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Provincial variation of carbon emissions from bituminous coal: influence of inertinite and other factors

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Abstract

We observe a 1.3 kg C/net GJ variation of carbon emissions due to inertinite abundance in some commercially available bituminous coal. An additional 0.9 kg C/net GJ variation of carbon emissions is expected due to the extent of coalification through the bituminous rank stages. Each percentage of sulfur in bituminous coal reduces carbon emissions by about 0.08 kg C/net GJ. Other factors, such as mineral content, liptinite abundance and individual macerals, also influence carbon emissions, but their quantitative effect is less certain. The large range of carbon emissions within the bituminous rank class suggests that rank-specific carbon emission factors are provincial rather than global. Although carbon emission factors that better account for this provincial variation might be calculated, we show that the data used for this calculation may vary according to the methods used to sample and analyze coal. Provincial variation of carbon emissions and the use of different coal sampling and analytical methods complicate the verification of national greenhouse gas inventories. Published by Elsevier Science B.V.

Keywords: Coal; Carbon emissions; Greenhouse gas; Inertinite; Macerals; Sulfur

1. Introduction

Because the carbon content of commercially traded coal is rarely reported, carbon emissions from coal combustion are estimated in national inventories of greenhouse gas emissions (EIA, 2000). These estimates are typically calculated from the tons of coal burned, the specific energy of the coal and a carbon emission factor specified according to the rank of the coal. The carbon emission factor is expressed as the mass of carbon emitted per unit of energy that, when multiplied by the total energy in a given amount of coal, provides an estimate of the amount of carbon emitted when that coal is burned.

Smith (1997) examines many of the carbon emission factors that have been applied to coal and concludes that these emission factors are fundamental to national greenhouse gas inventories and are also useful to plan ways to reduce greenhouse gas emissions. The importance of the carbon emission factor is also evident in the reference method specified by the Intergovernmental Panel on Climate Change to tabulate carbon emissions from coal combustion (Houghton et al., 1997). This method requires that different countries use the same, rank-specific, carbon emission factors to ensure that resulting national inventories are “transparent and verifiable.”

Early greenhouse gas inventories (Marland and Rotty, 1984; Marland and Pippin, 1990) used a single

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Table 1
Selected Australian coal data

Coal name	M_{as}[a]	M_r[b]	Ash[b]	VM[b]	C[b]	H[b]	N[b]	S[b]	O[b]	Gross MJ/kg[b]	Net MJ/kg[c]	Mott–Spooner[d]	V[e]	L[e]	I[e]	R_o[f]	kg C/net GJ[g]
Bee Creek thermal		1.6	15.0	14.0	74.6	3.59	1.42	0.42	3.4	30.00	29.17	−0.37	51	0	49	1.70	25.56
Blackwater export coking	10.5	2.0	8.0	26.5	78.5	4.50	1.89	0.54	4.6	32.00	30.96	0.16	58	2	40	1.05	25.35
Blair Athol thermal	16.0	7.5	8.0	27.2	70.0	3.80	1.61	0.25	8.9	27.30	26.28	0.12	30	3	67		26.63
Burgowan 12 washed thermal		1.8	7.8	29.9	76.5	4.97	1.54	0.72	6.7	32.40	31.26	−0.60	89	0	11		24.47
Burton export thermal	8.0	1.8	14.0	20.2	75.1	4.04	1.43	0.34	3.3	30.11	29.17	0.50	28	0	72	1.22	25.74
Callide domestic thermal	15.5	10.9	18.9	23.7	54.8	2.74	0.77	0.14	11.7	21.10	20.23	−0.68	25	1	73		27.10
Clermont thermal	13.0	5.0	9.5	27.5	70.9	4.10	1.54	0.34	8.6	27.84	26.81	0.38	37	4	58		26.44
CollinsvilleBowen raw thermal	8.0	1.5	9.0	26.0	78.3	4.65	1.52	0.72	4.3	32.00	30.94	0.41	58	3	39	1.10	25.31
Collinsville export coking		1.3	14.5	20.2	73.8	4.13	1.52	1.18	3.6	30.57	29.63	−0.40	41	0	59		24.89
Cook export coking	9.0	1.4	7.0	27.5	80.5	4.58	1.92	0.37	4.2	32.80	31.76	0.20	62	2	36		25.35
Cook export thermal	8.5	1.6	13.0	24.0	74.2	4.10	1.71	0.34	5.0	30.10	29.16	−0.29	36	3	61		25.45
Cullin–La–Ringo lota seam thermal		4.1	4.5	32.4	75.3	4.75	2.10	0.27	9.0	30.54	29.39	0.27	59	5	36	0.81	25.62
Curragh export coking	9.5	1.5	7.0	21.5	81.2	4.58	1.56	0.55	3.7	33.00	31.95	0.32	57	1	42		25.40
Daunia coking	8.0	2.1	9.0	21.5	78.9	4.18	1.42	0.36	4.1	31.82	30.85	0.06	49	0	51		25.56
Daunia thermal	8.0	2.2	12.5	20.0	75.8	3.84	1.28	0.34	4.0	30.82	29.92	−0.64	29	0	71		25.35
Dawson raw thermal		6.0	6.5	32.3	69.1	4.20	1.66	0.26	12.3	27.85	26.78	−0.48	64	2	34	0.65	25.82
Ebenezer washed thermal	10.0	4.0	14.0	39.0	66.4	5.08	1.15	0.49	8.9	28.05	26.83	0.30	88	12	0		24.75
Ensham semi-soft coking	9.5	4.3	8.3	29.5	73.4	4.46	1.75	0.52	7.3	30.15	29.06	−0.18	74	2	23	0.85	25.26
Ensham top thermal	10.0	4.0	10.0	26.5	72.9	4.21	1.63	0.52	6.7	29.30	28.27	0.31	33	3	64	0.81	25.79
Ensham thermal	10.0	4.0	13.0	25.5	69.9	4.15	1.58	0.50	6.9	28.05	27.04	0.45	34	3	62	0.81	25.85
German Creek east export coking	10.0	2.0	9.5	20.0	79.3	4.16	1.42	0.53	3.1	31.80	30.83	0.40	33	1	66		25.72
German Creek export coking	9.0	2.0	8.5	20.0	79.7	4.48	1.79	0.72	2.9	32.30	31.26	0.53	74	0	26		25.48
Goonyella export coking	10.0	1.0	8.9	24.0	79.8	4.60	1.71	0.54	3.4	32.66	31.62	0.27	66	1	33		25.24
Gordonstone export coking	8.0	2.0	6.5	34.0	78.0	5.12	1.92	0.64	5.9	32.10	30.92	0.64	77	4	19		25.21
Gregory export coking	8.0	2.0	6.5	33.5	77.9	5.03	1.92	0.64	6.0	32.45	31.29	0.05	78	3	19		24.88
Hail Creek coking	10.0	1.0	8.0	21.4	81.4	4.46	1.46	0.36	3.4	32.99	31.98	0.25	61	0	39		25.44
Jeebropilly export thermal	10.0	5.0	14.0	39.0	65.0	4.86	1.22	0.57	9.4	28.05	26.86	−0.85	89	9	2		24.19
Jellinbah east semi-soft coking	8.0	1.5	8.5	15.5	80.4	4.23	1.71	0.54	3.2	31.82	30.85	0.55	53	0	47		26.05
Moranbah North coking	10.0	1.5	8.5	26.5	77.5	4.50	1.62	0.45	5.9	32.50	31.47	−0.99	58	1	41		24.62
Moura export coking	10.0	2.0	7.8	27.5	78.7	4.60	1.62	0.45	4.9	32.57	31.51	−0.30	67	2	31	1.00	24.96
Moura Kcoal thermal	10.5	2.5	8.3	32.0	75.5	4.73	1.69	0.45	6.9	31.30	30.20	−0.20	65	3	32	0.80	24.99
New Hope export thermal	10.0	3.7	16.0	35.0	65.9	4.58	1.28	0.40	8.1	27.63	26.53	−0.15	78	6	16	0.73	24.85
Newlands export thermal	8.3	2.3	14.5	26.3	70.8	4.24	1.41	0.42	6.3	28.48	27.49	0.55	44	4	52	1.02	25.76
North Goonyella export coking	9.0	1.5	8.5	23.5	79.3	4.59	1.53	0.45	4.1	33.00	31.95	−0.44	72	1	27		24.82
Norwich Park export coking	10.0	1.0	9.9	17.2	80.1	4.10	1.78	0.62	2.5	32.50	31.57	−0.07	79	0	21	1.65	25.37
Oakleigh domestic thermal	10.0	5.7	15.0	39.0	63.8	5.15	1.27	0.48	8.6	27.20	25.92	0.20	94	5	1		24.62
Oaky Creek export coking		1.2	8.0	28.9	78.8	4.99	1.82	0.73	4.4	32.50	31.37	0.56	77	2	21	1.10	25.12
Oaky Creek Oaky North coking	9.0	1.2	9.0	24.0	79.1	4.58	1.98	0.54	3.6	32.83	31.79	−0.23	81	2	16	1.30	24.89
Peak Downs export coking	9.5	1.0	9.7	20.5	79.6	4.38	1.79	0.63	2.9	32.50	31.51	0.09	76	0	24	1.40	25.25
Riverside export coking	10.0	1.1	9.8	23.5	79.0	4.46	1.78	0.62	3.2	32.21	31.20	0.30	63	1	36	1.17	25.33
Saraji export coking	10.0	1.0	9.7	19.0	79.8	4.29	1.79	0.63	2.8	32.50	31.53	0.08	76	0	24	1.55	25.32
South Blackwater export coking	8.5	2.0	6.2	28.8	79.5	4.59	2.02	0.46	5.2	32.24	31.18	0.30	62	2	36		25.50
South Blackwater export thermal	8.0	2.0	12.0	25.5	73.9	4.30	1.72	0.60	5.5	29.73	28.73	0.21	36	2	61		25.71
South Walker Creek thermal	8.5	2.0	15.0	13.3	74.5	3.49	1.41	0.66	2.9	28.89	28.07	0.90	50	0	50	1.95	26.55
Theodore raw thermal	9.0	4.5	12.0	31.0	67.8	4.34	1.50	0.50	9.4	28.04	26.97	−0.48	68	3	29	0.67	25.14
Theodore washed thermal	11.0	4.5	10.0	31.6	70.1	4.45	1.54	0.43	9.0	28.79	27.70	−0.21	72	3	25	0.67	25.31
Valeria thermal	12.0	5.0	9.0	33.5	70.8	4.56	1.72	0.69	8.3	29.30	28.17	−0.45	46	11	43		25.12
Wandoan Deposits Austinvale washed thermal	15.1	8.3	15.0	39.1	59.1	4.60	0.77	0.23	12.0	24.69	23.47	−0.12	79	9	12		25.19

Table 1 (*continued*)

Coal name	M_{as}[a]	M_r[b]	Ash[b]	VM[b]	C[b]	H[b]	N[b]	S[b]	O[b]	Gross MJ/kg[b]	Net MJ/kg[c]	Mott–Spooner[d]	V[e]	L[e]	I[e]	R_o[f]	kg C/net GJ[g]
Wandowan Deposits Austivale raw thermal	12.8	7.9	23.9	36.1	51.8	4.23	0.75	0.27	11.2	21.59	20.46	0.11	88	4	21		25.29
Wards Well coking		1.1	8.4	22.2	80.3	4.43	1.72	0.45	3.6	32.66	31.66	0.14	61	0	39	1.25	25.36
Wilkie Creek Surat premium thermal	12.5	7.5	12.0	41.0	63.0	4.83	0.89	0.32	11.4	26.64	25.39	− 0.29	76	14	10	0.40	24.82
Yarrabee thermal	9.0	2.0	10.0	9.5	80.7	3.17	1.58	0.70	1.8	31.19	30.44	0.00	41	0	59		26.51

Note: The table is based on the raw data presented by Coxhead (1997).

[a] As-sampled moisture.

[b] Air-dried, whole-coal basis, where: M_r is the wt.% residual moisture, VM is the volatile matter, oxygen (O) is by difference, and wt.% H and O do not include hydrogen and oxygen in moisture.

[c] Net MJ/kg calculated according to: net MJ/kg = gross MJ/kg − 2.45(M_r + 9H).

[d] Mott–Spooner difference in MJ/kg (measured specific energy minus specific energy calculated after Mott and Spooner, 1940).

[e] vol.% vitrinite (V) liptinite (L) and inertinite (I) on a mineral-free basis.

[f] Mean maximum reflectance of vitrinite in oil.

[g] Carbon emission factor.

carbon emission factor for all coal. Observations by Winschel (1990), Grubb et al. (1991) and Hong and Slatick (1994) later showed that the carbon emission factor varies with coal rank. Accordingly, current inventories (Houghton et al., 1997; EIA, 2000) use rank-specific carbon emission factors for lignite, subbituminous, bituminous, and anthracite rank coal.

Jones et al. (1984) recognize provincial variation of coal properties where correlations observed in one coal field are not representative of the relationship in other coal fields. Hong and Slatick (1994) observe that carbon emission factors for US coal vary with both rank and geographic origin, which supports the idea of provincial variation of carbon emission factors. However, US coal is not well suited to demonstrate provincial variation since it rarely encompasses the wide range of rank (Ode and Gibson, 1960) and maceral content (Cook, 1975) observed elsewhere. Although efforts to understand how coal rank influences carbon emissions continue (Quick and Glick, 2000), the possible influence of maceral content on carbon emissions has not been widely considered. To evaluate this possibility, we examine carbon emissions from bituminous Australian coal with variable maceral content. Finally, we qualitatively compare carbon emissions from Australian and US coal and note some problems that need to be considered to obtain an accurate, quantitative comparison of carbon emissions from coal produced in different countries or regions.

2. Data

We examine data for 52 Australian and 44 US coal samples. All are of bituminous rank according to the Australian standard classification (SAA, 1987). The Australian data (Table 1) are from Coxhead (1997), who collected the information from mining companies to represent commercially available coal products from mines in Queensland, Australia. The US data are from Quick and Glick (2000), with some additional data for these coal samples from the Pennsylvania State University coal database (PSU, 1990) and the US Department of Energy coal database (Davis and Glick, 1993). Most of the US data originate from analyses of whole-bed channel samples collected from active coal mines.

The selected data records include values for maceral content, moisture, ash, volatile matter, elemental composition (C, H, N, O, and S) and specific energy with a Mott–Spooner difference (Given et al., 1986) within 1 MJ/kg. Other coal quality data considered include sulfur forms, ash composition, carbonate CO_2, crucible swelling and vitrinite reflectance.

2.1. The effect of analytical methods on assay results

Systematic differences between the Australian and US coal data are possible because they were obtained using different analytical methods. The Australian data were obtained using methods specified by the Standards Association of Australia, whereas the US data were obtained using methods specified by the American Society for Testing and Materials. Although different analytical methods should give identical results for values such as carbon content and specific energy, the equivalence of empirical measures of coal quality, such as ash, volatile matter and moisture, is uncertain. Results from such analyses are influenced by extrinsic factors such as crucible shape, heating rate, maximum temperature, and specimen particle size. These factors vary according to the standard test method used for the analysis (Gray, 1983; Thompson, 1986).

An example of how the analytical method influences the assay results is shown in Fig. 1a. Note the different kinds of moisture assays (as-sampled, residual, equilibrium and as-received) as well as the different results indicated for US and Australian coal. The difference between equilibrium moisture (US coal) and residual moisture (Australian coal) is largely due to the different analytical methods used to obtain these values. Equilibrium moisture is measured after the coal specimen has been conditioned at 97% relative humidity, whereas the residual moisture is measured after the coal has been conditioned at the lower, ambient humidity in the laboratory. However, the large difference between the as-received moisture (US coal) and the as-sampled moisture (Australian coal) cannot be attributed to different analytical methods. In this instance, both methods should indicate the moisture content of the coal sample when it was collected.[1] The comparatively high as-sampled moisture values reported for the Australian coal are noteworthy and are discussed below.

2.2. The effect of sampling strategy on assay results

Systematic differences between the Australian and US coal data are also possible because they were obtained using different coal sampling strategies (Hower et al., 1989; Spackman, 1989). For example, because most of the US coal was collected as in-ground channel samples, the resulting coal data are probably more variable than the Australian data, which are intended to represent the quality of coal in commercial shipments. Channel samples will reveal local differences between (and within) adjacent beds in a single mine. Such variation would not be observed in commercially shipped products in which coal from different beds or parts of a mine are blended together.

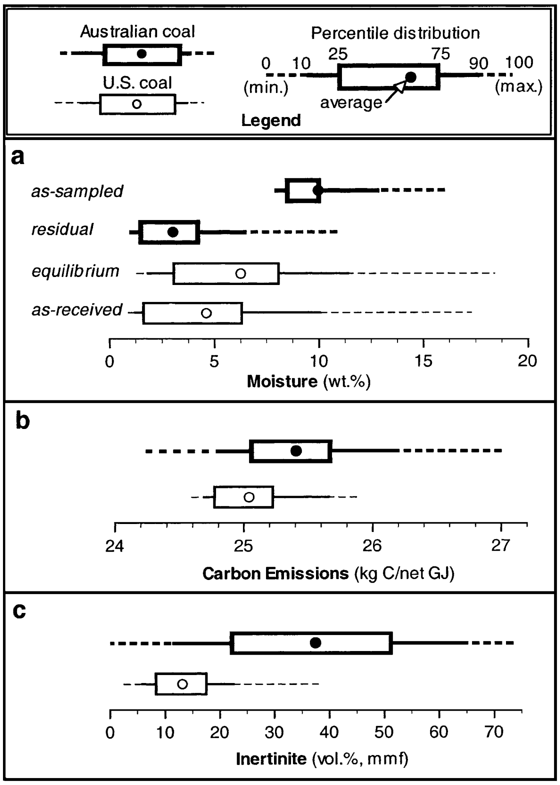

Fig. 1. Comparison of selected data for 52 Australian and 44 US bituminous coals: (a) Comparison of moisture values. *As-sampled* moisture is defined by SAA (1987); *residual* moisture is defined by SAA (1989) where it is called “air-dried moisture” (the name residual moisture is used here to avoid confusion with the much different “air-dry-loss moisture” sometimes reported in the US); *equilibrium* moisture is defined by ASTM (2000a); *as-received* moisture is defined by ASTM (2000b). (b) Qualitative comparison showing relatively high carbon emissions for Australian bituminous coal. Carbon emissions for Australian coal are calculated on a residual moisture basis, whereas carbon emissions for US coal are calculated on an as-received moisture basis. (c) Inertinite in the Australian coal shows a wide range of variation compared to the US coal.

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[1] Care was taken to minimize moisture loss between the time of collection and submittal of the US coal samples to the laboratory. As noted in ASTM (2000c, p. 10): “If the sample has been maintained in a sealed state so there has been no gain or loss, then the as-received basis is equivalent to the moisture basis as sampled.”

As noted above, the large difference between the as-sampled moisture values for the Australian coal and the as-received moisture values for the US coal (Fig. 1a) cannot be attributed to different analytical methods. Moisture in US bituminous coal is highest at the beginning of the bituminous ranks and declines as coalification advances through the bituminous rank stages (Damberger, 1971). Although this appears to be true for US coal, Fig. 2 shows that the Australian coal exhibits uniformly high moisture values throughout the bituminous rank stages. Moreover, we observe no clear relationship the between the as-sampled moisture content of Australian coal and other measures of coal quality (for example, ash yield or maceral content). McCutcheon and Barton (1999) likewise observe no relation between maceral and moisture content of Australian coal, although they do observe variation of moisture with mineral matter. Thus, although it is possible that the relatively high moisture content of the Australian coal (Fig. 2) is an inherent compositional feature, the lack of a relationship between moisture and other coal quality values suggests that these high moisture values are more likely caused by an external factor such as sampling strategy.

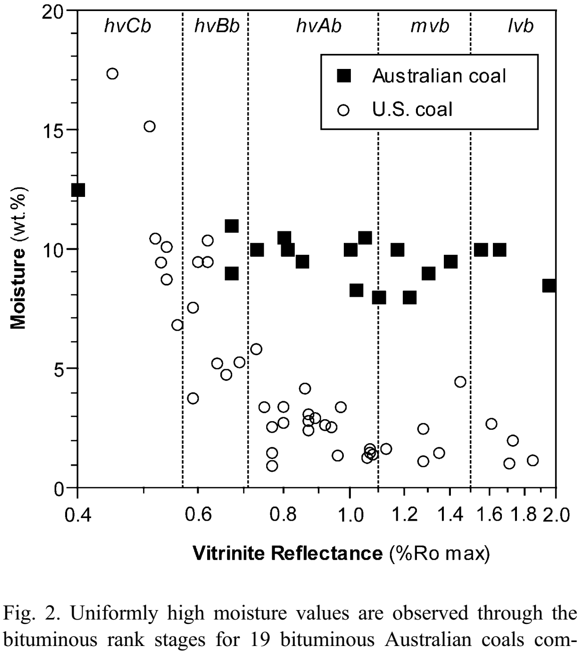

Fig. 2. Uniformly high moisture values are observed through the bituminous rank stages for 19 bituminous Australian coals compared to 44 bituminous US coals. The difference is attributed to sampling strategy (discussed in Section 2.2), which complicates the comparison of carbon emissions from US and Australian coal. Approximate reflectance limits for ASTM (1990) bituminous rank stages are modified from Davis (1984) where: *hvCb* is high volatile C bituminous, *hvBb* is high volatile B bituminous, *hvAb* is high volatile A bituminous, *mvb* is medium volatile bituminous, and *lvb* is low volatile bituminous.

Most of the Australian coal data represent coal products that have been washed at a coal preparation plant, whereas the US coal data represent samples collected at the mine prior to washing. Moisture usually increases during washing, especially where coal fines are recovered (Edwards, 1984). Evidence for moisture addition upon the washing of Australian coal can be found in Coxhead (1997); in the few instances where equilibrium moisture is reported, the equilibrium moisture values are lower than the as-sampled moisture values. Conversely, Fig. 1a shows that equilibrium moisture values for US coal are higher than the as-received moisture values. Thus, the comparatively high moisture values for the Australian coal (Fig. 2) probably result from sampling washed coal products rather than the in-ground coalbed.

3. Calculation of the carbon emission factor

Calculation of the carbon emission factor requires knowing both the carbon content and the specific energy. In this paper, the carbon emission factor is expressed as kilograms carbon per net gigajoule (kg C/net GJ) and is calculated from:

$$C_{\mathrm{ef}} = C\left(\frac{1000}{\mathrm{SE}_{\mathrm{net}}}\right) \tag{1}$$

where C_{ef} is the carbon emission factor, $\mathrm{SE}_{\mathrm{net}}$ is the net specific energy in MJ/kg, C is the percent carbon expressed as a decimal fraction, and the variables are on a whole-coal (moisture- and mineral-containing) basis. Note that we use net specific energy (sometimes called the lower heating value) rather than gross specific energy (sometimes called the higher heating value) in Eq. 1. As noted by Winschel (1990), the net specific energy does not include the latent heat of water vapor from combustion and consequently provides a better comparison of fuels used for combustion than gross specific energy. Water vapor from combustion originates from fuel moisture, as well as combustion of hydrogen in the fuel. Net specific energy (SE_{net}) is calculated from:

$$SE_{net} = SE_{gross} - 0.0245(M + 9H) \qquad (2)$$

where SE_{gross} is the gross specific energy reported from the laboratory, M is moisture, H is hydrogen (which excludes hydrogen in moisture), and the data are expressed on a whole-coal basis.

Sometimes, the moisture and hydrogen values used in Eq. 2 are not known. Lacking these values, the net specific energy cannot be calculated. In such instances, the carbon emission factor can only be directly calculated on gross energy basis (SE_{gross} rather than SE_{net} is used in Eq. 1).

For US coal, the difference between net specific energy and gross specific energy systematically varies with coal rank (Quick and Glick, 2000). Net specific energy is about 10% less than the gross specific energy at the lignite A stage of coalification, declining to about 2% less at the anthracite stage of coalification. Consequently, for US coal, the correlation between carbon emission factors expressed on a gross energy basis and carbon emission factors expressed on a net energy basis is largely a function of coal rank. Given the relatively high inertinite content of the Australian coal (Fig. 1c) the correlation observed for US coal is probably not representative of Australian coal. This is because Australian coal sometimes contains abundant inertinite, which contains less hydrogen than other macerals (Dormans et al., 1956). Assuming equivalent moisture content, Eq. 2 will show a smaller difference between net and gross specific energy for inertinite-rich coal than vitrinite-rich coal.

4. Comparison of carbon emissions from Australian and US coal

Eq. 2 shows that net specific energy is less than gross specific energy in proportion to the amount of moisture and hydrogen in the fuel. Because of the latent heat of water vapor, each 10% of moisture reduces the net specific energy by about 0.25 MJ/kg. Consequently, sampling strategy, which we suggest is responsible for the high moisture values for Australian coal shown in Fig. 2, influences carbon emissions. Other unrecognized systematic differences due to analytical methods or sampling strategy may also complicate any comparison of carbon emissions. Nonetheless, carbon emissions from Australian and US coal are compared in Fig. 1b.

Note that in Fig. 1b, carbon emissions factors for Australian coal are calculated using the data on a residual moisture basis, whereas emission factors for US coal are calculated using data on an as-received moisture basis. Although residual moisture and as-received moisture are measured using different analytical methods, they are used because they show the most similar results (Fig. 1a). Slightly higher carbon emission factors result if the data used in Eqs. 1 and 2 are expressed on an as-sampled or equilibrium moisture basis.

Given the likely bias due to different analytical methods and sampling strategies, the comparison of carbon emission factors shown in Fig. 1b is not definitive. An accurate comparison would also require weighting the results according to production tonnage or energy and restricting the comparison to the coal used for combustion. The comparison in Fig. 1b simply shows sample frequency, without differentiation of coal used for combustion or coal used for making coke. Nonetheless, Fig. 1b *qualitatively* illustrates that carbon emissions from bituminous coal from Queensland, Australia are higher and more variable than carbon emissions from bituminous US coal.

5. Macerals and carbon emissions: theoretical considerations

Eq. 1 (Section 3) shows that both carbon content and specific energy are required to calculate the carbon emission factor. Although such data are sometimes available for whole coals, values for the carbon content and specific energy of macerals are not. The carbon content of individual macerals can be directly measured using an electron microprobe (Gurba and Ward, 2000), but measuring the specific energy of macerals has not been accomplished. Fortunately, the specific gravity of different maceral groups systematically varies (Dormans et al., 1956), which allows the isolation of density fractions that are enriched in different macerals (Dyrkacz et al., 1984). Although specific energy values for isolated density fractions have not been measured, their elemental compositions can be determined (Choi et al., 1989), which allows the calculation of specific energy using the Mott and Spooner (1940) equation. Knowing both the carbon content and the specific energy allows the calculation of carbon emission factors for the maceral-enriched density fractions. This approach is used in Fig. 3 to suggest that different macerals contribute different amounts of carbon per unit of energy.

Fig. 3 suggests that carbon emissions from inertinite are greater than those from vitrinite and that liptinite group macerals have the lowest carbon emissions. Although informative, the approach used in Fig. 3 to estimate relative carbon emissions from different macerals has limited practical value. This is because the elemental analyses of specimens isolated by density gradient centrifugation are reported on a dry, ash-free basis, whereas the coal is mined and burned in a moist, mineral-containing condition. Furthermore, the Mott–Spooner calculations used to construct Fig. 3 do not reveal variation of the enthalpy of decomposition of different macerals. For example, since the heat of formation increases with aromaticity (Given et al., 1986) and inertinite macerals are more aromatic than vitrinite and liptinite group macerals (Choi et al., 1989), the enthalpy of decomposition should be relatively high for inertinite group macerals. Consequently, the actual range of variation is likely greater than that indicated by Fig. 3. Finally, the possible effect of incomplete recovery of maceral fractions as well as compositional changes due to demineralization, fine grinding or imbibed fluids during density gradient fractionation are uncertain. Despite these complications, the relationship shown in Fig. 3 is useful because it qualitatively illustrates how the maceral content of coal influences carbon emissions.

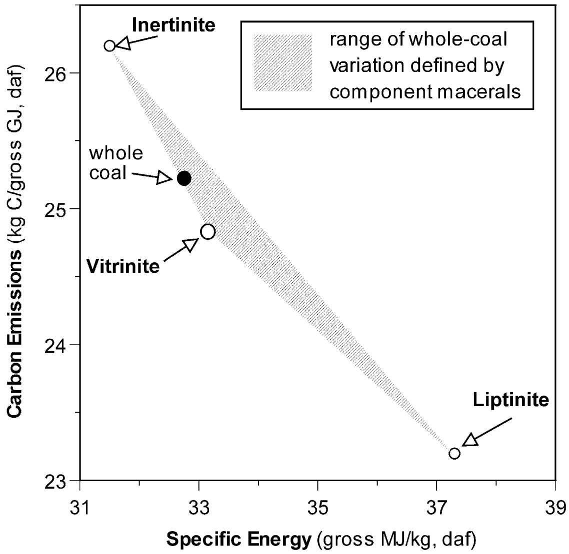

Fig. 3. Carbon emissions vary according to the relative abundance of vitrinite, liptinite and inertinite in a single bituminous rank US coal (West Virginia, Upper Kittanning coalbed). The figure was made using the Mott–Spooner equation (Given et al., 1986) and data on a dry ash-free basis (daf) from Choi et al. (1989). The significance and limitations of this approach are discussed in Section 5.

Quick and Glick (2000) observe that US coal with relatively abundant inertinite has higher carbon emissions than predicted from ASTM (1990) rank parameters. However, because US coal rarely contains much inertinite (Waddell et al., 1978), they conclude that carbon emissions from most US coal can be reasonably estimated from the coal rank alone. Unlike US coal, the Australian coal has more variable inertinite content (Fig. 1c), which makes the Australian coal data useful to evaluate the influence of inertinite on carbon emissions.

6. Inertinite and carbon emissions: correlation and complicating factors

If, as suggested by Fig. 3, abundant inertinite increases carbon emissions, then inertinite abundance should positively correlate with carbon emission factors determined for whole coals. Fig. 4 confirms the positive correlation between inertinite abundance and carbon emissions for bituminous Australian coal. Although Fig. 4 provides good evidence that inertinite in bituminous coal increases carbon emissions, the correlation is modest and deviations from the trend are apparent. A cross-plot (not shown) of the inertinite content vs. the squared residuals obtained from the relationship shown in Fig. 4 also indicates a mild systematic error. We suggest that the scatter around the best-fit line shown in Fig. 4, as well as the small systematic error indicated for the regression model, are caused by other factors, besides inertinite abundance, that influence carbon emissions.

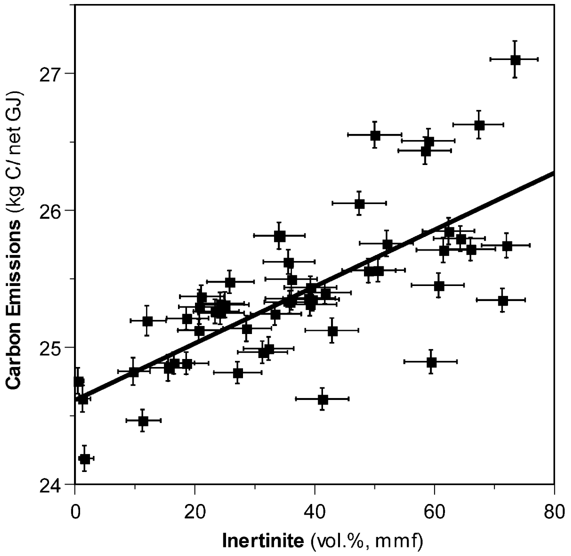

Fig. 4. Carbon emissions (C_{ef}) for 52 bituminous Australian coals increase with inertinite content (I) according to the equation $C_{ef} = 0.02075I + 24.62$. The correlation is modest (adjusted $R^2 = 0.52$, standard error = 0.39 kg C/net GJ), suggesting that other factors also influence carbon emissions from bituminous coal. Data are from Table 1. The error bars show 95% confidence limits due to the analytical precision associated with the measurement of inertinite and carbon emissions.

6.1. Precision

Error bars on Fig. 4 show 95% confidence limits due to analytical precision. As observed by Winschel (1990), the analytical error has a very small effect on the carbon emission factor. Although the error associated with the measurement of inertinite is slightly more significant, examination of Fig. 4 shows that the scatter about the regression line exceeds that which might be expected due to the limited precision of maceral analysis.

6.1.1. Estimating precision

Error associated with the measurement of inertinite content was estimated using tables and equations presented by Burstein (1971). Each estimate assumes 500 total maceral counts. Results from Burstein's binomial distribution model are similar to those obtained using the equations specified by SAA (1986) and ISO (1984) but increasingly differ as inertinite abundance approaches 0% or 100%.

The net cumulative effect of analytical measurement error on the calculated carbon emission factor was empirically determined. First, we consider the analytical precision associated with the measurement of the carbon, hydrogen, moisture and specific energy values used in Eqs. 1 and 2 to calculate the carbon emission factor. The influence of measurement error associated with each assay value was directly calculated by adding, and subtracting, half of the repeatability value listed in BSI (1981) from the corresponding assay result for a data record. The resulting data pair was then used in Eqs. 1 and 2, and the change to the carbon emission factor (the average, absolute deviation) was recorded as the expected standard deviation of the emission factor due to the error associated with a single assay. In this manner, we obtained a set a four expected standard deviations of the carbon emission factor for each data record corresponding to the individual effects of carbon, hydrogen, moisture and specific energy. Next, the calculated carbon emission factor of the data record was listed one million (10^6) times. Using this data set and the expected standard deviation for one of the assay values, we randomly generated a normally distributed million-member population to approximate the expected range of possible carbon emission factors. This process was repeated for the remaining three variables except that the initial population was the result of the previous iteration. The confidence interval was identified as the 2.5th and 97.5th percentiles of the final (fourth) million-member population for a given data record.

6.2. Other macerals

Another possible explanation for the scatter shown in Fig. 4 is that, besides inertinite, the relative amounts of vitrinite and liptinite also influence whole-coal carbon emissions. This is suggested by Fig. 3 in which the carbon emission factor for the parent coal plots within a triangular area defined by the three component maceral groups. Thus, carbon emissions appear to vary according to the relative amounts of all three maceral groups in a coal rather than any single maceral component. However, because the liptinite content of the Australian coal is uniformly low (Table 1), the variation of liptinite content is unlikely to be responsible for the scatter in Fig. 4, and inertinite content remains the dominant factor.

Less certain is the possible effect of compositional variation within individual maceral groups on carbon emissions. For example, the higher hydrogen content of detrovitrinite compared to telovitrinite (Brown et al., 1964) suggests that detrovitrinite will have lower carbon emissions than telovitrinite. Such variation is also suggested by methods used to predict the carbonization behavior of coal where certain inertinite group macerals are counted as reactive rather than inert components (Diessel, 1998).

6.3. Minerals

Whole-coal carbon emissions are also influenced by mineral content. Carbon dioxide released by the endothermic decomposition of carbonate minerals during coal combustion (Alpern et al., 1989) increases carbon emissions from coals that contain carbonate minerals. Direct measurement of the carbon dioxide liberated from coal with acid shows that carbonate minerals in the US coal contribute as much as 0.2 kg C/net GJ but average about 0.04 kg C/net GJ. The Australian data lack the measured values for carbonate carbon. However, using sulfur forms and ash composition data (Coxhead, 1997), and assuming that all the iron not in pyrite is present as siderite and all calcium is present as calcite, stoichiometric considerations suggest that carbonate minerals do not greatly influence carbon emissions from the Australian coal.

Although carbonate minerals appear to have a minor effect on carbon emissions from Australian and US coal, they may be more significant elsewhere. For example, Özdoğan (1998) suggests that consideration of carbonate mineral abundance should improve the prediction of carbon emissions from Turkish coal. Abundant carbonate minerals have also been observed in coal from Thailand (Ward, 1991).

For many coals, the total amount of mineral matter is probably more important than the amount of carbonate mineral present. Quick and Glick (2000) suggest that, for bituminous US coal, each 10% ash adds about 0.1 kg C/net GJ, presumably because the enthalpy of decomposition of mineral matter reduces the available heat of combustion. Nonetheless, the Australian coal averages about 10% dry ash without much variation, so the mineral abundance is not the main cause of the scatter shown on Fig. 4.

6.4. Sulfur

Sulfur in coal contributes about 9.3 gross MJ/kg sulfur (Given et al., 1986) and no carbon emissions. Consequently, as noted by Winschel (1990), high-sulfur coal produces lower carbon emissions than the otherwise similar low-sulfur coal.

A theoretical, sulfur-free carbon emission factor is calculated by first subtracting the sulfur's contribution to the gross specific energy according to:

$$\mathrm{SE}_{\mathrm{gross,sf}} = \frac{100}{(100 - S)}(\mathrm{SE}_{\mathrm{gross}} - 0.0926S) \tag{3}$$

where $\mathrm{SE}_{\mathrm{gross,sf}}$ is the gross MJ/kg on a sulfur-free basis, S is the wt.% sulfur on a whole-coal basis, and $\mathrm{SE}_{\mathrm{gross}}$ is the gross MJ/kg on a whole-coal basis. Next, the whole-coal wt.% carbon (C), hydrogen (H) and moisture (M) values are adjusted to a to a sulfur-free ($_{\mathrm{sf}}$) basis by multiplying these values by the factor,

$$\frac{100}{(100 - S)}.$$

A sulfur-free carbon emission factor is obtained by using the resulting values ($\mathrm{SE}_{\mathrm{gross,sf}}$, C_{sf}, H_{sf} and M_{sf}) in Eqs. 2 and 3. Results from these calculations are used in Fig. 5, which shows that for each wt.% sulfur in bituminous coal, carbon emissions decline by about 0.08 kg C/net GJ.

Note that the relatively high carbon emission factors indicated for Australian coal in Fig. 1b may be partly related to their low sulfur content. The average 0.5% sulfur in the Australian coal (Table 1) corresponds to an average 0.04 kg C/net GJ reduction of carbon emissions, whereas the average 1.3% sulfur in the US coal corresponds to an average carbon reduction of about 0.11 kg C/net GJ. Nonetheless, given the generally low sulfur content of the Australian coal (Table 1), the variation of sulfur content is not responsible for the scatter shown on Fig. 4.

6.5. Extent of coalification

Although the coals examined in this study are all classified as of bituminous rank, this class spans a wide range of coalification and the properties of any single bituminous coal will vary according to its

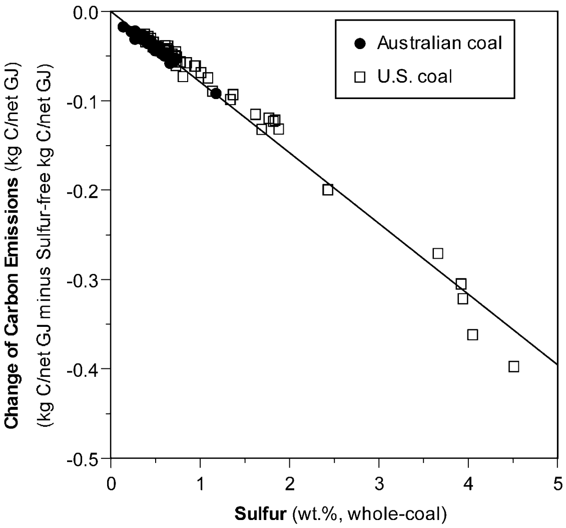

Fig. 5. Lower carbon emissions (ΔC_{ef}) are observed with the increasing sulfur (S) content of 52 Australian and 44 US bituminous coal samples. The equation for the regression line is $\Delta C_{ef} = -0.07913S$, which has an adjusted $R^2 = 0.97$ and a standard error = 0.01 kg C/net GJ. Note the low sulfur values for the Australian coal (average 0.5% sulfur). Calculation of carbon emissions on a sulfur-free basis is discussed in Section 6.4.

extent of coalification. Quick and Glick (2000) observe that carbon emissions from the US coal systematically vary through the bituminous rank stages by about 1.3 kg C/net GJ. We expect a similar systematic variation between carbon emissions and the extent of coalification for Australian bituminous coal. Such variation, if present, would explain nearly all of the scatter about the regression line for the Australian coal shown in Fig. 4.

7. Multiple regression analysis

In Section 6, we show that the inertinite content is positively correlated with carbon emissions (Fig. 4) and suggest that deviations from this trend are mostly caused by the variation of the extent of coalification through the bituminous rank stages. To test this idea, we use multiple regression analysis to find out if consideration of both the inertinite content and the extent of coalification, together provides a better prediction of carbon emissions than consideration of inertinite content alone.

Using ASTM (1990) rank-defining properties, Quick and Glick (2000) observe that carbon emissions from bituminous US coal are highest at the beginning, and at the end, of the bituminous rank class and are lowest near the middle (within the hvAb stage). However, the ASTM (1990) classification is unsuited to evaluate Australian coal. This is because Australian coal sometimes contains abundant inertinite that significantly increases the fixed carbon value (Strauss et al., 1976), which is used in the ASTM (1990) classification to distinguish the medium volatile bituminous and low volatile bituminous rank stages. Accordingly, we use available vitrinite reflectance data (Table 1) to evaluate the extent of coalification for 24 Australian coals.

The result of the multiple regression analysis, where vitrinite reflectance and inertinite content are used to predict carbon emissions, is illustrated in Fig. 6. Note that to accommodate the curvilinear variation of carbon emissions through the bituminous ranks (Quick and Glick, 2000), we include vitrinite reflectance squared as an explanatory variable in the regression analysis (Davis, 1973). The resulting predictive

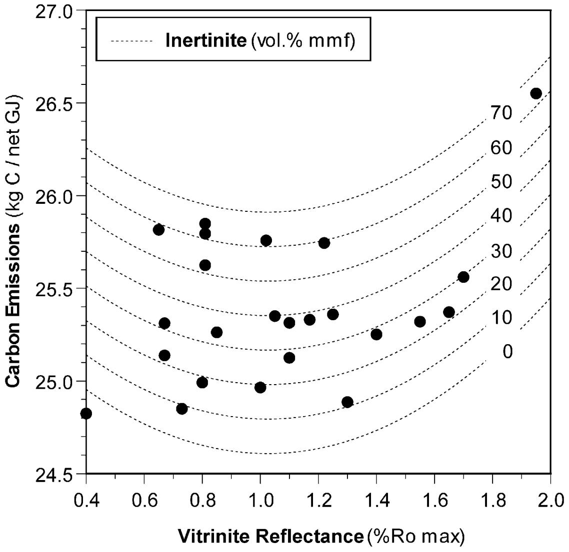

Fig. 6. Carbon emissions (C_{ef}) from 24 Australian bituminous coals vary with both inertinite content (I) and vitrinite reflectance (R_{o}), according to the equation: $C_{\mathrm{ef}} = 0.01861I - 1.814R_{\mathrm{o}} + 0.8848R_{\mathrm{o}}^2 + 25.54$, which has an adjusted $R^2 = 0.71$ and a standard error = 0.21 kg C/net GJ. The equation is the result of multiple regression analysis. Data used for the analysis are plotted on the figure.

equation (see caption, Fig. 6) shows an adjusted $R^2 = 0.71$ and a standard error = 0.21 kg C/net GJ. This is a much better prediction than that shown in Fig. 4, where inertinite alone is used to predict carbon emissions, which has an adjusted $R^2 = 0.52$ and a standard error = 0.39 kg C/net GJ.

Fig. 6 shows that only one data record below 0.6% reflectance and one data record above 1.7% reflectance are included in the multiple regression analysis. Nonetheless, the same trends are observed when the analysis is repeated without these two data records, which indicates that they are not unduly influential.

Examination of the regression equation listed in the caption for Fig. 6 shows that each 10% inertinite increases carbon emissions by about 0.19 kg C/net GJ. This corresponds to a total variation of 1.3 kg C/net GJ for the 0–72% range of inertinite content observed in the Australian coal data (Table 1). This equation can also be used to evaluate the potential effect of the extent of coalification on carbon emissions from Australian bituminous coal. For example, consider a hypothetical data set where inertinite content is fixed but vitrinite reflectance varies from 0.47 to 2.05, which corresponds to the approximate beginning and end of the bituminous rank class (Davis, 1984). Applying the equation listed in the caption for Fig. 6 to this hypothetical data set shows a potential 0.9 kg C/net GJ range of variation due to the extent of coalification within the bituminous rank class.

8. Discussion and conclusions

Our results show that inertinite abundance influences carbon emissions from bituminous rank coal. The effect of inertinite in coal belonging to other rank classes is uncertain. Because the optical properties of different maceral groups change at different rates during coalification (Smith and Cook, 1980), it seems likely that the composition of inertinite and its relative contribution to carbon emissions likewise change during coalification.

Coal data obtained using different analytical methods may systematically vary and lead to biased carbon emission factors. However, the significance of any bias due to analytical methods is not known. We suggest that sampling strategy is the primary cause of the different moisture values shown in Fig. 2, which, in turn, has a direct effect on the carbon emission factor calculated for the Australian and US coal. The use of different analytical methods and coal sampling strategies hinders the comparison of carbon emissions from coals produced in different countries or regions.

Fig. 6 empirically shows that higher carbon emissions are predicted for bituminous coal with abundant inertinite, which is consistent with the theoretical relationship illustrated in Fig. 3. We calculate that the 0–72% range of inertinite content for the Australian coal listed in Table 1 causes a 1.3 kg C/net GJ variation of carbon emissions. We also suggest that carbon emissions from bituminous Australian coal will systematically vary by 0.9 kg C/net GJ due to the extent of coalification within the bituminous rank class and predict a carbon emission minimum near 1.02% vitrinite reflectance. This is similar to the 1.3 kg C/net GJ variation of carbon emissions due to the extent of coalification of bituminous US coal (Quick and Glick, 2000).

Carbon emissions from bituminous Australian coal may vary by as much as 2.2 kg C/net GJ due to the combined effect of coalification and maceral abundance. The significance of this range of variation can be appreciated by noting that this is more than five times the difference between global emission factors (Houghton et al., 1997) applied to bituminous and subbituminous rank coals (25.8 and 26.2 kg C/net GJ, respectively). The variation of carbon emission within the bituminous rank class illustrates the doubtful validity of global, rank-specific carbon emission factors for estimating greenhouse gas emissions from coal.

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