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International Journal of Coal Geology 60 (2004) 169–196

International Journal of COAL GEOLOGY

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Geological controls on the sulphur content of coal seams in the Northumberland Coalfield, Northeast England

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Received 14 January 2004; accepted 18 May 2004
Available online 12 October 2004

Abstract

The sulphur content of coal is an important consideration when developing reserves for exploitation, driven by emission limits from power stations becoming more stringent. Variations in the sulphur content of Westphalian A and B coals from the predominantly freshwater Northumberland Coalfield, Northeast England, were studied according to their regional, stratigraphic and in-seam location. The observed variation in sulphur content spatially increases towards the source area away from more marine influenced areas, with increased sulphur content through time linked to changes in the general depositional environment as conditions became more marine-influenced. A model of basinal surface water and groundwater flow driven by post-depositional source area tectonism is thought to have played only a minor role in contributing secondary sulphur to the coal. However, the isotopic composition of coal pyrite shows a similar range in composition to that of pyrite and other sulphides from the North Pennine Orefield along the southern margin of the coalfield, suggesting an additional potential source of secondary sulphur, as sulphur-rich fluids were expelled northwards through the coal measures during early Permian Variscan transpression from the south. The Westphalian A and B are interpreted as third-order depositional sequences, defined by third-order maximum flooding surfaces. Each sequence is made up of several coal-bearing fourth-order parasequences, which tend to be more brackish to marine in character, on either side of the third-order maximum flooding surfaces when base level was relatively high. The lowest sulphur coals are confined to the lower to middle, relative low stand part of the Westphalian A third-order base level curve and the lowest part of the Westphalian B third-order base level curve. This difference is attributed to a more rapid rise of base level in the Westphalian B. The stratigraphic and spatial distribution of coal sulphur has been used as a guide to prediction of reserve identification for surface mining operations.

Keywords: Geological controls; Sulphur content in coal; NE England

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doi:10.1016/j.coal.2004.05.003

1. Introduction

The sulphur content of coal seams is a major factor in resource development and utilisation. Studies of the sulphur content of coal seams from within the same stratigraphic interval, deposited under a variety of marine and non-marine depositional environments, show that coals with marine roof rocks have higher sulphur contents than those with fresh- or brackish-water roof rocks (Ferm et al., 1979; Diessel, 1992; Chou, 1997). High sulphur coals are typically those in which sulphate-bearing seawater has infiltrated the original fresh- to brackish-water peat mire during or after deposition (Williams and Keith, 1963; Altschuler et al., 1983; Hunt and Hobday, 1984; Mastalerz et al., 1997). Thus, there is a strong environmental control on the sulphur content of coal seams (Phillips and Bustin, 1996; Dai et al., 2002). This relationship is emphasised by studies of modern peats subjected to marine influences where sulphur enrichment is mainly attributed to sulphate-reducing bacteria promoting precipitation of pyrite in peat (Casagrande et al., 1977; Cohen et al., 1984; Querol et al., 1989; Phillips et al., 1994).

Despite closure of all but one of the deep mines, Westphalian coal is still an important opencast resource in the Northumberland Coalfield of Northeast England (Fig. 1). Most opencast coal mined in the Northumberland Coalfield is used for power generation and lower sulphur coals are generally sought to comply with emission limits. Although the coals were deposited under predominantly freshwater conditions, their sulphur content, mainly in the form of pyrite, is variable and some high-sulphur coals have freshwater roof rocks, incompatible with a simple environmental control model. No systematic study of the geological controls on the sulphur content of coal seams in the Northumberland Coalfield has been undertaken to date. However, Spears et al. (1999), in their study of coal seams from four British Coalfield areas outside Northumberland, attributed their sulphur content to: (1) post-depositional controls, (2) contemporaneous channels providing direct access to peat mires and (3) source rock control.

Coals in the Northumberland Coalfield are high volatile bituminous coals (ASTM Class 2, Bituminous Group 3, High Volatile A (hvAb) coals). Coal rank increases to the south, in response to higher thermal gradients over shallowly buried basement rocks, and decreases towards the east. Ash yields range from 2.2% to 17%, but with the majority of the coals having ash of <10%. The sulphur content, which ranges from 0.6% to 8.4% S_{tot}, is present in the form of organic sulphur, sulphate and pyrite cubes, framboids, nodules, lenses parallel to bedding, locally deformed anastomosing veins and veinlets, and cleat-fills (Turner, 1999). The Westphalian A and B peat-forming mires in Northumberland evolved from low-lying, brackish- to freshwater wet forest mires and freshwater dry mires (Chen, 1990). They represent a limited range of peat-forming environments (Haquebard and Donaldson, 1969) dominated by forest moor with some open moor, reed moor and local limnic components (Chen, 1990; Navarre, 1998). Rapid lateral changes in peat-forming environments from reed moor to swamp forest, wet forest and dry forest over a distance of <10 km, have been documented by Navarre (1998) for the lower Westphalian B on the Northumberland coast. The dominance of bright coal lithotypes with a high vitrite and clarite content indicates the importance of arborescent vegetation (Chen, 1990).

In this paper, we examine the geological factors influencing the regional, stratigraphic, and between- and within-seam variations in sulphur content of coals in the Westphalian A and B Coal Measures in the Northumberland Coalfield of Northeast England (Fig. 1). This in turn has been used to categorise the potential an area may have to produce a low-sulphur coal. A sequence stratigraphic approach has been adopted for predictive modelling of the stratigraphic controls on variations in sulphur content of the coals, and isotopic analyses of various forms of pyritic sulphur in the coal have been employed to try and constrain the source of the sulphur. The surface and subsurface data on which this study is based includes new data provided by H.J. Banks from its own sites (Turner, 2002), existing National Coal Board data available from the British Geological Survey (National Coal Board Seam Records, 1946–1969; National Coal Board Northumberland Seam Maps, 1957) and additional new data from the Plenmeller opencast coal site, West Northumberland

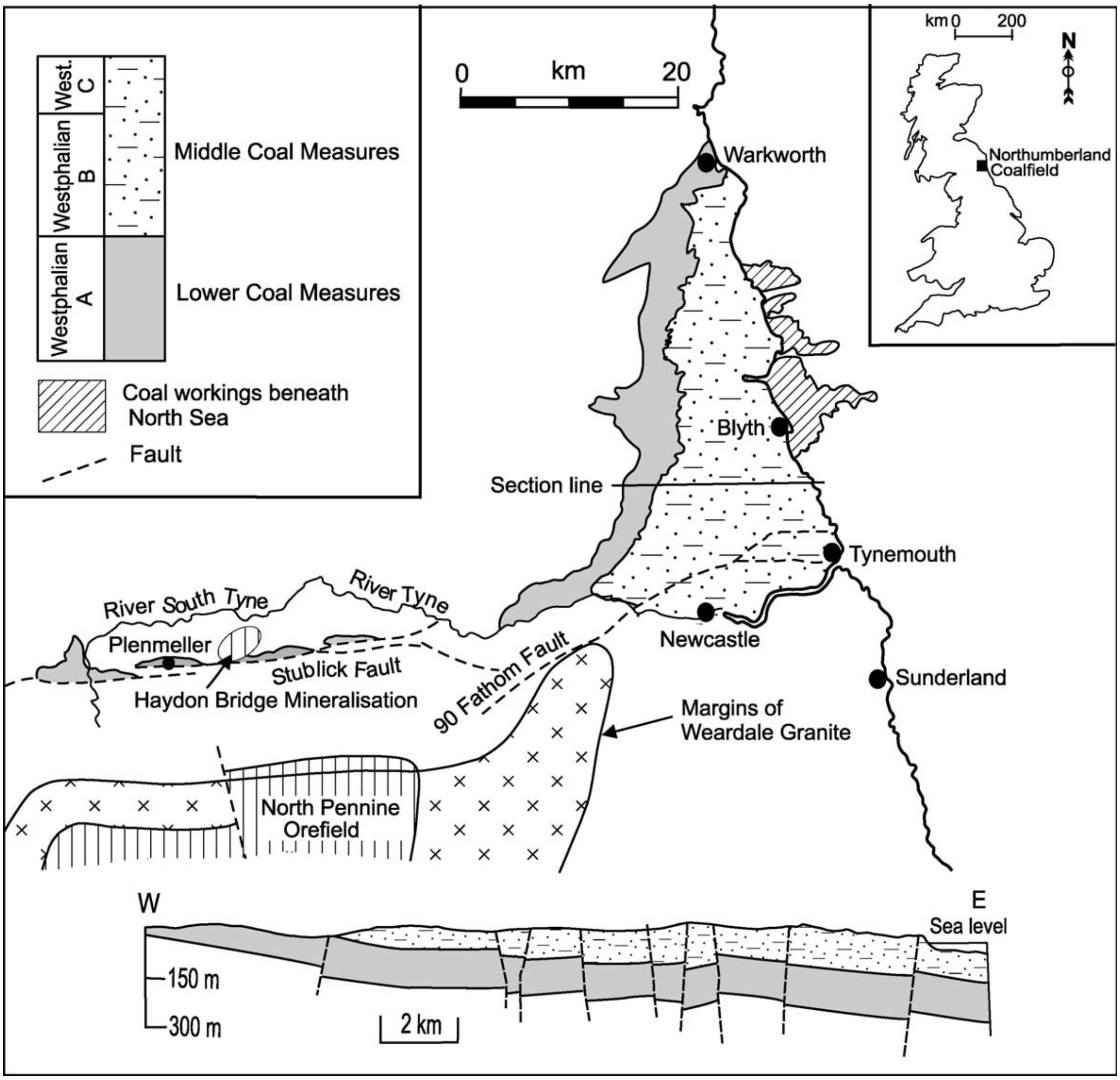


Fig. 1. Generalised geological map and cross-section of the Northumberland Coalfield showing the distribution of Lower and Middle Coal Measures and their relationship to Westphalian Stages.

(Turner, 1999), which has now been closed and restored.

2. Geological setting

In Northern England, early Carboniferous crustal extension, possibly induced by subduction and closure of a back-arc seaway, south of Britain, led to the development of fault-bounded half-graben basins (Leeder, 1988). The largest of these basins, the Northumberland Trough, developed along the line of the Iapetus suture, which marks the line of closure of the Pre-Atlantic Iapetus Ocean (Freeman et al., 1988; Scotese and McKerrow, 1990). The Northumberland Trough comprises the Solway basin in the west and the Northumberland basin in the east, separated by a shallow shelf (Fig. 2). The basin-fill shows an early to mid Carboniferous syn-rift phase of subsidence and sedimentation followed by a late Dinantian to West-

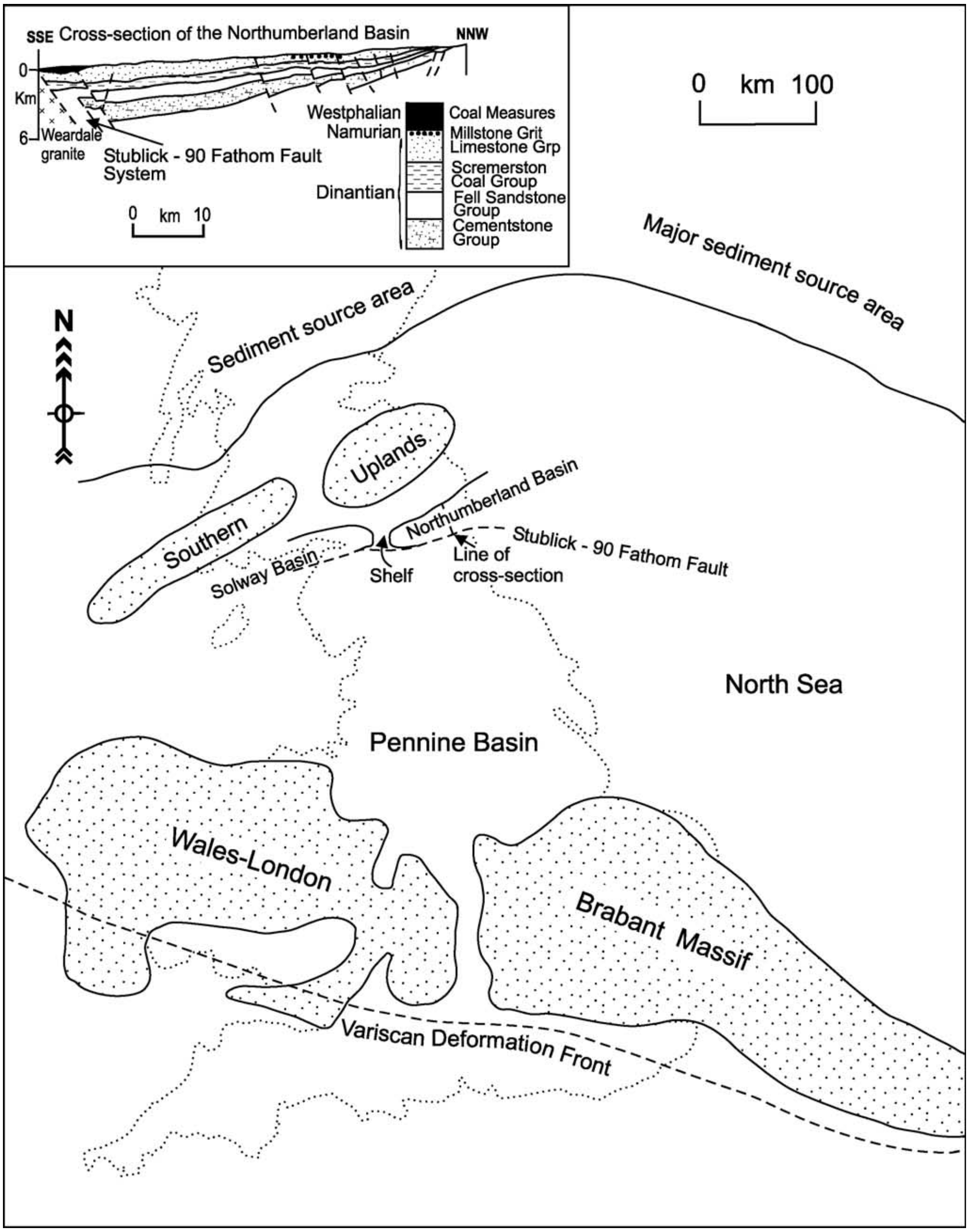


Fig. 2. Generalised palaeogeographic map of the Pennine Coal Basin showing the location of the Northumberland Basin, major sediment source areas and the final position of the Variscan tectonic front. The inset section top left shows the geometry and internal stratigraphy of the Northumberland Basin.

phalian post-rift thermal subsidence phase (Leeder and McMahon, 1988) (Fig. 2, inset). At least 4 km of sediment was deposited in the Northumberland basin during the Carboniferous, with the deepest part of the basin and maximum sediment thickness in the south, adjacent to the basin margin, Stublick-Ninety Fathom Fault zone (Fig. 2). Interaction between shallow marine shelf environments in the southwest and the sediment provenance terrain to the north and northeast led to frequent shifts of the contemporary shoreline, resulting in rapid lateral facies changes.

Sedimentation commenced in the Mississippian (lower Carboniferous) with deposition of local alluvial fan conglomerates that pass rapidly basinwards into arid, coastal alluvial plain channel sandstones, floodplain siltstones and dolomites of the Cementstone Group (Fig. 2, inset). Increasing humidity, coupled with tectonic uplift of the source area to the northeast, led to basinward progradation of sandy, bedload-dominated, perennial braided streams depositing the Fell Sandstone Group. As clastic supply from the northeast diminished, marine influences from the southwest increased, and transgressive–regressive cycles of limestone, shale and sandstone, capped by a seatearth and locally thick coals of the Scremerston Coal Group were laid down. A marine-dominated depositional phase followed, when vertically stacked 'Yoredale' type cyclothems of limestone, shale and sandstone with locally developed seatearths and thin coals, were deposited. The succession is divided into a Lower, Middle and Upper Limestone Group with a thick, laterally persistent limestone, the Great Limestone, at the top of the Middle Limestone Group defining the base of the Namurian (Johnson, 1980). Above the Great Limestone marine influences and limestones die out and the succession is dominated by fluvial sandstones, siltstones and mudstones including some 50 m of coarse Millstone 'grit' at the top (Fig. 2, inset), recording renewed tectonism in the hinterland.

The Limestone Group hosts extensive mineral veins of the North Pennine Orefield in Durham and adjoining areas of Northumberland. The veins mainly contain galena and sphalerite, with lesser amounts of pyrite and marcasite and occasionally chalcopyrite and pyrrhotite. The orefield gangue minerals are zoned with fluorite surrounded by barite, witherite or calcite. Mineral zonation is temperature-controlled with high temperature fluids in the centre of the orefield and lower temperature fluids along the margins. The Caledonian Weardale granite (Fig. 2, inset), intruded into lower Palaeozoic basement rocks, set up a hydrothermal convection system which squeezed hot brines out of the sedimentary basin shales (Sheperd et al., 1982), stripping out metals as they migrated through the Carboniferous, Lower Palaeozoic and Caledonian Weardale granitic rocks. The main phase of mineralisation is of Permian age, although potential mineralising fluids are thought to be still circulating today.

At the beginning of the Westphalian, Northumberland was located along the northern margins of the much larger equatorial Pennine Coal Basin which extended across central England and the southern North Sea into Europe (Fig. 2). Basin formation is attributed to regional strike slip (Dewey, 1982) or N–S directed extensional stress (Leeder, 1982) related to evolution of the Variscan orogen. Since the Pennine basin developed north of the active Variscan orogenic mountain front (Fig. 2), the basin-fill is relatively undeformed (Besly, 1988). Only the Lower and Middle Coal Measures, i.e. the Westphalian A, B and part of C, are preserved in Northumberland (Fig. 1). They comprise a 750-m-thick, predominantly alluvial succession, deposited on a broad, low-lying, coastal alluvial plain, transitional into paralic and more marine-influenced environments to the south and southwest (Fig. 3). Thus, the coastal plain was influenced by relative sea level fluctuations even though well separated from the shoreline and very distal (>200 km) from open marine influences. The alluvial plain was drained by predominantly low-gradient rivers of variable morphology flowing south and southwest, either directly or via numerous crevasse-splays which built out into fresh- to brackish-, shallow-water (<20 m deep) interdistributary lakes, as small delta systems (Fielding, 1984; Haszeldine, 1984; Chen, 1990; Guion et al., 1995; Fulton and Williams, 1988; Turner and Smith, 1995; O'Mara, 1995) (Fig. 3). Colonisation of the prograding delta top by peat vegetation was followed by a rise in base level and by increased accommodation space as lake waters drowned the peat mire, ultimately preserving it beneath lacustrine sediments as thin (<15 m), coal-capped coarsening-upward deltaic sequences of shale, siltstone and sandstone (Fig. 3, inset). The abundance of crevasse-splays implies frequent overbank flooding, which together with the low sinuosity of many trunk river channels, indicates that they were probably of the perennial type, characterised by fluctuating discharge.

During the Westphalian rates of sedimentation closely balanced rates of subsidence with only minor fluctuations. As a result shallow-water fluvio-deltaic

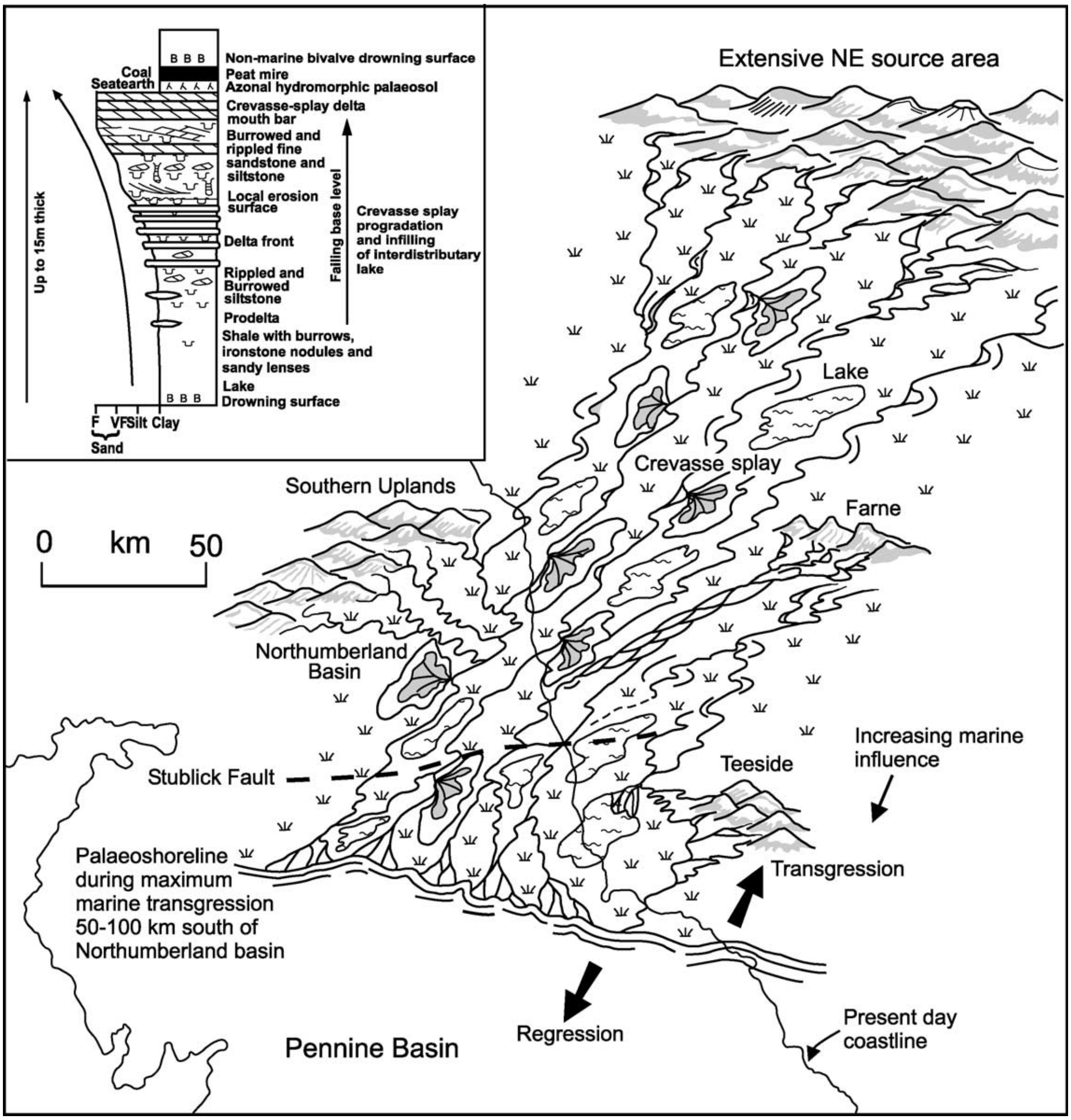
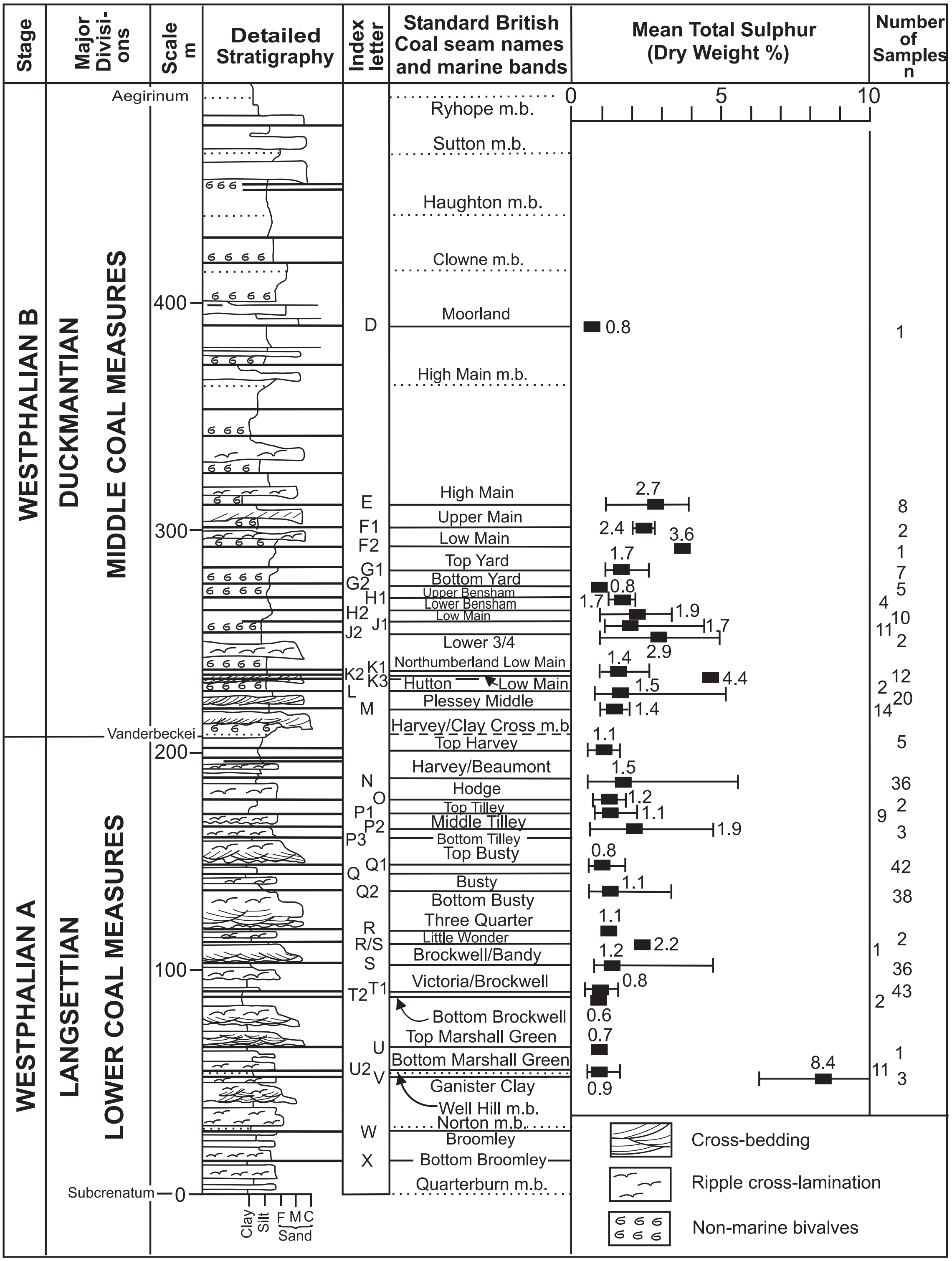


Fig. 3. Palaeogeographic model for the Westphalian A and B Coal Measures in Northumberland showing location of sediment source areas and approximate position of the palaeoshoreline during maximum marine transgression from the south. The inset section top left shows the main characteristics of a typical coal-capped coarsening-upward sequence produced by crevasse-splay progradation into an interdistributary lake.

conditions were maintained throughout deposition (Quirk, 1993; O'Mara and Turner, 1999), interrupted periodically by short-lived marine transgressions from the south and southwest, leading to the deposition of thin marine bands. These marine flooding events were of variable magnitude: those which record the greatest

Fig. 4. Stratigraphic variation in mean sulphur values for Westphalian A and B coal seams in the Northumberland Coalfield. Bars through boxes represent the range of values around the mean. Range bars terminated at 10% sulphur. The index letters for named coal seams are taken from the British Coal classification scheme. Note that the name of coal seams in Northumberland may differ from the county names assigned by British Coal. In this study, the county name has been adopted wherever possible.


Stage
Major Divisions
Scale m
Detailed Stratigraphy
Index letter
Standard British Coal seam names and marine bands
Mean Total Sulphur (Dry Weight %)
Number of Samples n
WESTPHALIAN B
DUCKMANTIAN
MIDDLE COAL MEASURES
WESTPHALIAN A
LANGSETTIAN
LOWER COAL MEASURES
Aegirinum
Vanderbeckei
Subcrenatum
Ryhope m.b.
Sutton m.b.
Haughton m.b.
Clowne m.b.
Moorland
High Main m.b.
High Main
Upper Main
Low Main
Top Yard
Bottom Yard
Upper Bensham
Lower Bensham
Low Main
Lower 3/4
Northumberland Low Main
Hutton
Low Main
Plessey Middle
Harvey/Clay Cross m.b
Top Harvey
Harvey/Beaumont
Hodge
Top Tilley
Middle Tilley
Bottom Tilley
Top Busty
Busty
Bottom Busty
Three Quarter
Little Wonder
Brockwell/Bandy
Victoria/Brockwell
Bottom Brockwell
Top Marshall Green
Bottom Marshall Green
Ganister Clay
Well Hill m.b.
Norton m.b.
Broomley
Bottom Broomley
Quarterburn m.b.
Clay
Silt
F M C
Sand
Cross-bedding
Ripple cross-lamination
Non-marine bivalves

marine encroachment and water depths are interpreted as maximum flooding surfaces, corresponding to the *subcrenatum*, *aegiranum* and *vanderbeckei* marine bands (Flint et al., 1995) defining the boundaries of the Westphalian A (Langsettian) and B (Duckmantian) successions (Fig. 4). Although these maximum flooding surfaces developed marine conditions over most of their aerial extent, in Northeast England they are mostly *Lingula*-rich mudstones, indicative of brackish rather than fully developed marine conditions. This is because marine conditions rarely penetrated this far north due to progressive shallowing northwards of the transgressions, and their dilution by freshwater fluvial systems draining southwards. Thus, base level rise was not of sufficient magnitude to impose truly marine conditions on Northumberland, where only the proximal expression of these marine bands is seen. Nevertheless, these marine transgressions were an integral part of the depositional system and they had a profound influence on fluctuations in base level and possibly on the sulphur content of the coal given that the principal source of this sulphur is dissolved sulphate in seawater.

3. Sulphur content of coals in the Northumberland Coalfield

Plots of organic sulphur and pyritic sulphur content against total sulphur content for the Top Busty, Bottom Busty and Beaumont coal seams in the Northumberland Coalfield show the organic sulphur content to be relatively constant compared to pyritic sulphur (Fig. 5). Thus, variations in the total sulphur content are largely controlled by pyrite in coal. In Northumberland, the average sulphur content for the coalfield is 1.67%, and there is a general increase in sulphur content towards the more marine-influenced coalfields to the south and west, except for the South Wales coalfield (Gayer et al., 1999). In this study, a high sulphur coal is defined as one that contains >1.3% total sulphur, as this is the cut-off value for the sulphur content of coals acceptable for local power generation.

Variations in the sulphur content of coal seams in Northumberland were examined according to: (1) regional location across the entire coalfield from west to east; (2) stratigraphic location throughout the Westphalian A and B succession; and (3) between- and within-individual seams.

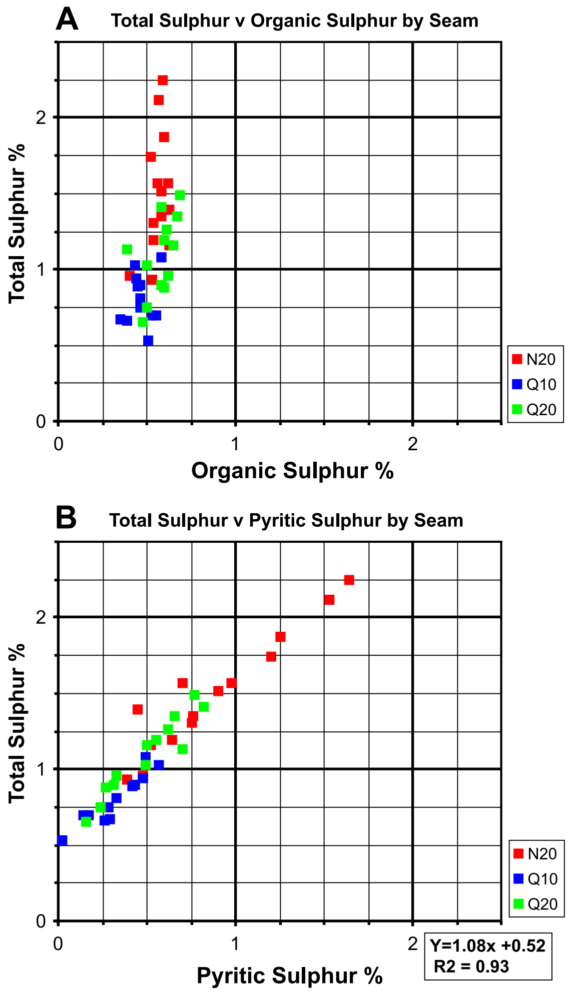


Fig. 5. (A) Total sulphur versus organic sulphur and (B) total sulphur versus pyritic sulphur for the Beaumont (N20), Top Busty (Q10) and Bottom Busty (Q20) coal seams from the Westphalian A of the Northumberland Coalfield.

3.1. Regional trends in the sulphur content of coal seams

Regional sulphur distribution maps of NE England have been compiled for eight coal seams from the Westphalian A (Fig. 6) for which there is a sufficiently large regional data base. The maps show a consistent reproducible regional trend with the sulphur content of all seams increasing towards the east and decreas-

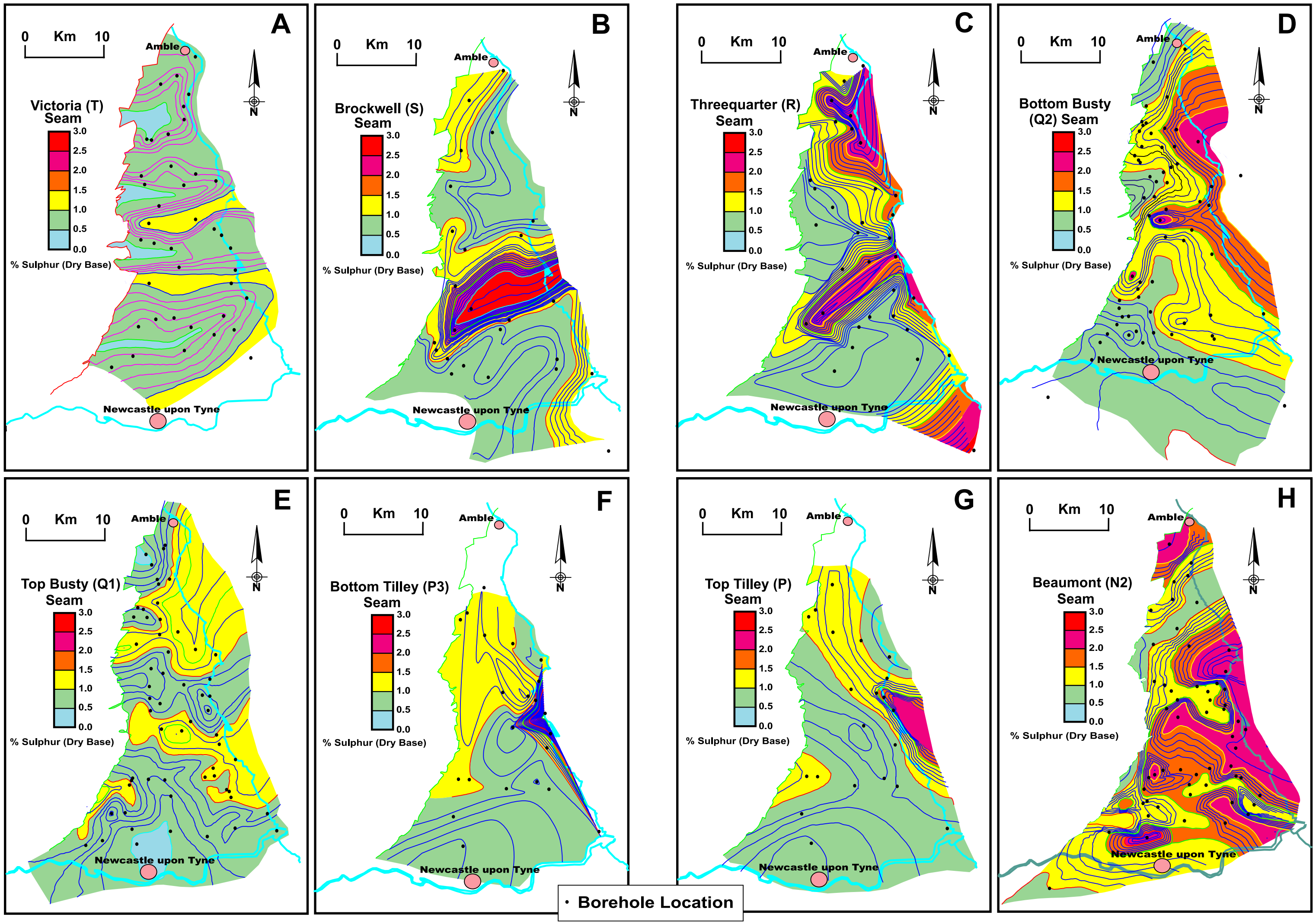


Fig. 6. Sulphur distribution maps showing the regional variation in sulphur content of eight coal seams from the Westphalian A of the Northumberland Coalfield. The sulphur content decreases across the coalfield towards the east and northeast with lower and higher values forming distinct zones trending approximately east–west.

ing towards the west across the entire coalfield area (Fig. 6). The Westphalian A Coal Measures, exposed at or near the surface in the west, dip into the subsurface to the east, hence the Coal Measures become younger eastwards, and only Westphalian B Coal Measures occur at the surface in the east and extend beneath the North Sea (Fig. 1). Thus, changes in stratigraphy across the coalfield may have an influence on regional sulphur trends in that younger Westphalian B coals appear to have higher sulphur contents (Fig. 4). However, if the link between high sulphur coals and marine conditions is correct, then the coals in the east and northeast should have lower, not higher, sulphur values, irrespective of stratigraphic position, given that they were deposited proximal to the source area and farthest away from marine and brackish water influences in the south. The fact that this is not the case in Northumberland suggests that extensive periodic flooding of peat mires by saline waters from the south was not the most significant factor controlling the regional distribution of sulphur in coal.

Local deviations from the regional trend are shown on the sulphur distribution maps where higher or lower sulphur values form zones of variable aerial extent, mostly trending approximately E–W (Fig. 6). Some high-sulphur zones extend into regionally extensive low-sulphur areas and may constitute a local problem that affects seam quality. For example, the Brockwell and Threequarter seams show pronounced high-sulphur zones (1–3%) covering areas of up to 50–60 km^2, and extending to the SSW for up to 10 km into regionally extensive low-sulphur (0.5–1%) areas (Fig. 6B,C). Both these high-sulphur zones occupy the same location and must have been a persistent feature through time. However, most high-sulphur values are restricted to an area adjacent to the Northumberland coast, where they appear to be fairly persistent features through time, even if not spatially confined to precisely the same areas. The origin of these high sulphur zones is uncertain, as they may reflect the limited data points on which the contouring is based. However, the maps for the Bottom Busty (Q2), Top Busty (Q1) and Beaumont (H) seams, which are based on a much more extensive set of data points (Fig. 6D,E,H), show similar trends to the other seam maps.

The regional increase in sulphur to the east and northeast, towards the sediment source area, suggests a possible source rock control on the sulphur content of the coals. Palaeocurrent and petrographic data indicate that the Northumberland Coal Measures were supplied with sediment from an extensive hinterland, ca. 250 km to the northeast, throughout the Silesian (Chisholm et al., 1996; Glover et al., 1996; Leng et al., 1999; Hallsworth and Chisholm, 2000) (Fig. 2), albeit with contributions from local uplands distributed around the compass (Haszeldine, 1984; Turner and Smith, 1995; O'Mara and Turner, 1999) (Fig. 3). Isotopic data indicates the presence of large areas of mature continental crustal rocks, possibly located in the Laurentian Shield, including lower Palaeozoic granitoids, Proterozoic or lower Palaeozoic sediments and low-grade metasediments, and minor Archean basement (Cliff et al., 1991).

In freshwater drainage systems, sulphate may be derived from source rock weathering, which provides the major source, and marine sulphate from seaspray which produces a flux of aerosol sulphate into the atmosphere (Granat et al., 1976; Youngsen, 1995). However, marine sulphate contributes much less than total river water sulphate (Hunt, 1987), especially given the distance from the coastline in the Northumberland basin. Sulphur may be readily oxidised and leached from sulphur-bearing source rocks and carried into the basin in solution mainly as terrestrial sulphate in the drainage system. Significant sulphate mobility occurs particularly when sulphur-bearing source rocks undergo uplift and erosion (Youngsen, 1995). Although local intrabasinal source rocks, such as Farne (Fig. 3), were uplifted and eroded at this time (O'Mara and Turner, 1999), they were probably not a significant source of upper Carboniferous sediment (Leng et al., 1999). Most sediment was supplied to the Westphalian Pennine basin from distant sources where uplift was active (Chisholm et al., 1996; Glover et al., 1996; Leng et al., 1999) (Fig. 7). Thus, sulphate-rich waters derived from these source areas could have enhanced the normally low-sulphate content of terrestrial waters (11.2 ppm, Youngsen, 1995), and on entering the basin contribute to the groundwater system, which is typically richer in sulphate than river water (Youngsen, 1995). The presence of syngenetic pyrite in the coal seams suggests that sulphate- and ferrous-enriched groundwater and overbank flood-

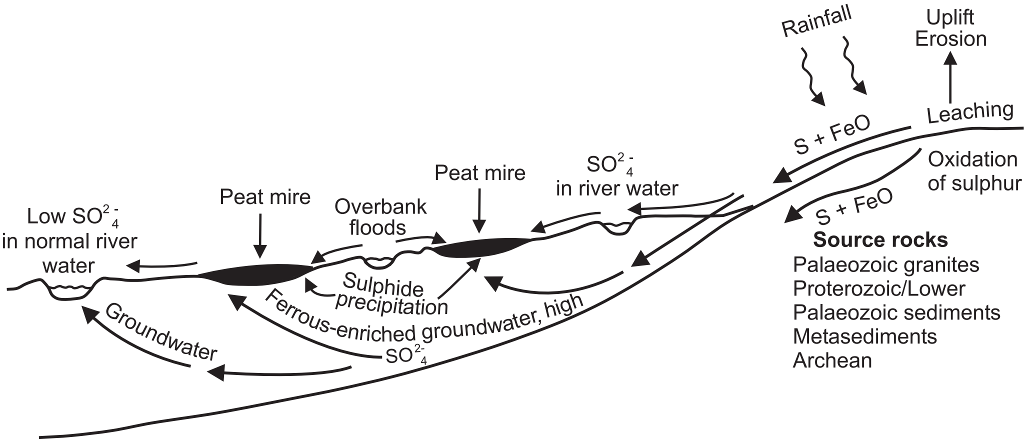


Fig. 7. Interpretive model for surface and groundwater flow paths along the northeast margin of the Pennine Basin, Northumberland, showing the relationship between sulphate, sulphide and pyrite in peat mires.

waters infiltrating the peat mire, precipitated sulphide, and that the pH of the mire was sufficiently high to promote sulphate-reducing bacterial activity (Shao et al., 2003). Such a model may help to explain the higher concentration of sulphate and hence sulphur in the coals closer to the basin margin as depicted in Fig. 7. Iron available to react with sulphur species may also have been sourced from the northeasterly provenance (continental crustal rocks contain up to 9.1 wt.% of FeO, Taylor and McClennan, 1985) and from more local, tectonically active sources such as the Southern Uplands (Leng et al., 1999), and brought into the basin along with the sediment load, attached to suspension load clays (Reidenouer et al., 1967; Love et al., 1983; Cohen et al., 1984). Spears et al. (1999) speculated that, under these conditions, coals affected by brackish-water flooding may have higher sulphur contents than those affected by marine flooding because of the greater availability of reactive iron from detrital sediment influx.

The high silica (23.4–50.9%) and alumina (14.1–39.0%) content of ash in some seams compared to others (Table 1) may indicate a stronger detrital influx into some of the peat mires at these times, or closer proximity to active channels. Moreover, clay (SiO_2 and Al_2O_3) is the dominant component in most ash

Table 1
Composition of ash from Westphalian A and B coal seams, Delhi Opencast Coal Site, Northumberland (see Fig. 4 for stratigraphic position of named seams)

Coal seam	SiO_2	TiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	Na_2O	K_2O	Cr_2O_3	MnO_2	P_2O_5	SO_3	Total seam dry-base ash (%)
Top Plessey	50.86	1.05	32.11	5.16	2.81	1.56	0.09	3.19	0.03	0.05	0.82	2.27	7.46
Bottom Plessey	23.37	0.51	14.14	29.98	12.48	5.27	0.20	0.73	0.02	0.40	0.41	12.50	6.22
Top Beaumont	40.54	0.75	25.18	18.21	4.85	2.61	0.12	1.69	0.03	0.20	0.24	5.59	20.72
Bottom Beaumont	27.74	0.72	19.57	22.42	11.69	5.18	0.18	0.72	0.04	0.29	0.33	11.12	6.53
Hodge	48.53	0.93	22.42	9.36	7.86	3.59	0.31	1.06	0.04	0.20	0.25	5.46	7.40
Top Tilley	54.04	1.33	38.96	1.99	0.32	0.82	0.08	1.65	0.04	0.01	0.12	0.65	5.20
Bottom Tilley	46.11	1.14	28.74	11.41	4.49	2.01	0.11	0.96	0.04	0.11	0.37	4.51	14.78
Tilley Stringer	31.91	0.58	23.63	23.32	6.43	4.12	0.96	0.76	0.06	0.18	0.29	7.77	12.23
Top Busty	46.75	1.05	28.57	8.95	5.10	2.66	0.12	1.19	0.04	0.12	0.22	5.23	7.42
Bottom Busty	37.02	0.80	24.31	19.20	6.88	3.38	0.15	0.74	0.03	0.20	0.24	7.05	8.86
Harvey Marine	41.11	0.76	25.70	18.97	4.38	2.59	0.14	1.69	0.03	0.13	0.19	4.30	14.30
Northumberland Low Main	41.97	1.09	26.64	11.49	7.48	2.75	0.11	1.37	0.04	0.23	0.29	6.53	8.99
Durham Low Main	45.38	0.97	28.48	13.23	3.38	1.92	0.11	2.36	0.03	0.11	0.27	3.77	15.70

samples (Table 1). Thus, if detrital clay acted as a source of iron for pyrite formation, then there should be a broad correlation between the sulphur and ash yields of the coal. Gayer et al. (1999) noted a weak to strong correlation between ash and sulphur for two coal seams in the South Wales coalfield. Markic and Sachsenhofer (1997) recognised a strong positive correlation between sulphur and ash contents in some Pliocene coal seams but no correlation in others. Plots of dry-basis ash versus pyritic sulphur for the Westphalian A Top Busty, Bottom Busty and Beaumont seams in Northumberland show no correlation (Fig. 8). This could be explained by the presence of raised mires which are able to deflect siliciclastic currents away from the mire (McCabe, 1987) and which typically give rise to coals with <10% ash (Diessel et al., 2000). Although raised mires have been recognised in the Westphalian B from coal petrography (Smith, 1968; Fulton, 1987), in Northumberland most mires drape and infill pre-existing topographic depressions (Haszeldine, 1989; Rippon and Spears, 1989) and the coal seams contain siliciclastic partings (coal splits) indicating contemporaneous clastic contamination of the mire. Likewise, the upward increase in sulphur content together with increasing vitrinite and higher ash yields in successive coal layers in some coal seams <3 m thick (Turner, 1999) argues for a planar rather than ombrogenous (domed) peat mire morphology (McCabe, 1984; Greb et al., 1999; Greb et al., 2002). Planar mires are more susceptible to periodic flooding, and typically give rise to coals with a higher ash content than ombrogenous peat-derived coals. This is not the case in Northumberland where the planar peat-derived coals have average ash contents of <10%, identical to the value given by Diessel et al. (2000) for ombrogenous peat-derived coals.

The lack of domed peats in Northumberland may reflect a seasonal tropical climate, when rainfall was not consistent enough (ever-wet) throughout the year to support raised mires (Cecil et al., 1985), or the proximal position of the coalfield away from significant marine influences. Evidence in support of a seasonal climate is the presence of desiccation cracks in seatearths (palaeosols), seasonal wetting of the soil profile and the abundant crevassing indicative of fluctuating channel discharge, consistent with a seasonal rainfall regime (Fielding, 1984). Marine transgressions are often accompanied by increased moisture, especially in equatorial climates (Heckel, 1995), thereby favouring ever-wet conditions essential for the formation of [modern] domed peats. Thus, whilst these ideal conditions may have existed further south, closer to marine environments (Smith, 1968; Fulton, 1987), to the north conditions were less suitable for ombrogenous mire develop-

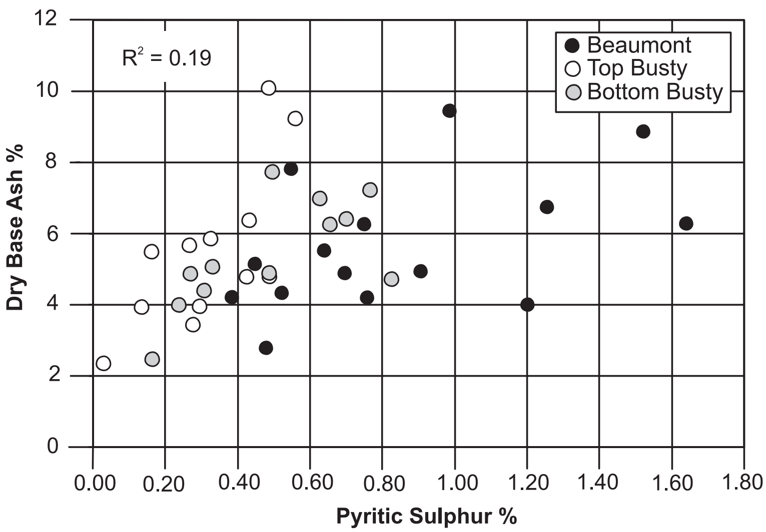


Fig. 8. Dry-based ash versus pyritic sulphur for the Beaumont, Top Busty and Bottom Busty coal seams for the Northumberland Coalfield.

ment. The most favourable site for mire development on coastal plains is estimated to be 40–100 km away from the palaeoshoreline (McCabe, 1984; Woodroffe et al., 1985; Michaelson and Henderson, 2000). In Northumberland, the lack of any direct marine influences on coal measure deposition suggests that the mires developed at least 100 km inland (Fig. 3). Accordingly, the lack of correlation between ash and sulphur suggests that the pyritic sulphur content of the coals was largely independent of any significant iron-enriched fine silt and clay washed into the mire from overbank floods. Thus, a significant source rock control on the pyritic sulphur content of the coal via this mechanism seems unlikely.

3.2. Post-depositional addition of sulphur to coal

The presence of epigenetic veins and cleat-filling pyrite (Love et al., 1983; White et al., 1994; Bouška et al., 1997; Gayer et al., 1999) suggests that some sulphur may have been added to the coal following deposition and cessation of organic peat reactivity (Hunt, 1987; Youngsen, 1995). In the UK, Westphalian Coal Measures cleat formation is thought to be primarily controlled by tectonism following uplift and release of overburden pressure (Rippon, 1996). Thus, pyrite-coating cleat surfaces post-dates cleat formation and coalification. The possibility of post-depositional addition of secondary sulphur into the coals is suggested by the presence of Lower Carboniferous (Mississippian) Limestone Group sediments and associated sulphide mineralisation of the North Pennine Orefield along the southern margin of the coalfield. The coals also contain unusually high concentrations of Ba of up to 2282 ppm (Turner, 1999) (Fig. 9), and the only nearby source of this Ba is the barite mineralisation in the North Pennine Orefield which extends northeastwards beneath the southwestern margin of the coalfield (Mills and Holliday, 1998) (Fig. 1). Galena has also been found in coals along the Northumberland coast and encountered on cleat and jointing during sample collection for this study.

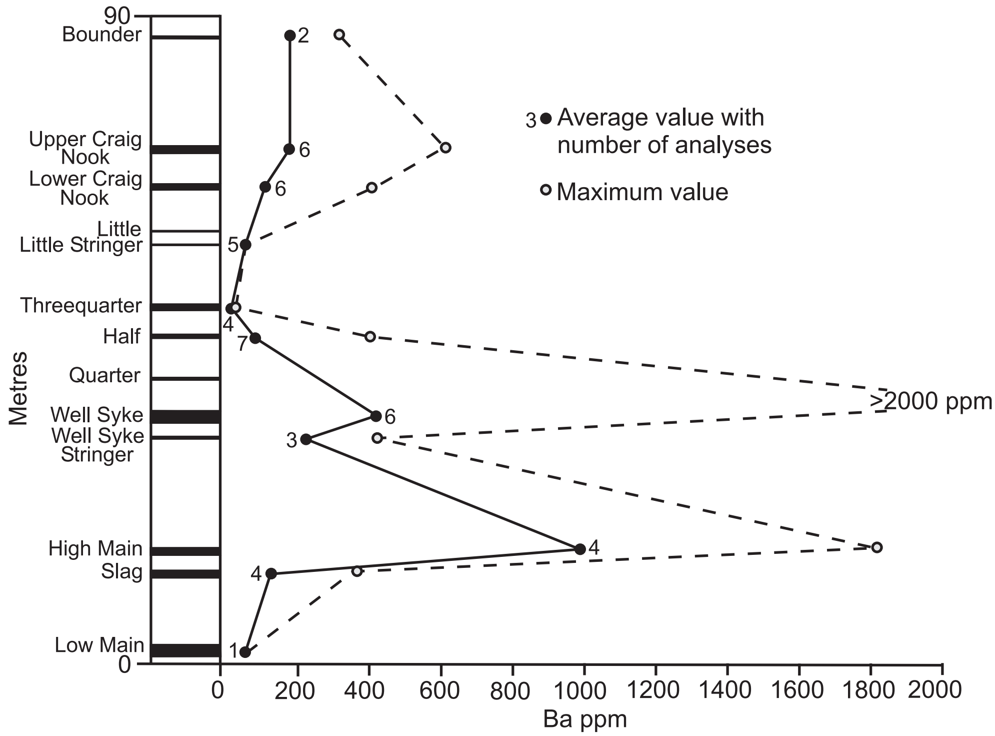


Fig. 9. Variation in barium content of Westphalian A coal seams from the Plenmeller Opencast Coal Site, Northumberland.

3.2.1. Sulphur isotopic composition and the source of sulphur

Sulphur isotope data for the North Pennine Orefield indicate that the sulphur in the Zn–Pb and Ba mineralisation was mostly derived from lower Carboniferous seawater sulphate (Edwards and Atkinson, 1987; Crowley et al., 1997). Age dating, based on U/Pb ratios, suggests that the first major phase of Pb–Zn mineralisation is late Carboniferous (Stephanian) to Permian in age (292±20–255±12 Ma, Dunham, 1966; Dunham et al., 1968), with subsequent phases, based on $^{40}Ar/^{39}Ar$ ratios, dated at 230 and 170 Ma (Dunham, 1990). Crowley et al. (1997) considered the barite mineralisation to be post-Carboniferous and probably lower Permian in age. The main mineralisation phase is associated with partial basin inversion during Variscan transpression (Chadwick and Holliday, 1991; Kimbell et al., 1989) and was emplaced at temperatures between 50 and 219 °C, with a temperature of <120 °C for barite (Dunham, 1990).

Armstrong and Purnell (1993) noted an anomalous increase in the conodant alteration index (CAI) and temperature increase up sequence in the Carboniferous of the southern Northumberland basin. They attributed this inverted thermal event to a short-lived extrabasinal heating event marking the onset of hydrothermal circulation and release of hot brines from the lower crust associated with the Pb–Zn mineralisation of the North Pennine Orefield. Mineralisation in the Haydon Bridge area (Fig. 1) along the southern margin of the coalfield occurred at 292±20 Ma from hot (CAI of 3 suggests 110 °C, Armstrong and Purnell, 1993) sulphate-rich NaCl brines (Sawkins, 1966; Smith and Phillips, 1974; Smith, 1980). Thus, the introduction of sulphur-rich fluids was associated with late Westphalian–early Permian Variscan transpressional tectonics from the south, which led to uplift, faulting and folding, and the imposition of a regional dip of up to 5° to the east and northeast in Northumberland (Mills and Holliday, 1998; Johnson and Dunham, 2001). As a result, late stage Variscan compressional tectonics, at about 280 Ma, may have induced the expulsion of hot sulphur-rich fluids from depth and large-scale fluid movement northwards and upwards through the Coal Measures towards the basin margin, possibly along basin margin faults (Mills and Holliday, 1998). These sulphide-rich brines, migrating down the regional dip may have acted as a source of both sulphur and iron, thereby adding to the post-depositional sulphur content of the coals. Evidence of post-depositional fluid movement in the Westphalian Coal Measures is seen in the sulphide mineralisation along fault planes, which acted as conduits for post-depositional fluid movement, Ba-rich fluids in the coalfield (Edmunds, 1975) and epigenetic barite mineralisation along the Stublick-Ninety Fathom fault system bounding the southern margin of the Northumberland Basin (Crowley et al., 1997). Post-depositional sulphide mineralisation commonly extends from fault planes into the adjacent Coal Measure strata and anastomosing pyrite veinlets in coal have been interpreted as mineralised fractures in the South Wales Coalfield by Gayer et al. (1991), who attributed them to a hydrothermal event.

A more rigorous assessment of the model can be made by comparing $\delta^{34}S$ isotopes from the North Pennine Orefield with those of pyrite from the coal in order to identify the potential source of sulphur in the coal. The isotopic composition of nine samples of pyrite and one sample of galena was determined in the Atmospheric Monitoring and Stable Isotope Laboratory at Royal Holloway, University of London, according to the laboratory procedures and analytical methods detailed in Grassineau et al. (2001). The samples, collected from fault planes, joints, cleats and fracture fills, were associated with four different coal seams formed in the same alluvial depositional environment: two from the Upper Westphalian A (Hodge and Bottom Beaumont) and two from the lowermost Westphalian B (Bottom Plessey and Northumberland Low Main) (Fig. 10). The Hodge and Bottom Beaumont seams have regional average sulphur contents of 1.2% and 1.5%, respectively, whilst the Bottom Plessey and Northumberland Low Main have identical average sulphur contents of 1.4%.

The sulphur isotope results in Fig. 10 are the first published data on the isotopic composition of sulphur in coals from the Northumberland Coalfield. Pyrite samples from the coal show a high variability in $\delta^{34}S$ values from −5.4‰ to +32.8‰ with an average of +5.1‰. A duplicate run of the unusually high sample of +32.8‰ (D6), carried out in order to be sure that it was the correct heavy value, showed a difference of 0.16‰. Such high values are not uncommon in low sulphur coals, especially those with non-marine roof rocks, and similar high values for pyritic sulphur have

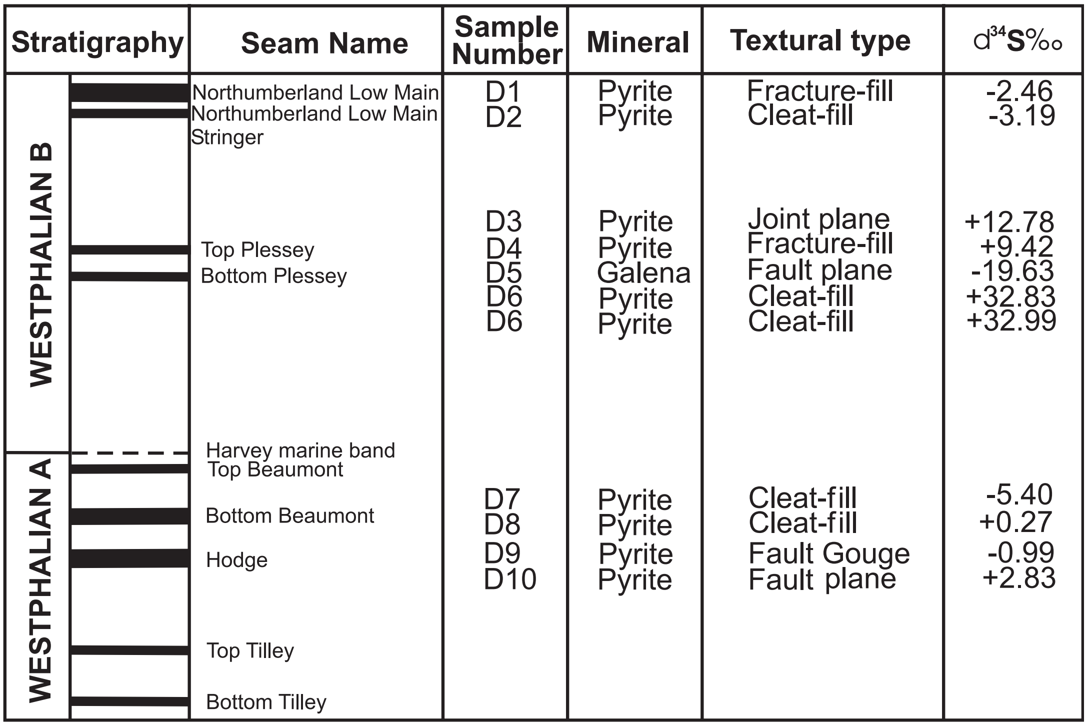


Fig. 10. Sulphur isotope composition of pyrite and galena associated with Westphalian A and B coal seams, Delhi opencast site, central Northumberland.

been recorded for Pennsylvanian coals in the USA (+28.5‰) and Australian Permian coals (+32.3‰) (Smith and Batts, 1974), both of which also show a similar range in sulphur isotope composition (−17.8‰ to +28.5‰ and −14.9‰ to +32.3‰, respectively) to Westphalian coals in Northumberland. The lowest δ^{34}S value of −19.6‰, obtained for a sample of galena from a fault plane below the Bottom Plessey seam, lies outside the range of the pyrites. Using the fractionation factor of Kajiwara and Krause (1971), the pyrite equilibrium for galena at 150 °C is 5.8‰, which give an adjusted value for galena of −13.8‰. This is much lower than previously reported values for galena from the North Pennine Orefield of up to −5.9‰ (Solomon et al., 1971) (Fig. 11). The large variation in δ^{34}S values from −5.4‰ to +32.83‰ is similar to that for pyrite δ^{34}S values (−45.4‰ to +0.8‰) for black shale-hosted antimony mineralisation in the northern Rhenish Massif, Germany, reported by Wagner and Boyce (2003), who consider such large ranges to be incompatible with local derivation of sulphur. This highly variable isotopic composition further suggests the possibility of more than one generation of pyrite (Dai et al., 2002; McKay and Longstaffe, 2003), consistent with the existing model for more than one phase of North Pennine Orefield mineralisation. Thus, sulphur may have been introduced in at least two stages related to coalification and burial depth: (1) whilst the peat was still spongy and permeable (pyrite veinlets, intercalated with the coal, and locally deformed by peat compaction); and (2) following lithification and jointing in the coal (cleat and joint pyrite).

Such high variability in δ^{34}S values is considered to be most typical of secondary pyrite sulphur with a marine sulphate source (Price and Shieh, 1979; Westgate and Anderson, 1984), whilst high values in excess of +18‰ are consistent with the post-depositional addition of sulphur (McKay and Longstaffe, 2003). The very high value of +32.8‰ exceeds the value of seawater sulphate (~+18‰ CDT for Mississippian seawater), suggesting that the system must have been open to addition of δ^{34}-enriched sulphur (McKay and Longstaffe, 2003). Whether such high

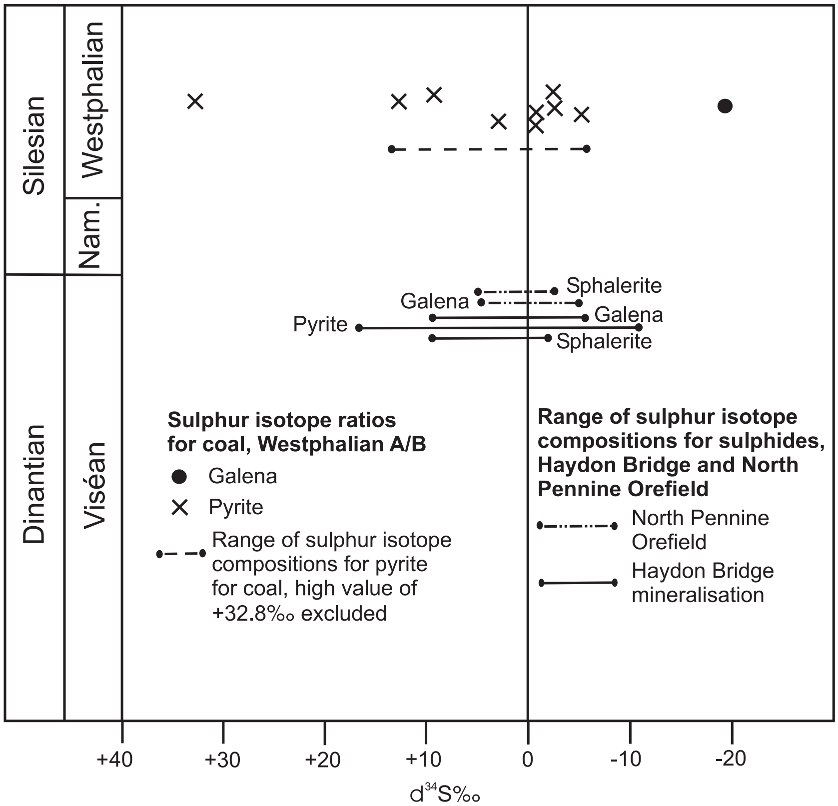


Fig. 11. Sulphur isotope values for coals from the Northumberland Coalfield, Pennine Orefield and Haydon Bridge. The sulphur isotope values for pyrite in the coals show a similar range to those for pyrite and other sulphides in the Pennine Orefield and Haydon Bridge.

values for pyrite are common throughout the coals remains to be tested by further work. However, if the very high δ^{34}S value (+32.8‰) is excluded, then the pyrite samples have relatively low negative to positive values ranging from a minimum of -5.4‰ to a maximum of +12.8‰. These δ^{34}S values are similar to the values of -3.1‰ to +8.0‰ (Edwards and Atkinson, 1987) and -10.5‰ to +16.9‰ (Crowley et al., 1997) for pyrite samples from the North Pennine Orefield and -5.9‰ to +9.4‰ for other sulphides from the North Pennine Orefield and related mineralisation at Haydon Bridge in West Northumberland (Crowley et al., 1997) (Fig. 3), which forms the northernmost limits of the orefield (Fig. 1). Thus, sulphur-isotope ratios for pyritic sulphur in the Westphalian A and B coals show a similar range to those for pyrite and other sulphides in the North Pennine Orefield; a relationship which argues for a similar origin and suggests the possibility that some of the sulphur may have been derived from a Mississippian (lower Carboniferous) marine sulphate source.

3.3. Sequence stratigraphy and stratigraphic variations in the sulphur content of coal

3.3.1. Sequence stratigraphic background

Early sequence stratigraphic modelling of British Coal Measures was based on general principles worked out for coal measures in the Eastern United States where the classic tripartitite subdivision into low-stand, transgressive and high-stand systems tracts is well documented (Heckel, 1986, 1995; Aitken and Flint, 1994; Bohacs and Suter, 1997). In this model, coals form mainly under increasing levels of accommodation space in the transgressive systems tract (TST), up to the maximum flooding surface at the base of the high-stand systems tract (HST), with deeply incised, laterally extensive palaeovalley-fills denoting a low stand sequence boundary unconformity. This model proved difficult to apply to British Coal Measures because unconformably based incised valley-fill sandbodies are absent due to: (1) the low-relief ramp setting of the alluvial plain, whereby lowering of sea level does not normally lead to erosion (Schumm, 1993; Shanley and McCabe, 1993; Posamentier and Allen, 1993); (2) the lack of any significant basinward shift of facies and stacking patterns indicative of a low stand sequence boundary (Van Wagoner et al., 1988; Holz, 1998; Holz et al., 2002); and (3) the relatively uninterrupted sedimentation during the Westphalian A and B, when subsidence always exceeded base level fall, hence the lack of significant unconformities, except those tectonically induced (Quirk, 1993; O'Mara and Turner, 1999).

In Northumberland the Westphalian A and B strata, which are approximately 500-m thick (Land, 1974; Smith, 1994), were deposited over 4 Ma (Leeder and McMahon, 1988). The Westphalian A is 205 m thick and the Westphalian B 295 m thick (Fig. 4). Thus, the Westphalian A and B were deposited over a time framework of approximately 1.64 and 2.36 Ma, respectively, and are considered to be genetic sequences in the terminology of Galloway (1989) or third-order depositional sequences (0.5–3.0 Ma, Mitchum and Van Wagoner, 1991; Duval et al., 1992) defined by the regionally widespread *subcrenatum*, *vanderbeckei* and *aegiranum* marine bands, or their proximal, brackish-water equivalents in Northumberland (Quarterburn, Harvey, Down Hill), which are interpreted as high magnitude third-order maximum flooding surfaces (Flint et al., 1995; O'Mara and Turner, 1999) (Fig. 12). The Westphalian A third-order sequence contains up to 19 higher frequency coal-bearing cycles with an average duration of ~86 ka/cycle, whereas the Westphalian B third-order sequence contains up to 21 higher frequency coal-bearing cycles with an average duration of ~112 ka/cycle, both values within the range for fourth-order parasequence cycles (0.1–0.5 Ma, Mitchum and Van Wagoner, 1991; Duval et al., 1992).

Fourth-order coal-bearing parasequence cycles in Northumberland (3–15 m thick) are bounded by transgressive, brackish-water *Lingula* bands and, more commonly, by non-marine bivalve beds (predominantly Anthraconaia and Carbonicola), which represent lacustrine flooding surfaces terminating mire formation (Fig. 12, inset). A progressive, more progradational trend followed as crevasse-splay deltas built regressive platforms for peat mire formation as base level began to rise prior to the next flooding event. The nature of these fourth-order flooding surfaces depends on their position within the third-order base level cycle. Those on either side of maximum flooding surfaces tend to be more marine-influenced in character (*Lingula*), whereas those in the middle part of the sequence, between maximum flooding surfaces, are characterised by non-marine bivalve bands (Fig. 12), consistent with a decrease in accommodation space.

The sequence stratigraphy of the Westphalian B Coal Measures in Northumberland and the southern North Sea is discussed in detail by O'Mara (1995). In summary, the Westphalian B third-order base level cycle above the *vanderbeckei* marine flooding surface is disrupted by the development of a tectonically induced low-stand incision (Fig. 12) overlain by a major fluvial sandbody (O'Mara, 1995), resembling a palaeovalley-fill (Aitken and Flint, 1994). This occurs about 30–40 m above the base of the sequence but is confined to an area of about 100 km^2 in the east–central part of the coalfield, extending a short distance offshore (Land, 1974). However, there is no significant incision of the underlying strata, probably due to the very slow rate at which accommodation space was created at this time (O'Mara, 1995). Thus, the lower part of the Westphalian B in this part of the coalfield includes a tectonically induced, local low stand, representing the lowest point on the third-order base level curve.

3.3.2. Stratigrapic variation and controls on sulphur content

Stratigraphic variations in sulphur content of Westphalian A and B coal seams in Northumberland show a significant increase in sulphur content upwards through the succession, albeit with one or two anomalous values (Figs. 4 and 12). The stratigraphic variation in maximum, minimum and average sulphur contents of individual coal seams in the succession, plotted in Fig. 13, shows the High Main coal to have the highest average sulphur content (4%), followed by the Hutton and Tilley seams. Nevertheless, the average and minimum values show a

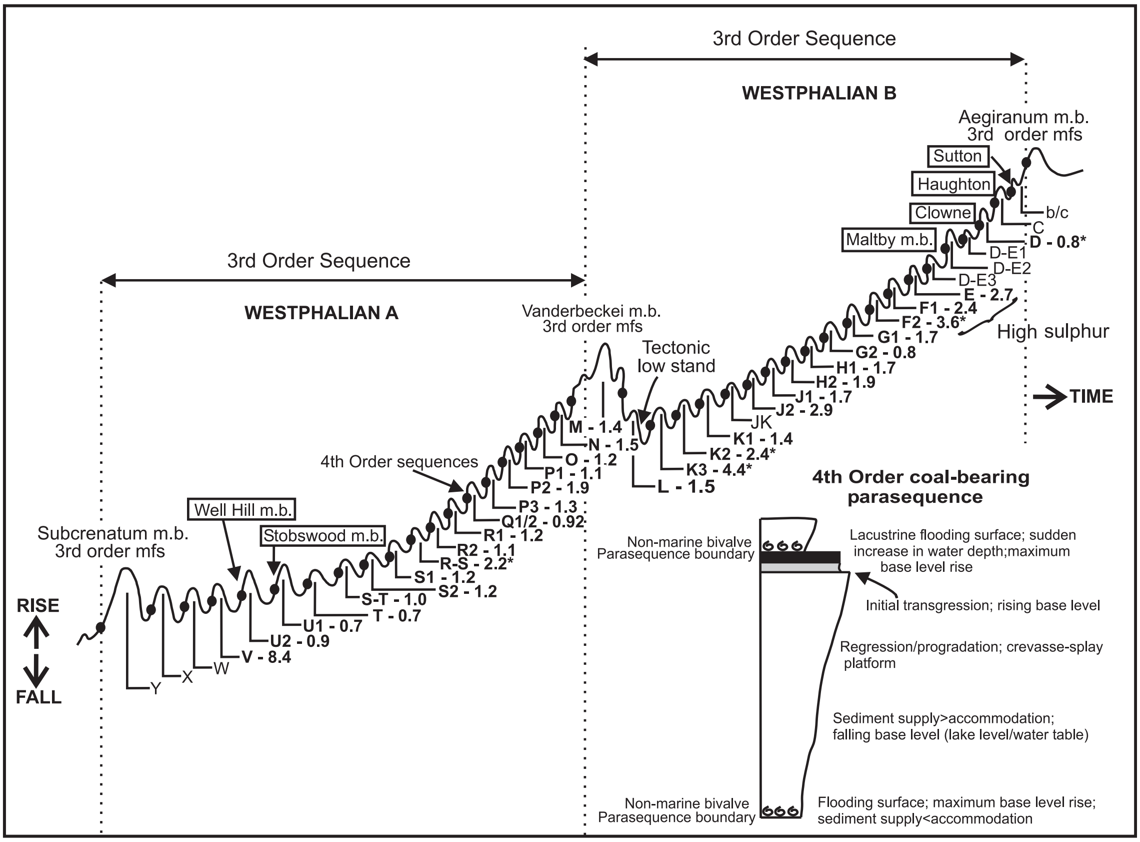


Fig. 12. Sequence stacking patterns for the Westphalian A and B Coal Measures in the Northumberland Coalfield showing third-order and fourth-order sequences, maximum flooding surfaces, seam location, index letter and average sulphur contents (see text for details). The name of coal seams for each index letter is given in Fig. 4. The section shows details of a typical fourth-order coal-bearing parasequence. The asterisk next to value indicates one sample only.

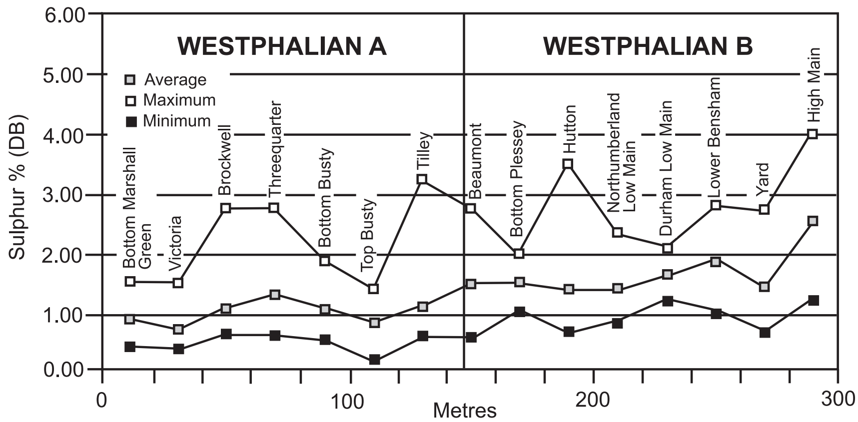


Fig. 13. Stratigraphic variation in the sulphur content of coal seams based on National Coal Board data (Coal Survey Seam Records for Borehole and Shafts in Northumberland, 1946–1969; Northumberland Coalfield Seam Maps, 1957).

broadly similar trend with the sulphur content increasing up the succession. This suggests a rise in sulphate salinities within an overall transgressive trend to sedimentation in which the third-order base level curves are superimposed on a possible late Carboniferous rise in relative sea level that may have been part of a lower second-order sea level rise (Fig. 14) equivalent to that between the Kinderscoutian and Myachkovian figured by Ross and Ross (1988) and Alekseev et al. (1996).

Variations in the sulphur content, plotted as average sulphur percentages for individual coal seams capping each fourth-order cycle, making up the Westphalian A and B third-order depositional sequences, are shown in Fig. 12. In the Westphalian A, the lowest sulphur coals (0.7–1.2%, average 0.9%) are mostly confined to the middle of the third-order sequence between the Marshall Green (U2) and Bottom Tilley (P3) seams, when base level was relatively low. The only exception is the Little Wonder/Stobswood (R-S) seam, which has a sulphur value of 2.2%, based on a single sample (Fig. 4). Approaching the third-order maximum flooding surface (*vanderbeckeii* marine band) at the top of the Westphalian A sulphur values increase with the seam immediately beneath the flooding surface, the Harvey/Beaumont (N), having a sulphur value of 1.5%. The Low Main/Marshall Green (U2) seam in the lower part of the sequence has a sulphur content of 0.9%, whereas the Ganister Clay seam (V) immediately below it has a much higher average sulphur content of 8.4%, probably due to the presence of the equivalent of the Well Hill marine band immediately above the Ganister Clay seam (Fig. 4). Sulphur values for the Broomley (W) and Bottom Broomley (X) seams at the very bottom of the sequence are not available. However, in west Northumberland, at the Plenmeller Opencast coal site, the Low Main/Marshall Green (U2) coal has a higher average sulphur content of 6.4% (average of 10 samples with a maximum value for individual samples of 14.1%, Turner, 1999, Appendix 3). These high values are attributed to a pyritic shale roof rock containing fish fragments and sporadic, large bivalves, interpreted as robust forms of Carbonicola typical of parts of the Langsettian (Turner, 1999). The shale has a strong gamma-ray signature and a Th/U ratio of <3, consistent with a brackish to marine influence on deposition (Adams and Weaver, 1958; Hollywood and Whorlow, 1993; Gayer et al., 1999), hence the greater availability of sulphate for pyritic sulphur. This interpretation is consistent with the clay mineralogy of the shales (Chen, 1990) and the high sulphur and strontium (518 ppm) values of the Low Main/Marshall Green coal, compared to the coals above (Turner, 1999, Appendix 3). In contrast, the next seam in the succession, the Victoria (T) seam, located above the last equivalent marine band (Stobswood), has a relatively low sulphur value of 0.8% (Figs. 4 and 12), as marine to brackish water influences on deposition declined and the shoreline shifted southwards concomitant with a fall in relative base level.

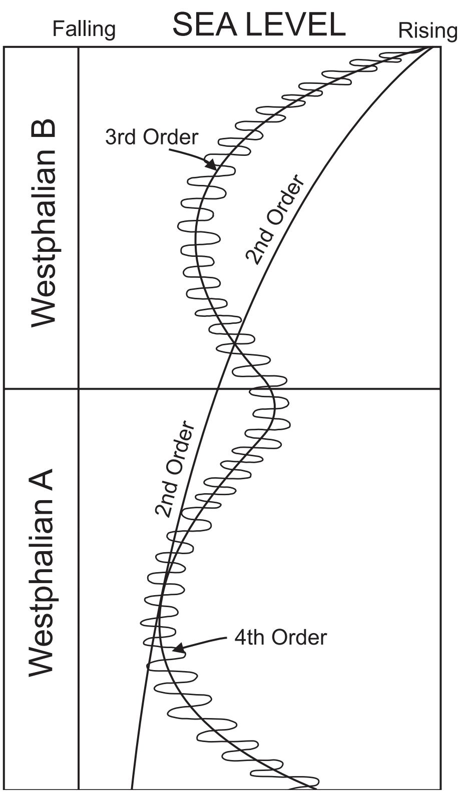


Fig. 14. Third order base level curves for the Westphalian A and B show an overall transgressive trend, which may have been superimposed on a possible late Carboniferous rise in relative sea level that may have been part of a lower second-order curve, equivalent to that between the Kinderscoutian and Myachkovian.

Coal seams in the lower Westphalian B, between the Middle Plessey (M) and Northumberland Low Main (K1) have relatively low sulphur values of

1.4%, apart from the Middle Low Main (K2) and Bottom Low Main (K3) coals with sulphur values of 2.4% and 4.4% (Fig. 12), respectively, both based on single samples. Although these values may reflect the inadequate sample base, a possible explanation for these high sulphur values is a more brackish water influence on deposition, as reflected in the black 'cannely' shale roof rock containing abundant pyrite nodules and tubes, a prolific fish and amphibian fauna, and bivalves, especially *Naiadites* sp. (Land, 1974). The remaining coal seams above K1, apart from the Bottom Yard (G2) coal, have high sulphur values and there is a generalised upward increase in sulphur, even within the relative low stand part of the third-order base level cycle, where low sulphur values would normally be expected. This is attributed to a more rapid rise of base level compared to the Westphalian A and lowermost Westphalian B, and the development of more marine-influenced, higher-sulphur coals, compared to the equivalent part of the Westphalian A third-order depositional sequence. This increasing marine influence in the upper part of the Westphalian B is seen in the number of closely spaced marine bands including the High Main (Maltby), Clowne, Haughton and Sutton just below the third-order maximum flooding surface (Figs. 4 and 12). These marine bands also show an increased abundance and diversity of faunal assemblages which implies that stratigraphic base level rise was not just higher than in the lower Westphalian B but of such a magnitude that conditions began to approach more normal open-marine salinities, and higher sulphate values, as marine conditions transgressed further northwards than before. This contrasts with the Westphalian A/B Coal Measures in South Wales where marine incursions become less frequent towards the top of the succession (Gayer et al., 1999).

The High Main (E), Upper (F1) and Lower (F2) Main coals, immediately below the High Main (Maltby) marine band, have consistently high sulphur values of 2.7%, 2.4% and 3.6%, respectively, whereas the Moorland (D) seam above, associated with the Clowne marine band (Fig. 12), has a lower sulphur value of 0.8%, despite the well documented influence on deposition exerted by the Clowne marine band (O'Mara, 1995). However, this value is based on just a single sample and the lack of data in this part of the Westphalian B succession reflects the generally uneconomic nature of the coals.

3.4. Sulphur variations between and within individual seams

3.4.1. Between seams

A detailed sampling programme and geochemical analysis of 12 middle Westphalian A coal seams exposed at the Plenmeller Opencast Coal Site, in the western part of the Northumberland Coalfield, was carried out as part of a British Geological Survey-funded research report (Turner, 1999, Appendix 3). Although all the seams at Plenmeller were analysed only five seams, which have at least six analysed samples per seam, each sample corresponding to individual coal layers, have been selected for assessment. Analyses for sulphur (S) and iron (Fe) have been plotted together for comparison in stratigraphic order from the base up (Fig. 15). The relationship between sulphur and iron provides a proxy indicator of whether pyritic or organic sulphur has the major effect on the total sulphur content of the coal: a similar trend implies that inorganic sulphur dominates to give pyrite, whereas an inverse relationship argues for a major organic sulphur affinity.

The Plenmeller Opencast site lies in the low sulphur western part of the Northumberland coalfield, between the *subcrenatum* and *vanderbeckei* maximum flooding surfaces in the middle, low stand part of the Westphalian A third order base level cycle. The data presented in Fig. 15 confirm that the coals have low sulphur contents (<1.3%), except for the anomalous Low Main/Marshall Green coal, mentioned previously. The iron versus sulphur plots for the Low Main, Well Syke and Upper Craig Nook seams all show a similar relationship with inorganic sulphur dominant, producing pyrite. The only striking departure from this trend is the Lower Craig Nook coal where there is a strong inverse relationship between S and Fe consistent with a major organic sulphur affinity (Fig. 15). This same pattern is seen in the uppermost part of the Half Seam; the lower part shows a typical inorganic sulphur affinity (Fig. 15). Thus, most seams have a consistent organic sulphur content, which rarely effects the total sulphur content of individual seams or parts of the same seam.

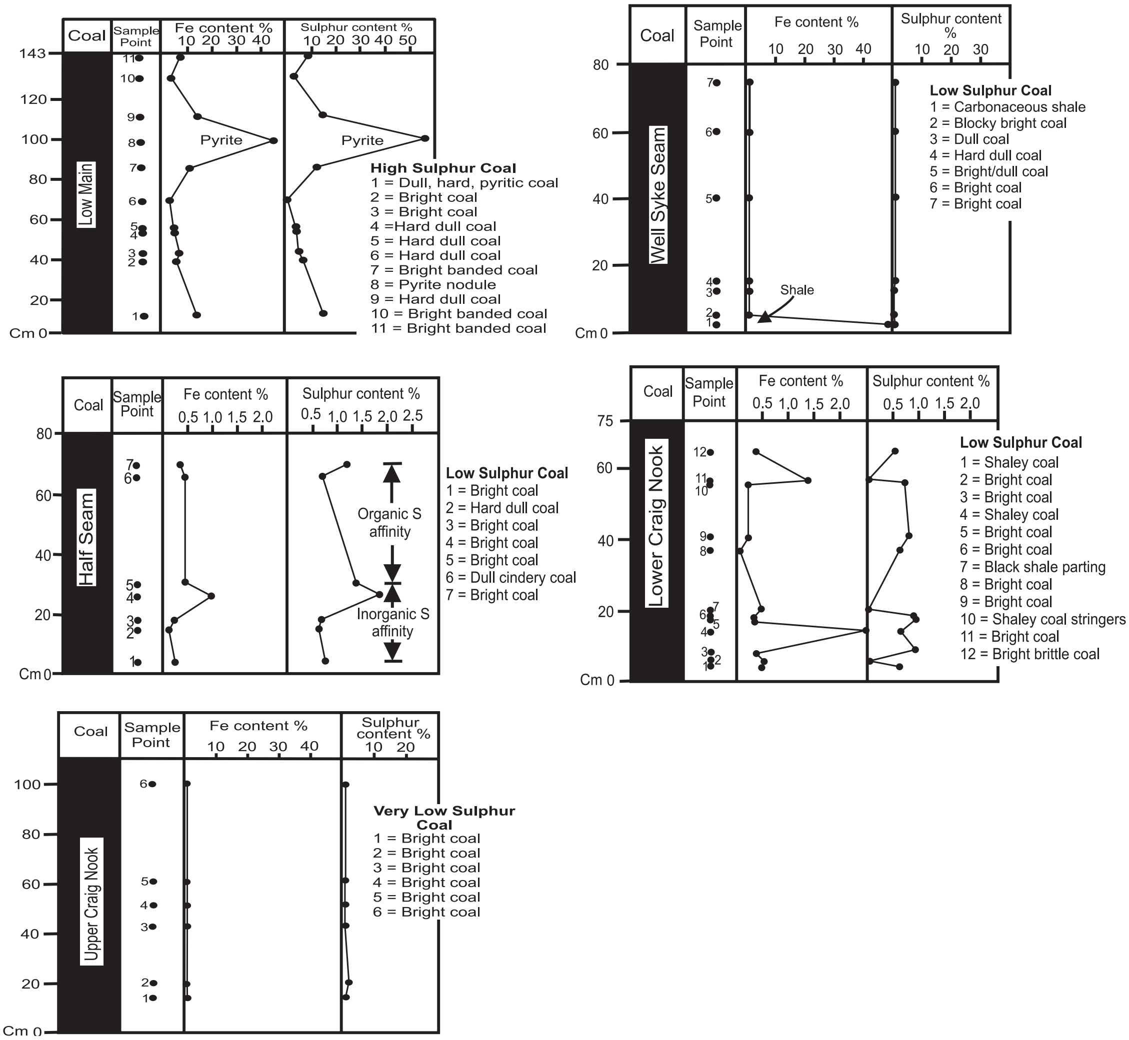


Fig. 15. Vertical distribution of iron and sulphur in five coal seams from the Westphalian A at the Plenmeller Opencast Coal Site, West Northumberland (based on unpublished data from Turner, 1999).

3.4.2. Within seams

Variations in sulphur content within individual seams at Plenmeller show no systematic trend common to all seams. The Low Main has a higher sulphur content at the top and bottom of the seam (Fig. 15), a feature noted by Gayer et al. (1999) for the Amman Rider and Bute seams in South Wales, and by Diessel (1992) and Banerjee et al. (1996) for transgressive coal seams. High sulphur in the top of the seam is attributed to the more marine-influenced roof rock. This is typical of the sulphur distribution seen in many marine-influenced coals (Williams and Keith, 1963; Casagrande et al., 1977; Petersen et al., 1998), and modern marine-influenced mires, although mires generally show a constant sulphur content or decrease in sulphur with increasing depth, the latter being typical of most coal seams (Phillips et al., 1994) irrespective of whether they are transgressive or regressive sequence stratigraphic coals (Diessel et al., 2000). The high total sulphur content of 12.84% at the bottom of the Low Main seam is associated with a dull, hard, pyritic coal ply. Sulphur values for the bottom and top of the seam are 24.7 and 4.4 times that of the lowest value (0.52%) for the seam, respectively.

The high sulphur value at the base of the seam is attributed to the presence of the equivalent of the Well Hill marine band, some 4 m below the floor of the seam, which imposed brackish-water conditions on initial peat accumulation, given the availability of reactive iron in the peat. The Well Syke and Upper Craig Nook all show a consistent internal trend, and apart from one high sample in the Half Seam and one low sample in the Lower Craig Nook they also show a consistent internal sulphur trend (Fig. 15).

4. Conclusions

The sulphur content of coals in the predominantly freshwater Westphalian A and B Coal Measures of the Northumberland Coalfield is variable and some high-sulphur coals (>1.3%) have fresh rather than brackish or marine roof rocks. Variations in the sulphur content of the coals are largely controlled by pyrite content. Rates of sedimentation closely balanced rates of subsidence during the Westphalian, hence shallow-water fluvio-deltaic conditions were maintained throughout deposition, interrupted periodically by brief, regionally extensive, marine transgressions from the south. These transgressive events, interpreted as maximum flooding surfaces, are represented by brackish water *Lingula*-rich mudstones in Northumberland. These maximum flooding surfaces divide the Coal Measures into a 205-m-thick Westphalian A third-order sequence, deposited over some 1.64 Ma and a 295-m-thick Westphalian B third-order sequence, deposited over some 2.35 Ma. The Westphalian A third order sequence contains up to 19 higher frequency fourth-order coal-bearing parasequence cycles with an average duration of ~86 ka/cycle, whereas the Westphalian B third-order sequence contains up to 21 higher frequency fourth-order parasequence cycles with an average duration of 112 ka/cycle.

Third-order maximum flooding surfaces record a maximum rise of depositional base-level when marine to brackish water influences were greater, peat mires were closer to the palaeoshoreline and fourth-order coal-bearing parasequences have a more brackish to marine character. In the middle, relative low-stand part of the sequence, the fourth order coal-bearing parasequences have a more non-marine character. The lowest sulphur coals in the Westphalian A are confined to the middle low stand part of the third-order sequence where brackish to marine influences are less. Coal sulphur values increase close to the lower and upper maximum flooding surfaces, defining the top and bottom of the sequence. In the Westphalian B, the lowest sulphur coals occur just above the maximum flooding surface in the lowermost part of the third-order sequence, contrary to predicted trends (Holz et al., 2002). This reflects local tectonism and a more rapid rise of base level compared to the Westphalian A. As a result brackish to marine influences were greater in the middle part of the Westphalian B sequence, and become more pronounced in the upper part, as evidenced by the more abundant marine bands approaching the *aegiranum* third-order maximum flooding surface (Fig. 12). Thus, stratigraphic variations in the sulphur content of coal seams show a generalised, but not consistent, increase in sulphur content upwards through the succession.

Superimposed on this generalised stratigraphic increase in sulphur content is a more significant regional variation. Regional sulphur distribution maps show a systematic increase in sulphur content across the coalfield to the east, towards the sediment source area, away from marine and brackish water influences in the south. Thus, extensive flooding of peat mires by saline waters from the south was probably not a significant control on regional sulphur distribution trends, and suggests, instead, a possible source rock control. Sulphur, oxidised and leached from source rocks, especially those actively undergoing uplift, may be carried into the basin by sulphate-enriched river water, which contributes towards the groundwater system on entering the basin. The sulphate- and ferrous-enriched groundwater infiltrating peat mires may produce sulphide. Likewise, source rock-derived iron, carried into the basin attached to clays, may infiltrate and contaminate peat mires from overbank flooding. Clays dominate coal ash, and if they acted as carriers then the sulphur and ash yields of the coals should show a broad correlation. No such correlation was found, and since the peat mires in the Northumberland Coalfield were predominantly planar rather than ombrogenous, and easily contaminated by detrital influx, a strong source rock control on the sulphur content of the coal, via this mechanism, seems unlikely.

Secondary epigenetic pyrite in the coal seams suggests post-depositional addition of sulphur into the coals, possibly from the North Pennine Orefield, along the southern margin of the coalfield. The sulphide and sulphate mineralisation in the orefield was emplaced in the late Carboniferous to early Permian, with most mineralisation being Permian in age. Sulphur-rich fluids may have been expelled northwards through the Coal Measures, towards the basin margin, about 280 Ma, following Variscan transpressional tectonism from the south. The isotopic composition of nine samples of pyrite and one sample of galena from the coals, show a high variability in δ^{34} values for pyrite of −5.4‰ to +32.8‰. The large variation in δ^{34}S with one high value of +32.8‰ is typical of secondary pyrite and suggests that: (1) local derivation of the sulphur is unlikely; (2) there is more than one generation of pyrite; (3) the pyrite had a marine sulphate source; and (4) the system must have been open to addition of δ^{34}S-enriched sulphur. The isotopic composition of coal pyrite shows a similar range in composition to the pyrite and other sulphides from Haydon Bridge and the North Pennine Orefield, which argues for a possible Mississippian (Lower

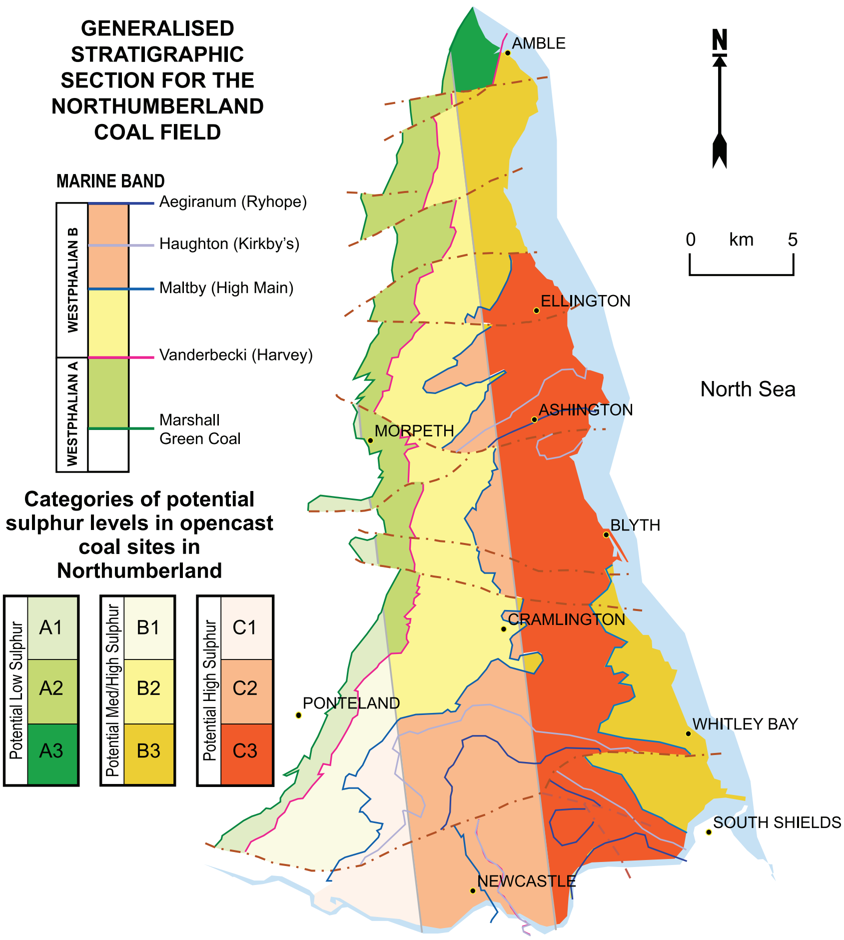


Fig. 16. Categorisation of the Northumberland Coalfield according to the stratigraphic and regional location of coals for potential opencast coal resources. Colours are based on stratigraphic location with the intensity of the colour due to overprinting of the regional effects of the variation in sulphur content (see text for details). No sulphur levels are shown on the map because it is intended to reflect a blend of coal from that particular section of strata rather than individual seams.

Carboniferous) marine sulphate source for some of the sulphur.

Plots of sulphur versus iron for five coal seams from the Plenmeller Opencast coal site, West Northumberland, show that inorganic sulphur dominates over organic sulphur, producing pyrite. Only one coal seam showed a strong affinity for organic sulphur; most seams have a consistent organic sulphur content. Between seam variations in sulphur content show no consistent trend. One seam had a higher sulphur content at the top and bottom of the seam, in accordance with observations on transgressive coal seams from elsewhere. High sulphur values in the top of the seams are consistent with observations on many marine influenced coals and modern marine-influenced peats, and the high sulphur value (12.8%) at the bottom of one seam reflects marine influences during initial peat deposition.

When the general regional increase in sulphur levels is superimposed on the main stratigraphic units, identified by marine band correlatives, a zonal classification scheme can be produced showing the potential of an open pit coal reserve to produce currently acceptable low sulphur fuel (Fig. 16). This approach has been used successfully as a guide to the exploration for new reserves in the Northumberland Coalfield.

Acknowledgements

We thank H.J. Banks and the British Geological Survey for providing data and financial support for the project, and Colin MacPherson for his helpful comments on the sulphur isotope data. The paper benefited from the helpful and incisive reviews of Rod Gayer and Alan Davis.

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