resemble either vitrinite or inertinite in reflected light. Etching may allow remnant liptinite macerals to be more readily identified.

Boghead coals, however, can be found at any coal rank. There are abundant references to cannel and boghead coals occurring in the same coalbed as humic coals (Ashley, 1918). Warwick and Hook (1995) noted subbituminous gelinite- and alginite-rich cannel lithologies associated with humic coals in the Eocene formations of Webb County, Texas, and adjacent areas in Mexico. Hook and Hower (1988) described the vertebrate-bearing cannel within the humic Upper Freeport coal in northeastern Ohio. Ashley (1918) devoted much of his report to the abundant Pennsylvanian cannel and boghead coals of West Virginia and Kentucky (see also Hower, 1995). In Kentucky, Pennsylvanian sapropelic coals occur in the same coalbed as high volatile A bituminous humic coals in the Skyline (Breathitt County) and Leatherwood (Perry County) coals, in the Mudseam coalbed (Elliott County), and the Upper Hance rider coalbed (Bell County) (Esterle, 1984; Gavett, 1984; Hutton and Hower, 1999). The latter cannel, a facies of the coalbed, is laterally continuous with bright, banded humic coal (Esterle, 1984). Pareek (2004) noted cannel coals within the Permian Gondwana coal measures of India and Sullivan (1959) found cannels in association with humic Pennsylvanian bituminous coals in Great Britain.

An excellent example of the occurrence of sapropelic coal at high rank is from southern China. These coals, often referred to as “stone-line coals,” are high-ash, anthracite-rank boghead coals, which occur in beds that range from 5 to 30-m thick (Yuan, 1999). These bogheads occur mainly in the early Paleozoic (including the Cambrian, Ordovician, and Silurian; Coal Industry Administration Bureau of Zhejiang Province, 1980; Fan and Yang, 1981; Li et al., 1982; Yuan, 1999). In them, the  $R_{max}$  of the homogeneous sapropelic groundmass is 3.75–8.40% (Zhu, 1983).

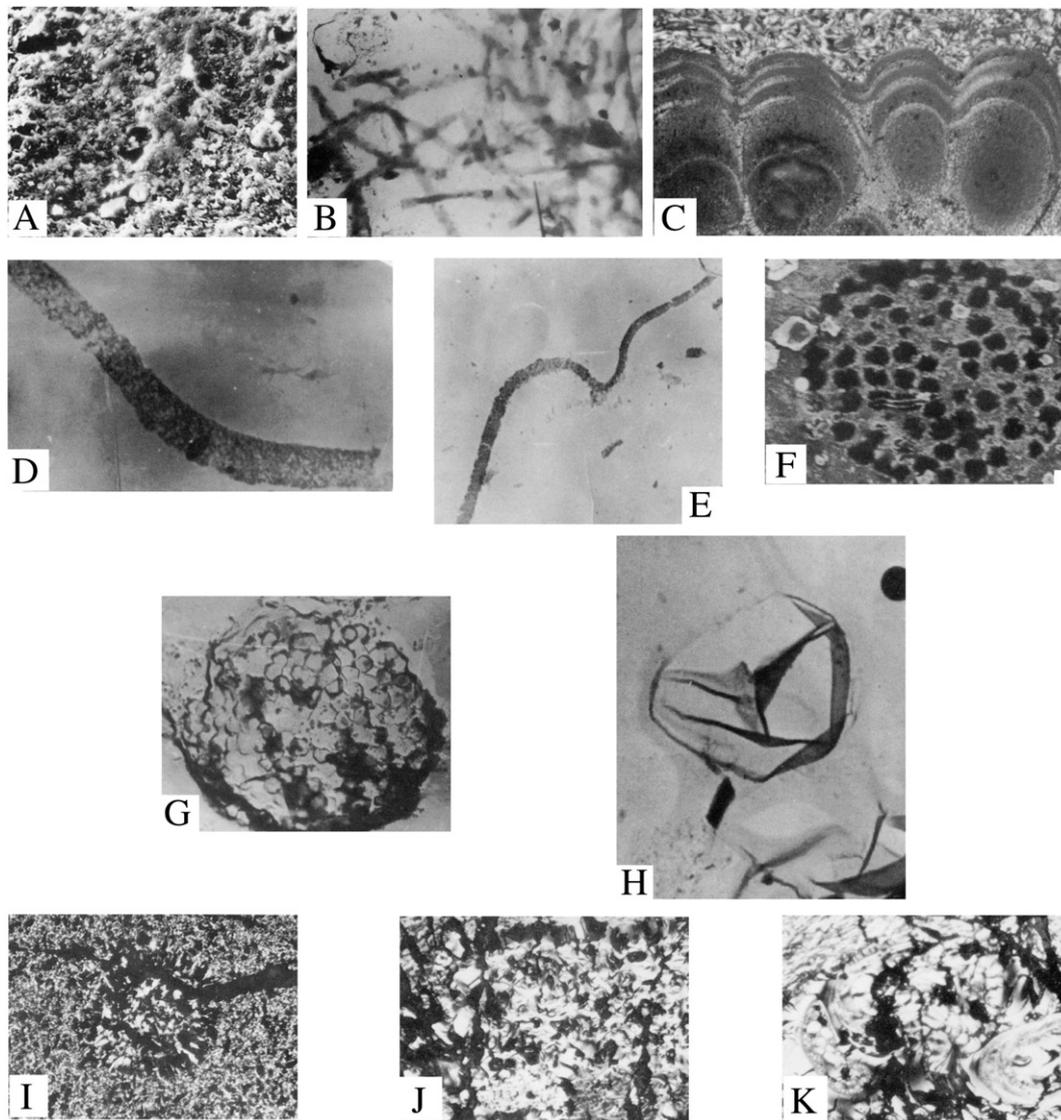
The organic matter in the bogheads is dominated by a sapropelic groundmass (Fig. 2A) derived from the degradation and alteration of algae and, to a lesser extent, cyanobacteria and acritarchs in shallow marine environments, as well as rare sponge spicules (Fig. 2B–H) (China National Administration of Coal Geology, 1996). The algae that formed the Early Cambrian boghead were mainly cyanophytes with

trace amounts of brown algae (China National Administration of Coal Geology, 1996; Li et al., 1982; Zhu, 1983). Cyanophytes and red algae contributed to the formation of the Silurian boghead anthracite, indicating a shallower marine environment in the Silurian than in the Cambrian period. Lu and Zhu (1983) found numerous microfossils in the Early and Middle Cambrian and Early Ordovician anthracite bogheads and their roof and floor rocks from western Hunan Province. The Cambrian fossils include filamentous algae (Fig. 2D, E), cyanobacteria (Fig. 2F), and acritarchs. The Ordovician anthracite bogheads contain cyanobacteria (Fig. 2G), and acritarchs. In addition to normal anthracite bogheads, natural coke, semi-graphite, and graphite that formed from anthracite bogheads occur in the sedimentary and metamorphic sequences of the region. Thermally-altered fine and coarse mosaic textures (Fig. 2I–J) and leafy and spherical anisotropic carbon (Fig. 2K) can be observed under the microscope.

### 1.3. Coal diagenesis and metamorphism

Coal diagenesis and metamorphism, collectively referred to as coalification, is a function of heat and pressure acting over time and is quantified as coal rank. Coalification involves the physical and chemical transformation from peat through lignite and subbituminous coal, to bituminous coal, and through bituminous coal to anthracite and meta-anthracite, and approaching graphite. Coal rank is measured by a progressive decrease in moisture and volatile functional groups with a consequent increase in the carbon content, vitrinite reflectance, and calorific value (although the latter slightly decreases at high ranks) of the coal (van Krevelen, 1993).

Among the factors controlling coalification noted above, heat is generally considered to be the most important (Hower and Gayer, 2002; Taylor et al., 1998). Increased heat coincident with greater depths of burial has been considered to be the primary factor in coalification (Hilt's Law, after Hilt, 1873). Higher rates of heat flow associated with nearby igneous intrusions, however, may be responsible for more localized increases in rank over areas of varying lateral and vertical extent, ranging from development of natural coke at contacts



**Fig. 2.** A. Spropelic groundmass in the Early Paleozoic anthracite boghead from Xupu, Hunan. (SEM; field width 85  $\mu\text{m}$ ). B. *Nocardia* ? sp. in the Middle Cambrian anthracite boghead from Pingli, Shaanxi (TEM; field width 15  $\mu\text{m}$ ). C. Conical columnar stromatolite (algae) in the Early Silurian anthracite boghead from Ankang, Shaanxi (reflected light, crossed polars, oil immersion; field width 375  $\mu\text{m}$ ). D. The filamentous alga *Oscillatoropsis minuta* in the Early Cambrian from Xupu, Hunan (TEM; field width 11.4  $\mu\text{m}$ ). E. The filamentous alga *Oscillatoropsis tenuis* in the Early Cambrian anthracite boghead from Xupu of Hunan (TEM; field width 8.3  $\mu\text{m}$ ). F. The cyanobacterium *Sphaerocongrus wufengensis* in the Middle Cambrian anthracite boghead from Xupu, Hunan (SEM; field width 4  $\mu\text{m}$ ). G. The cyanobacterium *Sphaerocongrus yinpingensis* in the Middle Ordovician anthracite boghead from Dongkou, Hunan (SEM; field width 4  $\mu\text{m}$ ). H. The acritarch *Leiosphaeridia* in the Cambrian anthracite boghead from Pingli, Shaanxi (TEM; field width 380  $\mu\text{m}$ ). I. Fine mosaic texture of the Early Silurian anthracite boghead from Ankang, Shaanxi (reflected light, oil immersion; field width 160  $\mu\text{m}$ ). J. Coarse mosaic texture of the Middle Cambrian anthracite boghead from Ziyang, Shaanxi (reflected light, oil immersion; field width 160  $\mu\text{m}$ ). K. Leafy and spherical anisotropic carbon in the Early Silurian anthracite boghead from Ziyang, Shaanxi (reflected light, oil immersion; field width 160  $\mu\text{m}$ ).

Panel A is from Zhang et al. (2003). Panel B is from China National Administration of Coal Geology (1996). Photo by Zhu and Lin. Panel C is from Chen et al. (1996). Photo by Zhu and Lin. Panel D is from Chen et al. (1996). Photo by Lin. Panel E is from Chen et al. (1996). Photo by Lin. Panel F is from Chen et al. (1996). Photo by Lin. Panel G is from China National Administration of Coal Geology (1996). Photo by Lu and Zhu. Panel H is from Chen et al. (1996). Photo by Lin. Panel I is from China National Administration of Coal Geology (1996). Photo by Shi. Panel J is from China National Administration of Coal Geology (1996). Photo by Shi. Panel K is from China National Administration of Coal Geology (1996). Photo by Shi.

between intrusions and coal seams (Gurba and Weber, 2001; Kisch and Taylor, 1966; Kwiecinska et al., 1992; Ward et al., 1989) to extensive areas of higher-rank coal within otherwise lower-rank coal deposits (Creaney, 1980; Dauly and Cook, 1988; Snyman and Barclay, 1989; Susilawati and Ward, 2006). Tectonically-driven geothermal fluids may also play an important role in coalification (Bayan and Hower, 2012; Gresley, 1896; Han et al., 1996; Hower and Gayer, 2002; Hower et al., 1993; Oliver, 1986, 1992; Ruppert et al., 2010; Uysal et al., 2000; Yang et al., 1996; among others).

The amount of time necessary to achieve a particular level of coal rank varies from less than a year in contact metamorphism to perhaps  $10^6$ – $10^7$  years for regional metamorphism (Bostick, 1979; Hood et al., 1975; Karweil, 1956). Teichmüller and Teichmüller (1966) cited the

contrast of middle Miocene high volatile bituminous phytoclasts from Louisiana and German low volatile Carboniferous coals at the same depth and current rock temperature as an example of the influence of time in coalification. Mississippian-age coals of the Moscow Basin<sup>2</sup> and Pennsylvanian-age coals of the Illinois Basin with  $R_{\text{max}}$  of ca. 0.4% are demonstrations that age alone is not a determining factor in coal rank. Geothermal fluids may complicate interpretations of time, as demonstrated by Harrison et al.'s (2004) estimate that anthracite rank in the Pennsylvania Anthracite Fields could have been achieved in 295 days. Barker (1991) and Giorgetti et al. (2000) demonstrated that time was

<sup>2</sup> Page: 20 Hower and O'Keefe (unpublished data from 2011) have measured a 0.38%  $R_{\text{max}}$  on a Moscow Basin (Belarus) coal supplied by Vladimir Seregin.

not a dominant factor in the organic metamorphism in the Cerro Prieto geothermal system, Baja California (Mexico)/California (USA). Pressure hinders metamorphism in closed systems (Carr, 1999; Dalla Torre et al., 1997). Given (1966), in his discussion of Teichmüller and Teichmüller (who in turn cited Weale, 1962), suggested that reactions during coalification involving polymerization of organic compounds could be influenced by confining pressure.

In the case of intrusions into or adjacent to coal, the source of the heating and of thermal fluids altering the coal is well known. Numerous examples are known from the US, China, Gondwana coalfields, and elsewhere (Barker et al., 1998; Cooper et al., 2007; Crelling and Dutcher, 1968; Dai and Ren, 2007; Dai et al., 2012a,c; Dapples, 1939; de Oliveira and Cawthorn, 1999; Finkleman et al., 1998; Ghose and Mukherjee, 1966; Ghosh, 1967; Han et al., 1996; Hickock and Moyer, 1940; Kisch, 1966; Lyell, 1847; Mastalerz et al., 2009; Querol et al., 2001; Rimmer et al., 2009; Schapiro and Gray, 1966; Schimmelmann et al., 2009; Schopf and Long, 1966; Stewart et al., 2005; Taylor, 1835; Wilkes, 1988; Yang and Han, 1979; Yang et al., 1996).

#### 1.4. Aims of the present work

This review of the fundamental difference between rank and type is necessary to educate and re-educate students and coal scientists on the nature of these two factors in coal formation. Building upon ideas already introduced, we first explore the issue of type, beginning with mire formation and maceral development, then the impact of rank upon type. Throughout, the limits of our present understanding will be defined and areas of suggested future research noted.

## 2. Environments of peat formation

Peat accumulates in mires, i.e., in wetlands with stagnant water. In mires, mainly helophytic plants grow, whereas aquatic plants thrive at deeper sites. For peat accumulation, the annual input of waterlogged helophytic plant remains must exceed the annual breakdown and decay. A peatland is a mire containing at least one peat layer. Peatlands or peat-accumulating wetlands commonly fall into two categories, fens or bogs (Wheeler and Proctor, 2000), although coastal mangrove communities and salt marshes may also be peat producing (Raymond et al., 2010, and references therein).

### 2.1. Environments of autochthonous peat formation

Autochthonous mire type is controlled by three main factors: 1) topography, being the result of geomorphologic and tectonic processes; 2) climate (including temperature), which influences water supply, vegetation type, and decomposition rates; and 3) nutrient load based on water supply and the resulting vegetation types and/or quantity and type of syngenetic mineral matter present. Based on these controlling factors, mires (and similarly the peatlands) are categorized as either A) fens or topogenous mires (controlled by topography, found in low-lying areas) or B) bogs or ombrogenous mires (controlled by climate, found in raised areas). Fens are kept moist primarily through the movement of nutrient-rich surface and ground water through the mire (making them minerotrophic), whereas bogs are exclusively fed by precipitation (making them ombrotrophic). As a result, the mineral input in bogs is low and the conditions are typically acidic (pH values <5.5), while fens are characterized by mineral-rich input and often neutral to alkaline pH conditions (Gore, 1983; Kaule and Göttlich, 1990).

#### 2.1.1. Controls on the locus of peat formation

Peat can only form in those environments that permit the rate of organic accumulation to exceed the rate of organic decay. In addition, peat of the type that will form a coal bed following diagenesis must accumulate in environments with minimal clastic influx, otherwise it will become diluted into organic muck and end up as organic-rich shale.

Thus, two independent parameters account for the above situations: (1) rate of peat formation (organic accumulation exceeding organic decay) and (2) restriction of clastic influx into the peat-forming environment (degree of dilution of the accumulated organic matter with siliciclastic sediment).

#### 2.1.2. Organic production

The initial formation of peat depends on the presence of quite specific physical conditions related primarily to paludification, i.e., the raising of local to regional ground-water base-level to the surface of the soil, or to terrestrialization, i.e., the relative lowering of the water table of a lake as it fills with plant remains (Cecil and Dulong, 2003; Gastaldo, 2010). This can be accomplished in several mutually interacting ways. Important in this process is climate, particularly in warm, frost-free tropical settings. Conditions must exist in which rainfall exceeds evapotranspiration on, say, a monthly average basis, for most of the year (Cecil and Dulong, 2003; Cecil et al., 1985; Moore, 1987; Wüst and Bustin, 2004), such that soil moisture remains high (Morley, 1981). Such climates are described as “humid” or “perhumid” (Cecil and Dulong, 2003; Thornthwaite, 1948). The power of rainfall to raise base level on a short term basis can be seen in the widespread surface paludification and flooding that accompanied the extreme rains in the upper Mississippi River valley in 1993, covering an area of more than 80,000 km<sup>2</sup>, driven by global-scale climate forcing factors (Trenberth and Guillemot, 1996; Wahl et al., 1993). This observation is not intended to mark the Mississippi Valley as an analog to a tropical peat-accumulating habitat; what it does show, however, is that rainfall alone, without any effects of sea-level rise, can elevate regional base-level and lead to widespread surface paludification. Rainfall also may sustain peat accumulation and is important in the development of domed, ombrotrophic mires in which nutrients enter the environment entirely through atmospheric deposition (e.g., Anderson, 1983; Lähteenoja et al., 2009; Posa et al., 2011). On the other hand, topogenous mires form in dependence on the interplay of the geomorphologic and hydrogeological regimes, and are only partly affected by the climate, i.e., this mire type occurs across climates, even in steppes (Taylor et al., 1998). In the upper part of the unusually thick (ca. 190 m) peat profile at Philippi, NE Greece, peat accumulation did not cease during Late Glacial times, when climate in this area was more arid in comparison to the Holocene temperate climate (Wijmstra, 1969), although it continued at lower rates (Christanis, 1987). The mineralogical composition of the substrate can also affect the persistence of standing water, particularly where climates are marginal for sustaining nearly permanently high water tables at the surface (e.g. Gastaldo, 2010). In certain instances, high concentrations of impermeable clays can lead to the development of aquicludes that impede drainage and can encourage the accumulation of organic matter.

Rising sea level causes the rising of the water table landward, and thus can initiate and sustain organic matter accumulation in marshes and mires (Cameron and Palmer, 1995; Cameron et al., 1989; Macintyre et al., 1995; Staub and Cohen, 1979). However, such environments in the modern world do not appear to be likely analogs for most Pennsylvanian coal beds (Cameron et al., 1989), although they may be analogs for parts of the extensive coal seams developed in the Wilcox Group of the US Gulf Coast (O'Keefe et al., 2005), as well as other coals worldwide. This is a topic that needs further investigation, as our understanding of coal deposits is in large part shaped by resource analyses, and coals that exemplify this type tend to be high ash, and thus poor resource rocks. In the few cases where they do host peats of high enough carbon content to develop into more than organic shales (e.g., Delaune and White, 2012), the areal extent of such deposits may be limited (Cameron and Palmer, 1995; Cameron et al., 1989; Staub and Cohen, 1979). Rather than being of a coastal origin, those smaller coal bodies in Pennsylvanian and younger landscapes that have been examined in detail often occupy low areas on landscapes, such as abandoned channels (Cardiff: Cady, 1915; Kerton Creek: Wanless, 1957; Roodhouse: Willman et al., 1975; examples in Nadon, 1998; Carlisle: O'Keefe, 2008; other Western Kentucky lignites:

Hower et al., 1990). Occasionally, widespread coals appear to have begun formation locally in abandoned channels (Pirtle: Wier, 1961; Herrin: Nelson, 1983; Upper Freeport: Hook and Ferm, 1988).

Despite the fact that modern paralic swamps do not seem to be good analogs for coal deposits, in many instances, Pennsylvanian coal has been attributed to landscape paludification driven by sea-level rise (discussed below). Many Pennsylvanian coals were of enormous areal extent, formed in peatlands that were contemporaneous throughout (that is, not time transgressive in their formation), requiring paludification over a vast area. Parts of the Pangean tropics that were in demonstrably drier climatic zones, such as in the western regions of the continent (Tabor and Poulsen, 2008), do not have coals and present no evidence that coal ever formed there, yet these areas were subject to the same rising global sea levels as the coal-bearing regions in the more central areas of Pangea (Cecil et al., 2003b). Therefore, sea-level change alone is not a driving force in peat accumulation.

### 2.1.3. Sediment restriction

Inhibition or elimination of sediment transport into the peat-forming environment, which is the second necessary criterion for accumulation of peat, also may be influenced by climate. Generally, preclusion or great reduction of sediment from the peatland has been attributed to mechanisms such as displacement of sediment deposition inland of coastal mires by rising sea level (e.g., Diessel, 1992; Flint et al., 1995); doming of the peat body, thus raising the surface above the sedimentary system (e.g., Ferm and Cavaroc, 1968; Greb et al., 1999); or baffling of sediment by vegetation growing at the margin of the peatland (e.g., Eble and Hower, 1995; Kravits and Crelling, 1981; Moore, 1991). These models implicitly or explicitly assume that peat formation takes place syndepositionally with local siliclastic deposition. They further imply a high volume of background sediment transport taking place around the peat-forming areas (e.g., Esterle and Ferm, 1986; Ferm and Staub, 1984; Horne et al., 1978; Kravits and Crelling, 1981; Staub and Esterle, 1994; Triplehorn and Bohor, 1986), which may sometimes spill into the environment of peat formation. These models all assume that sediment volume is related almost entirely to tectonics and proximity to sediment source areas (Ferm and Weisenfluh, 1989; Pashin, 1994; Wise et al., 1991), rather than increased weathering and transport due to increased rainfall. Thus, such interpretations require a default climate that is seasonally dry, because it is under these kinds of climates that soil binding by plant roots is limited enough that surface erosion occurs and siliclastics can move into landscape drainages (Cecil and Dulong, 2003). These models require further refinement, as it is now well known that a seasonal climate conflicts with the climatic conditions most favorable for the formation of thick, widespread peat deposits, wherein climatic seasonality is low, and rainfall is consistently high through much of the year, resulting in high rooting in soils, restricting sediment movement into stream drainages (Cecil and Dulong, 2003). This is a problem that needs further exploration, as it is now known that in ever-wet climates, similarly sediment-restricted peats occur adjacent to rivers with high-sediment load (e.g., in the Amazon Basin; Lähteenoja and Page, 2011; Lähteenoja et al., 2009), and these may be analogous to small, areally-restricted coals in the Mississippi Embayment. Clastics within coal beds probably have a number of different origins. Several of these are discussed herein. We will not consider clastic partings that can be identified unequivocally as resulting from volcanic ash falls (Spears, 1987), such as the parting in the Fire Clay coal bed of the Appalachian Basin (Greb et al., 1999), multiple ash layers in the Maning Formation Somerville Lignite (Raymond et al., 1997), or the Great Northern seam (Zhao et al., 2012).

There do appear to be clastic partings that originated from river flooding contemporaneous with peat accumulation. Such partings are generally associated with the marginal parts of some coal beds (e.g., Greb et al., 1999). However, many published examples of such deposits need to be re-examined in light of newer interpretations that

bring climate into consideration as a significant variable in the peat accumulation equation.

In contrast to the contemporaneity model, other interpretations distinguish widespread clastic partings in coal beds from those that appear to be limited to the margins of coal bodies (Cecil et al., 1985). A recently emerging model reinterprets widespread clastic partings not as “splits,” formed when siliclastics were introduced into mires during flooding from nearby rivers and streams, but as “merges,” formed when peat accumulation became interrupted over a widespread area. The different coal benches separated by merges are viewed as representing genetically distinct periods of peat formation (Greb and Eble, 2002; Greb et al., 1999, 2002). Examples are widespread clastic partings such as the so-called ‘blue band’ in the Herrin coal of the Illinois Basin (Nelson, 1983), or the so-called binder beds of the Pittsburgh coal of the central Appalachian Basin (Gresley, 1894). All appear to separate distinct intervals of peat formation (Eble et al., 2006; Mahaffy, 1985), and thus to represent really extensive disturbance surfaces probably related to flooding of the peatland surface by a change in regional water table (due either to a rapid change in base level or increased rainfall) (Cecil et al., 1985).

A different kind of clastic parting, long referred to as a “splay” (e.g., Burk et al., 1987; Kravits and Crelling, 1981; Treworgy and Jacobson, 1985), occurs in certain Middle Pennsylvanian coals of the Illinois Basin, and seems to have originated in ways other than through the introduction of clastic material into the peat mire during flooding from adjacent rivers. Riverine deposits may form large ribbon-like, no-coal, clastic zones within coal beds, with km-scale widths and lengths extending over 100s of km (Eggert, 1984; Hopkins, 1968; Wanless et al., 1969; Willard et al., 1995). For example, Elrick et al. (2008) and Elrick and Nelson (2010), have argued that “splits” in the Springfield coal of the Illinois Basin, proximal to the Galatia Channel, a peat-contemporaneous river deposit (Nelson et al., 2008), are actually sediments injected into and below the coal bed during the flooding stages associated with early transgression, during which the margin of the peat body was torn up by flooding and tidal action, resulting in floating peat mats and localized “flaps”. Such clastic partings are too limited in aerial extent to be splays, often extending only tens to a few hundred meters at most, neither are they ever rooted on top, which would be expected of a splay that was recolonized by vegetation (indicated by thick coal above the clastics), nor are upright tree stumps ever found buried at the base of these clastic partings, which would be expected to occur were they the result of catastrophic, sediment-laden floods. Finally, stringers of coal can be shown to run through these splits, connecting the benches of coal above and below them, and variably thick stringers of coal often come off the bottom part of the upper bench, running irregularly into the clastic parting; these observations indicate violent disruption of the margin of the peat body. In addition, these splits are composed of the same material with the same bedding features as the rhythmic, presumed tidalite sediments deposited on the top surface of the coal body adjacent to the channel.

In parts of coal bodies adjacent to contemporaneous river-channel deposits, active during peat accumulation, there also often are very fine, dark clastics intimately mixed with the coal, raising the mineral-matter content significantly. These clastics are clay-sized and appear to have been deposited contemporaneously with peat formation from streams and rivers that carried very little sediment (Elrick and Nelson, 2010). Such zones of clastic-enriched coal may extend several hundred meters from the area of no-coal, where the coal body is in irregular contact with river-channel siliclastic facies. Elrick and Nelson (2010) suggest that they represent fine grained sediments introduced into the peat body from the adjacent river, which was carrying only fines that flocculated and were deposited only near the channel margin. These partings are genetically different from “splay”-type deposits described above, which may also occur in the same coal beds. Rather than representing a large-scale mire disturbance by a splay, these represent clastic influx from streams and rivers that carried either a very low or very fine

sediment load that rapidly flocculated and was deposited upon introduction into the mire. Similar deposits are known from modern peat-forming environments, e.g., the Snuggedy Swamp (Staub and Cohen, 1979).

Other observations suggest, albeit indirectly, that active peat-forming mires experienced a limited influx of fine-grained clastic sediment. Greb et al. (2001) reported the creation of topographic irregularities in peat mires due to syndepositional tectonics, resulting in an inferred change from domed to planar geometry, which might be expected to attract flood waters bearing fine-grained siliciclastics. However, the subsequent planar peats have low amounts of fine-grained siliciclastics, suggesting limited clastic material in the background. Coals, or even benches within an apparently single coal bed, in their final phases of formation, may show changes in petrographic and palynological composition associated with the influx of fine-grained sediments (Greb et al., 1999; Jerrett et al., 2011), but without the introduction of a coarser siliciclastic fraction, again suggesting that such sediments either were not present in the depositional basin, or the flood intensities were too low to carry them out of the main drainage channels running through the mires. These interpretations are consistent with studies in modern tropical environments where streams draining humid to perhumid regions are virtually devoid of coarse siliciclastic sediment loads (Cecil et al., 2003a; Harris et al., 2008).

#### 2.1.4. Pennsylvanian coal and sequence stratigraphy

The cool climate mode that prevailed throughout much of the Pennsylvanian was similar to that in the Pleistocene and Holocene, and was dominated by glacial–interglacial cycles and their effects on sea level (Wanless and Shepard, 1936), sedimentary dynamics (Cecil and Dulong, 2003; Cecil et al., 2003a), and biotic patterns (DiMichele et al., 2010; Falcon-Lang and DiMichele, 2010). However, in representing a much longer time interval of nearly 20 million years, the Pennsylvanian “coal age” encompassed far greater ranges of variability in climate state and environmental fluctuation than has the Quaternary age (Bishop et al., 2010; Cecil, 1990; Eros et al., 2012; Fielding et al., 2008a,b; Montañez and Poulsen, 2013), including periods of nearly ice-free poles as well as intervals of intense glaciation. The physical and biological dynamics of this time were superimposed on the complex low-latitude landscape where a vast amount of Pennsylvanian peat formed (e.g., Cecil et al., 2003a; Cleal et al., 2009; Gastaldo et al., 1993; Greb et al., 1999, 2003, 2008; Staub, 2002). The Euramerican portion of this landscape included the Variscan–Appalachian mountain ranges, subparallel to the equator (Opluštil and Cleal, 2007), creating large areas of both cratonic and intermontane basins for peat formation in Europe and southeastern Canada (Bashforth et al., 2010; Calder, 1994; Opluštil and Cleal, 2007; Piedad-Sánchez et al., 2004; Roscher and Schneider, 2006), and the large cratonic basins in the United States, in which many of the major coal beds, particularly in the Middle Pennsylvanian, developed on flat landscapes often of vast areal extent (Belt et al., 2011; Calder and Gibling, 1994; Cecil et al., 1985, 2003b; Greb et al., 2003; Heckel, 1995, 2008; Staub, 2002).

Climatic fluctuations, sea-level fluctuations, changes in siliciclastic sediment flux, and the formation of peat and limestone, were not independent variables (Cecil and Dulong, 2003). Rather, they all reflect ties to one another and to high-latitude ice dynamics, which itself may have been dependent on orbital forcing factors and atmospheric CO<sub>2</sub> levels (e.g., Birgenheier et al., 2010; Cecil and Dulong, 2003; Heckel, 2008; Horton and Poulsen, 2009; Montañez and Poulsen, 2013; Peyser and Poulsen, 2008; Poulsen et al., 2007; Rosenau et al., 2013; Royer et al., 2004; Rygel et al., 2008; Wanless and Shepard, 1936), or even unknown astronomic factors.<sup>3</sup> Certain aspects of tectonics also may have strong linkages to prevailing climate, which has been shown to play an important role in erosion rates, rates of uplift, and rates of basinal

subsidence (e.g., Harris and Mix, 2002; Hay, 1996; Montgomery et al., 2001; Whipple, 2009). As a consequence of the inter-dependence of these major variables, one might expect that the repeated patterns evidenced by the “cyclothems” of Euramerican Pennsylvanian coal basins reflect systemic controls.

For the purposes of this review, the coal bed is the portion of these cyclic patterns that is of particular interest. Many, if not most, of the economically viable large coal seams in the Pennsylvanian have a ravinement surface on top of them and are overlain by marine rocks. By definition, the ravinement is a parasequence boundary and the base of the transgressive systems tract. The coals themselves contain evidence of variations in level of decay, water table, etc. throughout the seam, indicating that peat accumulation was not simply driven by transgression and progressive inland paludification (DeMaris, 2000; Gastaldo et al., 1993; Rosenau et al., 2013). Much remains to be learned concerning the effect of orbital cycles on tropical paleoclimate and peat accumulation and the role of Milankovitch orbital cycles in controlling sea level and paleoclimate. Critical questions include:

- What controlled the initiation and continuance of peat formation?
- What controlled its preservation? and
- What was the areal extent of a peat-forming landscape at any given instant in time?

There are also changes through time in the details of the factors controlling peat formation, which affected the extent and quality of coal beds, resulting in distinct temporal patterns (Section 2.1) (e.g. Cecil, 1990; Cecil et al., 1985; Schutter and Heckel, 1985).

#### 2.2. Macerals forming in the mire

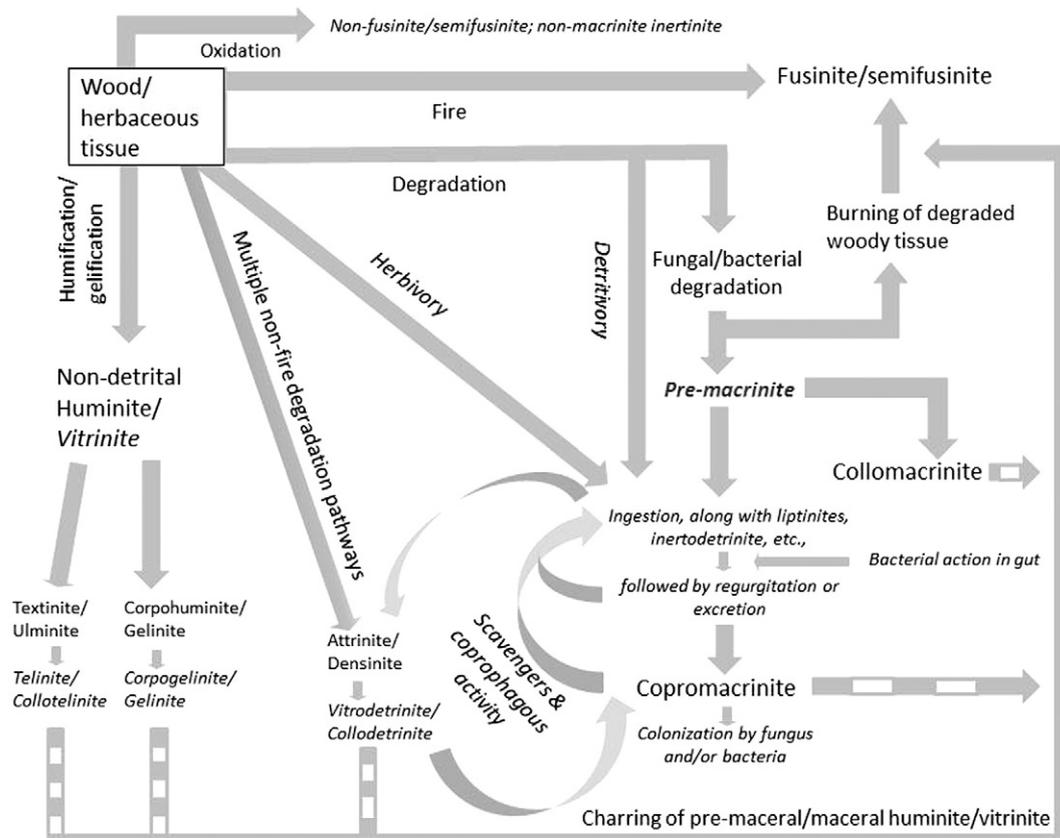
Time, climate, vegetation, and geological setting are inter-related and inter-dependent prerequisites for coal formation, and coal rank progresses irrespective of, and unaffected by, coal type and grade (Cairncross, 2001). Maceral composition is controlled by peat-forming vegetation type, as well as by the Eh and the pH conditions, and the microflora of the peat, which all are, in part, controlled by the climatic conditions and sedimentary substrate upon which the peat develops, as well as the sedimentary influx into the peat; all factors are inter-related rather than independent (Cook, 1975).

Figure 3, from Hower et al.'s (2013a) continuing discussion of the origin of certain inertinite macerals, shows the interrelationships between the woody or herbaceous starting material and the resulting vitrinite or inertinite macerals. The pathways can be complex; as stated earlier with respect to determining the origin of macerals (Hower et al., 2009) “provenance begets the maceral, but provenance has a diffuse time line...let the macerals be the guide, but temper this with the full complement of the available petrographic and biological knowledge.”

##### 2.2.1. Huminite/vitrinite

Woody and parenchymatous tissues and their components can form huminite macerals under a variety of pH and Eh conditions (Sýkorová et al., 2005) in both topogenous and ombrogenous mires. Corpohuminite and textinite formation has been demonstrated to begin within as little as five years in wood deployed in a moist seep (O'Keefe et al., 2011; their figure 13C). As rank increases, the tissue remnants are referred to as “huminite macerals” until they reach a reflectance of about 0.4% R<sub>r</sub>, and are referred to as “vitrinite macerals” from a reflectance of about 0.5% to 7.0% R<sub>r</sub> (ICCP, 1998; Sýkorová et al., 2005). This is a nomenclatural artifact, and NOT one maceral changing into another. This nomenclatural artifact, and thus confusion of coal rank and coal type, has been perpetuated by the ICCP in treating the macerals separately (ICCP, 1998; Sýkorová et al., 2005), rather than as a rank-driven continuum of aspects of the same material (Fig. 3). The source of huminite and vitrinite macerals are identical; and the gelification processes active during the transition from peat to lignite (brown coal) are the same. Some

<sup>3</sup> See debate over passage of the solar system through the galactic plane (Overholt et al., 2009; Shaviv and Veizer, 2003).



**Fig. 3.** Developmental pathways of helophytic-derived macerals. Liptinite and non-wood-origin inertinite (resin-derived secretinite and funginite) are not directly included as end products. Funginite, as fungus, is part of the process. The charring of pre-maceral huminite or huminite or vitrinite macerals is not necessarily a continuing step in maceral formation and the end product, while located close to primary fusinite/semifusinite on the chart, has a different origin than primary fusinite/semifusinite. This distinction is a necessary part of the description of the macerals. The naming of the inclusion-free *collomacrinite* and the complex *copromacrinite* is a convenience in the description of the maceral origins and does not represent formally accepted maceral nomenclature.

From Hower et al. (2013a).

confusion enters in due to floral turnover within mires. The finer features of huminite/vitrinite macerals change significantly from the Devonian to the Permian, and again from the Triassic to the Cretaceous, reflecting plant evolution. Huminite/vitrinite macerals from the Early Devonian were derived primarily from lower vascular (and some non-vascular) plants, while the Late Devonian–Pennsylvanian saw a transition to woodier varieties, the Pennsylvanian to Permian had a transition to gymnospermous plants, the Triassic to Cretaceous had a transition to primarily gymnosperms, and the upper Cretaceous to Miocene saw a transition to angiospermous constituents in many regions, with gymnospermous input often restricted to members of the Taxodiaceae. Coeval with these transitions, variations in maceral aspect are present, with less-well preserved tissues giving way to better-preserved cellular tissues, and an admixture of less well-preserved tissues, often representing angiospermous input. Preservation of these tissues is significantly impacted by eH, climatic setting, and faunal evolutionary history. As saprophytic and decomposing fungi, bacteria, and insects adapted to evolving mire plants, vitrinite/huminite tissue preservation generally becomes diminished, and an increase in detrohuminite/detrovitrinite group macerals is seen. Regardless of taxonomic origin, huminite/vitrinite macerals are categorized into three varieties: Telohuminite/telovitrinite (Fig. 4A–H) is indicative of little aerial (aerobic) decay and is formed from cell walls; detrohuminite/detrovitrinite (Fig. 4I–P) is indicative of significant aerial decay and is formed from remnants of woody material after attack by both saprophytic and decomposing organisms; and gelohuminite/gelovitrinite (Fig. 4Q–X) form from non-liptinitic cellular contents or materials that have migrated into void spaces (ICCP, 1998; Sýkorová et al., 2005). At higher ranks, distinguishing vitrinite varieties can

become challenging, although this is aided by etching, which can reveal cellular structures still present even into the anthracite rank (Fig. 5).

### 2.2.2. Pseudovitrinite

In investigations of the coking properties of central Appalachian coals, Thompson et al. (1966), Benedict et al. (1968), and Thompson (2000) found that a portion of the vitrinite group was anomalously poorly-reactive during the coking process, noting several microscopic characteristics that distinguished it from the more reactive vitrinite:

- higher reflectance
- slitted structures
- unusual fracture patterns
- remnant cell structure
- paucity or absence of syngenetic pyritic inclusions, and
- higher relief.

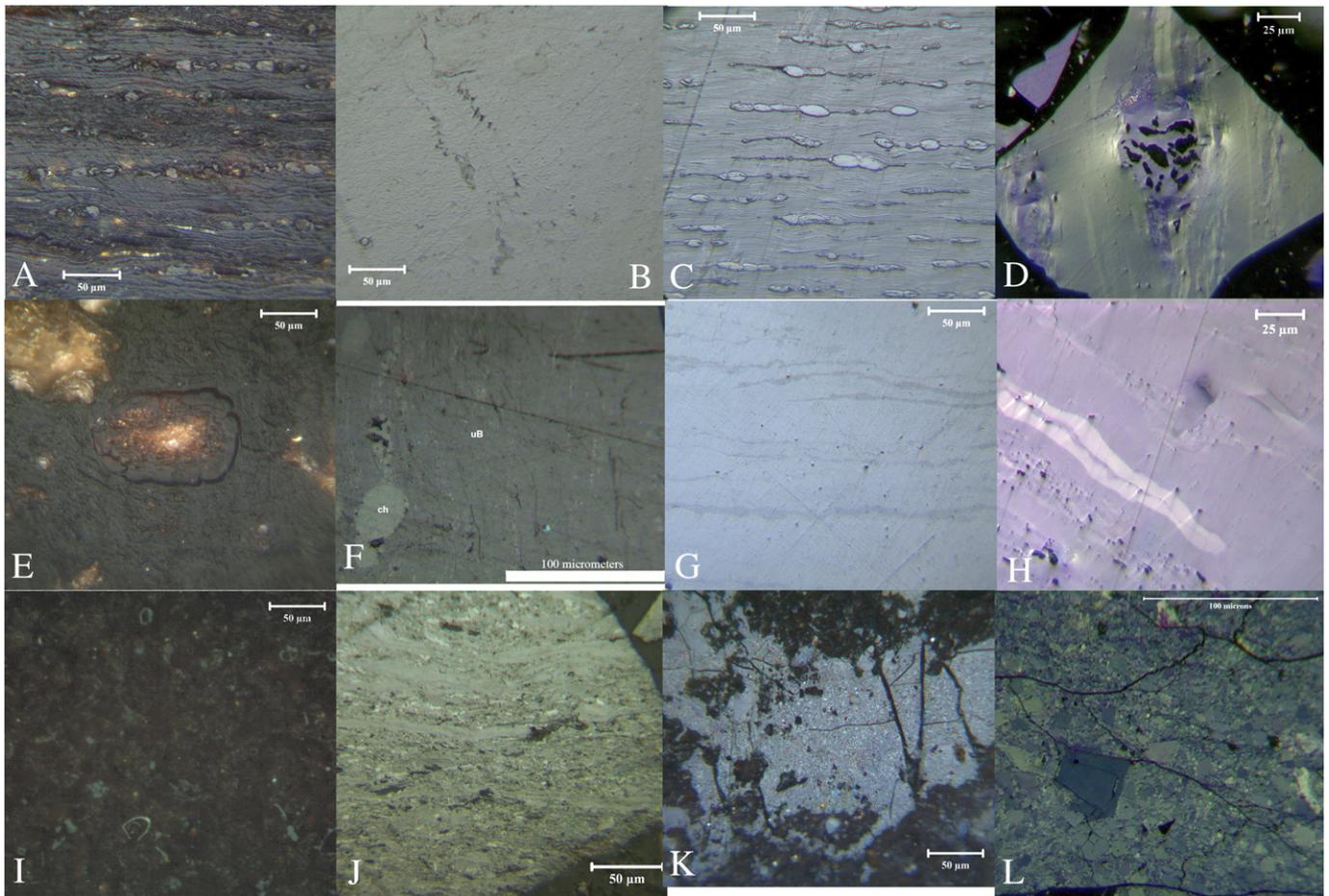
Consequently, they proposed the term “pseudovitrinite” to represent this vitrinite category. However, Kaegi (1985) suggested that only the slitted structures be used as a differentiating mechanism between pseudovitrinite and collotelinite, due to the difficulty when assessing changes in reflectance in blend samples; that remnant cell structure is already used as a distinguishing criterion between collotelinite and telinite; and the fact that the other distinguishing features proposed do not hold for all coals. While the distinctive characteristics of pseudovitrinite are obvious in many central Appalachian coals, they are less obvious in Illinois Basin coals of the same rank and age, limiting Benedict et al.’s (1968) conclusion that pseudovitrinite is present in coals of all ranks and is an inherent maceral in all coal seams. Pseudovitrinite, characterized by the slit

structure and slightly enhanced reflectance, is frequently observed in South African Permian-age coals (Kruszewska, 2003).

Although both collotelinite and pseudovitrinite may have originated from the same or similar woody precursors, the exact origin of pseudovitrinite remains uncertain. Mackowsky (personal communication cited by Smith, 1980), suggested a pseudovitrinite origin from primary oxidation during the biochemical phase of coalification, and Kaegi (1985) determined that pseudovitrinite falls between vitrinite and oxyvitrinite (a low temperature oxidation product). Benedict and Berry (1964) conducted controlled oxidation studies, producing pseudovitrinite-like microfeatures with similar anomalous chemical properties as high-pseudovitrinite coals. Bengtsson (1987) determined that pseudovitrinite caused the delayed release of volatile matter, and Kruszewska (1998) determined that pseudovitrinite had a delayed, but ultimately comparable reactivity to collotelinite, with cryptopseudovitrinite observed at 350 °C. Johnson et al. (1987), using photoacoustic microscopy, determined that vitrinite and pseudovitrinite were thermally different entities up to about 92% carbon, with the differences more pronounced in the lower ranks. Spackman (1970), however,

found a wide divergence of pseudovitrinite behavior in physical and chemical tests. Gurba and Ward (1998), Ward and Gurba (1999), Ryan et al. (2003) and Mastalerz and Drobnik (2005) have studied vitrinite forms, including pseudovitrinite, with respect to reflectance contrasts, chemical composition, and gas adsorption properties. Ward and Gurba (1999), using light-element electron microprobe analysis techniques, have shown that the different vitrinite macerals, including pseudovitrinite, in coals of the Gunnedah Basin, Australia, all have essentially the same chemical composition (C, O, N, and S) within the same coal sample, despite differences in reflectance and texture.

Should pseudovitrinite be placed within the vitrinite group? All other vitrinite macerals are distinguished by differences in source material (and degree of degradation/disaggregation and/or degree of gellification), while pseudovitrinite may originate from the oxidation of a similar source material (Smith, 1980). In the Stopes–Heerlen maceral-classification system and the subsequent ICCP System 1994 (ICCP, 1998, 2001; Sýkorová et al., 2005), a significant difference in the degree of oxidation is the basis for distinguishing between maceral groups, for example, between the vitrinite and inertinite groups, rather



**Fig. 4.** Examples of “typical” humite/vitrinite macerals through a rank series from peat (leftmost image) to lignite/brown coal (image second from left) to bituminous coal (image second from right) to anthracite (image on right). Fig. 4-1: A–D, textinite/telinite series; E–H, ulminite/collotelinite series; I–L, attrinite–vitrodetrinite series. Fig. 4-2: M–P, densinite/collodetrinite series; Q–T, corpohuminite–corpogelinite series; and U–X, gelinite series. A. Textinite. Neogene peat from Centralia Washington, USA; photo by Hower. B. Textinite. Cenozoic lignite from Tahlesel, Bohemia, Germany (now the Czech Republic); from the Vanderbilt coal collection; photo by Hower. C. Telinite with corpogelinite. Kimmeridgian of New Jersey, USA; photo by Hower. D. Telinite. Asturian of Schuylkill Co., PA, USA; Photo by Hower. E. Ulminite. Peat from Kerry Bog, Ireland; Photo by Hower. F. Ulminite with corpohuminite. Middle Eocene Claiborne Lignite from Kentucky, USA; photo by O’Keefe. G. Telinite with faint cutinite. Triassic of Virginia, USA; Photo by Hower. H. Telinite with sporinite from a megaspore. Asturian of Schuylkill Co., PA, USA; Photo by Hower. I. Attrinite. Recent Indonesian Peat. Photo by Esterle. J. Attrinite. Miocene Eel River Coal, Mendocino Co., California, USA; Photo by Hower. K. Vitrodetrinite. Permian Heshan coalfield, Guangxi, China; Photo by Hower. L. Vitrodetrinite. Permian of Guangdong, China; Photo by Hower. M. Densinite. Modern Loose Compost, Rowan County, KY; Photo by O’Keefe. N. Densinite. Miocene of Slovakia; Photo by Hower. O. Collodetrinite. Aptian Wulantuga Coal of Inner Mongolia, China; Photo by Hower. P. Collodetrinite. Permian of Chongqing, China. Photo by Hower. Q. Corpogelinite. Recent Indonesian Peat. Photo by Esterle. R. Corpogelinite. Paleocene Buelah Zap from North Dakota, USA; Photo by Hower. S. Corpogelinite. Pennsylvanian of Bell County, KY; Photo by Hower. T. Corpogelinite. Permian of South Africa; Photo by Hower. U. Gelinite. Modern drift wood from Holden Beach, North Carolina, USA; Photo by Hower. V. Gelinite. Cenozoic brown coal from Dambroich bei Bonn, Germany; Vanderbilt Coal Collection; Photo by Hower. W. Gelinite. Asturian from Tioga County, Pennsylvania, USA; Photo by Hower. X. Gelinite. Asturian Buck Mountain Coal, Schuylkill County, PA, USA; Photo by Hower.

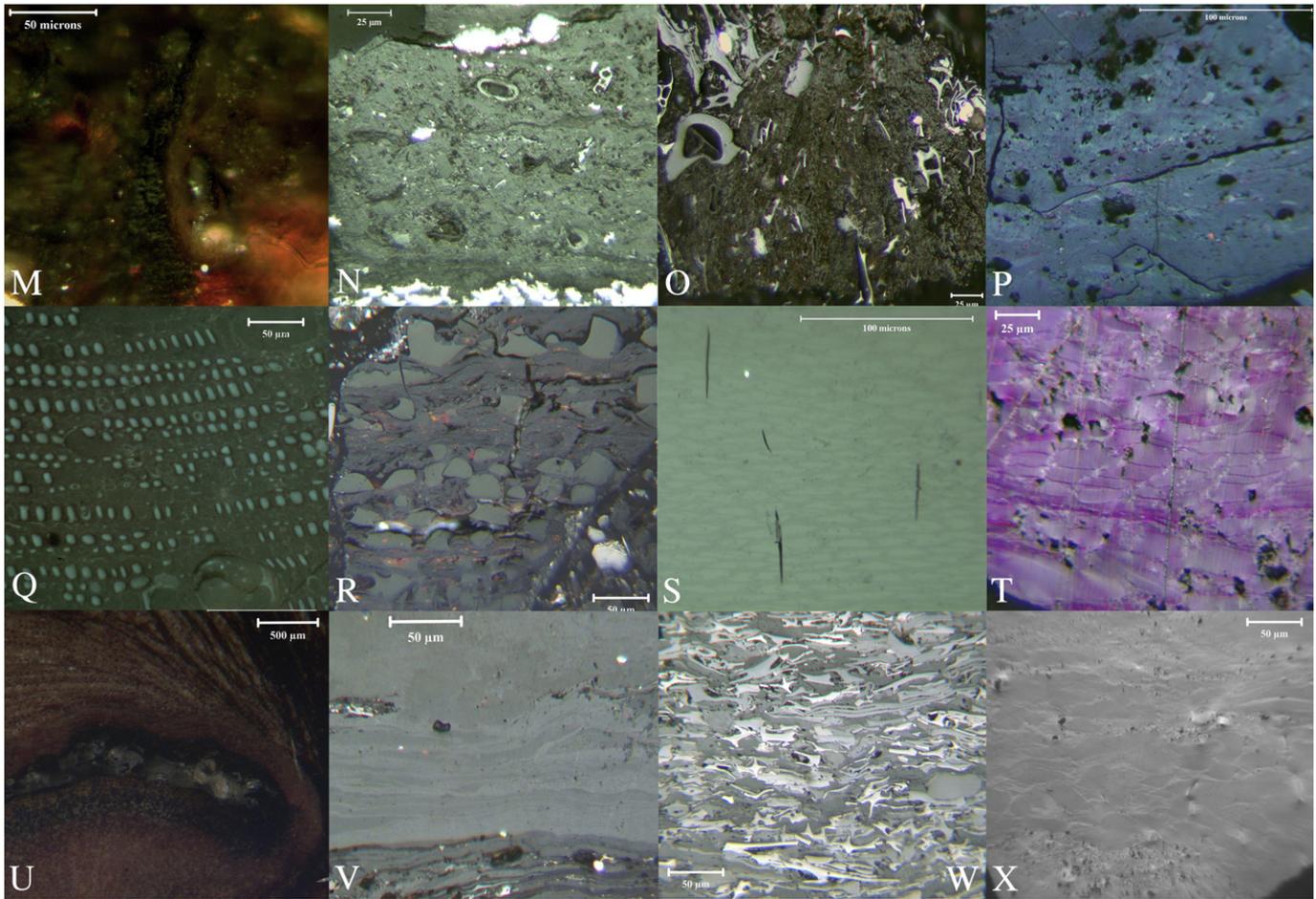


Fig. 4 (continued).

than between macerals within the same group. As such, Smith (1980) and Diessel and Gammidge (1998) considered pseudovitrinite to be more appropriately classified as a separate maceral group rather than as a member of the vitrinite maceral group, but Kruszewska (1998) places pseudovitrinite as part of collotelinite, as it is likely that pseudovitrinite formed as collotelinite and underwent mild oxidation at a later stage during coalification. The lack of variation in chemical composition between vitrinite and pseudovitrinite noted by Ward and Gurba (1999) also supports inclusion of pseudovitrinite in the huminite/vitrinite maceral group.

### 2.2.3. Inertinite

The inertinite maceral group is highly diverse, and includes semifusinite, fusinite (pyrofusinite, degradofusinite, rank, and primary fusinite), funginite (including hyphae and mycelia; part of what was previously named sclerotinite), secretinite (part of what was previously named sclerotinite), macrinite, micrinite, and inertodetrinite. The origin of inertinite is somewhat controversial, with three schools of thought found in the literature: 1) fire origin (Abu Hamad et al., 2012; Demchuk, 1993; Diessel, 2010; Scott, 1989; Scott and Glasspool, 2007; Scott and Jones, 1994); 2) oxidation by microbial activity and dehydration in an aerobic environment (Cook, 1981; Hower et al., 2009); and 3) biochemical alteration (Sen, 1999; Teichmüller, 1974). Scott (1989, 2000) determined that a range of temperatures in wildfires, above and below ground, would result in a complete range of charred to uncharred material with different reflectance values, and that there is “no need to invoke ‘oxidative processes’ other than fire” (Scott, 1989, p. 469) in the formation of fusinite. However, Schopf (1975) noted

that a charcoal origin can occasionally be proven for fusain, but this does not prove that all fusain formed due to conflagration or isolated peat fires. Sen (1999) proposed that the precursor of fusinite originates from the residual lignin-rich cell walls of plant debris, and semifusinite from the partial removal of the cellulosic layer. In peat, pyrofusinite or primary fusinite has a high reflectance, but other inertinite macerals have a low original reflectance, supporting the view that some inertinite is derived from biochemical alteration of plant material (Teichmüller, 1974) and original and altered fungal material (O'Keefe et al., 2011) within the peat environment.

While not arguing the fire pathway from woody and herbaceous tissue to the inertinite macerals fusinite and semifusinite (Bustin and Guo, 1999; Demchuk, 1993; Guo and Bustin, 1998; Hudspeth et al., 2012; Marynowski et al., 2011; McParland et al., 2007; Petersen, 1998; Scott, 1989, 2000, 2002; Scott and Glasspool, 2005, 2006, 2007; Scott and Jones, 1994; Scott et al., 2000; Winston, 1993), and including the (likely) similar transformation of resins and corphuminite precursors to secretinite (Hower et al., 2008a,b; Lyons et al., 1986), it must be recognized that some of the macerals classified as inertinite do not have an origin within the plant kingdom. The inertinite maceral funginite is produced by fungi, and macrinite is the result of severe alteration by non-plant entities (fungi, bacteria, and animals [Hower et al., 2009, 2011b]). In essence, the accepted inertinite classification (ICCP, 2001) is too broad, conflating macerals with largely combustion origins (fusinite, semifusinite, secretinite) with macerals representing the agents (funginite) or products (macrinite, possibly micrinite) of degradation. That being said, there are transitional forms of fusinite and semifusinite which originated from degraded woody material, as with

the example in Fig. 6. In the latter case, the fusinite resembles cell structures derived from fungal and bacterial degradation as discussed by Varossieau (1949) and Klaassen (2008).

In reality, the origin of inertinite can be considered to be varied, with multiple pathways possible (Fig. 3; Hower et al., 2013a,b,c): i) a fire origin for fusinite (specifically pyrofusinite, but possibly other fusinite forms) and possibly some occurrences of semifusinite, secretinite and inertodetrinite; ii) a biochemical and/or microbial origin for funginite, macrinite, secretinite, and micrinite; iii) an oxidation and/or biochemical origin for semifusinite, secretinite, and inertodetrinite; and iv) an allochthonous environment for inertodetrinite and micrinite following the break-up of partially coalified matter due to water movement. In addition, oxidation and microbial attack may occur pre- and/or post-fusinitization by fire (Rayner and Boddy, 1988), producing semifusinite and fusinite (more commonly) with at least two distinct phases of formation, and heat from microbial action may result in enhanced reflectance values, or zonation. Scott (1989, 2002) notes that inertinite macerals other than fusinite may have formed along different, or consecutive pathways, noting that decaying plant litter and wood under fungal attack may char at slightly different rates.

**2.2.3.1. Fusinite and semifusinite.** Fusinite is accepted to be a brightly-reflecting fire-derived maceral (Bustin and Guo, 1999; Guo and Bustin, 1998; Scott, 1989, 2002; Scott and Glasspool, 2007). Fusinite appears white under reflected light and generally has reflectance values higher than 6%  $R_r$  (ICCP, 2001). Fusinite displays well-preserved cellular structures, reflective of its rapid origin during combustion events. It does not change morphology or reflectance significantly during coalification.

Semifusinite, an intermediate maceral between huminite/vitrinite and fusinite, having formed in the peat by weak humification, dehydration, and redox processes, may show vague or partially visible cell lumens derived from parenchymatous and xylem tissues of stems, herbaceous plants, and leaves (ICCP, 2001). According to the ICCP (2001), Gondwana semifusinite is leaf-derived and Carboniferous semifusinite is wood-derived. Due to the significant proportion of semifusinite in Gondwana coals, an oxidative origin was proposed by Cook (1981), an option disputed by Scott (1989). Scott (1989, 2000) and Scott and Jones (1994) suggest that low-temperature fires will give rise to large quantities of lower reflecting semifusinite (reflectance values less than 2%), and high-temperature fires, and possibly prolonged exposure, will result in high reflecting fusinite and possibly inertodetrinite particles. Diessel (2010) supports the concept of incomplete combustion being the main cause of inertinite formation globally. Scott and Glasspool (2007) propose a 2% reflectance (mean random in oil) boundary between fusinite and semifusinite,<sup>4</sup> but this raises a concern regarding Smith and Cook's (1980) finding that "inertinite cannot be considered immutable with rank" (p. 646). Smith and Cook (1980) determined a dynamic coalification path for inertinite, with an inertinite coalification jump of 0.7 to 2% over the mean-random-vitrinite-reflectance rank range of 0.2 to 0.9%. In South African coals, a range of semifusinite macerals occur, subdivided into reactive and inert semifusinite,<sup>5</sup> based on the observed shade of gray relative to vitrinite; reactive semifusinite is  $\leq 0.3\%$  higher in reflectance than the maximum vitrinite reflectance (slightly lighter shade of gray, with definite structure, and a mottled appearance under crossed-polars) and inert semifusinite has a brighter appearance, but with a lower reflectance than fusinite (Falcon and Snyman, 1986; Hagelskamp and Snyman, 1988; Kruszewska, 1989).

<sup>4</sup> That is, inertinite with a mean random reflectance in oil above 2% is fusinite, and that under 2% is semifusinite.

<sup>5</sup> Reactive semifusinite has been shown to be as reactive as vitrinite under certain conditions in South African coals (Falcon, 1986), and may explain why some inertinite-rich coals behave in a comparable manner during coking compared to vitrinite-rich coals of a comparable rank (Kershaw and Taylor, 1992).

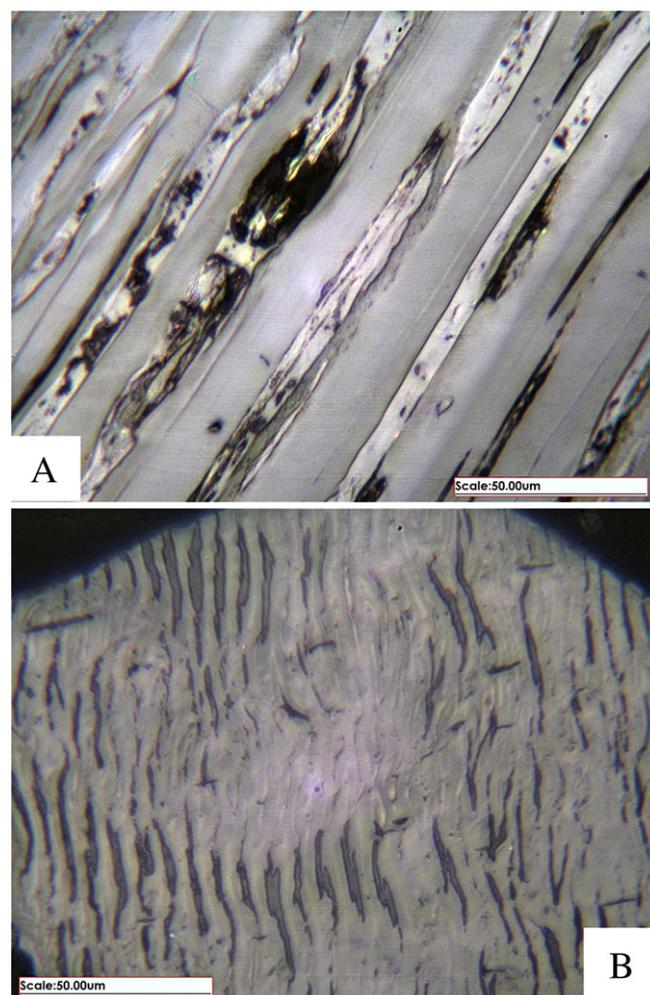
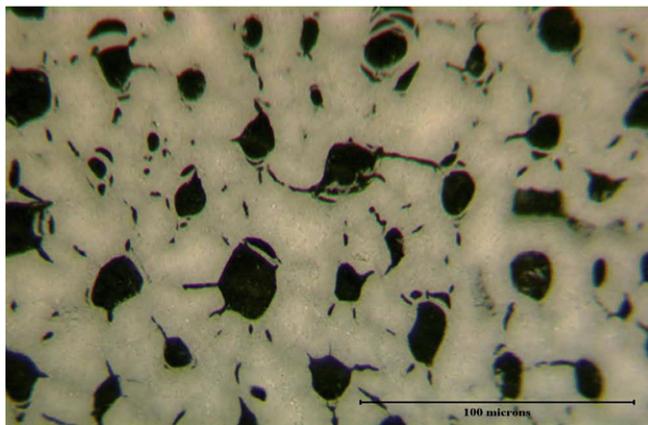


Fig. 5. Examples of vitrinite macerals in etched anthracite coal. A is telovitrinite; B is telovitrinite with resinite. Photos by Eble.

Degradofusinite, a term coined by Teichmüller (1944, 1950) for low-reflecting semifusinitic material, forms in depositional environments due to the desiccation of variably gelified humic matter which leads to the retention of oxygen, or during the decomposition of wood as a result of wood-decomposing fungi, or during sub-surface oxidation of coal (Taylor et al., 1989, 1998). Degradofusinite is common in the Carboniferous coals of Upper Silesia and in most Permian coals of Gondwana (Taylor et al., 1998). Scott and Glasspool (2007), though, are of the opinion that the formation of degradofusinite has not been proven. Some of what has been described as degradofusinite is properly considered to be macrinite (Hower et al., 2009, 2011a,b, 2013a,b), however, many forms occur as larger pieces or as altered zones within or surrounding huminite/vitrinite macerals, such as that figured by Taylor et al. (1998) in their Figure 4.28c.

Rank fusinite refers to fusinite that formed during the geochemical coalification of huminitic–liptinitic cell tissues. Cell walls of woods may be protected against microbial attack by the impregnation of resin, cutin, suberin, etc., thus preserving the cellulose (Taylor et al., 1998). Scott (2002) is of the opinion that the rank and primary fusinite subdivisions are misinterpretations of the original plant chemistry, as charring explains the origin of these fusinite forms. However, Teichmüller (1974) provides evidence for the formation of rank fusinite, as well as degradofusinite and primary fusinite. Primary fusinite refers to strongly reflecting fusinite that is believed to represent the original tissue structure of plants. Scott (1989) discounts aerobic decay for fusinite formation due to a perceived lack of modern fusinite-like material. However,



**Fig. 6.** Fusinite showing evidence of significant degradation prior to combustion. Note the delaminated cell walls and internal cracking. China (Permian, Yunnan Province) coal sample CN 87 02 – fusinite. Refigured from figure 8 of [Dennen et al. \(2010\)](#).

[Taylor et al. \(1998\)](#) cite several instances of present day vegetation that can form primary fusinite, such as the leaves of *Acrostichum aureum* (mangrove fern) in Florida peats ([Cohen, 1968](#)) and fertile shoots of the *Equisetum arvense* (horsetail) ([Koch, 1966](#)).

Pyrofusinite is the inertinite attributed to formation in wood or peat fires. Incomplete combustion due to oxygen deficiency will result in fossil charcoal ([ICCP, 2001](#); [Scott, 1989, 2000, 2002](#); [Scott and Glasspool, 2007](#); [Scott and Jones, 1994](#)). Strongly charred gymnospermous wood with thin cell walls, high relief, and strongly fluorescing resins can be preserved intact where the botanical structure has been in-filled or appears as fine, brittle fragments. Lower temperature exposure will result in lower reflecting fusinite, perhaps with thicker cell walls. Horizons of thick fusinite lenses are apparent in the Permian Witbank-Highveld (South Africa) coals, frequently in-filled with carbonate minerals, making extremely hard material. In the Cretaceous Wulantuga coal (Inner Mongolia, China) ([Dai et al., 2012b](#)), many fine fusinite fragments are distributed in the low-rank coal, suggesting a secondary deposit of the fusinite. Alternatively, the original botanical material was a herbaceous plant which shattered easily when burnt, subsequently being deposited as fine splinters of fusinite. Fine fusinite associated with vitrinite bands could have originated from wind-blown material deposited in subaqueous environments.

**2.2.3.2. Secretinite.** Following the classification by [Lyons et al. \(1986\)](#) of secretion-derived inertinite, the [ICCP \(2001\)](#) published documentation separating sclerotinite macerals into funginite and secretinite. Some of what was called sclerotinite (*sensu stricto*) refers to durable forms of fungal mycelia. Funginite, discussed in more detail below, can occur as single cell structures, or bi- and multiple-cells, hyphae (thin tubular forms), and mycelia (clusters of tubular forms), and essentially consists of chitin ([ICCP, 2001](#)). Secretinite is considered to be non-cellular, commonly round, vesicled to non-vesicled, and equate to elongate bodies, derived from highly oxidized or burnt resin or humic gels ([ICCP, 2001](#)). In Carboniferous and Permian coals, secretinite may have formed from cell secretions or oxidized resins of higher plants, which may have survived peat or seam fires ([Taylor et al., 1998](#)).

**2.2.3.3. Inertodetrinite.** Inertodetrinite consists of redeposited debris of fusinite, semifusinite, and other inertinite macerals ([Taylor et al., 1989](#)), and can include small amounts of redeposited vitrinite. Inertodetrinite showing graded bedding may indicate redeposition in an aqueous/subaqueous environment, and a more mixed redeposition may indicate wind transportation where the peat surfaces may have dried out or were burned as a result of peat fires. Inertodetrinite may occur as discrete fragments, or, more commonly in South African coals, as

allochthonous conglomerates, sometimes parallel to bedding, or mixed deposition. Small, highly reflecting particles of inertodetrinite may have been a product of very high temperature charcoal that fragmented ([Scott and Jones, 1994](#)) and was deposited proximally or transported considerable distances.

**2.2.3.4. Macrinite and funginite: the role of microbes in peat transformations.** As will be discussed in more detail below, fungus is an important agent in the development of degraded wood and maceral assemblages. In modern peats, aerobic bacterial and fungal activity decreases within a meter of the surface as pore waters become anoxic ([Baker, 1970](#); [Bradley, 1966, 1970, 1973](#); [Given et al., 1983](#); [Latter et al., 1967](#); [Schopf, 1952](#); [Visser, 1964](#); [Waksman, 1944](#); [Waksman and Stevens, 1929](#); [Wüst et al., 2001](#)). Among others, [Graham \(1962\)](#), [Pirozynski \(1976\)](#), [Stubblefield and Taylor \(1988\)](#), [Dix and Webster \(1995\)](#), [Pfister \(1997\)](#), [Volk \(2001\)](#), [Carlile et al. \(2001\)](#), [Lutzoni et al. \(2004\)](#), [Webster and Weber \(2007\)](#), and [Taylor et al. \(2009\)](#) have discussed fungus form and function.

There are an estimated  $3.5 \times 10^9$  bacteria/cm<sup>3</sup> and 600 m of fungal mycelium/cm<sup>3</sup> in a mixed moor (transitional mire) peat ([Latter et al., 1967](#)) and up to 2 km of fungal mycelium/cm<sup>3</sup> in a forest soil ([Read and Boyd, 1986](#)). The absence of fungal hyphae in coal is not necessarily an indication of the lack of fungal activity. Actinomycete and Gram-positive bacteria degrade fungal chitin and chitinase belonging to both groups has been recovered from histosols in Japan ([Belova et al., 2006](#); [Schrempf, 2001](#); [Terahara et al., 2009](#)). In addition, brown rot fungi autolyze (self-digest) their hyphae ([Carlile et al., 2001](#); [Pujana et al., 2009](#)).

The importance of fungus, seen in coal as the maceral funginite, as an agent in degradation has been discussed by [Waksman \(1930\)](#), [Barghoorn \(1949, 1952\)](#), [Teichmüller \(1958\)](#), [Beneš \(1959, 1960a,b, 1969\)](#), [Beneš and Kraussová \(1964, 1965\)](#), [Hower et al. \(2010a\)](#), [Taylor and Krings \(2010\)](#), [Hower and Ruppert \(2011\)](#), [O'Keefe et al. \(2011\)](#), and [O'Keefe and Hower \(2011\)](#). Fungus is known from coals through all of the major coal-forming periods, including the Pennsylvanian ([Andrews and Lenz, 1943](#); [Beneš, 1959, 1969](#); [Cash and Hick, 1879](#); [Coulter and Land, 1911](#); [Duparque and Delattre, 1953a,b](#); [Herzer, 1893](#); [Hook and Hower, 1988](#); [Krings, 2001](#); [Krings et al., 2009](#); [Lapo and Drozdova, 1989](#); [Lesquereux, 1880](#); [Renault, 1903](#); [Richardson, 2010](#); [Richardson et al., 2012](#); [Stach, 1935](#); [Stach and Pickhardt, 1957, 1964](#); [Schopf, 1952](#); [Stubblefield and Taylor, 1984](#); [Taylor and Osborn, 1996](#); [Taylor and Taylor, 1997](#); [Thiessen, 1925a,b](#); [Wagner and Taylor, 1981, 1982](#) (their figure 3c)).

The degradational origin of certain macerals, including the highly macerated and degraded forms of the huminite/vitrinite macerals and macrinite, has long been established. In particular, the fungal- and bacterial-related origins of macrinite (not always by that name) have been discussed by [Waksman \(1930\)](#), [Barghoorn \(1949, 1952\)](#), [Duparque and Delattre \(1953a,b\)](#), [Stach \(1956\)](#), [Teichmüller \(1958\)](#), [Beneš \(1959, 1960a,b, 1969\)](#), [Beneš and Kraussová \(1964, 1965\)](#), [Lapo and Malán \(1981\)](#), [Cross and Phillips \(1990\)](#), [Eble et al. \(1994\)](#), [Belkin et al. \(2009, 2010\)](#), [Pujana et al. \(2009\)](#), [Sweeney et al. \(2009\)](#), [Hower et al. \(2010a, 2011b, 2013a\)](#), [Taylor and Krings \(2010\)](#), [Hower and Ruppert \(2011\)](#), [O'Keefe and Hower \(2011\)](#), and [Dai et al. \(2012b\)](#), in particular, their figure 9). For example, [Teichmüller's \(1958\)](#) sapropelic "Ried-Moor Kohle" facies had more fungi than less-degraded facies, an observation repeated in various forms by [Waksman \(1944\)](#), [Straka \(1960\)](#), [Schwab \(1962\)](#), [Wallace and Dickinson \(1978\)](#), [Moore and Hilbert \(1992\)](#), [Esterle and Ferm \(1994\)](#), and [Moore et al. \(1996\)](#).

The fungal association with wood and associated plant parts (resins, leaf cuticles, roots, etc.) begins while the tree is still standing and continues beyond the death and burial of the remains of the tree. Fungus plays a role in diseases of living, but potentially stressed, trees ([Beute and Benson, 1979](#); [Loo, 2009](#)). Fungus can also invade and ingest resins ([Hower et al., 2010a](#)). Mycorrhizal fungi are associated with plant roots and, as such, can play a role in the breakdown of the peaty soil for the

nutritional benefit of the host living plant (Hower et al., 2011a; Moore et al., 1996; Waksman, 1944).

Within the cycle of the standing tree to the degraded peat soil, fungi such as the modern white-rot fungus *Phanerochaete chrysosporium*, are highly efficient degraders of lignin (Huang et al., 2010) and are vital contributors to soil lignin transformations (Blanchette, 1984a,b, 1991; Otjen and Blanchette, 1982, 1984; Thevenot et al., 2010). The breakdown of cellulose by brown-rot fungi and, in particular, lignin by white-rot fungi allows arthropod detritivores to more easily ingest wood (García Massini et al., 2012; Hower et al., 2011a,b, 2013a). Microbial decomposition is a surface process; bacteria and fungi attack the surfaces of plant cells and plant cell walls. Thus any process that reduces the particle sizes of organic debris increases the rate of microbial decomposition. Invertebrate detritivores influence the rate of decomposition and decomposition processes by shredding plant detritus and increasing the surface area to volume ratio of organic debris (Lavelle, 1987; Lavelle and Spain, 2001; Swift et al., 1979).

A number of terrestrial arthropod coprolite classifications based on size, shape, density and contents have been proposed for ancient peat (Baxendale, 1979; Habgood et al., 2004; Raymond et al., 2001; Scott and Taylor, 1983). Both the size and content of arthropod coprolites from peat may be important for understanding the origin of macerals. All workers classifying coprolites in peat have distinguished between small fecal pellets mostly 30–40 µm in diameter, probably produced by oribatid mites or small collembola, and larger coprolites up to 8 mm in diameter (Baxendale, 1979; Habgood et al., 2004; Raymond et al., 2001; Scott and Taylor, 1983). Based on the widespread occurrence of small fecal pellets in tunnels found in pieces of wood, oribatid mites may have been the primary terrestrial arthropods involved in wood decomposition in Pennsylvanian through Triassic wetlands (Cichan and Taylor, 1982; Kellogg and Taylor, 2004; Labandeira et al., 1997). However, millipedes are also known to bore into wood, and some of the larger coprolites from the Lewis Creek coal-ball deposit contain wood fragments (Scott and Taylor, 1983).

Coprolites from Pennsylvanian permineralized peat fall into two categories based on contents. Most are filled with vegetative debris or seeds, and are brown to black in color. This category includes most fecal pellets, which probably derived from oribatid mites and small collembola, as well as coprolites, which probably derived from larger collembola, millipedes, and Paleozoic detritivorous insects (Scott and Taylor, 1983 and references therein). Coprolites filled with spores and pollen are golden or amber-colored (Baxendale, 1979; Taylor and Scott, 1983). These coprolites are less common than brown to black coprolites, and range in size from fecal pellets, 30–40 µm in diameter, to large coprolites, 5–6 mm in diameter (Raymond et al., 2001). The earliest spore-filled coprolites come from the Early Devonian Rhynie Chert (Habgood et al., 2004).

Coprolites within coals, coal balls, and modern peat provide a record of the fate of the partially digested remains of plant tissues (Chaloner et al., 1991; D'Rozario et al., 2011; Kubiena, 1955; Labandeira and Phillips, 2002; Lesnikowska, 1990). Bacteria (Bradley, 1966; Lavelle and Spain, 2001), nematode larvae (Botella et al., 2010), and fungi (Baxendale, 1979; Baxter, 1975; Beute and Benson, 1979; D'Rozario et al., 2011; Loo, 2009) are known to be associated with coprolites. The role of arthropods' or other predators' herbivory or detritivory of leaves (Baxendale, 1979; Lesnikowska, 1990; Raymond et al., 2001) and stems (Beute and Benson, 1979; Chaloner et al., 1991; D'Rozario et al., 2011; Labandeira and Phillips, 2002; Rolfe, 1985; Rothwell and Scott, 1983; Scott, 1977; Taylor and Scott, 1983) is established at least from the Pennsylvanian, with seed ferns and tree ferns being the most common targets in the Pennsylvanian and Permian based on coprolites and damage to plant tissues (D'Rozario et al., 2011; Hower et al., 2013a; Labandeira and Phillips, 2002; Lesnikowska, 1990; Raymond et al., 2001; Rothwell and Scott, 1983). Evidence for herbivory includes wounds with signs of a reaction to the damage (Chaloner et al., 1991; Lesnikowska, 1990).

Further bacterial decomposition can occur in the gut of the detritivores (Lavelle and Spain, 2001). Studies of modern environments such as Mud Lake, Florida, demonstrate that cyanobacteria can pass through the digestive system of chironomid larvae, *Cladocera*, ostracods, and copepods with minimal damage (Bradley, 1966, 1970, 1973; Bradley and Beard, 1969). The latter environment, with little or no organic material outside of what is produced in the lake, is an extreme example of gyttja (organic-rich mud) as a coprogenic material as described by Hansen (1959) after von Post (1862). Intact spores (Habgood et al., 2004; Labandeira, 1998; Scott, 1977), undamaged parenchyma cells (Taylor and Scott, 1983), and fragments of wood and seeds (Baxendale, 1979; Taylor and Scott, 1983) have been found in Paleozoic coprolites. The cycle can be repeated many times as coprophilous fungi colonize and digest coprolites (Greif and Currah, 2007; Harper and Webster, 1964; Kuthubutheen and Webster, 1986; Massee and Salmon, 1901, 1902; Page, 1939; Richardson, 2002; Wicklow and Hirschfield, 1979). Both coal-ball deposits that have been studied in detail show evidence of coprophagy by terrestrial arthropod detritivores (Raymond et al., 2001; Scott and Taylor, 1983). Hower et al. (2011b, 2013a) identified macrinite with abundant fusinite, liptinite, vitrinite, and funginite inclusions as a consequence of one or more of digestive/excretion/(with or without) coprophilia cycles. Simple macrinite, without inclusions, may represent the preservation of the white-rot and/or brown-rot fungus processing of wood, without the subsequent digestion and excretion.

**2.2.3.5. Microbial and resin interactions.** Resins, insects, and fungus are part of a balance of predators (insects), accidental or intentional pathogens (fungus or bacteria; fungi are both saprophytes and facultative parasites (Barron, 2003)), and the attempts at protection on the part of the tree (resin). Insects may serve as the entry point of pathogenic fungus into the tree (Christiansen et al., 1999; Loo, 2009), with an example being fungi *Ophiostoma ulmi* and *O. novo-ulmi* carried by bark beetles, resulting in Dutch Elm disease (Loo, 2009; Martín et al., 2010; Moser et al., 2010).

Plants produce essential oils (De Alwis et al., 2009; López et al., 2007), alkaloids (Holmes et al., 2008), enzymes, lignin (Panov and Terziev, 2009), terpenoids (Bednarek and Osbourn, 2009; Collado et al., 2007; De Alwis et al., 2009; Dutta et al., 2009), or tannins (Exarchos and Given, 1977; Given, 1984; Given et al., 1983) as protection against microbes. Fungi alter their environment by producing antibiotics (Bose, 1953; Holding and Franklin, 1965; Waksman, 1944; Wallace and Dickinson, 1978; among others) or by re-using terpenoids against other fungal species (Rikkinen and Poinar, 2000). Resiniculous fungi, such as *Chaenothopsis bitterfeldensis*, use resin as a food source (Rikkinen, 1999; Rikkinen and Poinar, 2000; Tibell and Titov, 1995).

As discussed by Hower et al. (2010a), notations of fungal associations with resins, with or without insects, are numerous in the resin and amber literature but have been relatively ignored in coal petrology studies. Funginite, as the fossil spores, sclerotia, and hyphae of fungal bodies, has long been recognized in fossil amber (Berkeley, 1848; Thomas, 1848) and in coal (Jeffrey and Chrysler, 1906).

**2.2.3.6. Gondwana perspectives on inertinite.** Coals in the Permian Karoo Sequence of southern Africa (Tanzania southwards, including Mozambique, Zambia, Namibia, Botswana, Zimbabwe, Malawi, Swaziland, and South Africa) typically have high inertinite content, with variable semifusinite and vitrinite contents, and are low in sulfur compared to Laurasian coals (Cadle et al., 1993; Hower et al., 2012). Coals in the main Karoo Basin (South Africa) formed in similar tectonic environments to Carboniferous coals in the USA (Appalachian, Illinois, and Michigan Basins) and the Wolfgang and Blair Athol Basins of Australia (Cadle et al., 1993), in that they formed on the distal or passive margin of a retroarc foreland basin. The coal-forming period spanned the early to mid-Permian, during which the palaeoenvironment changed from glacial, to marine, fluviodeltaic, fluvio-lacustrine, to

dry–wet desert conditions (Cadle et al., 1993; Falcon, 1989). The post-glacial climatic setting resulted in a distinct floral assemblage, different from the Northern Hemisphere Laurasian coals. The coal-forming peat mires of southern Africa were colonized by sub-arctic mosses; cold-temperate conifers; cool-temperate deciduous forests with *Gangamopteris* and *Glossopteris* floras; and less dominant lycopods, ferns, cordiaaleans, and early gymnosperms (Falcon, 1986, 1989). In the main Karoo Basin, coals accumulated in fresh to brackish mires with low subsidence rates and well-drained mires, resulting in oxidation of the peat and microbial degradation, and hence the high inertinite content on a regional scale. However, according to Falcon (1986), as much as 60% of the inertinite group macerals may be classified as semi-reactive macerals. Hagelskamp and Snyman (1988) discussed semi-reactive inertinite in detail and determined that humified and partially vitrinitized components underwent partial oxidation during rafting prior to redeposition, giving rise to low-reflecting inertinite (Fig. 7). The predominance of detrital macerals (such as inertodetrinite) and carbominerite over massive and cellular macerals (such as vitrinite and semifusinite) may indicate a hypautochthonous deposition of the peat rather than in situ oxidation due to water-level fluctuations (Hagelskamp and Snyman, 1988), however, these deposits require further work using modern methodologies.

In South Africa, two metamorphic events affected the rank of the coal (Snyman and Barclay, 1989). There is a general west–east increase in coal rank across the main Karoo Basin (De Jager, 1983), with vitrinite reflectance values around 0.55% in the western, Free State coalfield (Snyman and Barclay, 1989), to meta-anthracite (>4.0%) (Cadle et al., 1993) in the eastern Vryheid Kwazulu-Natal coalfields and Swaziland. Localized contact metamorphism was a result of dolerite intrusions (sills and dykes), which intruded the entire Karoo Basin during the Jurassic (Drakensberg Formation), following the close of the coal-forming period. Snyman and Barclay (1989) determined that the coal was of a lignite stage of coalification at the time of the intrusions, as indicated by the narrow contact aureoles and low thermal diffusivity of the coal (implying that the magmatic heat was consumed in water evaporation). Superimposed burial metamorphism determined the current rank of the coals, with the easterly increase related to the fairly steep geothermal gradient towards the east. In many locations across southern Africa, the dolerite intrusions resulted in devolatilization of the coal (fairly extensive in the Witbank Coalfield, South Africa), or pockets of semi-anthracite (Zimbabwe) and anthracite (Namibia) (Cairncross, 2001), but these tends to be localized. Natural graphite occurs between the coal and the dolerite intrusions in some localities (Fig. 8).

Along with the west–east trend in rank across South Africa, there is a general increase in vitrinite content and sulfur, and a decrease in mineral matter (Cadle et al., 1993). The higher vitrinite content (average of 35% by vol. in the No. 4 seam (Cadle et al., 1993)) is related to the more distal setting of the peat mires, and accelerated rates of subsidence; the higher rank is related to the palaeotopography and the thickness of the Earth's crust in the Kwazulu-Natal Basin. The very high inertinite content (>75% by vol. in the No. 4 seam) and lower rank of the Free State coals is probably related to the elevated stable position of the coalfield, lower geothermal heat, and greater oxidation and microbial degradation of the organic matter.

Beyond the stable cratonic shelf, which typified much of the Karoo Basin in South Africa, localized intra (such as the Waterberg Coalfield, or the Tanzania coalfields) or intercratonic (such as the Livingstonia Coalfield, Malawi) graben or half-graben basins enabled the accumulation of sediments and vegetation for longer periods of time in embayments (Cairncross, 2001; Falcon, 1989). These embayments resulted in more extensive sequences of coal seams, frequently with higher vitrinite contents, towards the mid-to-late-Permian. Coal rank ranges from subbituminous to bituminous medium rank A–D in these embayments, with a predominance of medium rank C bituminous coals, and isolated coking coal deposits (such as Wankie Coalfield, Zimbabwe;

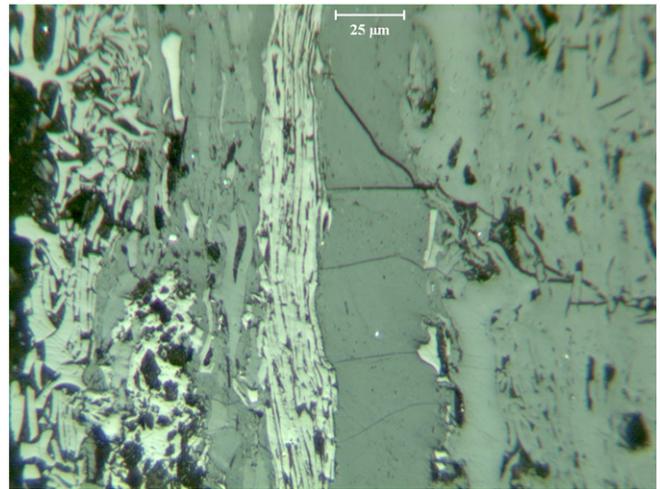


Fig. 7. Low-reflecting inertinite in a Permian Gondwana coal from Mozambique (image Moz 01 09). Photo by Hower and Wagner.

Tshikondeni Coalfield, South Africa; Moatize Coalfield, Mozambique). Regional metamorphic controls on rank were a result of higher geothermal gradients, due to thinner crust, across the intracratonic belts and rifted basin coalfields (Cairncross, 2001). In addition, localized contact metamorphism caused significant localized changes in coal rank. While

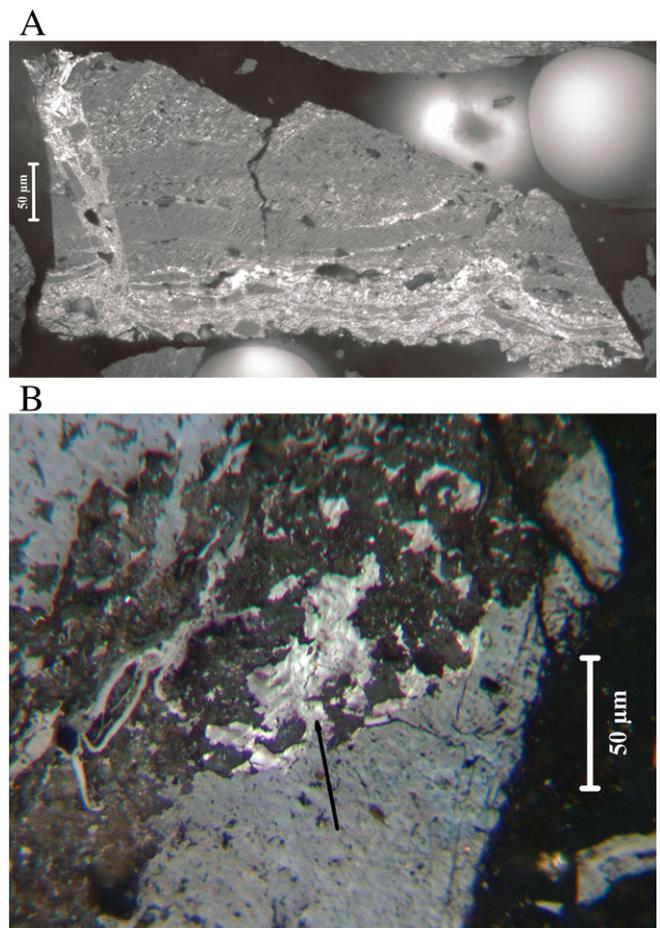


Fig. 8. A. Natural graphite occurring in a coal adjacent to a dolerite intrusion in South Africa. B. Pyrolytic carbon in Permian Matla coal, Witbank coalfield, about 0.7%  $R_{\text{mean}}$ . Photos by Hower and Wagner.

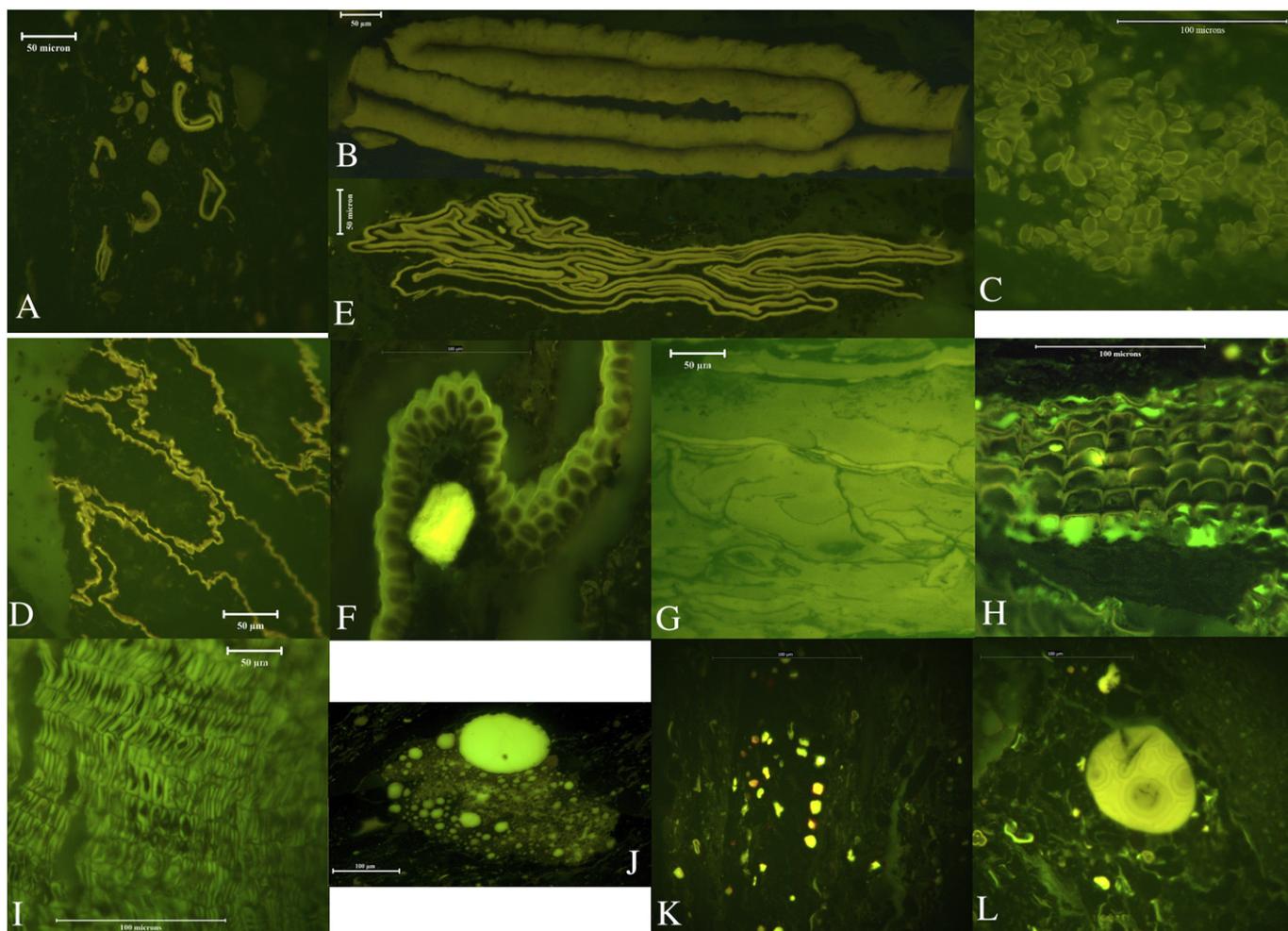
inertinite and high mineral matter tends to typify the southern African coals, frequent deposits of high vitrinite (up to 80%) and low ash (below 10%, generally associated with inertinite rather than vitrinite) occur. This can largely be attributed to upper deltaic plain/braided plain depositional environments versus lacustrine/lagoonal settings (Semkiwa et al., 1998). Mid-Triassic coal seams occur in the Beaufort area of South Africa (Molteno Formation), to the south of the main Karoo Basin, where low-volatile bituminous to anthracite, vitrinite- and mineral-rich seams occur, but are not extracted.

#### 2.2.4. Liptinite macerals

Liptinite macerals, which derive from hydrogen-rich plant and algal materials, include sporinite, composed of plant spores and pollen; cutinite, composed of plant cuticles and cuticular layers; resinite, composed primarily of resin, but also of waxes, balsam, latex, oil, etc.; alginite, derived from algae; suberinite, formed from corkified cell walls found primarily in bark; bituminite, a decay product of algae and plankton; chlorophyllinite, a rare maceral composed of chlorophyll that is preserved only under the most anaerobic conditions; liptodetrinite, or detrital bits of liptinite; and exsudatinite, a secondary maceral that forms from other liptinite macerals and perhydrous vitrinite during coalification and flows to fill voids (Taylor et al., 1998). Fluorinite, previously

listed as a separate maceral, is now agreed by ICCP to be a type of resinite (W. Pickel, personal communication to J. O'Keefe on 28 March 2006).

Terrestrial-sourced liptinite macerals (most sporinite, cutinite, resinite, and suberinite) are more common in coal than alginite, bitumenite, and exsudatinite, although each may occur in specific coals. Alginite is an important indicator of standing water in mire systems, especially those containing oligotrophic to dystrophic lakes (Taylor et al., 1998). The occurrence and aspect of several of these macerals is known to change through time. Sporinite generally has two forms (Fig. 9), miospores/pollen and megaspores. Through palynology, it is well known that specific types of sporinite are indicative of depositional environments. For example, *Taxodium* sp. (bald cypress) pollen indicates deposition in acidic, freshwater mires with standing water for at least part of the year, while *Nypa* sp. (Nipa palm) pollen indicates deposition in a brackish water setting, either a mangrove swamp or a tropical salt marsh. Resinite, too, may be an indicator of depositional environment or plant contributions to mires. For example, fluorinate-type resinites are indicative of substantial contributions of essential-oil rich plants, such as *Myrica* (myrtle) to the mire, and resinites in ducts within textinite/telovitrinite are indicative of gymnospermous plants, and thus acidic mire conditions. Cutinite macerals change significantly through time, and range from the "typical" Carboniferous toothed forms, to very thick, multicellular forms seen in waxy



**Fig. 9.** Changing aspects of liptinite through time. All images are in blue-light illumination. A. Sporinite. Miospores in the Carboniferous Moscow Basin; Seredin collection coal; Photo by Hower. B. Sporinite. A megaspore in the Permian from China; Photo by Hower. C. Sporinite. Pollen from the middle Eocene Claiborne Formation of Kentucky, USA; Photo by O'Keefe. D. Cutinite. "Classic" toothed cutinite from the Carboniferous of Mongolia; USGS World Coal Quality Collection; Photo by Hower. E. Cutinite. Thin cutinite; Photo by Hower. F. Cutinite. Thick, layered cutinite from the middle Eocene Claiborne Formation of Kentucky, USA; Photo by O'Keefe. G. "Barkinite"-type suberinite. Liptinitic material separated by very thin cellular divisions; Photo by Hower. H. Suberinite. Thin form from the middle Eocene Claiborne Formation of Kentucky, USA; Photo by O'Keefe. I. Suberinite. Thick form from the middle Eocene Claiborne Formation of Kentucky, USA; Photo by O'Keefe. J. Resinite. Jurassic from the Sinai Peninsula; Photo by Hower. K. fluorinite-type Resinite in textinite through the middle of field of view. Middle Eocene Claiborne Formation of Kentucky, USA; Photo by O'Keefe. L. Resinite. Zoned resinite from the Middle Eocene Claiborne Formation of Kentucky; Photo by O'Keefe.

leaves found in Cretaceous–modern miros, to very thin forms seen in many modern sub-tropical to temperate tree leaves (Fig. 9). Suberinite has been the subject of intense debate in recent years, as there has been question of whether very robust forms from Asia, termed “barkinite” are a separate maceral or not (Hower et al., 2007; Sun, 2002, 2003, 2005; Zhong and Smyth, 1997). While it has been generally accepted that suberinite as a maceral is absent prior to the rise of angiosperms, it is now clear that from the Permian–Cretaceous many transitional gymnospermous and proto-angiospermous plants likely had corkified bark tissue. Suberinite may, in fact, be present much earlier in wetland environments, as it evolved as a waterproofing mechanism. Molecular phylogenies for the evolution of the suberin macromolecule point toward its existence more than 400 Mya. It is well known that *Equisetum* at least through the Jurassic has a clear Casparian strip (Channing et al., 2011), which, like cork tissues, is composed of suberin. This feature likely extends through the lineage of Equisetales. It is also well known that suberin has an overall low preservation potential (Versteegh and Riboulleau, 2010), therefore, we may be either not recognizing it in the fossil record or it may not be preserved. This is another area where significant research is needed.

### 3. Coal rank

An ideal coal rank parameter should exhibit a distinct change with respect to the metamorphic temperature. Unfortunately, no single parameter is useful over the entire coal rank range. Petrographers rely on vitrinite reflectance but it can be problematical in the lower coal ranks. Chemical rank parameters are a function of the maceral and mineral contents and the non-mineral inorganic geochemistry, although this can be overcome if electron microprobe techniques are used to evaluate the composition of particular maceral components (e.g. Gurba and Ward, 2000; Ward et al., 2005). Even traditional analyses based on vitrinite concentrates, as proposed by Pattiesky and Teichmüller (1960), are suspect if comparisons are made between samples with variations in organic sulfur, as an example. In general, though, alternatives to vitrinite reflectance are moisture and heating value in low-rank coals, with heating value being used up to about the high volatile A/medium volatile bituminous boundary; volatile matter in medium volatile bituminous through semi-anthracite ranks, with carbon (ultimate analysis) also used in some national systems; and hydrogen in anthracite. Theoretically, Vicker's microhardness could be used (Hower et al., 2008b), however this is not routinely accomplished. Microhardness, as with reflectance, is measured on inclusion-free vitrinite and, as with the use of hydrogen, it has the advantage of exhibiting a sharp increase through the anthracite rank range.

#### 3.1. Mechanisms of coal rank

Coal rank is generally considered to be a function of some combination of heat, pressure, and time.<sup>6</sup> The relationship between coal rank and depth of burial, with the implication of higher temperatures at greater depth, is commonly known as Hilt's Law (Hilt, 1873). In many corners of the world, this concept, in some form, dominated the discussions of coal metamorphism. Indeed, much of the discussion in Taylor et al. (1998; with a strong bias towards pre-1990 literature) centers on depth-of-burial and geothermal-gradient models. In North America, however, some geologists advocated tectonic pressure models (Stadnichenko, 1934; White, 1925; Wood et al., 1969). Their objections to heating were based in part on the realization that known igneous intrusives had no apparent influence on western Pennsylvania coal rank (Stadnichenko, 1934) but, also, knowingly or not, on misinterpretations of coal rank in

the Bernice Field (Sullivan Co., Pennsylvania) semianthracite (Hower, 1997).

Damberger (1991) noted that the influences of pressure and heating cannot be fully resolved because (1) coals were more deeply buried in the past, (2) paleogeothermal gradients in tectonically active areas were greater in the past, and (3) thermal fluids influenced coal rank. Gresley (1896) anticipated the latter argument, particularly in opposition to the pressure-related hypotheses. The notion of hydrothermal influences on coal metamorphism generally did not gain traction until the 1980s with the observations of Oliver (1986, 1992). Just within the Appalachians, connections between the flow of thermal fluids and coal metamorphism were made with respect to the Pennsylvania Anthracite Fields (Bayan and Hower, 2012; Daniels and Altaner, 1990; Daniels et al., 1990, 1994, 1996; Harrison et al., 2004; Hower and Gayer, 2002; Hower et al., 1993; Juster et al., 1987; Ruppert et al., 2010); western Pennsylvania bituminous coals (Zhang and Davis, 1993); the Valley Fields, Virginia (Lewis and Hower, 1990); and the Central Appalachian bituminous coals (Collins, 1993; Evans and Battles, 1999; Goldhaber et al., 2003; Hearn et al., 1987; Hower and Rimmer, 1991; Hower et al., 1991; Ruppert et al., 2010; Sakulpitakphon et al., 2004); and Alabama (Pashin et al., 1990; Winston, 1990).

The timing of coal metamorphism can range from near instantaneous in the case of igneous intrusions, to thousands of years (or less) in geothermal systems, to millions of years for burial diagenesis (Barker, 1989). Barker (1991) argued for a  $10^0$ – $10^1$  year organic metamorphism event in the California (USA)/Baja California Norte (Mexico) Cerro Prieto geothermal system. While most coal metamorphic events would likely not have such a brief duration, Harrison et al. (2004) did argue that the Pennsylvania Anthracite Field metamorphism could have had a duration of less than 300 days. In any case, the time near the peak metamorphic temperature, for example within 15 °C sensu Hood et al. (1975), is a critical factor in any consideration of thermal metamorphism.

#### 3.2. General physical changes to coal with rank

As coals increase in rank, there is a general decrease in moisture, increase in heating value, and overall increase in density. Coal's macroscopic luster generally becomes glossier as rank increases and fracture becomes more smoothly conchoidal. These physical changes are the result of geochemical changes addressed more fully below.

Together, the geochemical transformations of coal, regardless of type, result in changes in coal cleat. Coal cleat varies with rank such that spacing between cleats decreases as rank increases from lignite to medium volatile bituminous, then increases slightly again in anthracites as cleats begin to anneal (Law, 1993). Coupled with these rank-induced changes in coal cleat are variations associated with type. In general, vitrains tend to have closer cleat spacings than do durains (Stach et al., 1982; Taylor et al., 1998). Additionally, low ash coals have more closely spaced cleats than high ash coal. An example of these variations can be seen in the middle Eocene low-rank coals in the upper Mississippi Embayment, USA. Cleats in the high-ash Carlisle lignite ( $R_o \sim 0.23\%$ ) are spaced more widely than those in the somewhat lower ash and more mature Fancy Farm lignite ( $R_o \sim 0.33\%$ , and both are more widely spaced than the low-ash portions of the Weakly lignite ( $R_o \sim 0.35\%$ ). Fuller treatment of the interplay of coal type, and rank on cleating can be found in Law (1993), Laubach et al. (1998), and Dawson and Esterle (2010).

#### 3.3. Maceral changes with rank

Maceral changes with rank are well-documented in the literature (Teichmüller, 1989). The increase in vitrinite reflectance with increasing metamorphic grade is a function of the increasing aromatization of vitrinite molecules with progressive metamorphism (Teichmüller and Teichmüller, 1958; Section 4, below) and the pressure-related alignment of the aromatic molecules (Hower and Davis, 1981; Levine and

<sup>6</sup> Coal rank, as noted in Section 1.1, is one of the fundamental parameters controlling coal quality. For further discussion of the technological aspects of coal rank variation, see van Krevelen (1993), arguably the most fundamental presentation of the topic; Jasieńko (1978); Taylor et al. (1998, chapter 9), and Suárez-Ruiz and Crelling (2008).

Davis, 1989a,b). Although the reflectance of liptinite macerals converges with vitrinite reflectance though the medium to low volatile reflectance ranges, the macerals do not disappear, they just become more difficult to distinguish against the vitrinite background (Taylor et al., 1998, p. 122). Turner and Randall (1923) and Turner (1925, 1930a,b, 1932) identified sporinite in anthracite following heat etching of the polished surface. The form shown in Turner (1925), his Fig. 11) is clearly the Pennsylvanian spore *Punctatisporites minutus* (identification by Cortland Eble in 1994, unpublished data). Turner (1925) also showed several examples of resinite in the latter paper.

Sporinite is seen in Fig. 10A and B, views of the same anthracite particle, and Fig. 10C. In all cases illustrated, the sporinite can be clearly distinguished from the vitrinite (Fig. 10A and B) or the surrounding detrital macerals and clay (Fig. 10C). At the anthracite stage, liptinite macerals display a higher maximum reflectance and lower minimum reflectance than the surrounding vitrinite; the random reflectances of liptinite and vitrinite being the same (Hower, 1978; Hower et al., 1994a,b).

Funginite, perhaps because of its apparent rarity compared to vitrinite and also due to its lesser abundance in Pennsylvanian coals compared to Mesozoic and younger coals, has been rarely, if ever, described in anthracite. The funginite in the anthracite illustrated on Fig. 10D has the internal structure commonly seen in Cenozoic funginite.

#### 4. Organic geochemical changes with rank

The process of coalification encompasses physical and chemical changes that begin shortly after deposition and burial and continue during thermal maturation. Since vitrinite is the dominant maceral group in most coals, most studies of the organic chemistry of coal have been on the transformation of vitrinite group macerals.

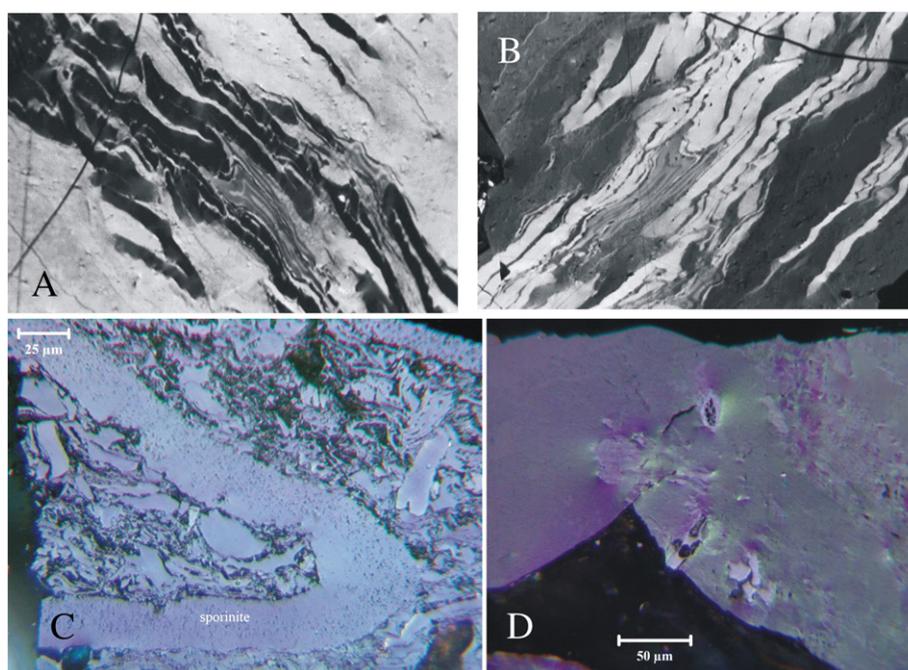
The processes of wood transformation begin with elimination of cellulose and transformation of lignin (Fig. 11; Drobnik and Mastalerz, 2006; Hatcher et al., 1981; Russell and Barron, 1984). Transformation of lignin initiates with the cleavage of aryl ether bonds, including hydroxyl and methoxyl groups and the cleavage of  $\beta$ -O-4 aryl ethers (Hatcher and Clifford, 1996). These reactions produce phenolic-OH,

resulting in the formation of catechol-like structures. This, in turn, leads to alkylation of aromatic rings (Botto, 1987). Another important early coalification process is the cleavage of aryl-O bonds in lignin, specifically methoxyl groups attached to the aromatic rings through a demethylation process (Hatcher et al., 1981; Stout et al., 1988). Consequently, the chemical structural composition of subbituminous coal is that of the lignin precursor after losing its methoxyl groups through demethylation and dehydroxylation and after the loss of its side chain hydroxyls. Retention of plant structures in low-rank coals indicates that maceration of the wood structure does not necessarily accompany the chemical transformations.

Evolution from subbituminous coal to bituminous coal involves transformation of catechol-like structures to phenols (Hatcher and Clifford, 1996) that subsequently condense to form aryl ethers or dibenzofuran-like structures. Aromatic ring condensation is indicated by increased abundance of naphthalenes and fluorenes. Ring closure and aromatization of the alkyl side chains result in increased aromaticity. With progressive coalification, phenolic structures are lost in favor of benzene-like structures. These benzene-like structures (with their associated aliphatic side-chain carbons) may undergo condensation and form polycyclic aromatic ring systems present in higher rank coals (e.g., Fakoussa and Hofrichter, 1999).

Semi-anthracite and anthracite are characterized by highly aromatic structure. At the onset of the semi-anthracite rank, a sudden molecular orientation develops and small aromatic stacks aggregate into clusters (Oberlin et al., 1980; Rouzand et al., 1991). Interlayer spacing gradually decreases into anthracitic rank and further on the metamorphic progression to graphite (Wilks et al., 1993).

Those chemical and structural transformations of organic matter result in progressive changes in the composition of coal. With regard to elemental chemistry, increase in carbon content and decrease in oxygen content are most prominent characteristics (Stach et al., 1982). The loss of oxygen is particularly pronounced at the subbituminous C/B rank, although individual oxygen groups have their unique maturation path, often increasing at early stages of coalification and diminishing to disappearance later (Drobnik and Mastalerz, 2006; Petersen et al., 2008). Hydrogen content of about 5.0% persists through ranks including



**Fig. 10.** A & B. Orthogonal views sporinite in Pennsylvanian anthracite ( $R_{\max} > 5\%$ ), Southern Anthracite Field, Pennsylvania. Scale = ca. 250  $\mu\text{m}$  on long axis. Oil immersion, 50 $\times$  objective, crossed-polars. C. Sporinite in European anthracite ( $R_{\max} = 3.89\%$ ). Scale = 25  $\mu\text{m}$ . Oil immersion, 50 $\times$  objective, crossed-polars with wavelength plate. D. Funginite (upper center) in vitrinite in European semianthracite ( $R_{\max} = 2.86\%$ ). Scale = 50  $\mu\text{m}$ . Oil immersion, 50 $\times$  objective, crossed-polars with wavelength plate. Photos by Hower.

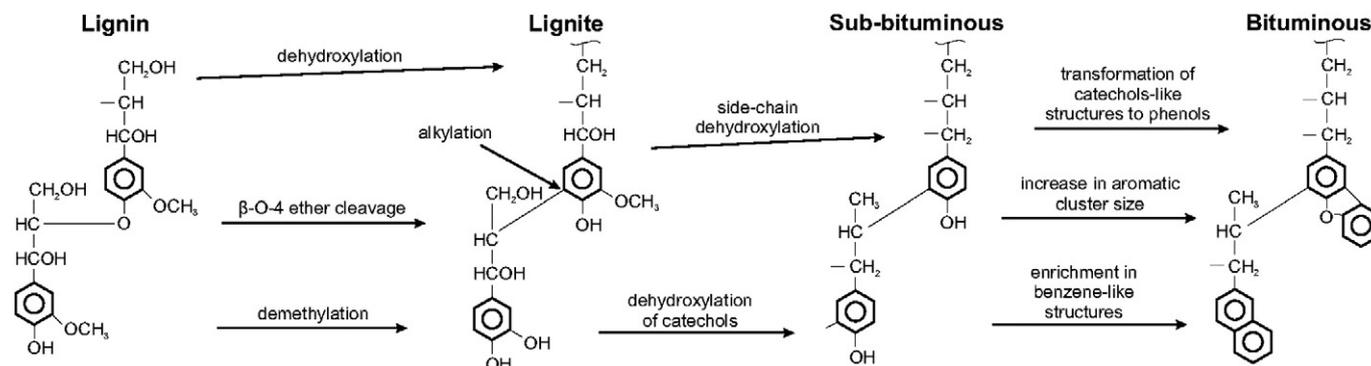


Fig. 11. Chemical transition from wood to peat, showing the elimination of cellulose and transformation of lignin. Modified after Hatcher and Clifford (1996).

medium volatile bituminous and decrease in low volatile bituminous coal and anthracite (Strapoć et al., 2011). Nitrogen content in most coals is below 2% and does not show a consistent relationship with rank.

Although maceral groups follow these general trends in elemental composition, each has its own maturation path because of differences in original chemical composition. In general, macerals increase in aromaticity from liptinite to vitrinite to inertinite (Dyrkacz et al., 1981; Given, 1984; Hatcher and Clifford, 1996; Hower et al., 1994; Maroto-Valer et al., 1998; Mastalerz and Bustin, 1993b; Nip et al., 1992; Rimmer et al., 2006; Stankiewicz et al., 1996; Walker and Mastalerz, 2004). Therefore, the maceral groups respond differently to the coalification processes, as demonstrated in Fig. 12 for carbon and oxygen contents determined in-situ by electron microprobe on individual macerals (Mastalerz and Bustin, 1993a,c). Similar trends are shown for macerals in Australian coals by Ward et al. (2005). Such data demonstrate that sporinite reaches the same carbon content as vitrinite at vitrinite reflectance of ~1.35%, and that sporinite matches the carbon content of semifusinite at vitrinite reflectance of ~1.70%. The abundance and distribution of functional groups also evolves with progressive coalification. Fig. 13 demonstrates the differences between vitrinite, sporinite, and semifusinite with increasing rank (Mastalerz and Bustin, 1993b). The main difference between vitrinite of lignite and subbituminous ranks compared to vitrinite of the higher ranks is the appearance of aromatic hydrogen bands in the 700–900  $\text{cm}^{-1}$  out-of-plane region, and their subsequent increase in contribution with rank. Aliphatic hydrogen bands in the 2800–3000  $\text{cm}^{-1}$  stretching region are very small

in huminite/vitrinite of lignite and subbituminous ranks, prominent in vitrinite of high and medium volatile bituminous ranks and decrease in contribution at higher ranks. In comparison to vitrinite, aliphatic hydrogen bands in sporinite show consistent decrease in intensity from subbituminous to bituminous rank. Similar to vitrinite, sporinite experiences a distinct increase in aromatic hydrogen bands in the 700–900  $\text{cm}^{-1}$  region. Like vitrinite, semifusinite, experiences gradual loss of aliphatic hydrogen in 2800–3000  $\text{cm}^{-1}$  going from high-volatile to anthracite rank, although absolute intensities of the bands of this region are significantly smaller in semifusinite compared to vitrinite. These aliphatic bands are undetectable in semifusinite of lignite rank. There is also significant difference between semifusinite of lignite and semifusinite of other ranks with regard to the aromatic hydrogen bands in the 700–900  $\text{cm}^{-1}$  region; they are very small in lignite and prominent in high volatile bituminous and higher ranks.

Based on the chemical and structural data, numerous chemical models of coal have been proposed over decades, beginning with Fuchs and Sandhoff (1942). The models differ because of the specifics of the chemical data used. However, it is commonly accepted that the coal is a macromolecule built of condensed aromatic rings joined by ether, alkoxy and sulfur bridges, and having hydroxyl groups, methoxyl groups and carboxyl groups attached to the aromatic nuclei. Comprehensive reviews of molecular modeling of coal structure can be found in van Krevelen (1993) and Vandenbroucke and Largeau (2007).

Hydrocarbons are only minor constituents of coal, especially in lignite and subbituminous coals. However, their composition provides

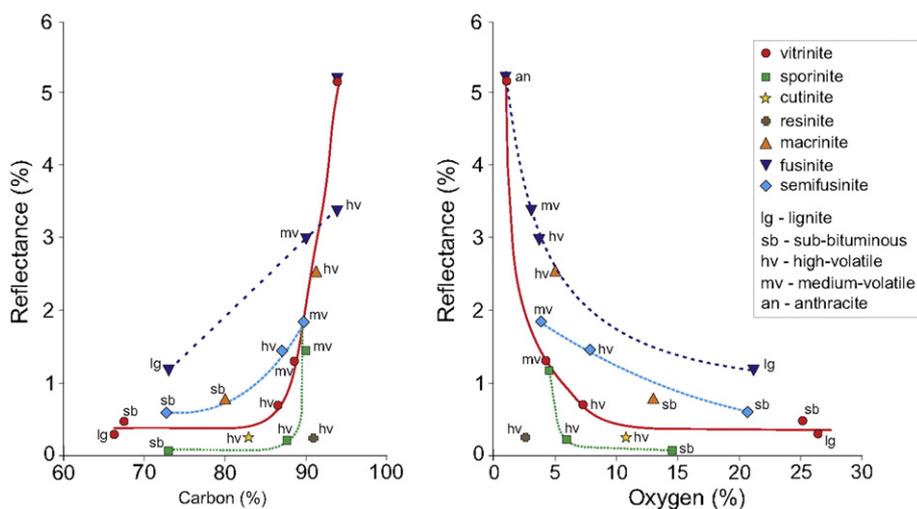
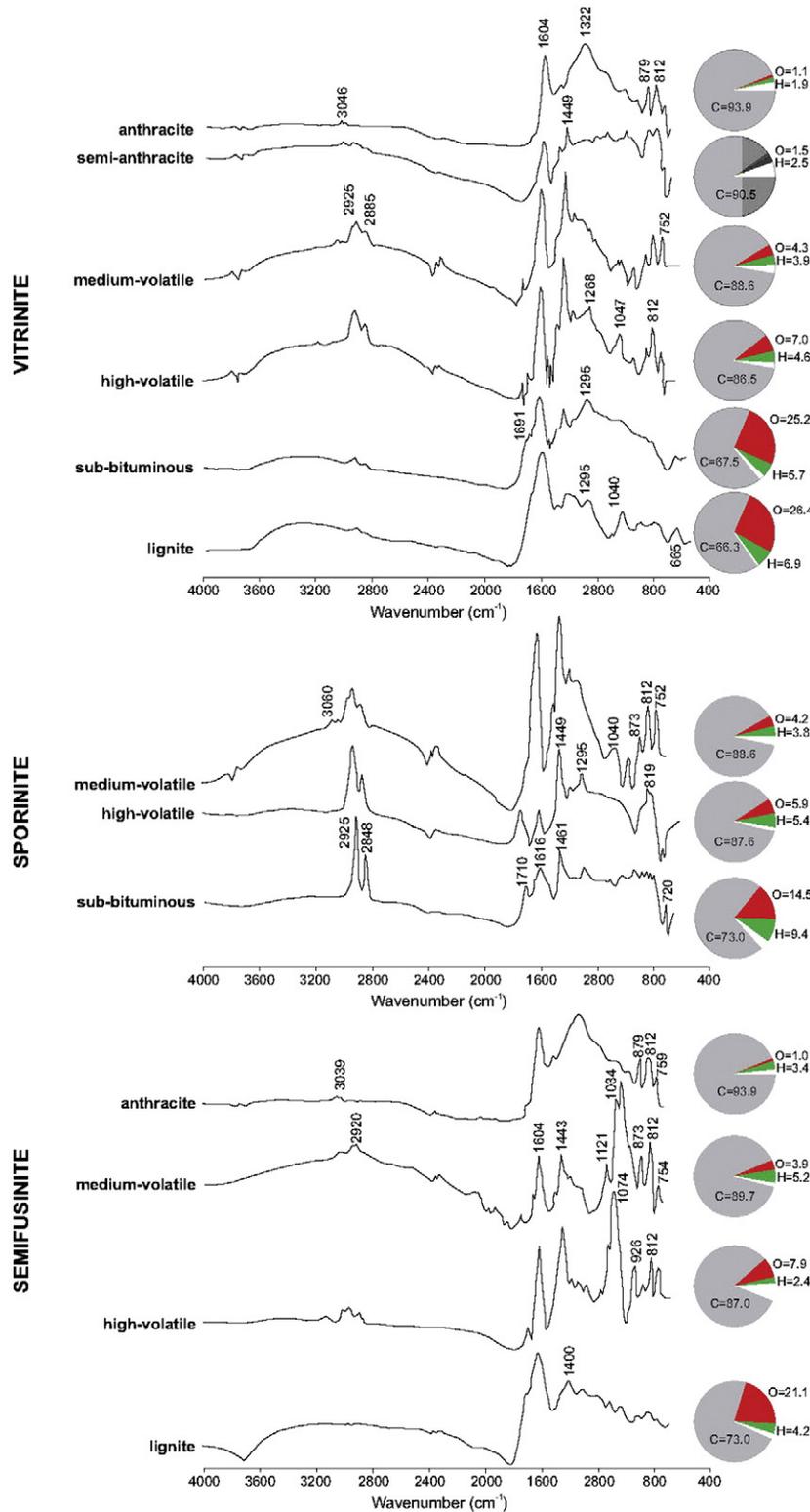


Fig. 12. Carbon and oxygen content of individual macerals of different coal ranks. Each point of the diagrams represent average value of numerous analyses obtained with electron microprobe. Note different paths of vitrinite, sporinite, semifusinite, and fusinite. Other macerals (macrinite, resinite, cutinite) were available only sporadically and this is why are represented only once or twice.

Modified from Mastalerz and Bustin (1993a).

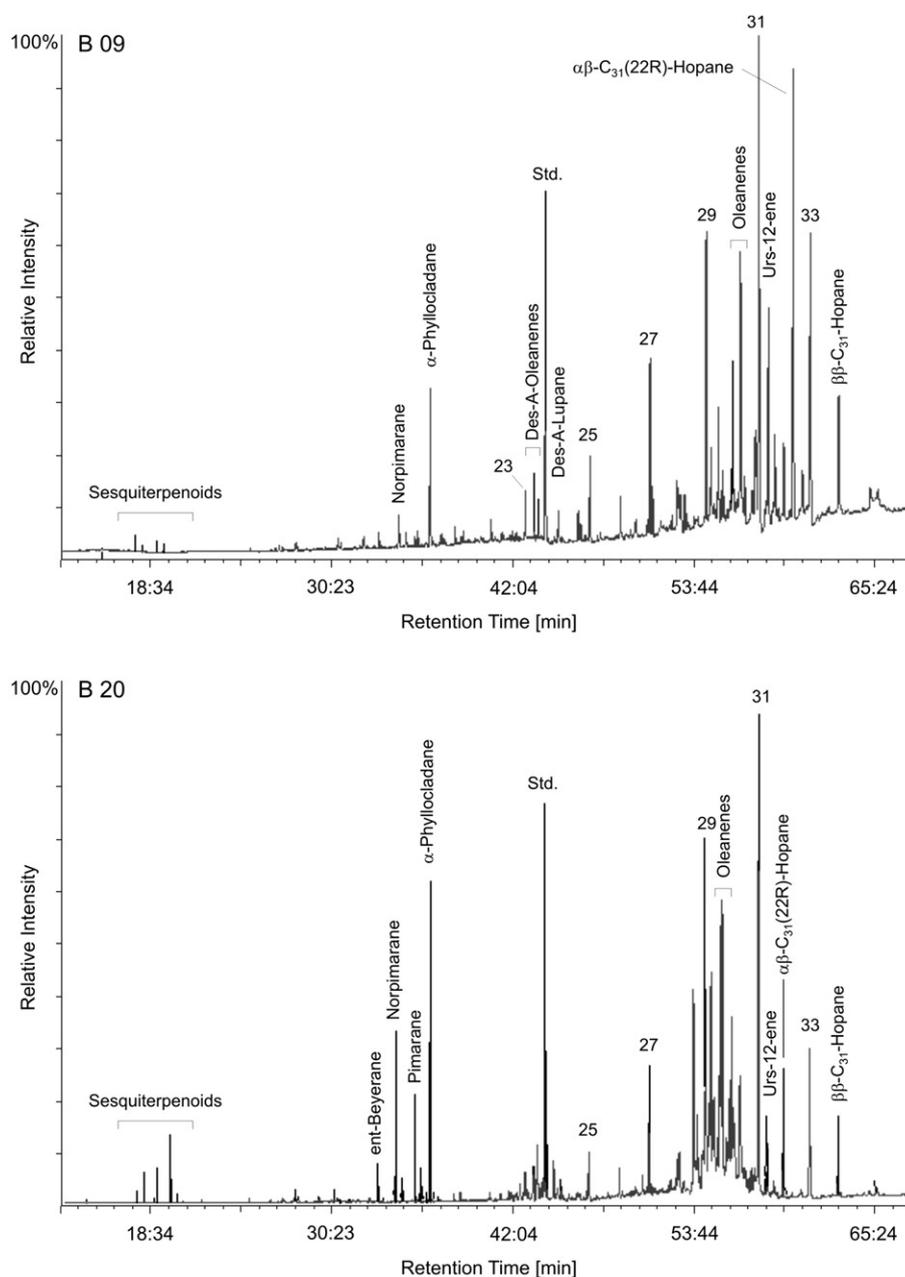


FTIR spectra and elemental composition of vitrinite, sporinite and semifusinite from lignite to anthracite.

**Fig. 13.** Micro-FTIR spectra of vitrinite, sporinite, and semifusinite of coals of different ranks. Pie diagrams next to them represent elemental composition of these macerals. Modified from Mastalerz and Bustin (1993b).

information on depositional environment and changes in peat-forming vegetation (Bechtel et al., 2003, 2008; Otto and Wilde, 2001; Otto et al., 1995, 1997; Stefanova et al., 2002). Examples of the compositional diversity of hydrocarbon composition found in subbituminous coal are shown in Fig. 14. Relationships between petrography-based facies

indicators and biomarker composition indicate that the preservation of plant tissue (as shown using the tissue preservation index, or TPI; Diessel, 1992) is strongly influenced by the contribution of conifers to peat formation, whereas gelification index (GI) reflects bacterial activity, that is in turn governed by the water-table within the mire



**Fig. 14.** Gas chromatograms (total ion current) of the saturated hydrocarbon fractions of the samples B 09 and B 20 from the Bourgas coal seam. *n*-Alkanes are labeled according to their carbon number. Std. = Standard (deuterated *n*-tetracosane). Refigured from Bechtel et al. (2005).

(Bechtel et al., 2004, 2005). The diagnostic biomarkers of gymnosperms (i.e. diterpenoids) and angiosperms (oleanane-, ursane-, and lupane-derivatives) decrease in intensity with increasing rank. They are usually not found in bituminous coal. Instead *n*-alkanes, hopanes, biphenyls and polycyclic aromatic hydrocarbons become predominant.

The content of  $17\beta$ ,  $21\beta(\text{H})\text{-C}_{31}$  hopane relative to the sum of  $17\beta$ ,  $21\beta\text{-C}_{31}$  hopane plus  $17\alpha$ ,  $21\beta(\text{H})\text{-C}_{31}$  hopane can be used for rank assessment in lignite and subbituminous coal. Within lignite the measured ratios are in the range of 0.5 to 0.7 (Mackenzie et al., 1981). Lower values (<0.5) are reported from subbituminous coal. Within the subbituminous stage, the  $\beta\beta$ -hopanes disappear, and the ratio of 22S over  $22\text{S} + 22\text{R}$  isomers of the  $\alpha\beta\text{-C}_{31}$  hopanes (Mackenzie et al., 1982) can be used for rank assessment up to vitrinite reflectance values of 0.6% ( $R_r$ ). In high-rank coals, information is mainly based on the methylphenanthrene index (MPI 1; Radke and Welte, 1983) and the empirical relationship between MPI 1 and vitrinite reflectance (Radke

et al., 1984). This parameter is valid up to vitrinite reflectance values of 2.0% ( $R_r$ ).

## 5. The impact of type and rank on coal as a source rock and reservoir rock

Coal can be both a hydrocarbon source rock and also a reservoir rock. Indeed, in many coalbed methane plays it acts as both simultaneously, with methane both being generated and adsorbed into pores. Coal has long been known to be a source rock. Its potential as a source rock depends in part upon type, in that more-hydrogen rich coals, such as those rich in alginite (torbanites), cutinite, and also suberinite (Mukhopadhyay and Hatcher, 1993; Powell et al., 1991), have a greater generating potential than less-hydrogen rich coals. This is, however, rank dependent, as liptinite macerals (see above) become less aliphatic with increasing rank. A full overview of the oil generating potential of

coals can be found in Wilkins and George (2002), with additional information in Levine (1993).

Coalbed methane resources are more difficult to tie to either type or rank, as they can be of biogenic, thermogenic, or mixed origin (Levine, 1987). Methane is a product of the decomposition process of primarily woody material. In general, coalbed methane is of biogenic origin from the peat to the low end of the high-volatile A bituminous rank, and its production occurs in both humic and sapropelic coals (Clayton, 1998) due to the interaction of microorganisms with organic matter and CO<sub>2</sub> in the peat or coal seam coupled with the location of the water table (Moore, 2012). After this point, thermogenic methane is produced (Clayton, 1998; Flores, 1998), however, biogenic gas may remain adsorbed in the coal micropores, thus yielding mixed gas upon production. Additionally, biogenic gas can continue to form at any rank where conditions are right for microorganism to colonize the system; thus, there can be multiple generations of methane of multiple origins present in a single seam. Many excellent reviews detailing coalbed methane systems exist, most notably Moore (2012) and Flores (2014).

## 6. Conclusions

Coal rank and type are fundamentally separate parameters. Coal type is directly related to the environment of peat formation, the climate of peat formation, and the decompositional history of the mire being studied. In general, coal type can be defined as being either humic or sapropelic. Humic coals are those that develop from primarily helophytic plant materials and may be either autochthonous, forming in place, as is the case of many domed peats; or allochthonous, transported to the site of deposition, such as may be seen in coals formed in log-jams. Sapropelic coals develop from primarily aquatic plant materials, or from helophytic remains that have undergone significant decomposition (e.g., detrohuminite/detrovitrinite and macrinite). Both humic and sapropelic facies of a single coal are present in the rock record. It is not uncommon for coals to begin as a collection of alginite, sporinite, and humo-/vitro- and inertodetrinite in a depression, and as the depression fills to become increasingly humic as the mire transitions from being topogenous to being ombrogenous.

Coal rank provides a reflectance overprint upon coal type. In general, vitrinite macerals become increasingly reflective as rank increases, as do liptinite and inertinite macerals. By the top end of low volatile bituminous, it becomes increasingly difficult to differentiate vitrinite and liptinite macerals. By the anthracite rank, this becomes challenging. Etching of high-rank coals allows for individual maceral identifications to be made, and thus for coal type to be identified. The use of white light with crossed polars also allows the differentiation of liptinite macerals. Both humic and sapropelic coals occur as anthracites, notably in the Zhejiang Province of China.

Our understanding of the depositional settings of peat and coal are robust, especially in North America, Australia, China, and Europe. Significant work remains to be done in Africa, central Asia, and in South America. Beyond specific locations, there are numerous questions remaining about the effect of Milankovitch and other orbital cyclicity on the initiation, accumulation, and preservation of peats, as well as on coals' occurrence within existing sequence stratigraphic models. Existing models for peat formation need to be reexamined and possibly revised in light of new data on the formation and preservation of tropical, temperate, and cool-climate peats.

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