

Selected references for submission with RE Sources scoping comments for GPT

- ⌘ A RAIL EMISSION STUDY FUGITIVE COAL DUST ASSESSMENT AND MITIGATION
- ⌘ Bibliography of coal dust articles from BNSF filing with Transportation in 2010
- ⌘ Biological Effects of Unburnt coal in the Marine Environment (2)
- ⌘ Bulk Carrier Structure Sept. 2012
- ⌘ Bunker-spill-Risk 2001
- ⌘ Extreme Waves and Ship Design 2007
- ⌘ Coal dust dispersal around a marine coal terminal in BC
- ⌘ MINIMIZING GROUNDWATER CONSUMPTION FOR REQUIRED FUGITIVE DUST CONTROL PROGRAMS
- ⌘ The Role of Chemicals in Controlling Coal Dust Emissions
- ⌘ Unexpectedly high mortality in Pacific herring embryos

A RAIL EMISSION STUDY: FUGITIVE COAL DUST ASSESSMENT AND MITIGATION

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ABSTRACT

A four-year study on fugitive coal dust emissions has produced estimates of coal loss during rail transport and developed suppression techniques that can reduce dusting from rail cars by 95 to 99%. The critical issues of emission characterization and material loss quantification had to be resolved before cost effective dust control strategies could be implemented and evaluated. Laboratory assessments, computer-based simulations, and field experiments were used to model and quantify coal dust emissions. These methods revealed coal losses along a ~500 mile-long rail corridor of up to 0.6 tons/car, with typical losses of 0.2 to 0.4 tons/car from metallurgical coals occurring under sunny, dry and windy conditions. A combination of load-top grooming, surfactants, and chemical binding agents proved to be the most effective method for reducing fugitive coal dust emissions during transit.

INTRODUCTION

Fugitive coal dust from in-transit coal cars does not appear to violate ambient air-quality standards. In fact, track-side monitoring of PM-10/TSP yielded no firm basis for remedial action. At issue, however, is the railroad's goal to reduce coal dust emissions and their impact as a nuisance pollutant.

Most of the evidence of fugitive coal dust emissions comes from anecdotal reports of dust plumes or the observations of coal deposition along the rail corridors. Without any standards of objectivity, coal dust complaints have given rise to the perception of significant a coal dust problem. Accordingly, a study was designed to relate the perceived problem (i.e., visual emissions) to the existence of quantifiable material losses (i.e., material losses that may represent significant environmental impact and/or financial consequences).

Previous attempts to quantify material losses produced mixed and controversial results, (Brown and Speichert, 1976; Guarnaschelli, 1977; Hardy Associates, 1979; Cope, 1980; McCoy, 1980; Williams, et al., 1982; Nobel, et al., 1983; Morrison, Hershfield Ltd., 1983; Cope, et al., 1984; Swan Wooster Engineering Co. Ltd, 1985; Environmental Sciences Ltd., 1985; Cope, et al., 1986; Wituschek, et al., 1986; Stewart, et al., 1987; Mikula and Parsons, 1988). Therefore, the characterization and quantification of losses along Norfolk Southern's (NS) rail corridors were identified as critical issues to be resolved before prescribing effective control strategies. Since early 1991, NS and Simpson Weather Associates (SWA) have conducted numerous laboratory and field-rail experiments to assess the magnitude of material losses and develop techniques to mitigate fugitive coal dust emissions during transit. A coal shipper, CONSOL also contributed to the field studies. This paper presents an overview of the study's ongoing efforts and results to date.

GENERAL STUDY APPROACH

The Norfolk Southern Rail Emission Study (NSRES) was conducted within one rail corridor, through which primarily export metallurgical (met) coal was transported. The choice of the rail corridor was based on its variety of terrain, relatively heavy volume of coal traffic, and the number of coal-dust complaints received. Metallurgical coal was chosen since, in most cases, it is considered more dusty than steam coal.

Field Trials

In an attempt to overcome some of the problems encountered in previous studies, the NSRES employed a number of independent field measurements to 1) act as quality-assurance checks within data sets, 2) to identify and understand aberrant measurements, and 3) to corroborate findings between data sets. Much of the early field data was gathered using a specially designed research caboose. As the study progressed, the instrumentation became more compact, thus reducing the need for the research caboose.

Scale Weights

The first of the field data sets is car weights. These weights were measured using static, decoupled, electronic scales. The scales have a reported accuracy of 0.01%. The weights were taken of selected cars before transit and then again after transit. As a reference, a scale monitor car that traveled with each weighing experiment was weighed at both locations to determine a scale correction factor. In addition, a tarped coal car was used, on occasion, as a second reference. It was assumed that no coal was lost during transit from the tarped car, and moisture loss and gain was minimized. To accurately evaluate the weight changes in coal cars moving from mine to port, moisture variations were taken into account. To account for moisture changes, a water budget was developed containing all known variables of moisture movement in and out of coal cars. Measured rainfall and estimated evaporation values were assigned to the water budget variables so that moisture changes could be used to adjust the scale weight differences. Moisture change correction factors were also empirically generated from coal samples collected in the field. In spite of all the precautions taken to assure accurate scale weights, an uncertainty in coal losses ± 200 lbs. still remains. This is most likely due to inherent scale inaccuracies and moisture changes that cannot be precisely measured, such as water dripping out the bottom of hopper doors. Because similar problems with scale weights have been encountered in other railroad work, we decided not rely on scale weight changes as the sole determinate for material losses. Rather, we used scale weights and three other methods jointly to arrive at a material loss estimates. These other methods are described below.

Load-top Volume Changes

The second method used to estimate material losses involved measuring the volume changes on the top of the coal loads from the mine to port. For the first several field trials, a series of photographic transects were taken in selected coal cars at various points along the rail corridor. Scaled photographs of the same cars were compared throughout the trip and material losses were calculated based on volume losses within a given car. Coal within each car settling was taken into account and samples were taken to obtain bulk densities for the mass-loss calculations. It should be noted, that as a part of these calculations, we assumed that no coal was detrained from the top, flat portion of the coal load during transit. Because of this assumption, mass-loss calculations based on volume losses tended to *underestimate* actual material losses.

The photographic method of calculating, while general successful, encountered problems related to the changes in bulk densities of coal as it dries and drifts and inadequate measurements in the fronts and rears of cars where significant erosion and redeposition can occur during transit. In addition, the photographic method was very labor intensive. Consequently, another method was developed to estimate volume changes and evaluate redistribution of coal within a car. This method, called the Coal Car Load Profiling System (CCLPS), used three cameras to produce a digital contour map of the coal surface and calculate volume changes from mine to port within a given car. Recently, the CCLPS data gathering process has evolved into an infrared laser-based system which is smaller, faster, and does not require special lighting as did the three-camera technology.

Real-time Observations

To characterize the nature of fugitive dust emissions and develop an understanding of the wind erosion processes on coal cars during transit, an instrument package was designed to monitor a variety of environmental parameters in real time as the cars moved down the rail corridor. The instrument package, Rail Transport Emissions Profiling System (RTEPS) measured the following variables: wind speed, wind direction, rainfall, coal surface

temperature, coal temperature and moisture at two different depths, fugitive emissions (using a real-time aerosol sensor, or RAS), air temperature, and relative humidity. All of these data were collected and stored in a data logger attached to RTEPS and were retrieved via a lap top computer at various locations along the corridor. A time-lapse video camera was also part of RTEPS to provide visual records of emission events.

Passive Collection

To directly sample detrained material in transit, passive collectors were designed and built to mount on the rear sill of test cars. The passive collectors were sampled at various stops along the rail corridor to help identify the dustiest portions of the trip.

Dust Suppression Techniques

Once it was determined how much coal was being lost during transit, several mitigation techniques were evaluated, including:

- water only (40 to 100 gallons/car, depending on the experiment);
- grooming ("rounding" of the load profile) only;
- water and compaction;
- surfactants only;
- surfactants plus binding agents;
- binding agents only; and
- tarped cars (used as control cars for various experiments).

Experiments were also conducted where the average train speeds were decreased, and where trips were run mostly at night to decrease emissions. While lower train speeds and coal surface temperatures produced less stress on the coal loads and therefore lower emissions, such operational constraints were neither sufficiently effective nor practical and therefore were not seriously considered as permanent mitigation techniques. In addition, several load profile modifications were used alone, and with the treatments listed above, to abate fugitive dust emissions. Initially, a "normal" profile had a trapezoidal cross-section as shown in Figure 1a. After it was shown that profile modification alone significantly reduced emissions, the "bread-loaf" or groomed profile became the norm (Figure 1b). Other grooming/loading options included loading the coal flat, at or below the car sill level, loading lower than normal, and reshaping the top of the load into the "bread-loaf" shape. For clarification, the following definitions are given for surface treatments.

Normal profile: for the first sixteen field trials, cars that had a trapezoidal cross-section (Figure 1a); for the last fourteen field trials, cars that had an arcuate or "bread-loaf" cross-section (Figure 1b).

Groomed profile: any car that had an arcuate cross-section, or was modified to eliminate angular or trapezoidal cross-section.

Untreated cars: cars that may or may not be groomed, but received no additional water spray, surfactants, nor chemical binders.

Treated cars: cars that may or may not be groomed, but did receive additional water spray and/or surfactants, and/or chemical binders.

RESULTS

Laboratory Evaluations

Using the relative dusting index generated from the SARTDX experiments, coals were ranked according to their dusting potential. The final overall rankings were based on combining three dusting parameters: 1) wind speed

threshold (WST), or the lowest wind speed at which emissions were detected; 2) maximum real-time aerosol monitor (RAM) readings; and, 3) total integrated emissions (IE), the calculated area under the entire emissions curve.

Interestingly, when the overall dustiness rankings based on the above three parameters were compared to what the rankings would have been based only on moisture content and fines content, the rankings were found to be discordant. While it is assumed that moisture content and size consist do play a role in a coals' dusting potential, it is clear that other factors (e.g., coal chemistry, moisture migration through the coal, and angle of repose) can play an equally important or even dominant role in dusting during transit.

For the 19 different coals tested in the SARTDX experiments, the inherent coal moisture contents ranged from 2.8 to 11.4%. In order to test all coal samples under the same conditions, it was necessary to dry all samples to approximately 1.5% moisture content ($\pm 0.5\%$). It is fully recognized that such drying procedures do not reflect actual field conditions, as moisture contents vary significantly from mine to mine. However, the drying process allowed for marked and consistent delineations between the different coals' dusting potential, which was the objective of the SARTDX experiments. Figures 2 a and b, below, show SARTDX wind tunnel plots for two coals. Coal # 1, (Figure 2a) displays a moderate tendency to dust, while Coal # 2 (Figure 2b) shows a much greater propensity to dust during transport. This is displayed in the upper parts of the graphs, along the "Mini-Ram" axis.

Field Studies

Scale Weight Changes

During the field trials, 317 cars were weighed. For the earlier field experiments, a normal profile for a fully loaded coal hopper was trapezoidal in cross-section, had a smooth flat top-surface, and was stacked approximately eighteen to twenty-four inches above the car sill. After taking moisture changes into account, the normally loaded, untreated cars lost an average of 0.36 tons (± 0.1 tons), $n = 52$. The range for the scale-weight losses was from 0 to 0.6 tons, and some cars actually showed a weight gain--due to water uptake during transport. The greater losses occurred during the most severe (hottest and driest) conditions in the summer months, when wind and train speed averages were highest compared to other field trials.

Those cars that were loaded at or below the sill appeared to loose less coal in most cases, compared to normally loaded and untreated loads, but this difference was not statistically significant. Furthermore, these loading techniques reduced the load capacity for each car by 10 to 15%. Since loading at or below the sill gave mixed dust control results and reduced the load capacity, this dust suppression strategy was abandoned.

For the most recent field trials, the normal load-out procedure was changed to a "bread loaf" profile. The change in profile produced a measurable reduction in the weight losses for the untreated cars, with an average of approximately 0.20 tons (± 0.1), down from the 0.36 tons for ungroomed cars. While load profile changes produced significant decreases in weight losses, further reduction in material losses (95 to 99% from untreated cars, based on passive collection) was achieved by applying surfactants and/or binding agents to the groomed profiles.

RTEPS Data

The RTEPS instrument package offered an independent and corroborative perspective of material losses compared to the scale weight changes and passive collection. RTEPS was not designed to quantify material losses, but to record in real time the intensity and frequency of dusting "events." We emphasize that the emissions are a relative measure (relative to no emissions), and do not represent material losses. There is a strong positive correlation between frequent, intense dusting events during the course of a trip and its scale weight changes and passive collection. Furthermore, the higher the average coal surface temperatures, wind speeds and train speeds, the more frequent and intense the dusting events became (Fig. 3). While riding behind the coal trains in the research caboose, it was clear that dusting increased when coal cars passed through tunnels, over trestles, and

close to topographic interfaces. RTEPS data also showed that emissions were most frequent during accelerations between fifteen and thirty miles per hour. The most frequent and intense emissions occurred when the study trains passed other trains moving in the opposite direction at track speeds.

Load-top Volume Changes

The original photographic method for estimating volume changes produced material loss estimates of 0.11 to 0.76 tons, with an average of 0.31 tons ($n = 31$). For these same cars, scale weight losses averaged 0.36 tons, thus providing some credence to the claim that the photographic method underestimates material losses. An example of "before" and "after" transects are shown in Figure 4. The photographic method also laid the foundation for an automated volume-change detection system such as CCLPS. As CCLPS becomes further developed, we hope to obtain more and more reliable results from our volume/mass-loss calculations.

Trip Stress Index (TSI)

In order to compare the stresses from trip to trip, an index was devised from information collected with RTEPS. Air temperature, coal surface temperature, and wind speed were combined to arrive at a Trip Stress Index (TSI), allowing direct comparison of the stresses from each trip. A relationship between passive collection and TSI was revealed through data analyses and is discussed below.

Passive Collection

Over the course of the thirty field trials, a total of 360 passive collector samples have been taken. The combination of profile modification and chemical sprays has resulted in a 95 to 99% reduction in coal losses compared to normal trapezoidal load profiles according to passive collection data. Statistical analyses of passive collection show that treated cars can be distinguished from untreated cars with a 99.9% confidence level. Table 1 depicts the average passive collection over all trips for untreated versus treated cars. The 153 passive collector samples not shown were either collected during "experimental" treatments, or there was no direct comparison available for treated versus untreated cars for a given experiment.

There appears to be no useful correlation between scale weight changes and passive collection on a car-by-car basis, likely due to the inherent scale inaccuracies and moisture content variations. This is another reason not to rely on the scale weight changes alone for material loss estimates, but instead, to apply independent loss estimates techniques. However, a clear relationship between passive collection and TSI is revealed in Figure 5. This relationship appears to be exponential. On the other hand, the data suggest that there is some threshold above which passive collection (i.e., fugitive emissions) significantly increases.

Surface Treatment Evaluations

As previously mentioned in the "Methodology, Surface treatments" section, a variety of surface treatments were tested during the study for their dust suppression capabilities. Using untreated cars as the reference for judging the success of treatments, results from RTEPS show that water-only treatments, whether sprayed on at the mines or en route, suppressed fugitive emissions for a maximum of only two to three hours under stressful conditions during a thirty-six to seventy-two hour trip. In fact, untreated surfaces actually emitted less dust than water-only treated cars under certain conditions (e.g. freezing temperatures). This was the case for both groomed and ungroomed cars. Grooming alone reduced passive collection and scale weight losses from an average of 0.36 tons to 0.20 tons during the most stressful trips. When profile grooming was combined with chemical treatments, even greater reduction in fugitive emissions was realized, up to 95% over untreated cars.

CONCLUSIONS

A total of thirty field trials have been conducted to date for the NSRES.

Analyses and stratification of a 360,000-car database yielded a standard deviation of about 6 tons in dump weights, masking any meaningful signal for weight losses for the NSRES.

Material losses based on scale weight changes for ungroomed, untreated cars averaged about 0.36 tons/car under high stress trip conditions.

Material losses based on scale weight changes for groomed, untreated averaged about 0.20 tons/car in the high stress trip conditions.

Intensity and frequency of emissions are greatest when the train is accelerating between 15 and 30 miles per hour, and when passing on-coming trains.

Increased fugitive emission events are associated with tunnels, trestles, and topographic interfaces.

The relationship between the Trip Stress Index and passive collection indicated that there is a stress threshold above which fugitive emissions significantly increase.

Based on passive collection, material losses from groomed, treated cars were reduced by up to 95% over untreated and ungroomed cars.

ACKNOWLEDGMENTS

This work was primarily funded by the Norfolk Southern Corporation, with additional assistance from CONSOL.

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Table 1. Passive Collection for Untreated Versus Treated Cars

UNTREATED CARS AVERAGE (g)	TREATED CARS AVERAGE (g)
n = 113	n = 94
131	5

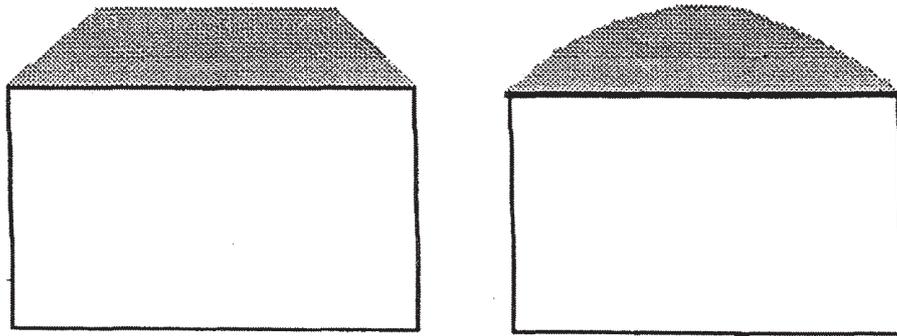


Figure 1a (left) and 1b (right). Cross Sections of Coal Hoppers with Trapezoidal Profiles (1a) and Rounded Profiles (1b)

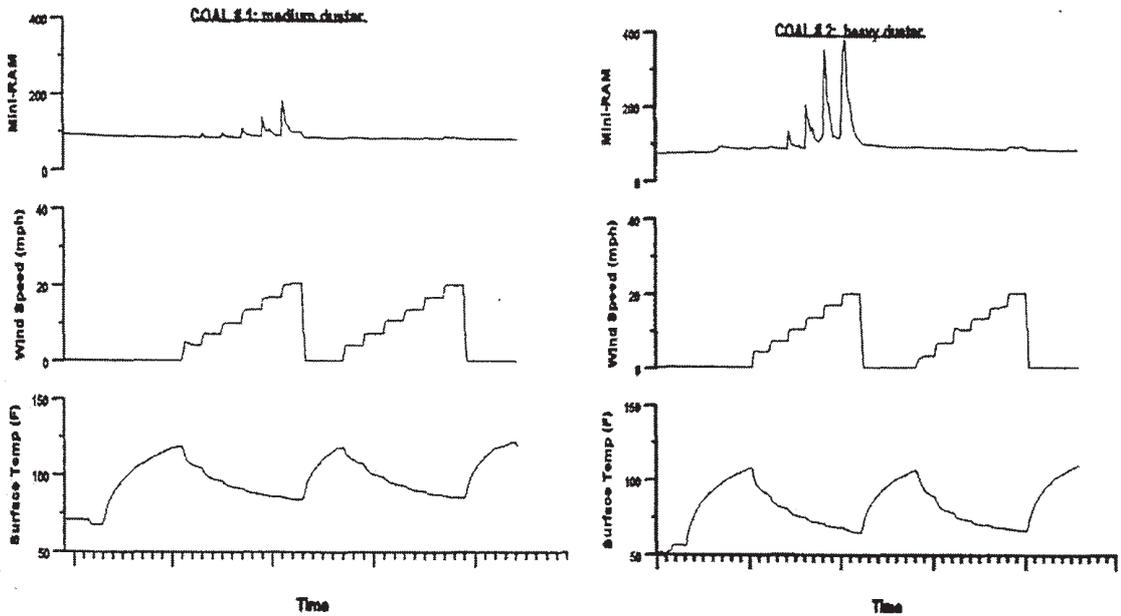


Figure 2a (left) and 2b (right). Graphical Difference Between "Medium" and "Heavy" Dusty Coals According to SARTDX Procedures

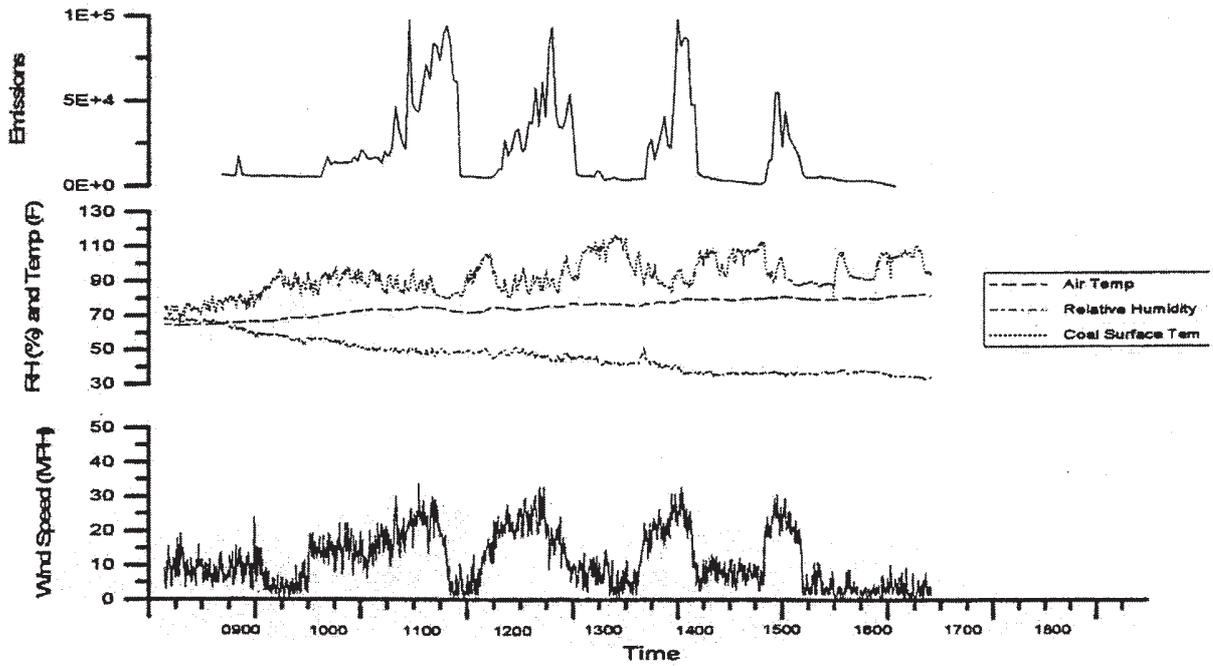


Figure 3. A Typical RTEPS Trip Profile Showing the Correlation Among Emissions, Coal Surface Temperature, and Wind Speed

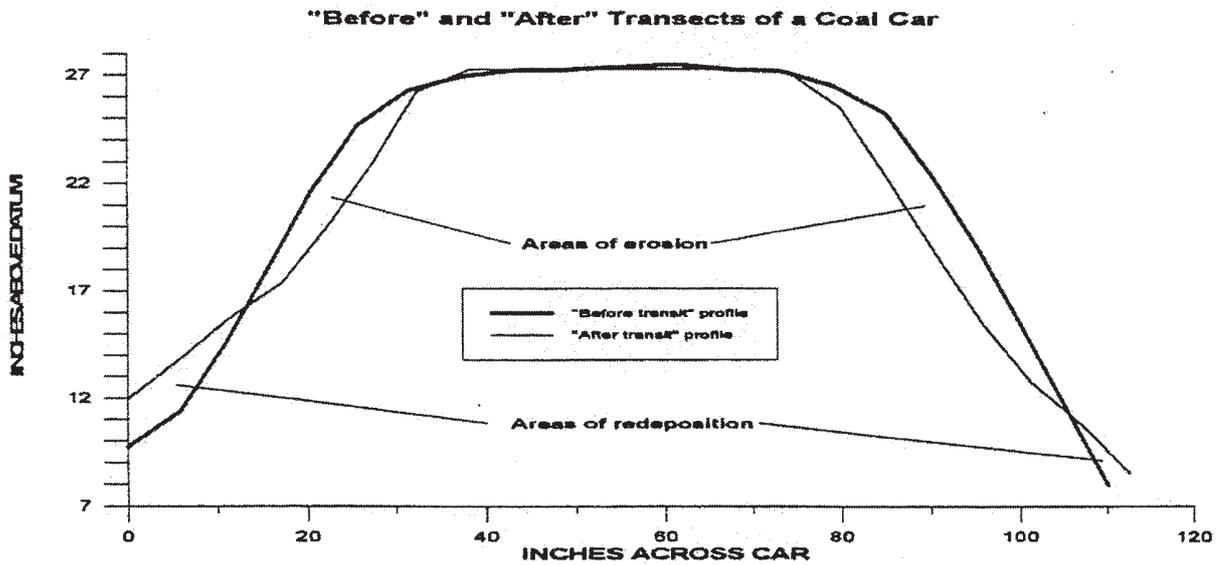


Figure 4. An Example of a Photographic Transect Across an Untreated Car Showing Areas of Erosion and Deposition

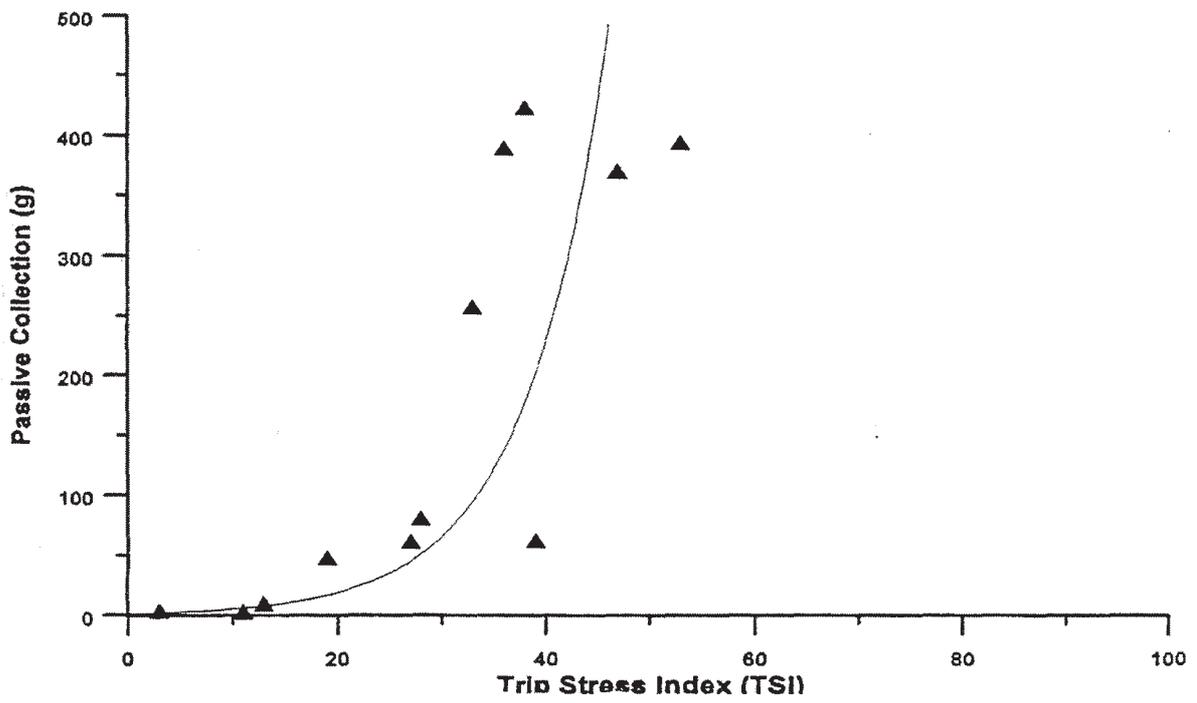


Figure 5. Trip Stress Index Versus Passive Collection

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August 5, 2010

VIA ELECTRONIC FILING

Ms. Cynthia Brown
Chief, Section of Administration
Office of Proceedings
Surface Transportation Board
395 E Street, SW
Washington, DC 20423-0001

Re: *Petition of Arkansas Electric Cooperative Corporation for a Declaratory Order,*
STB Finance Docket 35305

Dear Ms. Brown:

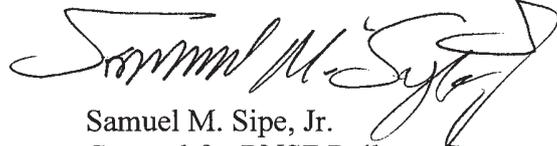
This letter responds to requests for BNSF Railway Company to provide additional information on two matters raised by Board members at the July 29, 2010 hearing in the above-referenced case.

First, in response to a request from Vice Chairman Mulvey, attached is a list of academic and industry articles and reports related to coal dust that we have been able to locate. We have identified whether the materials on our list were already included in the record in this proceeding, are readily available on the internet, or are being provided to the Board as attachments to this letter. We have seen references to a few other articles and reports, but have not included them on our list if we have been unable to locate a copy.

Second, Commissioner Nottingham asked BNSF to advise the Board whether BNSF loaded the railcars above the sill during its removal of coal dust that had accumulated near certain waterways in 2008. We confirmed that BNSF did not load the railcars above the sill. We note that one of the articles on the attached list addresses the question raised by Commissioner Nottingham as to whether loading coal below the sill would reduce coal dust emissions. This article indicates that this loading technique reduces the load capacity of each railcar by more than ten percent without a statistically significant reduction in coal dust emissions. *See* Edward M. Calvin, G. D. Emmitt & Jerome E. Williams, *A Rail Emission Study: Fugitive Coal Dust Assessment and Mitigation*, *Proceedings for the Seventh Annual Environment Virginia '96 Symposium*, 44, 48, Lexington, Virginia (April 11-12, 1996).

Please contact the undersigned if you have any questions regarding this letter.

Sincerely,



Samuel M. Sipe, Jr.
Counsel for BNSF Railway Company

cc: Chairman Daniel R. Elliott III
Vice Chairman Francis P. Mulvey
Commissioner Charles D. Nottingham
Parties of Record

Coal Dust Articles and Reports

Aurecon Hatch, Coal Leakage from Kwik-Drop Doors: Coal Loss Management Project Queensland Rail Limited Reference H327578-N00-EE00.08 Revision 1 (July 21, 2009) (available at <http://www.qrnetwork.com.au/media-and-community-centre/environmental-policies/Coal-loss-management.aspx>).

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BIOLOGICAL EFFECTS OF UNBURNT COAL IN THE MARINE ENVIRONMENT

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Abstract Unburnt coal is a widespread and sometimes very abundant contaminant in marine environments. It derives from natural weathering of coal strata and from anthropogenic sources including the processing of mined coal, disposal of mining wastes, erosion of stockpiles by wind and water, and spillage at loading and unloading facilities in ports. Coal is a heterogeneous material and varies widely in texture and content of water, carbon, organic compounds and mineral impurities. Among its constituents are such potential toxicants as polycyclic aromatic hydrocarbons (PAHs) and trace metals/metalloids. When present in marine environments in sufficient quantities, coal will have physical effects on organisms similar to those of other suspended or deposited sediments. These include abrasion, smothering, alteration of sediment texture and stability, reduced availability of light, and clogging of respiratory and feeding organs. Such effects are relatively well documented. Toxic effects of contaminants in coal are much less evident, highly dependent on coal composition, and in many situations their bioavailability appears to be low. Nevertheless, the presence of contaminants at high concentrations in some coal leachates and the demonstration of biological uptake of coal-derived contaminants in a small number of studies suggest that this may not always be the case, a situation that might be expected from coal's heterogeneous chemical composition. There are surprisingly few studies in the marine environment focusing on toxic effects of contaminants of coal at the organism, population or assemblage levels, but the limited evidence indicating bioavailability under certain circumstances suggests that more detailed studies would be justified.

Introduction

Coal is one of the oldest and most widespread anthropogenic contaminants in marine and estuarine environments. This review addresses the question of whether unburnt coal represents an environmental risk. The review arose from a request to assess the potential ecological effects associated with proposed storage and shipping of coal from an existing port. Coal is a heterogeneous material and different forms vary in their physical and chemical properties. In the course of this study, it was found that there was considerable information on the chemical composition and physical properties of coal, as might be expected for a major industrial feedstock. While some common components of coal, such as polycyclic aromatic hydrocarbons (PAHs) and trace metals might become environmental contaminants and have the potential to cause adverse biological effects at sufficiently high concentrations, it was surprising that there was relatively little information on the bioavailability of contaminants from coal, or on biological effects at the levels of organisms,

populations or assemblages directly related to coal, either in the laboratory or field. This lack of information on the ecological effects of unburnt coal was unexpected in view of the common occurrence of coal in the marine environment and the continuing importance of coal as a source of heat and as an industrial feedstock.

Coal has been traded by sea at least since Roman times. Its co-occurrence with iron ore in the English Midlands was one of the factors that made possible the large-scale production of iron and laid the foundations for the industrial revolution of the late 18th–19th centuries. From then until the 1960s coal was the world's single most important source of primary power. In the late 1960s this role was taken by oil, but the imbalance is likely to swing back again because of the relative sizes of remaining reserves of coal and oil (equivalent to 200 yr and 40 yr for coal and oil, respectively, at current rates of production; World Coal Institute 2004). The current global political climate is also encouraging oil-importing countries to reduce their reliance on oil and become more self-sufficient in energy production and reserves of coal are much more widespread geographically than those of oil.

Global coal production and consumption

In 2002, total global production of hard coal (bituminous and anthracite — the different types of coal are described below) was 3837 million tonnes (Mt) and that of brown coal/lignite 877 Mt (these and other economics data were taken from the Web sites of the World Coal Institute 2004, Coalportal 2004 and Australian Coal Association 2004). In contrast to oil-exporting countries, major producers of hard coal have a wide geographical distribution, as shown in Table 1, although the quality (rank) of coal varies greatly. Production of brown coal is dominated by Germany, Greece and North Korea but, because of its lower economic value and relatively high water content, it is usually consumed close to the point at which it is mined. Transport of coal by sea (including international trade) is dominated by hard coals, and bituminous types in particular. The latter are

Table 1 Production and export of hard coal in 2002 by country

Country	Total production, Mt (%)	Exports, Mt	
		Thermal	Coking
PR China	1326.0 (34.6)	72.0	13.8
U.S.	916.7 (23.9)	16.2	18.3
India	333.7 (8.7)	0	0
Australia	276.0 (7.2)	91.3	107.5
South Africa	223.0 (5.8)	67.7	0.9
Russia	163.6 (4.3)	36.1	9.0
Poland	102.6 (2.7)	19.1	3.5
Indonesia	101.2 (2.6)	65.6	7.4
Ukraine	82.9 (2.2)	n.d.	n.d.
Kazakhstan	70.6 (1.8)	n.d.	n.d.
Canada	67.9 (1.8)	3.4	23.4
Colombia	39.4 (1.0)	34.4	0
Total	3837	435.0	195.4

Data from World Coal Institute 2004 and Coalportal 2004, values for exports are estimates. Percentages based on total world production.

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used for electricity generation ('thermal' coal) and for industrial processes, particularly the manufacture of steel ('coking' coal). Anthracite is the least abundant of the world's coal stocks and consequently represents only a very small part of world trade in coal, despite its high energetic and economic value.

Just over 60% of current coal consumption is used to produce heat and power, including about 39% of global electricity generation. A further 16% is used in the steel industry, in blast furnaces fuelled by coal and coke. Domestic uses and non-metallurgical industries (including the manufacture of cement) each represent about 5% of total consumption. Many of the world's largest economies rely on coal to generate 50% or more of their electricity, including the United States (50%), Germany (52%) and, significantly, the emerging economies of China (76%) and India (78%). Between 1995 and 2020, world energy demand is predicted to rise by 65% and fossil fuels are expected to meet 95% of this increase. Much of the coal used in power generation, however, is of low rank (lignite and sub-bituminous types) and its relatively low economic value and high water content make it unattractive for international trade. Consequently, more than 60% of the coal used for electricity generation globally is consumed within 50 km of its source.

Roughly 14% (630.4 Mt) of world production of hard coal is currently traded internationally, 69% for power generation and 31% for metallurgical use. This compares with a trade of 427.4 Mt in 1994, an increase of 47.5% over the last decade. Australia is the world's largest exporter of hard coal with 21% (91.3 Mt) of thermal and 55% (107.5 Mt) of coking coal sent to more than 35 countries, principally Japan (90.2 Mt) and other Asian countries (Republic of Korea 25.3 Mt, Taiwan 17.2 Mt, other Asian nations 11.0 Mt), but also Europe (31.8 Mt), India (13.6 Mt), north Africa, the Middle East and South America. Other important exporters of thermal coal are China, Indonesia, South Africa, Russia, Colombia, Poland and the U.S., whereas Canada, in addition to these countries, also exports coking coal. Exports of both categories are expected to rise in the near future, with Australia increasing exports of coking coal and all exporting countries increasing their exports of thermal coal. The major coal importing countries are Japan (estimated 91.8 Mt in 2002), the Republic of Korea (44.4 Mt), Taiwan (42.6 Mt), Germany (31.6 Mt), the United Kingdom (22.5 Mt) and other European Union states (153.8 Mt).

These figures illustrate several relevant points. First, the amount of coal traded by sea is huge, even in an era that we commonly think of as being dominated, in terms of energy production, by oil and gas. Second, the exporters and importers of hard coal are often separated by large distances, for example Australia and Europe. Third, the centres of production and consumption of coal, and the shipping routes connecting them, continue to shift as the centres of industrial production and power consumption change. In particular, the rising industrial outputs of China and India are likely to bring continuing changes to global trade in coal. China's exports and imports of hard coal have tripled over the last decade while India's imports have more than doubled.

Coal forms the backbone of heavy industry and electricity generation in many countries. To ensure an uninterrupted supply, utilities and industrial facilities that need to run continuously often stockpile coal for 30–90 days of consumption (Davis & Boegly 1981a). For example, it is estimated that approximately 100 Mt of coal are stockpiled in the U.S. alone (data for 1997 cited by Cook & Fritz 2002). For logistical reasons, coal stockpiles are commonly located close to waterways and therefore represent a major source of coal particulates and leachates to the aquatic environment.

The need for information on ecological effects of coal in the marine environment

The need to assess the effects of unburnt coal in the marine environment may arise from new sources of contamination or from remobilisation of coal already present and incorporated into sediments. Development of new coal mines and associated coal washing facilities (or the continued operation of existing mines) near the coast brings the possibility of environmental contamination,

and requires assessments of environmental risk. Coal storage and loading facilities at ports are also potential sites of contamination, often on a very large scale. For example, the world's largest export coal handling facility at the Port of Newcastle, New South Wales, Australia, has storage area for 3.5 Mt of coal (Australian Coal Association 2004). Coal travels from trains or storage areas to ships via conveyors and in very large volumes. The Port of Hay Point in Queensland, Australia, for example, can load in excess of 20,000 t h⁻¹ (R. Brunner, Ports Corporation of Queensland, personal communication). During the transfer, storage and loading operations there is potential for loss of coal to the surrounding environment through spillage and wind and water erosion. Many coal-handling ports operate best-management practices to reduce these fugitive losses, but an assessment of the appropriate level of reduction requires an understanding of the mechanisms of coal's environmental effects. Measures that are adequate to prevent unacceptable reduction in water clarity, for example, might not be considered adequate if exposure to coal had a demonstrably adverse toxic effect on aquatic organisms.

In the past, control of contamination by particulate coal around mines and ports was less strict than it is today and sediments in these areas are likely to contain a substantial legacy of historical coal contamination. Capital dredging may remove these sediments, resuspending some of the coal into the water column and transferring the remainder to spoil disposal areas (Birch et al. 1997). Changes to patterns of water movement, for example following deepening of navigation channels to accommodate vessels of larger draught or infilling of intertidal areas for port or other developments, could also lead to erosion and remobilisation of coal-bearing sediments (French 1998). Again, assessment of the associated environmental risks requires understanding of the mechanisms of effect.

Scope of the review

The present review focuses on the ecological effects of unburnt coal in the marine environment. The effects of products of combustion of coal, such as fly ash, which have been reviewed elsewhere (e.g., Duedall et al. 1985a,b, Swaine & Goodarzi 1995), and the by-products of coking and coal gasification are not considered. Also excluded are effects of materials that may be added to coal to improve its handling characteristics during transport and storage, such as glycol or chlorinated water used to create slurries for transfer by pipeline, and potential hazards from 'synfuels' (combinations of coal with oil emulsions, used for power generation, coking and steel manufacture in some countries). Effects of spoil from coal mines, including acid mine drainage, are also outside the present scope because, again, they have been extensively reviewed elsewhere (e.g., Evangelou 1995, Geller et al. 2002) and because their effects derive not just from the presence of coal but also (perhaps mainly) from associated rocks and minerals (although coal-pile leachates may be generally similar in quality to acid mine drainage: Davis & Boegly 1981a). While the focus of this review is the marine environment, information on the quality and ecotoxicology of stockpile leachates is also considered. Although leachates are generally derived from freshwater (rainfall or water sprayed to suppress dust) they provide a potential conduit for coal-related contaminants to enter the marine environment. Also included are studies of physical effects of coal on freshwater organisms, since the mechanisms of effect are likely to be the same in saline waters. The Discussion attempts to evaluate and synthesise the information from the perspective of environmental risk assessment and mitigation. Although this approach may deviate from a typical scientific review, a 'risk assessment' format may be useful for those faced with assessing and managing effects of coal in the marine environment.

The review begins with an overview of coal types, because there are differences among them in their potential ecotoxicological effects. Sources and the distribution of coal in the marine environment are then discussed. Consideration of effects of coal on marine organisms begins with

physical effects such as smothering and abrasion. Next, chemical information on coal is reviewed in relation to its role as a potential source of contaminants to the marine environment. Following this, the rather limited range of information on effects of coal-derived contaminants at the biological levels of the cell, organism, population and assemblage is described. This description focuses specifically on coal-derived contaminants, rather than reviewing the general literature on effects of the contaminants concerned. The Discussion addresses the question of whether unburnt coal presents a problem in the marine environment, identifying scenarios (such as chemical environmental conditions and type of coal) for which it is or is not likely to pose an ecological hazard, and others for which we have insufficient knowledge to make an assessment. Finally, management options for mitigating potential environmental effects and directions for future research are briefly considered.

Where possible, published, widely accessible sources of information have been used and the 'grey literature' avoided. However, the paucity of information on many aspects of coal's environmental effects has made some reference to grey literature unavoidable. For up-to-date background information on current production and trade in coal, reference is made to relevant sources on the Internet, many of which are provided by bodies representing the coal industry.

Types of coal

Variation in age and conditions of formation gives rise to a range of types of coal, classified into four broad categories ('ranks'). These vary in their chemical composition (and, therefore, their potential for biological effects), their energy content and, ultimately, their use. Alternative systems of coal classification are summarised by Ward 1984. Lignite ('brown coal') is the least mature rank and contains relatively little carbon and energy, and a relatively large proportion of water and volatile matter. It represents about 20% of world reserves of coal and is mainly used for power generation. The second type of low-rank coal, sub-bituminous, has a higher carbon content (71–77%), lower water content (10–20%) and is used for power generation, production of cement, and various industrial processes. It ranges in appearance from dull and dark brown to shiny and black, and in texture from soft and crumbly to hard and strong. It represents about 28% of world coal reserves. Of the 'hard' coals, the less organically mature form, bituminous coal, is used for power generation ('thermal' or 'steam' coal) and manufacture of iron and steel ('coking' coal). It represents 51% of world coal reserves. Bituminous coal varies in content of volatile matter, whereas the most organically mature and highest ranked coal, anthracite, always contains less than 10% volatile matter and is capable of burning without smoke. It is hard, has a high carbon content (ca 90%) and has various domestic and industrial uses. Although it is the most valuable form of coal, it constitutes only 1% of world coal reserves.

Other important determinants of coal quality, and its corresponding utility, relate to its mineral content. For example, sulphur, chlorine and phosphorus occur in substantial amounts in some coals and have the potential to generate corrosive acids upon oxidation or heating. Other coals may have high contents of metals and metalloids. These chemical properties not only affect the behaviour of a specific type of coal in its intended use, but also significantly determine its behaviour in the environment.

Sources and distribution of particulate coal in the marine environment

Coal enters the marine environment through a variety of mechanisms (Figure 1), including natural erosion of coal-bearing strata (Shaw & Wiggs 1980, Barrick et al. 1984, Barrick & Prah 1987). Papers by Short et al. (1999), Boehm et al. (2001), Mudge (2002) and Van Kooten et al. (2002) feature a debate about the source of background hydrocarbon contamination in the Gulf of Alaska,

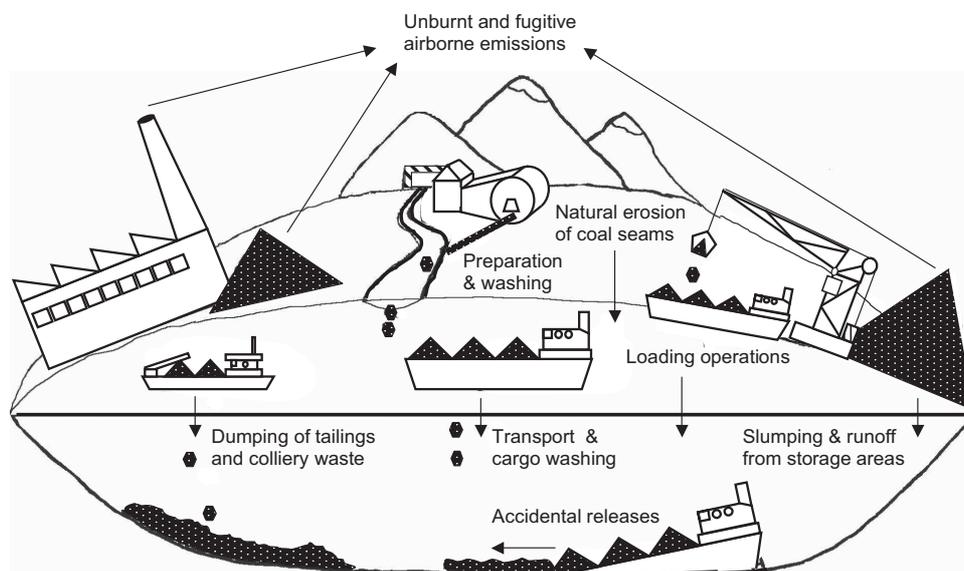


Figure 1 Sources of coal to the marine environment.

with one group suggesting oil-based sources and the other suggesting coal. Mudge (2002) concluded from a multivariate statistical assessment of the relative contributions of coal, oil seeps, shales and rivers that the hydrocarbons probably derived from a mixture of sources, whose contributions varied across the sampling area.

Anthropogenic inputs of coal occur at several stages of the coal utilisation sequence (Figure 1). These include: disposal of colliery waste into intertidal or offshore areas (Eagle et al. 1979, Santschi et al. 1984, Norton 1985, Limpenny et al. 1992, McManus 1998); wind and water erosion of coastal stockpiles (Sydor & Stortz 1980, Zhang et al. 1995); coal-washing operations (Pautzke 1937, Williams & Harcup 1974); spillage from loading facilities (Sydor & Stortz 1980, Biggs et al. 1984); 'cargo washing' (the cleaning of ships' holds and decks after offloading dry bulk cargoes by washing with water and discharging over the side; Reid & Meadows 1999); and the sinking of coal-powered and coal-transporting vessels (French 1993a, Chapman et al. 1996a, Ferrini & Flood 2001).

As a result of these various inputs, unburnt coal occurs very commonly in marine sediments (Goldberg et al. 1977, 1978, Griffin & Goldberg 1979, Tripp et al. 1981) and may represent a considerable proportion of the sediment. The abundance of coal in the marine environment is likely to be greatest adjacent to storage and loading facilities in coal producing and importing countries, around spoil grounds receiving colliery waste, along shipping lanes and in areas receiving terrestrial runoff from catchments where coal mining occurs (French 1993b, Allen 1987). In sediments off the northeast coast of England, for example, subject to inputs of coal from natural weathering and dumping of colliery waste, coal represented up to 27% of combustible matter by dry weight (Hyslop et al. 1997).

Coal can be a common contaminant even away from such point sources and over larger spatial scales. Goldberg et al. (1977) found that coal, coke and charcoal together represented up to 1.9% by dry weight of the surficial sediments in Narragansett Bay, Rhode Island, U.S., mostly consisting of particles $<38 \mu\text{m}$. The bay has a large human population and intense industrial activity. The angular shape of the coal particles suggested that they had been introduced directly to the coastal environment, rather than being transported via rivers. Goldberg et al. suggested that this material was derived from coal-burning ships or coal burning on adjacent land. Similar results were found

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for Chesapeake Bay (Goldberg et al. 1978), where likely sources included coal mining in the adjacent catchment of the Susquehanna River, in addition to coal-burning ships and domestic and industrial uses on the adjacent land. Goldberg et al. (1978) noted that the local power company formerly dredged the lower parts of the river, separated the coal, and used it for power generation. This practice stopped once dams were constructed on the lower river, trapping coal and other sediments until they were periodically resuspended and flushed out of the dams by storms. An extreme example of the wide geographical extent of human inputs of coal to the marine environment is provided by the presence of coal clinker at depths of 3000–5000 m in the Venezuelan Basin in the Atlantic (Briggs et al. 1996).

Geographical patterns of input and distribution of coal in the marine environment vary at a range of scales. Long-term, historical changes include shifts in centres of coal production, such as the rise and fall of British coal output in the 19th and 20th centuries and the more recent rise in coal production in China. Accompanying these are changes in trading routes for coal. For example, in 1982, the seaborne trade in thermal coal was 61 Mt in the Atlantic and 25 Mt in the Pacific, with respective values for coking coal of 48 and 73 Mt. By 2002, following average annual growth in seaborne trade of 8% for thermal coal and 1.8% for coking coal, the equivalent figures were 170 and 233 Mt for thermal coal and 60 and 114 Mt for coking coal (World Coal Institute 2004). In addition to these long-term changes in sources of coal contamination, concentrations at a particular location may also show short-term changes related to factors such as seasonal variation in fluvial discharge (French 1993c).

In addition to direct inputs to estuarine and coastal environments, coal may be transported from its source (natural or anthropogenic) by rivers. This process may introduce a variable lag between production and contamination, depending on the distance involved and the storage capacity of the river system (Allen 1987). Man-made alterations to patterns of river flow, such as dams, may increase this lag. Hainly et al. (1995) estimated that the three dams on the Susquehanna River, which flows into the Chesapeake Bay, had accumulated about 250 Mt of sediment, of which about 20 Mt (8%) was coal. Remobilisation of this stored coal (and other contaminants), for example when the dams reach the end of their functional lives, may provide a significant source of contamination to downstream environments. Similarly, contaminated estuarine or coastal sediments may act as a source of future contamination through remobilisation. French (1993a, 1998) suggested that new inputs of coal to the Severn Estuary (United Kingdom) must derive from erosion of contaminated sediments already in the estuary because production of coal in the nearby Welsh coalfields had effectively stopped by the time of his study. Estimates indicated that mudflats and saltmarshes in the estuary contained 10^5 – 10^6 t of coal (Allen 1987, French 1998). As rising sea levels bring about increased rates of erosion of intertidal flats and saltmarshes in many parts of the world (e.g., Adam 2002, Scavia et al. 2002), remobilisation of historic contaminants, including coal, may become increasingly important.

Because coal generally has a lower specific gravity than many other components of sediments (the specific gravity of coal varies with its ash content, ranging from 1.2–2.9 g cm⁻³; Alpern 1977, compared with 2.65 g cm⁻³ for quartz; Brady & Weill 2002), transport by water movement may result in larger particles of coal being transported and deposited with smaller, denser particles of sands and gravels. Settling times and, therefore, transport distances will also be greater for a given particle size. In intertidal sediments of the Severn Estuary, coal particles (silt to sand size) were most abundant in the finest-textured sediments (Allen 1987).

Physical effects of coal on marine organisms

Most of the mechanisms whereby particulate coal may exert a physical effect on organisms are shared with other types of suspended and deposited sediments and have been reviewed elsewhere.

For example, Moore (1977) reviewed the effects of particulate, inorganic suspensions on marine animals and Airoidi (2003) reviewed the effects of sedimentation on biological assemblages of rocky shores. These effects are therefore only dealt with briefly here. Moore (1977) made the distinction, from the perspective of biological effects, between scouring by larger particles, such as sands, and the turbidity-creating effects of smaller particles, such as silts and clays. Many animals and plants living on rocky shores trap sediments and, thereby, influence rates of sediment transport, deposition and accretion (Airoidi 2003), and this is equally true for animals living in soft sediment habitats (e.g., Wolanski 1995, Young & Harvey 1996, Esselink et al. 1998, Norkko et al. 2001). The reviews by Moore (1977) and Airoidi (2003) show that, conversely, sediments affect the abundance and composition of marine organisms and assemblages when in suspension and following deposition. Effects may be lethal or sublethal and may act directly (e.g., by abrasion, scour or smothering) or indirectly (e.g., by alteration of the nature of the substratum or by modification of processes of predation or competition). Airoidi (2003), however, pointed out that the mechanisms by which sedimentation affects marine organisms are often poorly understood and that different aspects of sediment transport, such as burial, scour and turbidity, are often confounded and not explicitly differentiated in studies.

Direct effects

Increased concentrations of suspended particulate coal in the water column may cause abrasion of animals and plants living on the surface of the sea bed or on structures such as rocks or wharf piles (Emerson & Zedler 1978, Kendrick 1991, Hyslop et al. 1997 and see references in Moore 1977 and Airoidi 2003). The probability and severity of this effect will depend on the concentration, size and angularity of the coal particles and on the strength of water currents (Newcombe & MacDonald 1991, Lake & Hinch 1999). Newcombe & MacDonald (1991) pointed out that the particle dose to which an organism is exposed (a function of the concentration of suspended material and the duration of exposure) is a more relevant measure of stress than concentration alone but that duration is often not reported in studies of the effects of suspended sediments. Mean suspended solids concentrations of 1000–3000 mg l⁻¹ have been recorded in coal pile runoff in Canada, exceeding Canadian water quality criteria (10 mg l⁻¹) by three orders of magnitude (Table 4, p. 84: Fendinger et al. 1989, Curran et al. 2000). Because of its generally lower specific gravity, larger particles of coal will be transported further by a given current speed than particles of quartz sand, potentially producing greater abrasion. Hyslop & Davies (1998) tested the hypothesis that reduction in occurrence and biomass of the green alga *Ulva lactuca* on shores receiving inputs of colliery waste was due to physical scouring. Laboratory tests compared the effects of three size categories of waste (<0.5 mm, 0.5–2.0 mm and >2.0 mm) under still and turbulent conditions. Over 8 days, plants gained weight when no colliery waste was present but lost weight in the presence of waste. Maximal weight loss occurred in the presence of waste of grain size 0.5–2.0 mm (vs. <0.5 mm and 0–2.0 mm) under turbulent conditions, suggesting that the coarse sediment acted as an abrasive and may have been responsible for the removal of components of the ephemeral algal flora of shores receiving colliery waste in northeast England. In contrast to the effect on macroalgae, the distribution of animals on the same shores was not affected by the presence of the waste (Hyslop et al. 1997).

Particles of coal in suspension will also reduce the amount and possibly the spectral quality (Davies-Colley & Smith 2001) of light that reaches the sea bed or other underwater surfaces, in a manner similar to other suspended particles (Moore 1977). This, in turn, may affect growth of plants such as seaweeds, seagrasses, and microalgae on the surfaces of sediments and rocks (e.g., Duarte 1991, Preen et al. 1995, Vermaat et al. 1996, Terrados et al. 1998, Longstaff & Dennison 1999, Moore et al. 1997). Again, the magnitude of this effect will depend on the amount and size

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of coal particles in suspension (which will, in turn, depend on rate of supply and patterns of water movement), duration of exposure and existing water clarity (Newcombe & MacDonald 1991).

Deposition of coal dust on the surface of plants above and below water may also reduce photosynthetic performance. Mangroves growing around South Africa's largest coal-exporting port, Richards Bay, accumulate deposits of coal dust on both upper and lower leaf surfaces and on branches and trunks (Naidoo & Chirkoot 2004). The presence of the dust reduced photosynthesis, measured as carbon dioxide exchange and chlorophyll fluorescence, by 17–39%. There was no evidence that coal particles were toxic to the leaves, but mangroves closest to the source of the dust appeared to be in poorer health than those further away. The amount of dust accumulated on leaves varied among mangrove species, with *Avicennia marina*, which has relatively hairy leaves, accumulating more than *Bruguiera gymnorhiza* or *Rhizophora mucronata*.

Suspended particles in general may clog feeding and respiratory organs of a wide range of marine animals, reducing efficiency of feeding and respiration and possibly damaging the organs (see reviews by Newcombe & MacDonald 1991, Newcombe & Jensen 1996, Wilber & Clarke 2001), or, as in the case of some bivalve molluscs, cause reduction in the rate or efficiency of feeding or cause it to cease altogether (see discussions in Moore 1977, Bayne & Hawkins 1992). Moore (1977) provided a taxonomic review of information on the effects of suspended sediments on animals. Groups generally intolerant of higher levels of suspended sediments include sponges, some scleractinian corals, serpulid polychaetes, bivalve molluscs and ascidians, but there is considerable variation in tolerance within each group. It is reasonable to assume that coal will have similar effects across the same range of taxa, but this has not been determined.

In a study of the effects of coal on ventilation and oxygen consumption in the Dungeness crab (*Cancer magister*), there was no measurable effect over an exposure period of 21 days relative to crabs living in clean water (Hillaby 1981). In this experiment, however, the coal was mixed into the sand in the bottom of the aquaria, and was not kept in suspension, so the response to suspended material may not have been measured. Furthermore, coal-amended sediments were allowed to equilibrate for at least 15 days in flow-through tanks prior to beginning the experiments, which may have allowed fine particles of coal to be flushed out of the sediment. The lack of significant differences in oxygen consumption between treatments was, in part, due to large within-treatment variability because variance within treatments increased with exposure duration and proportion of coal to sand. A previous study (Pearce & McBride 1977, cited by Hillaby 1981), in which some coal remained in suspension throughout the duration of the experiment, reported that particles of coal progressively accumulated in the crabs' gills. This accumulation may have affected respiration and oxygen uptake although these were not measured in the experiment.

In a freshwater study involving an early example of *in situ* toxicity testing, Pautzke (1937) exposed young trout (*Oncorhynchus mykiss gairdneri* (cited as *Salmo gairdneri*) and *Oncorhynchus clarkii* (cited as *Salmo clarkii*) to suspended coal washings (a mixture of crushed coal and associated quartz, slate and other impurities) by confining them in mesh cages in a contaminated stream. He reported 100% mortality among *O. mykiss* after 2.5 h and among *O. clarkii* after 0.5 h. There was no mortality among fish of either species exposed to the same mine water without the washings (neither exposure nor control treatments were replicated). The concentration of suspended material in the contaminated stream was not explicitly stated (a figure of 3 oz gallon⁻¹ [22.5 g l⁻¹] was given but it is unclear whether this was in the test area of the stream or closer to the washing area). The dead fishes showed heavy secretions of mucus from the skin and gills, to which particles of coal adhered. Coal and slate particles were also found in the stomachs. Pautzke also noted a 'haemorrhagic appearance' to the heart and liver. In a later study, suspended particles of coal of 200 mg l⁻¹ were reported to reduce growth rate in *O. mykiss*, but did not kill them (Herbert & Richards 1963, cited in Gerhart et al. 1981).

Suspended sediments can also cause mortality of eggs and larvae of fishes and benthic invertebrates (Auld & Schubel 1978, Wilber & Clarke 2001). Eggs, larvae and, in some species, adult fishes exhibit rapid increases in adverse effects as the duration of exposure to suspended sediments increases, implying the existence of a threshold concentration resulting in adverse effects (Newcombe & Jensen 1996). However, it should be noted that these studies did not specifically assess effects of suspended coal, so extrapolations must be made with caution.

As coal settles out of suspension onto the sea bed, its most direct effect is likely to be smothering of animals and plants. Around wharves where coal is loaded and unloaded the accumulation of spilled coal may be considerable. French (1998) found a horizon of coarse coal debris 10 cm thick in the sedimentary record at Lydney Harbour on the Severn Estuary (United Kingdom), representing spillage from a former coal wharf. Examples of the effects of rapid deposition of sediment on benthic macrofaunal assemblages have been reported for soft sediments (McKnight 1969, Peterson 1985, Fahey & Coker 1992, Smith & Witman 1999) and rocky intertidal areas (Daly & Mathieson 1977, Littler et al. 1983). Several of these studies reported high levels of mortality among the animals affected and many reported that species distributions in affected areas were related to the degree of sediment deposition (reviewed by Airoidi 2003). Circumstantial evidence also indicates that adverse effects of sediments can be due to inhibition of larval settlement and recruitment (Airoidi 2003). In the case of soft-sediment benthos, mortality is likely to be greater when the deposited sediment is different to that naturally present at the site (Maurer et al. 1986). Because of coal's often relatively low density, larger particles of coal may be deposited with smaller, denser particles of sands and gravels (Barnes & Frid 1999) and effects on benthic organisms may be correspondingly large.

Rocky shores affected by sediments are mainly occupied by four groups of organisms with different life-history traits: (1) long-lived, sediment-tolerant species; (2) opportunistic species able to recolonise rapidly following mortality resulting from burial and scour; (3) species that migrate into and out of the affected area as the degree of burial changes; and (4) species that trap sediments and are able to tolerate burial and scour. Characteristics that appear to confer the ability to tolerate burial and scour include the regrowth of upright portions from surviving basal structure; opportunistic cycles of reproduction and growth or vegetative reproductive capability; apical meristems that enable growing parts to remain above the sediment surface; tough, wiry bodies or thalli; erect morphology that reduces the settlement of sediment; physiological characteristics that confer tolerance of darkness, anoxic or hypoxic conditions and high concentrations of sulphides (Airoidi 2003). Similarly, on soft-sediment shores, experimental studies show that some species of seagrass respond to moderate rates and depths of sediment burial by increasing the shoot internodal and leaf-sheath length, rate of development of new leaves and vertical growth (Marbà & Duarte 1994, Duarte et al. 1997). Field studies of seagrass assemblages along gradients of siltation found that species richness and biomass declined rapidly when the silt and clay content of the sediment exceeded a threshold (Terrados et al. 1998, Bach et al. 1998). It may be assumed that deposition of large amounts of fine coal particulates will elicit similar effects.

Indirect effects

Deposition of coal on the sea bed will cause changes in the physical environment, particularly the character of the substratum, and give rise to indirect effects on benthic organisms. These may include infilling of rocky crevices that act as important habitats for benthic organisms such as crabs and lobsters (Shelton 1973) and reduced sediment stability due to the relatively high erodibility of coal particles, making the sediment less suitable for animals to live in. Moore's (1977) taxonomic review of effects of sediment deposition on soft-sediment benthos includes such indirect effects caused by alteration of habitat. Conversely, in naturally homogeneous sediments, such as fine muds,

the presence of coarser particles of coal may increase the heterogeneity of the sediment, allowing a larger range of animals to inhabit it. The previously mentioned coal clinker found at depths of 3000–5000 m in the Venezuelan Basin in the Atlantic, for example, provided a hard substratum for colonisation by an often-abundant, suspension-feeding anemone, *Monactis vestita* (Briggs et al. 1996). Accumulations of coal particles on sandstone ridges off the Mediterranean coast of Israel enhanced the otherwise limited availability of hard substrata (Siboni et al. 2004). Particles were colonised by barnacles, bryozoans and serpulid polychaetes and grazed by a variety of gastropods. A potential extension of this effect is provision of substrata for recruitment and establishment of substratum-specific, non-indigenous species (Carlton 1996), particularly in ports where coal is loaded and unloaded and where international shipping provides a vector for the introduction of exotic species (Carlton 1985).

Indirect physical effects may also be biologically mediated (see discussion by Chapman 2004). Reduction in growth and abundance of plants as a result of reduced water clarity with consequent effects on primary consumers, inhibition of recruitment or removal of adult competitors, predators or grazers, selection of tolerant species and a host of other factors may give rise to a range of indirect physical effects of the presence of suspended and deposited sediment in the marine environment (reviewed by Moore 1977 and Airoldi 2003). Reduced water clarity can also reduce the feeding efficiency of visual predators such as fishes (see Wilber & Clarke 2001 for a recent review). Equivalent effects due the presence of coal are presumably likely, but no examples were found in the literature.

Chemical properties of coal

From a chemical standpoint, coal is a heterogeneous mixture of carbon and organic compounds, with a certain amount of inorganic material in the form of moisture and mineral impurities (Ward 1984). In addition to its predominant elemental building block, carbon, coal contains a multitude of inorganic constituents that may greatly affect its behaviour in, and interactions with, the environment. Unburnt coal can be a significant source of acidity, salinity, trace metals, hydrocarbons, chemical oxygen demand and, potentially, macronutrients to aquatic environments (Tables 2–6), which pose potential hazards to aquatic organisms (Cheam et al. 2000). Trace metals and polycyclic aromatic hydrocarbons (PAHs) are present in amounts and combinations that vary with the type of coal (Tables 2 and 3). For a detailed review of trace metal content of coals, the comprehensive monograph by Swaine (1990) is recommended. A fraction of these compounds may be leached from coal upon contact with water, such as during open storage or after spillage into the aquatic environment (Figure 2). Whether or not these can be leached from the coal matrix and affect aquatic organisms will depend on the type of coal, its mineral impurities and environmental conditions, which together determine how desorbable these potential contaminants are. For example, leaching of metals and acids strongly depends on coal composition, particle size and storage conditions and is accelerated in the presence of oxygen or oxidising agents and if coal remains wet between leaching events (Davis & Boegly 1981a,b, Querol et al. 1996).

Acid-generating potential

One of the most common environmental problems in the handling of many coals is the generation of acid leachates. Rainwater runoff from coal piles can be highly acidic due to the oxidation of pyrite impurities to sulphuric acid, leading to pH values as low as pH 2 (Table 4, Scullion & Edwards 1980, Davis & Boegly 1981b, Fendinger et al. 1989, Carlson & Carlson 1994). The acidity of coal leachates is primarily a function of a coal's sulphur content, such that highly pyritic coals (sulphur content >3%) generally have low pH values of around 2, whereas sulphur-poor coals

Table 2 Inorganic chemical properties of particulate coal; concentrations by dry weight

Origin	U.K.	Germany	Canada	Spain	U.S.	U.S.	U.S.	U.S.	U.S.	U.S.	Western U.S. coals	New South Wales and Queensland	China	SW China	South Island coals*	New Zealand	North Island coals*	ANZECC ISQ-low	ANZECC ISQ-high
Rank	B	L, B	B	SB, B	B	B	B	SB	SB	SB	SB, B	SB, B	A	SB, B	SB, B	SB, B	SB, B		
N %	1-2				0.9-1.4										1.2-1.4				
S %	0.5-4			2.2-9.5	2.0-5.2	1.1	1.1	4.6							0.5-3.0				
Cl %	0.01-1	0.14-0.25	0.003-0.116					1.06							0.06-0.33				
Ag ppm					0.01-0.06	0.02-0.08	0.016	0.01-0.07							0.003-0.11		0.012-0.19	1	3.7
As ppm	1-73	1.5-50	0.2-240	5.3-35.7	1.8-100	1.0-120	1.0-120	0.34-9.8							<1.5-27.5		<2.25	20	70
B ppm	0.5-160	2-236	9-360	141-436	5-120	12-230	12-230	16-140							10-342		47-708		
Ba ppm	<6-500	45-350	10-1000	36-91	72-420	5.0-750	5.0-750	160-1600							8.3-148		21-81		
Cd ppm	0.02-5	0.02-21		0.26-0.33	0.10-0.60	0.1-65	0.1-65	0.10-0.60							<0.3-0.9		<0.7-1.7	1.5	10
Co ppm	0.4-60	7-30	0.2-21	4-12.5	1.5-33	2.0-34	2.0-34	0.6-7.0							<0.1-14.0		0.4-6.8		
Cr ppm	1-45	4-80	2.1-95	11-38	10-90	4.0-60	4.0-60	2.4-20							0.4-20.9		0.6-14.1	80	370
Cu ppm	5-240	10-60	0.2-52	7-16	5.1-30	5-44	5-44	3.1-23							0.95-9.4		1.0-13.6	65	270
F ppm	5-500	20-370	31-890	104-33,265	50-150	29-140	29-140	19-140											
Fe ppm					1780	1780	1780	14,900											
Ga ppm	3-10			5-10	2.9-11	0.8-10	0.8-10	0.8-6.5							0.14-3.0		0.2-5.8		

Table 3 Organic chemical properties of particulate coal; all concentrations in ppm (= $\mu\text{g g}^{-1}$) dry weight

Name	Rank	Total aliphatics	Total aromatics	Total PAHs	ANT	NAP	PHE	BaP	BaA	BbF	CHR	FL	FLT	PYR	Reference
'U.S. coal'	B	670-960	200-2160												1
W Washington coal	L, SB, B, A	0.2-3350 ^a													2
Coal or coke particle	B	3.4-29.9	17.6-62.4	1.43	2.74	4.94	1.27	1.46	1.32	1.53	1.53	1.62	1.65		3
Pennsylvania and Maryland coals	B				0.34-0.65	0.08-0.46				0.25-1.17	0.12-0.36		0.04-0.41		4
Virginia coal				1081.8	2.571	3.268	26.803	1.978	6.281	15.332 ^b	7.218	9.190	7.051		5
ANZECC & NOAA				4	0.085	0.16	0.24	0.43		0.384		0.6	0.665		6, 7
ISQG - Low = ERL				45	1.1	2.1	1.5	1.6	2.8			5.1	2.6		6, 7
ANZECC & NOAA															
ISQG - High = ERM							0.56	0.37	0.34			0.75	0.49		pers.
MOEE LEL															commun.

Abbreviations: Quotation marks = geographical origin not specified; empty cells = not analysed; ^a = total n-alkanes in $\mu\text{g g}^{-1}$ of organic carbon; ^b = sum of chrysene and triphenylene; L = lignite; SB = sub-bituminous; B = bituminous; A = anthracite; ERL = effects range low; ERM = effects range mean; ISQG = interim sediment quality guideline; LEL = lowest effects level; ANT = anthracene, NAP = naphthalene; PHE = phenanthrene; BaP = benzo(a)pyrene; BaA = benzo(a)anthracene; BbF = benzo(b)fluoranthene; CHR = chrysene; FL = fluorene; FLT = fluoranthene; PYR = pyrene; ANZECC = Australian and New Zealand Guidelines for Fresh and Marine Water Quality; NOAA = NOAA Screening Quick Reference Table for Organics; MOEE = Ministry of the Environment and Energy guideline (Ontario, Canada) for particulate bound PAH, as personal communication Kim Irvine (16 June 2004).

References: 1 Tripp et al. 1981, 2 Barrick et al. 1984, 3 Chapman et al. 1996a, 4 Fendinger et al. 1989, 5 Bender et al. 1987, 6 ANZECC 2000, 7 Buchman 1999

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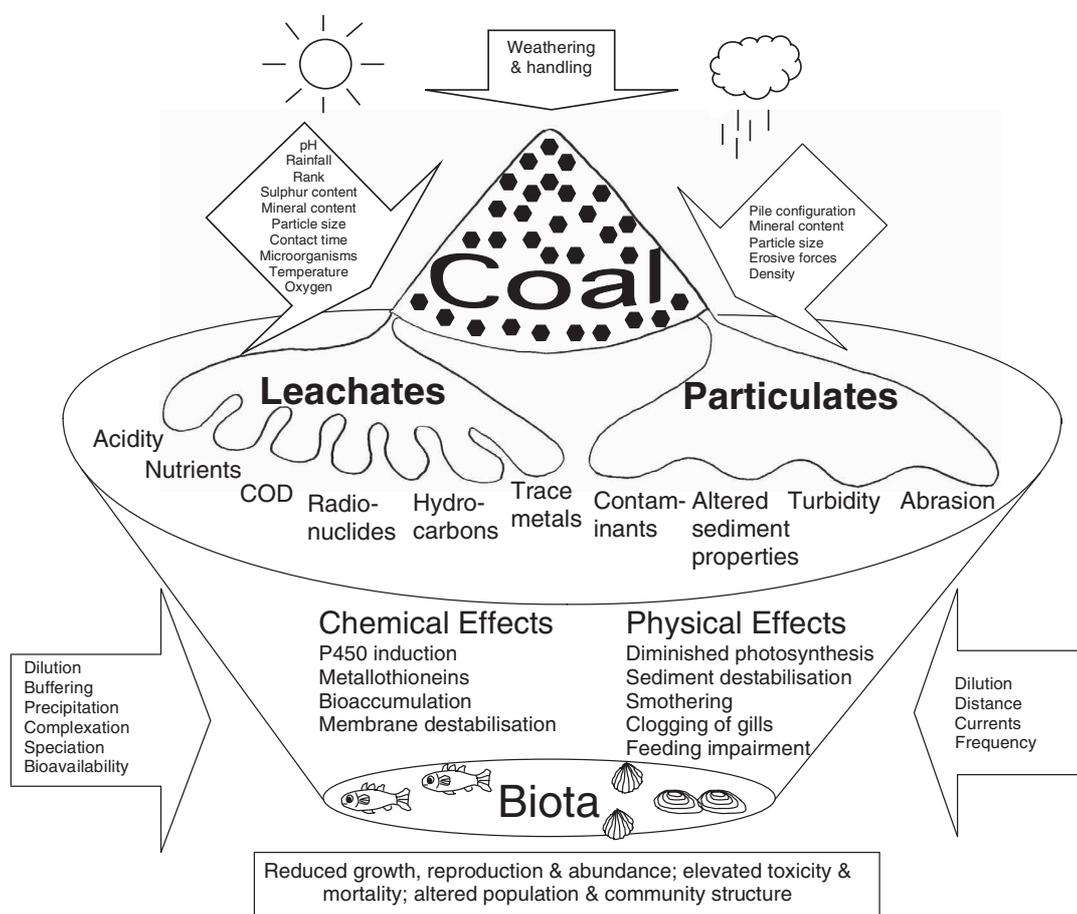


Figure 2 Factors affecting behaviour and effects of unburnt coal in the marine environment (COD = chemical oxygen demand). Influential factors in boxed arrows.

(sulphur content 1–2%) produce more pH neutral runoff (Davis & Boegly 1981b, Tiwary 2001, Cook & Fritz 2002). Higher sulphur content in coal also leads to higher suspended solids concentrations in runoff due to the breakdown of the coal matrix during oxidation. Furthermore, longer leaching duration increases suspended solids concentrations by promoting microbial degradation of sulphur compounds (Stahl & Davis 1984). The strong acid-producing potential of coal pile runoff has been confirmed in numerous studies of simulated or actual leaching of coal stockpiles (Hall & Burton 1982, Tease & Coler 1984, Swift 1985, Tan & Coler 1986, Carlson 1990, Cook & Fritz 2002) and has been shown to exert negative effects on terrestrial vegetation (Carlson & Carlson 1994), groundwater quality (Carlson 1990, Cook & Fritz 2002) and stream invertebrate communities (Swift 1985). Apart from sulphur content, a number of other factors are likely to influence coal leachate pH, including age and particle size of coal, rainfall frequency and amount, coal moisture content and the presence of sulphur-oxidising bacteria (Davis & Boegly 1981a). Conversely, acidity of coal pile leachates can be greatly diminished by promoting the growth of sulphate-reducing bacteria within coal piles (Kim et al. 1999). In the marine environment, significant impacts of acidic leachates are unlikely, due to the vast buffering capacity of seawater bicarbonate, except perhaps for constricted and poorly flushed embayments and estuaries. Water quality guidelines such as

Table 4 Physicochemical properties of coal leachates. Inorganics

Type of leachate	Coal type and origin	Sulphur % dw	Condi- tion	TSS mg l ⁻¹	TDS mg l ⁻¹	Filter pore µm	pH	EC µS cm ⁻¹	SO ₄ mg l ⁻¹	Al mg l ⁻¹	As µg l ⁻¹	Cd µg l ⁻¹	Cr µg l ⁻¹	Cu µg l ⁻¹	Fe mg l ⁻¹	Hg µg l ⁻¹	Ni µg l ⁻¹	Pb µg l ⁻¹	Se µg l ⁻¹	Zn µg l ⁻¹	Ref.
Coal leachate	E U.S.	4.6	Lab			0.45	2.1–3.8	187–4320	213–4060				0.18–55	n.a.	6–1470		n.a.	n.a.		n.a.	1
Coal leachate	SB W U.S.	1.1	Lab			0.45	4.6–8.3	27–670	35–189				0.16–49	n.a.	<0.10		n.a.	n.a.		n.a.	1
Stockpile runoff	n.a.	n.a.	Field			Unfilt	3.1–4.6	122–1716	100–1640				n.a.	500–2500	1.0–71.0					200–3100	2
Stockpile runoff	n.a.	1–3	Field	8–2300 (470)	2500–1600 (7900)	?	2.3–3.1 (2.79)		1800–9600 (5160)	66–440 (260)	5–600 (170)	<1	<5–11 (7)	430–1400 (860)	240–1800 (940)	<0.2–2.5 (0.4)	740–4500 (2590)		<1–30 (6)	2300–16,000 (6680)	3 in 4
Stockpile runoff	n.a.	>3	Field	38–2500 (420)	1200–7500 (3150)	?	2.5–3.1 (2.65)		870–5500 (5080)	22–60 (43.3)	6–46 (20)	<1–3 (2)	<5–11 (7)	10–460 (230)	62–480 (265)	3–7 (4)	240–460 (330)		<1–1 (1)	1100–3700 (2180)	3 in 4
Stockpile runoff	n.a.	n.a.	Field	2188 ± 3402		Unfilt			3.2 ± 3.4			1.9 ± 1.5	24 ± 24	35 ± 28	15.5 ± 14.4		50 ± 30	22 ± 18		280 ± 300	5
Stockpile runoff	Coal from 11 power plants	n.a.	Field		247–44,050 (12,600)	?	2.1–7.8		133–21920 (6880)				0–15.7 (2.74)	1600–3400 (2100)	0.06–93,000 (10,800)					6–23,000 (5890)	6 in 4
Stockpile runoff	?	?	Field		9330–14,900 (11,700)	?	2.2–5.8						100–750 (260)	100–6100 (1690)	10–5250 (1140)					2400–26,000 (5900)	7 in 4
Stockpile runoff	?	?	Field		4600–11,600 (6500)	?	2.4–2.9 (2.7)		1100–6900 (4100)	48–75 (62)				150–1000 (420)	150–1000 (420)						8 in 4
Ground-water under coal pile	E U.S.	Sulphur-rich	Field	>10,000		?	2.2	Up to 8480	Up to 22,200	Up to 1100				Up to 9560							9, 10
Simulated runoff	E U.S.	2–5	Lab	322–3300		?	1.7–3.2	3300–24,000													11
Stockpile runoff	E U.S.	2–5	Field			?	1.9–2.0	2800–19,000													11
Coal leachate	SB U.S.	'Low'	Lab			0.45	7.0–7.7	15–350	40–120			<0.2–0.8	0.2–0.5	0.1–0.6	0.0017–0.0025			<0.5		0.4–2.1	12
Coal leachate	SB & B Spain	2.2–9.5	Lab			Centri-fuged	n.a.		0–1851		0–345	7–52	0–700	100–2100	1–7900		100–21,000	0–6	1–180	100–42,000	13

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Coal leachate	W U.S.	Low	Lab	?	0-0.1	0.05-0.08	0-0.1	0.10-1.8	0-20.5	0-0.30	0-0.95	0.1-1.9	14
Coal leachate	W U.S.	Low	Lab pH 7.3	?	0.5-2.35	0-0.18	0.01-0.95	0.7-4.2	1.6-7.9	0.4-1.9	2.4-3.5	0.7-4.2	14
Coal leachate	W U.S.	Low	Lab pH 4.8	?	1.76-6.08	0.02-0.57	0-0.22	0.9-3.5	2.9-21.1	0-0.5	1-0-1.2	0-5.0	14
Coal leachate No. 2	W U.S.	Low	Lab	?	0.05-0.95	0.1-0.7	0.09-0.27	1.50-6.27	0.06-0.51	0-0.41	0-2.1	1-13.5	14
Coal leachate	E U.S.	High	Lab	?	0-1.9	2.30-4.79	0.43-1.95	6.44-63.9	180-323	0.01-0.22	0-2.2	65-220	14
Coal leachate	MW U.S.	0.6	Lab	?	598			0.32					15
Coal leachate	MW U.S.	2.9	Lab	?	33,766			15,700					15
Stockpile runoff	E U.S.	2.7-4.1	Field U.S.	?	1.3-17.0	0.002-6.49	1.4-3.1	1050-20,000	796-13,625	19.9-1872		3820-19,4350	16
Guidelines													
CWQG (FW)					FW 6.0-9.0 SW 7.0-8.7	FW 0.017 SW 0.12	FW 5.0 SW 1.0-8.9*	FW 2-4 SW 1.5-56*	FW 0.1	FW 25-150	FW 1-7	FW 1	FW 30
ANZECC (FW)					6-9	n.e.	13-24*	1.0	1.4	0.6	11	3.4	11
ANZECC (SW)					n.e.	n.e.	n.e.	4.4	1.3	0.4	70	4.4	n.e.
NOAA SQuiRT acute					n.e.	n.e.	FW 0.75 SW n.e.	FW 4.3 SW 16-570*	FW 13 SW 4.8	FW 1.4 SW 1.8	FW 470 SW 74	FW 65 SW 210	FW 120 SW 90
NOAA SQuiRT chronic					n.e.	n.e.	FW 0.087 SW n.e.	FW 2.2 SW 11-74*	FW 9 SW 3.1	FW 0.77 SW 0.94	FW 52 SW 8.2	FW 2.5 SW 8.1	FW 120 SW 81

Abbreviations: n.e. = not established; n.a. = not analysed; ? = no information available; B = bituminous; SB = sub-bituminous; unflt = unfiltered; ± = mean ± standard deviation; (parentheses) = mean; TSS = total suspended solids; TDS = total dissolved solids; E = eastern; W = western; MW = midwestern; FW = freshwater; SW = sea water; EC = electrical conductivity; * = trigger value depends on speciation of element; CWQG = Canadian Water Quality Guideline for Protection of Aquatic Life; ANZECC = Australian and New Zealand Guidelines for Fresh and Marine Water Quality (2000); trigger level for protection of 95% of species; NOAA SQuiRT = NOAA Screening Quick Reference Tables for Inorganics in Water; quotation marks = concentration not specified.

References: 1 Davis & Boegly 1981b, 2 Scullion & Edwards 1980, 3 Cox et al. 1977, 4 Davis & Boegly 1981a, 5 Curran et al. 2000, 6 Nichols 1974, 7 Anderson & Youngstrom 1976, 8 Featherby & Dodd 1977, 9 Carlson 1990, 10 Carlson & Carlson 1994, 11 Fendlinger et al. 1989, 12 Gerhart et al. 1980, 13 Querol et al. 1978, 14 Coward et al. 1978, 15 Cook & Fritz 2002, 16 Swift 1985, 17 Environment Canada 2002, 18 ANZECC 2000, 19 Buchman 1999

ANZECC (2000) recommend a general runoff pH to receiving waters (the guidelines do not specify whether marine or freshwater) of between 6–9, primarily to minimise corrosion (presumably of metal equipment, though this is not stated). For preventing adverse biological effects, Perkins (1976) recommends that materials introduced into saltwater portions of coastal waters should not change receiving water pH by more than ± 0.1 pH units from ambient conditions and at no time should they alter pH beyond the range of 6.7–8.5. Notwithstanding, there are no published studies that document drastic changes of seawater pH as a result of unburnt coal discharges.

Chemical oxygen demand

Coal pile leachates may have an increased chemical oxygen demand (COD) due to fine, suspended coal dust particles (Srivastava et al. 1994). Featherby & Dodd (1977) measured COD values of 37–161 mg l⁻¹ in coal pile drainage, and it is likely that COD of unfiltered stockpile runoff may exceed 1000 mg l⁻¹ when accompanied by elevated suspended solids concentrations.

Salinity

Coal pile runoff is often saline, due to salts formed during the oxidation and dissolution of mineral components of coal (e.g., sulphate from pyrite oxidation). While coal-generated salinity may not be important for the marine environment from a mass-loading perspective, the elemental composition of coal pile runoff may differ from sea water. Total dissolved solids (TDS) concentrations as high as 44 g l⁻¹ and electrical conductivity (EC) exceeding 8000 $\mu\text{S cm}^{-1}$ have been measured in runoff of sulphur-rich coal piles (Nichols 1974, Carlson & Carlson 1994; see Table 4). Nevertheless, on average, TDS concentrations of coal pile leachates do not exceed 15 g l⁻¹ (Table 4), which makes them less saline than typical sea water (ca 35 g l⁻¹). Because of the naturally high salinity and conductivity of sea water, the salinity inputs emanating from coal storage piles are not likely to have significant ecological effects on marine organisms. However, coal pile salinity may affect terrestrial and freshwater biota before reaching the marine environment. For example, death of terrestrial vegetation was observed at EC values above 4000 $\mu\text{S cm}^{-1}$ for soil solutions (Rendig & Taylor 1989).

Nutrients

There appear to be very few published data on the delivery of macronutrients into the aquatic environment from unburnt coal. Notwithstanding, coal does contain nitrogen and phosphorus in measurable quantities and a fraction of these nutrients appear to be leachable. Nitrogen makes up approximately 1–2% (by weight) of mineral-free coal (Table 2) and is commonly associated with the organic compounds present, because no nitrogen-bearing minerals are known in coals (Ward 1984). Gerhart et al. (1980) measured nitrate concentrations of 0.075–0.166 mg l⁻¹ and 0.02–0.12 mg l⁻¹ in filtered leachates of low-sulphur, sub-bituminous coal (1.6% and 0.8% by weight, respectively), with a mean of approximately 0.07 mg l⁻¹ for the 0.8% coal leachates. Ammonium nitrogen was approximately 0.15 mg l⁻¹ and dissolved organic nitrogen was 1.35 mg l⁻¹. The greatly elevated nitrate levels of some coal mine drainages are due to the use of explosives (Tiwary 2001). Oxidation of coal, combined with heating to 270°C, releases water-soluble organic nitrogen compounds such as phenanthridine, phenyl pyridine, pyridine, and azaarenes such as quinoline and derivatives (Francis 1961, Barrick et al. 1984). Most nitrogen, however, is released only upon heating, either as ammonium or nitrogen oxides.

UNBURNT COAL IN THE MARINE ENVIRONMENT

Although phosphorus is an important element in living cells, its concentration in coal is generally low. Most coals contain between 10–2000 ppm phosphorus (Table 2; Francis 1961, Swaine 1990, Rao & Walsh 1997), usually present as inorganic apatite, $\text{Ca}_5(\text{PO}_4)_3(\text{F}, \text{OH})$, $\text{Ca}_{10}\text{F}_2(\text{PO}_4)_6$, or bound in aluminium hydroxides (Ward 2002), although some coals, such as Alaskan coal, contain concentrations up to 1% (Rao & Walsh 1997, 1999). Phosphorus content is often correlated with fluorine (Francis 1961), but there is disagreement whether P is typically associated with the organic or the mineral fraction. Gerhart et al. (1980) measured 0.02–0.12 mg l⁻¹ of total phosphorus in filtered leachates containing 0.8% sub-bituminous coal. This included between 0.01 and 0.1 mg l⁻¹ dissolved phosphorus and <0.001–0.008 mg l⁻¹ reactive phosphorus. Ward (2002) found up to 60% of the phosphorus in south Australian coals to be leachable by water washings, although he did not report concentrations. Phosphorus showed variable distribution in these coal deposits, being present in the water-exchangeable fraction in some coals but in the acid-soluble fractions in others. Querol et al. (1996), in a sequential leaching experiment, found between 88–94% of the phosphorus contained in four Spanish coals (P content 68–200 ppm) to be leachable by nitric acid digestion, whereas little phosphorus was mobilised by water or ammonium acetate extractions of the same coal samples. Other than the studies reported above, there are few published data describing how leachable phosphorus is under typical environmental conditions as encountered in stockpiles or water-submerged coal.

Trace metals

As a decomposition product of ancient plants, coal contains virtually every element found in living plant tissues, including trace metals (Table 2). Metals may be present as dissolved salts in pore waters, as metallo-organic compounds, or as mineral impurities (e.g., iron in pyrite, FeS, and zinc in sphalerite, ZnS). Information on trace elements in coal has been reviewed comprehensively by Swaine (1990) and Swaine & Goodarzi (1995), including environmental aspects during mining and combustion, though, unfortunately, not during storage and transport. Every type of coal contains a sizable inorganic fraction, which affects its abrasive properties, stickiness, corrosion potential and release of trace metals (Ward 2002). The forms in which potentially toxic trace elements are held in coal, and the extent to which these may be released, vary among coals and greatly depends on the mineral matter present and, to a lesser extent, on coal rank. Many studies have indicated links between the minerals present in coal and the concentration of particular trace elements (reviewed by Ward 2002). For example, As, Cd, Pb, Hg, Sb, Se, Tl and Zn are often associated with sulphides and, therefore, show strong correlations with, for example, pyrite content of coal. Chromium and a number of other elements tend to associate with aluminosilicates, and strontium and barium are often found in the presence of carbonates and aluminophosphate minerals. The low pH of sulphur-rich coal pile leachate favours dissolution of metals such as Fe, Cu, Mn, Cr and Zn (Anderson & Youngstrom 1976). Trace metal concentrations in runoff from stockpiles of sulphur-rich coal can be so high as to endanger groundwater quality, in the absence of buffering capacity of the environment (Cook & Fritz 2002). For example, stormwater runoff from coal piles has been measured to contain more than 100 mg l⁻¹ of aluminium and several mg l⁻¹ of copper, iron and zinc (Table 4). A study by Curran et al. (2000) of coal storage piles in Ontario, Canada, found total (i.e., dissolved and suspended) metal concentrations to exceed Canadian water quality guidelines for Al, Cd, Cr, Cu, Fe, Pb and Zn (Table 4). While the metal loadings from stockpile runoff represented a relatively small input to the lake into which it drained, the authors suggested that there was potential for localised effects. Comparison of leachate metal concentrations with other international guidelines listed in Table 4 shows that for virtually any metal, exceedances of the guideline values can be found for at least some coal samples.

Although it has been stated that high concentrations of arsenic and selenium are typically associated with coal-pile leachate (Cook & Fritz 2002), there is little evidence available in the published literature to support the contention that As and Se are especially elevated in leachates (Table 4). However, in areas with As-rich coal, such as southwest China (As content of up to 3.5%; Table 2, Ding et al. 2001), poisoning of humans (arsenosis) is a serious health concern, although the toxicity derives primarily from contact with coal combustion products rather than from unburnt coal. Most coals contain 0.5–80 ppm As and 0.2–10 ppm Se (Swaine & Goodarzi 1995).

Coward et al. (1978) conducted elaborate leaching studies on coals from the western and eastern U.S. to determine the dominant factors affecting leaching of metals (Table 4). In their factorial design, the investigators changed a number of variables believed to affect leachate quality in coal storage piles, including pH, temperature, particle size, oxygen saturation, contact time and flow. While most variables showed some effect for some metals, raising the temperature and lowering the pH generally increased leachate concentrations of most metals measured. Furthermore, sulphur-rich eastern coal had a much lower leachate pH than western coal and, on average, leached considerably higher amounts of metals such as Cd, Co, Cu, Mn, Ni and Zn. Metal leaching from western coals could be increased by lowering the pH of the leaching solution or adding complexing agents such as EDTA. The leachate data collected in Table 4 support the interim conclusion that high sulphur content and low leachate pH closely correlate with, and thus are useful indicators of, elevated metal concentrations in coal leachates.

The most comprehensive coal leaching study to date was conducted by Querol et al. (1996), who determined leachability of over 40 trace metals from four Spanish sub-bituminous to bituminous coals by sequential extraction with water, ammonium-acetate and nitric acid (HNO_3). By monitoring the release of trace metals and the distribution of the predominant mineral phases in coal (including different types of sulphides, sulphates, carbonates and aluminosilicates) after each extraction step, Querol et al. were able to determine the likely mineral phases with which different elements were associated. Elements associated with sulphides, sulphates and organic matter showed the highest extraction efficiencies (up to 90% after HNO_3 treatment). These included As, B, Be, Mn, Mo, Ni, Pb, Se, Sr, Tl, U, V, Y, Zn and heavy rare-earth elements. In contrast, elements with strong aluminosilicate affinity (e.g., Sn, Sb, Rb and Ta) had lowest leachability, and elements with intermediate affinity to aluminosilicates had intermediate mobility (Ba, Cd, Co, Cr, Cs, Cu, Ga, Ge, Li, Zr and light rare-earth elements). The major leachable fraction in coals was found in the nitric acid fraction, which mobilised the organic matter and sulphide-associated elements. However, all elements with strong organic and sulphide-sulphate affinities also had a water-leachable fraction, whose size depended on the degree of weathering of the coal sample. This study clearly showed that mobility of trace elements in coal is controlled by their affinities to organic matter and mineral impurities.

Metal toxicity to animals and plants is crucially dependent on the dissolved form, or speciation, of the metal, specifically the free metal ion concentration (Hall & Anderson 1995). Low pH values generally increase toxicity by increasing the free metal ion concentration (Gerhardt 1993, Wood 2001). For example, increasingly acidic pH increases toxicity of Cd, Fe, Zn and Pb to many aquatic invertebrates (Gerhardt 1993), and aluminium toxicity to plants occurs at soil pH below 4.5 (Andersson 1988). For this reason, metal-rich and acidic coal leachates potentially represent a compounded stressor for aquatic organisms. In addition to uptake and associated toxicity from dissolved metals, a certain fraction of metals may be accumulated from particles by ingestion (Wang & Fisher 1999) or direct contact.

In a study of effects of colliery waste dumped on the sea bed off the coast of northeast England, Hyslop et al. (1997) found that concentrations of metals in waste washed up on the beach were much lower than those in coal and waste prior to dumping (Table 6). Concentrations of Cr, Cu, Ni and Pb in the washed-up coal were well below sediment quality guidelines for the protection of

aquatic life. The fact that much of this metal content apparently leached out while the material was on the sea bed suggests that it might have been biologically available. Uptake of contaminants derived from coal by sediment-living animals and plants raises the possibility that contaminants may be transferred to higher levels of the food web and, if uptake is by mobile animals, exported to other areas (Wang 2002, Blackmore & Wang 2004).

Hydrocarbons

Unburnt coal contains a large fraction of volatile organic hydrocarbons, and organic compounds from coal pile runoff include aliphatic and aromatic hydrocarbons (Tables 3 and 5; Barrick et al. 1984, Stahl et al. 1984, Fendinger et al. 1989). In fact, the majority of the organic carbon in coal is believed to exist in the form of large, 5- or 6-membered rings of aromatic molecules (Neff 1979), and aromaticity increases with rank or coalification. Conversely, during the diagenetic coalification process, the oxygen content of coals is lowered, which results in lower concentrations of hydroxylated aromatic compounds, such as phenols, which are most prevalent in lignitic coals (Schulz 1997). Among the aromatic compounds, polycyclic aromatic hydrocarbons (PAHs) are of particular environmental interest, because they can be mutagenic or exert narcotic toxicity when present in bioavailable form. Studies of aquatic sediment contamination in the state of Washington (U.S.) have found high PAH concentrations within a few kilometres of industrial facilities or river systems draining coal-bearing strata (Barrick & Prahl 1987). However, it should be noted that chemical alteration of coal, by refining or coking processes, tends to greatly concentrate PAHs (in addition to *de novo* synthesis at high temperatures), such that sediment PAH signatures may have been swamped by these 'processed coal' sources.

Sediment contamination by coal can be readily distinguished from other sources using molecular markers such as azaarenes, alkylated phenanthrenes (primarily 1- or 3-methylphenanthrene), and chrysene and picene derivatives (Barrick et al. 1984). Other dominant coal components include pristane, C₁₉ and C₂₀ tricyclic diterpanes, retene, tetrahydrochrysenes and hydropicenes. In an extensive analysis of 16 Washington coals, Barrick et al. (1984) found that increasing rank (i.e., lignite to anthracite) increased the proportion of low molecular weight n-alkanes relative to other aliphatic hydrocarbons. Increasing rank furthermore increased the proportion of unsubstituted PAHs relative to alkylated homologues (e.g., naphthalene vs. methyl-naphthalenes) and increased 3-ring azaarenes relative to 2-ring azaarenes. Lower rank coals tended to be dominated by retene and had low quantities of the PAH phenanthrene. Interestingly, Barrick et al. (1984) found only trace amounts of unsubstituted 4- to 7-ring PAHs in Washington coals (except in highest ranked coals). These high molecular weight PAHs, which include well-known compounds such as fluoranthene, pyrene and benzo(a)pyrene, are generally indicative of combustion sources and tend to dominate the PAH signature of marine sediments near urban areas (Latimer & Zheng 2003). Coal hydrocarbon signatures can furthermore be distinguished from petroleum by the absence of an unresolved complex mixture (UCM) bulge in their gas chromatograms. Gerhart et al (1980) measured phenol concentrations of 0.16 mg l⁻¹ in leachates of sub-bituminous coal from the Western U.S., while Featherby & Dodd (1977) found considerably lower concentrations (<0.001–0.012 mg l⁻¹) of phenol in coal pile drainage from a Canadian power plant.

In studies of simulated rainfall on coal stockpiles, the highest concentrations of PAHs occurred during the 'first flush' events, when concentrations of suspended solids in the runoff were highest (Fendinger et al. 1989). PAHs that commonly occur in measurable concentrations in coal leachates include naphthalene, phenanthrene, chrysene, fluoranthene and pyrene (Table 5). In a leaching study of four types of coals (spanning lignitic to bituminous rank), Stahl et al. (1984) noted that more PAHs were leached from sulphur-rich bituminous coals than from low sulphur sub-bituminous coal or lignite, although no correlation between sulphur content and leachate concentration was found

Table 5 Physicochemical properties of coal leachates. Organics

Type of leachate	Coal type or origin	Sulphur % dw	Study location	Suspended solids mg l ⁻¹	Filtration procedure	ACE µg l ⁻¹	BaA µg l ⁻¹	BaP µg l ⁻¹	BKF µg l ⁻¹	CHR µg l ⁻¹	FLT µg l ⁻¹	FL µg l ⁻¹	PHE µg l ⁻¹	NAP µg l ⁻¹	PYR µg l ⁻¹	DOC mg l ⁻¹	Σ Arom µg l ⁻¹	Ref.
Stockpile runoff	n.a.	n.a.	Field Canada	2188 ± 3402	Unfiltered		71 ± 85 ^a	47 ± 49 ^a	208 ± 254 ^a	284 ± 339 ^a	140 ± 173 ^a						1	
Stockpile runoff	n.a.	n.a.	Field Canada	dissolved	Whatman #1		0.9 ± 0.2	0.7 ± 0.2	4.5 ± 2.1	6.1 ± 3.3	3.1 ± 1.4						1	
Simulated rain runoff	B Illinois #6	>2%	Lab	extracted 1 kg coal	Glass wool	1	0.6	0.6	3	40–48	4	5	20–33				2	
Simulated rain runoff	B Kentucky	>2%	Lab	extracted 1 kg coal	Glass wool					11			8				2	
Simulated rain runoff	SB Nerco Montana	<2%	Lab	extracted 1 kg coal	Glass wool					0.7–6							2	
Simulated rain runoff	L Montana	1–3%	Lab	extracted 1 kg coal	Glass wool					0.6–1.2							2	
Simulated rain runoff	? Texas	?	Lab	extracted 1 kg coal	n.a.	15			16			14					3 in	
Simulated rain runoff	Montana		Lab	9 kg coal													2	
Simulated rain runoff	Pennsylvania & Maryland	2–5%	Lab	extracted 6.8 kg coal	Whatman GFC	0.06–0.47						0.15–0.29	0.06–0.19	0.04–0.97	0.01–0.33	1–28	0.9–16.7	4
Simulated rain runoff	Pennsylvania & Maryland	2–5%	Lab	0.08–2.26	Suspended			0.12–11.20	0.5–5.76	0.10–2.78	0.10–5.15		0.05–3.58	0.07–5.22	0.10–5.15	0.4–323	4	
Stockpile runoff	Pennsylvania & Maryland	2–5%	Field U.S.		Filtered											29	34–95	4
Stockpile runoff	Pennsylvania & Maryland	2–5%	Field U.S.		Suspended												5–42	4

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Stockpile runoff	Canada	Field	100	Settled SW leachate	0.285	0.233	0.417 ^c	0.466 ^b	0.353	0.28	2.037	0.408	0.373	5
Simulated runoff	SB	Low	0.8% w/w	0.45 µm									13-27	6
SW leachate	Virginia	n.a.	1-10 mg l ⁻¹	Filtered						<0.001	0.2	0.1		7
Guidelines														
MOEE			10		144		46	102		95		85		in 1
ANZECC												FW 16		8
												SW 70		
CWQG					5.8	0.018	n.r.	0.04	0.04	3	0.4	FW 1.1	0.025	9
												SW 1.4		
NOAA												FW 30	FW 2300	10
SQuiRT-acute, SW												SW 7.7	SW 2350	

Abbreviations: n.a. = not analysed; ? = no information available; ^a = total concentration in µg g⁻¹ organic carbon; ^b = including triphenylene; ^c = sum of benzo(b)fluoranthene and benzo(k)fluoranthene; B = bituminous; L = lignite; SB = sub-bituminous; ± = mean ± standard deviation; FW = fresh water; SW = sea water; FLT = fluoranthene; PHE = phenanthrene; BaP = benzo(a)pyrene; PYR = pyrene; CHR = chrysene; NAP = naphthalene; FL = fluorene; BaA = benz(a)anthracene; BkF = benzo(k)fluoranthene; ACE = acenaphthene; DOC = dissolved organic carbon; Σ Arom. = total aromatic hydrocarbons; MOEE = Ministry of the Environment and Energy guideline (Ontario, Canada) for particulate bound PAH; taken from Curran et al. (2000), with corrected units; ANZECC = Australian and New Zealand Guidelines for Fresh and Marine Water Quality (2000); trigger level for protection of 95% of species; CWQG = Canadian Water Quality Guidelines for the Protection of Aquatic Life (2002); NOAA SQuiRT = NOAA Screening Quick Reference Tables for Organics.

References: 1 Curran et al. 2000, 2 Stahl et al. 1984, 3 Wachter & Blackwood 1978, 4 Fendinger et al. 1989, 5 Campbell & Devlin 1997, 6 Gerhart et al. 1980, 7 Bender et al. 1987, 8 ANZECC 2000, 9 Environment Canada 2002, 10 Buchman 1999

Table 6 Concentrations of trace metals in colliery waste and tailings dumped at beach and offshore sites in northeast coast of England, and of waste washed up on the beaches of the area. Sediment quality guidelines from several sources are shown for comparison. All values are $\mu\text{g g}^{-1}$.

Type of material	Cd	Cr	Cu	Hg	Ni	Pb	Zn	Reference
Colliery waste <90 μm	2	13	90	1	52	110	100	Norton 1985
Colliery waste 90–500 μm	2	5	120		43	230	540	Norton 1985
Tailings <500 μm	0.72–0.79	18–29	30–50	0.05–0.10	38–40	52–72	54–68	Limpenny et al. 1992 ^a
Waste on shore	0.01	0.38	0.54		0.45	0.65	0.1	Hyslop et al. 1997
Whole sediment	0.6	51	54	0.24	34	75	190	Limpenny et al. 1992 ^b
ANZECC ISQG–low	1.5	80	65	0.15	21	50	200	ANZECC 2000
ANZECC ISQG–high	10	370	270	1	52	220	410	ANZECC 2000
NOAA/Environment Canada ISQG	0.7	52.3	18.7	0.13	n.e.	30.2	124	Persaud et al. 1993 ^c
NOAA/Environment Canada PEL	4.2	160	108	0.7	n.e.	112	271	Persaud et al. 1993 ^c

Abbreviations: Empty cell = not analysed; n.e. = not established; ISQG = Intermediate Sediment Quality Guidelines; PEL = Probable Effect Level

Notes: ^a range of 2 values, ^b maximum value from 11 samples taken near beach disposal point, ^c derived from the same source as NOAA guidelines; Buchman 1999.

in a subsequent study (Fendinger et al. 1989). Stahl et al. (1984) also noted that many of the organic compounds known to exist in raw coal, such as alkenes, alkyl benzenes, alkyl phenols, were not detected in their laboratory leachates, which was attributed to poor extraction efficiency of the leaching medium (distilled water). Schulz (1997) showed that extraction of phenolic compounds from coal dust by aqueous solutions could be greatly enhanced by the addition of surfactants such as lecithin. In general, the hydrocarbon concentrations (primarily PAHs) that have been measured in filtered leachates have been much less than $50 \mu\text{g l}^{-1}$, and are typically less than $5 \mu\text{g l}^{-1}$. It is likely that the overall poor solubility of PAHs limits substantially higher concentrations in leachates. Assuming a maximum concentration of $50 \mu\text{g l}^{-1}$ for an individual PAH compound, Stahl et al. (1984) estimated an average-sized coal stockpile (ca 10^5 t) to release 2–3 g of that PAH as runoff yr^{-1} . The derivation of this number is obscure, however.

Because PAHs are poorly water soluble and highly hydrophobic, they have a high affinity for particles, and especially for the hydrophobic domains of organic matter or condensed forms of carbon (Bucheli & Gustafsson 2000). Thus, coal runoff containing suspended coal has considerably higher PAH concentrations than filtered leachates (Tables 3 and 5). It should also be noted that PAHs leached from particulate coal are subject to volatilisation, photodegradation and bacterial degradation that could diminish dissolved concentrations before they reach the receiving waters.

Even though the toxicity of PAHs is well recognised (Di Toro & McGrath 2000, Di Toro et al. 2000), water quality guideline values exist for only a handful of compounds (Table 5) and differ greatly between different sets of guidelines. PAHs as a class of contaminant share a similar mode of action (narcosis), so that the toxicity of mixtures of PAHs should be additive (Verhaar et al. 1992). Thus, while concentrations of individual PAHs in coal leachates may be below the respective EC_{50} (i.e., the effects concentration leading to a response in 50% of the test organisms), it is possible

that the summed concentrations of PAHs may approach toxic levels for undiluted leachate. While additivity of PAH toxicity has been corroborated in laboratory studies with mixtures of pure compounds (Swartz et al. 1997), this phenomenon has not been confirmed for PAHs in coal pile leachates. Furthermore, the majority of PAHs in coal pile leachates has been found to be associated with particulate material, whereas dissolved concentrations are commonly low ($<5 \mu\text{g l}^{-1}$). For example, Leppard et al. (1998) found that more than 95% of the water-borne PAHs in a coal-impacted harbour were associated with suspended flocs. This is likely to reduce their bioavailability, as discussed below. To date, there is no published evidence of direct PAH toxicity to marine invertebrates from particulate coal or coal leachates.

Radioactivity

Unburnt coal contains uranium and thorium, and a variety of radioactive isotopes from the natural decay series of ^{238}U , ^{235}U and ^{232}Th , along with traces of ^{40}K (Swaine 1990). Concentrations of Th and U for most types of coal range between 0.5–10 ppm and 0.5–20 ppm, respectively (Swaine 1990), and are generally similar to or lower than concentrations in soil and other sedimentary strata. Nevertheless, some coals in India may contain up to 100 ppm U, and up to 2% U has been measured in coal from Colorado and adjacent regions in the eastern Rocky Mountains (summarised in Swaine 1990, Tadmor 1986). Highest concentrations of radioactive elements have typically been measured in lower rank coals, such as lignite. Hedvall & Erlandsson (1996) summarise average activity mass concentrations of 50, 20 and 20 Bq kg^{-1} for ^{40}K , ^{238}U and ^{232}Th for unburnt coal. Most studies concerned with radioactivity from coal have focused on the release of radionuclides during the combustion process, which tends to concentrate heavier radioactive elements in the fly ash. Average activity mass concentrations in escaping coal fly ash, estimated by Hedvall & Erlandsson (1996), range between 70–1700 Bq kg^{-1} for eight radionuclides. There are no explicit studies in the literature on the aqueous leachability of radioactivity from unburnt coal, such as from storage piles, but it is reasonable to assume that the released radioactivity will be lower than in fly ash, where the entire coal matrix is destroyed. McDonald et al. (1992), conducting a nationwide survey of radioactivity in coastal U.K. sediments, found a 710-fold concentration of ^{238}U relative to sea water in sediments at a site receiving coal spoils from a local colliery, compared with concentration factors of approximately 100 for sediments away from direct industrial inputs. Concentration factors for ^{210}Pb and ^{210}Po were approximately 1900, compared with 300–650 for a nearby, coal-free sediment sample. While the study reported concentration factors for marine biota (seaweed, mussels and winkles; no species names given) at other sites, no bioaccumulation data were collected for the colliery site. However, assuming similar bioavailability of these radioactive elements in coal-laced sediments as in other sediments, concentration factors in biota would be expected to be 10 times lower for ^{210}Pb and ^{238}U and of similar magnitude or up to an order of magnitude higher for ^{210}P . Given that concentrations of radioactive elements in coal are of a similar order of magnitude as in soil or shale, and assuming a similarly low bioavailability, biological effects from the traces of radioactivity in coal can be considered highly unlikely.

Toxic effects of unburnt coal and leachates on aquatic organisms

In marked contrast to coal's well-documented potential to cause adverse physical effects in aquatic organisms, as reviewed above, there is surprisingly little published evidence demonstrating direct toxic effects of unburnt coal to marine organisms and communities (published information on effects of unburnt coal on aquatic organisms is summarised in Table 7). This paucity of evidence seems to uphold the hypothesis that unburnt coal is an ecotoxicologically relatively inert substance (Chapman et al. 1996a). On the other hand, the scarcity of evidence for toxic effects of coal in the

Table 7 Effects of unburnt coal, including leachates, on aquatic organisms

Species	Experiment type	Exposure condition	Coal type	Coal concentration	Assumed stressor	Observed effect	Reference
Freshwater algae	Lab	Centrifuged leachate	Sub-bituminous, Montana, U.S.	Coal concentration, 3–20% v/v	Volatile organic compounds	Stimulation of algal growth by coal leachates and changes in species composition. Growth inhibition by coal distillates in closed containers, which disappeared upon aerating containers, possibly due to removal of volatiles. In field mesocosms, 1–20% v/v distillate concentrations increased algal and bacterial numbers and killed zooplankton	Gerhart et al. 1980
Green alga (<i>Ulva lactuca</i>)	Lab	Suspended colliery waste	NE England, U.K.	29% by weight in waste, 1 g l ⁻¹ suspended waste	Abrasion by particulates	Reduced growth in presence of waste and water movement but increased growth with waste under still conditions	Hyslop & Davies 1998
Duckweed (<i>Lemna minor</i>) (freshwater)	Lab & field	Unfiltered slurry	Western Coal No. 1, U.S.	16.6–83.3 g l ⁻¹	Metals, suspensoids	Similar growth over 16 d compared to controls; greater bioaccumulation of some metals (Ba, Co, Cu, Zn).	Coward et al. 1978
Mangrove (<i>Avicennia marina</i>), South Africa	Field	Airborne coal	n.d.	n.d.	Light reduction	Reduced CO ₂ exchange by 17–39%, decreased photosynthetic performance	Naidoo & Chirkoot 2004
Deposit-feeding polychaete (<i>Arenicola marina</i>), NE England	Field	Deposited colliery waste	NE England, U.K.	11% of sediment by weight	Physical destabilisation of sediment by particulates	Worms avoided ingesting coal particles during deposit feeding (possibly on the basis of particle size); avoidance of contaminated sediments in choice tests; reduced abundance	Hyslop & Davies 1999
Crab (<i>Cancer magister</i>)	Lab	Deposited (and suspended?) coal	n.d.	Up to 50% of sediment by weight	Smothering of gills by particulates	Accumulation of coal in gills at higher concentrations	Pearce & McBride 1977

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Crab (<i>Cancer magister</i>)	Lab	Deposited (and suspended?) coal	n.d.	Up to 75% of sediment by volume	Smothering of gills by particulates	No measurable difference in ventilation and oxygen consumption over 21 d relative to controls	Hillaby 1981
Marine predatory snail (<i>Hexaplex trunculus</i>), Israel	Lab & field	Coal sediment	n.d.	n.d.	Cd from direct contact	Snails from coal-impacted site had 1.8 times higher Cd concentrations in hepatopancreas, up to 3.6 times increased epithelial permeability, reduced lysosome transport and 3 times higher metallothionein levels than coal-free controls	Siboni et al. 2004
Oyster (<i>Crassostrea virginica</i>)	Lab	Suspended coal dust including leachate (pre-equilibrated for 2 weeks)	<40 µm	0, 1 and 10 mg l ⁻¹	PAH	No significant adverse effect on oyster survival, shell growth or pumping activity after 28 d, but clams in highest treatment had slightly reduced shell growth (non-significance due to large variance). No significant accumulation of PAHs in tissues of depurated oysters, despite observable ingestion of coal. However, note again high variance in tissue levels after 28d.	Bender et al. 1987
Deposit-feeding bivalve (<i>Macoma balthica</i>), Alaska	Field	Coal in sediments	Erosion of exposed coal seams, Kachemak Bay, Alaska, U.S.	n.d.	Saturated and unsaturated hydrocarbons	Animals from naturally coal-contaminated site contained an array of hydrocarbons characteristic of the coal in the sediment, but animals were not depurated prior to analysis, so ingestion and assimilation could not be distinguished	Shaw & Wiggs 1980
Intertidal assemblages of rocky and sandy shores, NE England	Field	Deposited colliery waste	NE England, U.K.	27% of sediment by weight	Physical abrasion, destabilisation of sediment by particulates	Reduced number of macroalgal species on contaminated rocky shores, and of macroinvertebrates on sandy shores	Hyslop et al. 1997

Table 7 (continued) Effects of unburnt coal, including leachates, on aquatic organisms

Species	Experiment type	Exposure condition	Coal type	Coal concentration	Assumed stressor	Observed effect	Reference
Benthic faunal assemblages, NE U.K.	Field	Colliery waste dumped on the shore	NE England, U.K.	n.d.	Suspended and deposited particulates	Infilling of crevice habitats of crabs and lobsters, reduced abundance and diversity of soft-sediment assemblages, with only mobile polychaetes, amphipods and ophiuroids present around one of the larger beach dumpsites	Shelton 1973
Subtidal soft-sediment benthic assemblages, NE England	Field	Colliery waste dumped on sea bed	NE England, U.K.	Up to 20% in waste, and waste represents >70% of sediment	Suspended and deposited particulates	Reduced diversity and abundance at sites with large amounts of waste present, but effects of colliery waste confounded by disposal of dredge spoil at same site and by differences in water depth. No evidence of uptake of metals by commercially harvested fish and crustaceans	Norton 1985
Subtidal soft-sediment benthic assemblages, NE England	Field	Colliery waste dumped on sea bed	NE England, U.K.	1.8–5.7% of sediment by weight	Suspended and deposited particulates	Reduced abundance and diversity at former waste disposal sites compared with control site, with variable evidence of recovery among impacted sites, but one impacted site had higher diversity than the control (6 months after cessation of dumping)	Johnson & Frid 1995
Intertidal soft-sediment benthic assemblages, NE England	Field	Colliery waste dumped on shore	NE England, U.K.	Up to 20% in waste, no data given on concentration in sediment	Suspended and deposited particulates	Reduced abundance and diversity at former waste disposal sites compared with control site and undumped site with nearby, natural source of coal, with variable evidence of recovery among impacted sites	Barnes & Frid 1999

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Intertidal soft-sediment benthic assemblages, British Columbia, Canada	Field	Coal in sediments	Bituminous coal from wrecked collier	4.35% of sediment (as TOC)	Deposited particulates	Changes in benthic assemblage at sites where coal present, attributed to grain-size effects rather than the toxicity of coal-derived PAH (which showed little correspondence with sediment toxicity)	Chapman et al. 1996a
Benthic faunal assemblages, Svalbard	Field	Coal in sediments	Svalbard	n.d.	Fine sediment	High concentrations of PAH present in sediments, presumed to be derived from runoff from coal stores and general industrial activity. Low faunal diversity and dominance by species characteristic of organically enriched areas attributed to effects from deposition of fine-grained glacial sediments, untreated sewage inputs and garbage dump leachate.	Holte et al. 1996
Stream invertebrate assemblages, South Wales, U.K. (freshwater)	Field	Suspended and deposited stockpile runoff	South Wales, U.K.	n.d.	Siltation by coal particulates, low pH, trace metals	Reduced faunal abundance and diversity in both pH-affected and sediment-affected stretches of the stream, with variable susceptibility among taxa, with Ephemeroptera, Plecoptera and Trichoptera most susceptible and burrowers, e.g., chironomid larvae and oligochaetes least	Scullion & Edwards 1980
Stream invertebrate assemblages, eastern U.S. (freshwater)	Field	Coal-pile runoff	Eastern U.S.	n.d.	Trace metals, low pH	Reduced faunal diversity attributed to combination of effects of periodic drought and coal-pile runoff	Swift 1985

Table 7 (continued) Effects of unburnt coal, including leachates, on aquatic organisms

Species	Experiment type	Exposure condition	Coal type	Coal concentration	Assumed stressor	Observed effect	Reference
Fathead minnow (<i>Pimephales promelas</i>)	Lab	Suspended coal, spiked with phenanthrene	Sub-bituminous, Decker, Montana, U.S.	10–20 mg l ⁻¹	Abrasion	No changes to gut and gill epithelium and no changes to growth rate over a 14-d period; pronounced mucus and coal accumulation in gut, but rapid gut clearance after exposure; no difference in phenanthrene uptake between coal treatments and particle-free controls	Gerhart et al. 1981
Fathead minnow (<i>Pimephales promelas</i>)	Lab	Centrifuged and uncentrifuged leachates	Sub-bituminous, Montana, U.S.	6.3 g l ⁻¹ centrifuged; 25 g l ⁻¹ uncentrifuged	PAHs	100% mortality in uncentrifuged leachate after 96 h. No increased mortality of juveniles or adults exposed to centrifuged leachate for 3–24 weeks; growth rate similar to controls, but onset of maturity delayed; 36% spawning success in leachates vs. 90% in controls; Some qualitative differences in GC analyses of tissue extracts	Carlson et al. 1979
Bullhead catfish, rainbow trout (<i>Ictalurus nebulosus</i> and <i>Oncorhynchus mykiss gairdneri</i>)	Lab	Coal heavy distillate and derived fractions	Mixed with water	10 mg l ⁻¹	Hydrocarbons	Pathological responses (hyperplasia and engorgement of blood vessels of gill tissue, changes to mitochondria and rough endoplasmic reticulum), particularly in rainbow trout	Stoker et al. 1985
Rainbow trout (<i>Oncorhynchus mykiss gairdneri</i>)	Lab	Suspended coal		120–600 mg l ⁻¹	Mechanical irritation	Cough rate increased twofold	Hughes 1975

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Rainbow trout (<i>Oncorhynchus mykiss gairdneri</i>)	Lab	Suspended coal washings	Nottinghamshire, U.K., coal washings	200 mg l ⁻¹	Mechanical irritation	No toxic effects but reduced growth over a 10-month period	Herbert & Richards 1963
Rainbow trout (<i>Oncorhynchus mykiss gairdneri</i>)	Lab	Centrifuged leachate	Sub-bituminous, Montana, U.S.	6.3 g l ⁻¹ centrifuged;	PAHs	No pronounced differences in liver parameters: liver weight, hepatic microsomal protein, DNA content and AHH activity after 28 d; no consistent increase in hepatic AHH activity and cytochrome P450 content over 21 d upon exposure to coal steam distillates	Carlson et al. 1979
Chinook salmon (<i>Oncorhynchus tshawytscha</i>)	Lab	Suspended coal	n.d.	60–500 mg l ⁻¹	PAHs	Increased CYP1A1 and ribosomal protein L5 expression in liver	Campbell & Devlin 1997
Steelhead and cutthroat trout (<i>Oncorhynchus mykiss</i> and <i>O. clarkii</i>), eastern U.S. (freshwater)	Field	Suspended coal	Washington, U.S.	22.5 g l ⁻¹ (?)	Mechanical irritation by particulates	Mortality after 0.5–2.5 h exposures to suspended coal washing in a stream	Pautzke 1937
Eight fish species, including brown trout (<i>Salmo trutta</i>) South Wales, U.K. (freshwater)	Field	Suspended coal washings	SE Wales, U.K.	TSS up to 1530 mg l ⁻¹	Suspended solids	Fish densities of all species except trout declined downstream of discharge; no spawning in main trunk of river; poor growth of trout	Williams & Hareup 1974

n.d. = no data.

marine environment could also reflect a limited research effort in this area. Indeed, most studies on the effects of unburnt coal on aquatic biota have been done in freshwater. Given the extensive compositional heterogeneity of coal and the diversity of weathering and exposure conditions, it seems improbable that coal as a whole can be labelled as 'toxicologically benign'. One general problem of toxicological studies is that toxic effects may occur at various biological and temporal scales, from molecular to ecosystem-wide, or from short-term to chronic, which makes results dependent on the chosen scale of observation. Furthermore, it is often difficult, if not impossible, to separate toxic effects from physically induced stress. For example, several field studies in freshwater have observed reductions in abundance, diversity, growth and reproduction of fishes and macroinvertebrates, which were attributed to physical stress to organisms by suspended coal particles (Herbert & Richards 1963, Williams & Harcup 1974, Scullion & Edwards 1980). In the absence of a direct and definable causal mechanism, chemical toxicity cannot be ruled out.

The following discussion assesses factors that influence the bioavailability of toxicants in coal and then reviews information on the response of aquatic biota to unburnt coal at three different biological levels: (1) cellular, (2) organism and (3) populations and assemblages. Toxicological effects, like physical effects, can act both directly and indirectly. Indirect effects include responses of populations to changes in the abundances of predators, prey or competitors caused by contaminant toxicity (Chapman 2004). Chapman (2004) pointed out that indirect effects are mainly associated with ecosystem function rather than structure but that function is rarely determined in ecological risk assessments.

Factors that influence coal toxicity — dilution, buffering and bioavailability

As the previous discussion has shown, coal contains a plethora of compounds that may be leached upon contact with water and that have the potential to cause toxic effects to aquatic biota. The amount of material that is leachable crucially depends on the coal type, mineral impurities and leaching conditions. For example, low-rank coals, such as brown coals, lignites and sub-bituminous coals, contain a large fraction of mineral matter in pore waters and associated with the organic compounds of the coal macerals, which are often readily leachable with water. In higher rank coals, such as bituminous coals and anthracite, expulsion of moisture and associated dissolved minerals, combined with changes in the chemical structure of the organic matter, tend to remove these non-mineralised inorganics, so that the majority of inorganic impurities are typically present as particulate minerals, discrete from the coal macerals (Ward 2002). Whether potentially toxic components of coal actually exert a negative impact on aquatic biota is determined by their bioavailability and the concentration they attain in the receiving environment. Thus, even though trace metals may be leached from coal piles, their concentrations after dilution by large volumes of water, such as coastal seas, may become negligible compared with other sources. Furthermore, formation of insoluble salts upon contact with sea water, complexation by dissolved organic matter in sea water or in coal leachates (Table 5), adsorption onto particle surfaces, or redox reactions that result in changes of speciation or solubility may render metals leached from coal biologically unavailable. Metals and metalloids that are readily soluble under low pH conditions, such as Cd, Cu, Pb, Zn and As may become insoluble upon contact and dilution of leachates with alkaline sea water. On the other hand, particle-bound metals and metalloids that are soluble under alkaline conditions, such as Cr and Se, may become solubilised upon contact with sea water. With the exception of a study by Duedall et al. (1982) on compacted coal waste material, no attempts have been made to measure the release of metals from coal deposited on the sea floor. Biggs et al. (1984) speculated that the combination of acidic and low redox conditions in sediments could favour metal release from coal and subsequent uptake by benthic organisms but no experimental studies have attempted to test this hypothesis.

Analogous to trace metals, bioavailability and toxicity of organic constituents of coal are largely dependent on the degree to which they are adsorbed onto suspended or deposited particles (Meador et al. 1995). Because only a few studies have documented direct effects of organic compounds in coal (Table 7), it has been argued that coal constitutes essentially an inert contaminant (Chapman et al. 1996a). Coal particles may bind and stabilise PAHs either by incorporation into the solid coal matrix or by strong adsorption to surfaces that probably act quite similarly to activated carbon (Ghosh et al. 2000, 2001, Talley et al. 2002). The presence of coal in aquatic sediments can therefore result in elevated and highly variable PAH and total organic carbon (TOC) concentrations to which equilibrium partition theory does not apply. Thus, even though PAH concentrations in such coal-laced sediments may exceed sediment quality guidelines (Long et al. 1995, ANZECC 2000; see Table 3), the majority are in an unavailable form by being tightly bound to coal particles (Chapman et al. 1996a, Paine et al. 1996). On the other hand, some studies have found benthic organisms to accumulate elevated concentrations of PAHs in the vicinity of coal-coking plants (Uthe & Musial 1986) and coal-fired power stations (Eadie et al. 1982), although the accumulated PAHs may have originated from combustion or coalification processes rather than from unburnt coal. In a Canadian study on environmental impacts of coal mines and coal-fired power stations on sediment quality, Cheam et al. (2000) observed highest toxicity to chironomids and amphipods from sediment that was collected downstream of a colliery and that contained high levels of PAHs relative to other sites sampled (e.g., $4 \mu\text{g g}^{-1}$ naphthalene, $3.4 \mu\text{g g}^{-1}$ by dry weight of phenanthrene and several other PAHs with concentrations between $0.1\text{--}1.0 \mu\text{g g}^{-1}$ by dry weight). In contrast, other invertebrates tested (mayflies and tubificids) showed no or little decrease in survival, growth and reproduction. Because sediment trace metal concentrations in colliery sediment were similar to other sites, the observed toxicity may have been the result of PAH contamination.

Despite the strong partitioning of PAHs to particulate matter, PAHs do desorb from coal into clean water to a measurable extent. For example, several $\mu\text{g l}^{-1}$ of total PAH have been measured in filtered coal leachates (Table 5; Stahl et al. 1984, Fendinger et al. 1989), yet these concentrations generally fall well below water quality guidelines. PAHs potentially derived from coal have been found to accumulate in animal tissues by Paine et al. (1996), who measured elevated PAH levels in the hepatopancreas of *Cancer magister* in an inner harbour area contaminated by coke and pitch particles. However, it is quite likely that the main sources of these accumulated PAHs were modified coal products (coal tar, coke) rather than unburnt coal. Furthermore, despite being measurable in animal tissues, these concentrations did not lead to differences in growth, size, catch-per-unit effort or condition. Interestingly, low molecular weight PAHs (LPAH) dominated in crab hepatopancreas, in contrast to the sediment PAH signature, which was dominated by high molecular weight PAHs. The dominance of LPAHs in crab tissue may indicate that it is mostly the very hydrophobic, high molecular weight PAHs (HPAHs), and not all PAHs in coal, that are unavailable.

It is currently not known what happens to coal-associated contaminants in organism guts, where surfactants and other forms of dissolved organic carbon may enhance solubilisation of particle-bound contaminants (Mayer et al. 1996, 2001, Voparil & Mayer 2000). For example, Mayer et al. (1996) found that digestive fluids of deposit feeding macrofauna, such as the sediment-ingesting lugworm *Arenicola marina*, and the holothurian *Parastichopus californicus*, solubilised up to 200 times more PAHs and up to 2400 times more copper than would be predicted from water-solid partitioning with clean sea water. Consequently, toxic components that are adsorbed to coal particles in water and appear to be unavailable might become available upon ingestion and contact with gut fluid. Because different invertebrate species may differ greatly with regard to specific gut fluid composition and the potential for solubilising organic contaminants or trace metals (Mayer et al. 2001), it is currently impossible to assess accurately the risk of dietary exposure to coal-bound contaminants for invertebrates inhabiting coal-contaminated sediments.

The likelihood of acidic and saline coal leachates affecting terrestrial and aquatic communities will depend greatly on the dilution and buffering capacity of the receiving environment. In light of the enormous buffering capacity of sea water, the changes in pH and salinity in the marine environment due to coal leachates are likely to be undetectable. Impacts of high acidity and salinity will therefore be restricted to the terrestrial environment surrounding, and the culverts and any streams draining, the coal storage and handling facilities. It seems probable that if acidic coal pile leachate enriched in dissolved metals comes into contact with sea water, the ensuing changes in pH and ionic strength will facilitate flocculation and precipitation of metals and other previously dissolved or suspended components (Salomons & Förstner 1984). This material is likely to settle out at the point where it enters the marine environment or in nearby sheltered areas. Thus, accumulation of particle-bound metals could potentially be predicted for the mixing zone of leachates in estuaries (e.g., Williamson & Morrisey 2000, Chapman & Wang 2001).

Effects at the cellular level

Coal dust has been shown to affect PAH-metabolising enzymes in livers of chinook salmon (*Oncorhynchus tshawytscha*) (Campbell & Devlin 1997). Exposure of fish to coal suspensions of 60–500 mg l⁻¹ for 8 days increased expression of CYP1A1, an enzyme of the hepatic cytochrome P450 system, which plays a central role in the cellular detoxification of xenobiotic compounds such as PAHs. Furthermore, coal dust induced the expression of a ribosomal protein, L5, whose function, however, remains unknown. The induction of CYP1A1 has been used as a sensitive biomarker for the exposure of animals to organic contaminants such as PAHs. Expression of this gene does not denote toxicity or cellular damage but rather a cellular biochemical response to toxic compounds. However, the activation of its cytochrome P450 system represents a double-edged sword for an animal from a toxicological standpoint. On the one hand, CYP1A1 converts hydrophobic compounds to more water-soluble derivatives that may be excreted whereas, on the other hand, while doing so, it may generate more toxic, mutagenic by-products.

Whereas the previous study observed a clear cellular response of fishes to coal leachates, Carlson et al. (1979) found no change in liver parameters in rainbow trout (*Oncorhynchus mykiss gairdneri*) exposed for 28 days to aqueous leachates of sub-bituminous coal from Montana (U.S.): liver weights, microsomal protein and DNA content, and aryl hydrocarbon hydroxylase activity (AHH) varied similarly over 28 days of exposure compared with control fish. Exposure of trout to steam distillates of coal for 21 days resulted in no PAH bioaccumulation nor significant increases in hepatic mixed function oxidase (MFO) activity (i.e., AHH activity and cytochrome P450 protein content) compared with controls. The reason for this may lie in differences in the type of coal used or the method of preparing leachates. Whereas Carlson et al. (1979) centrifuged their leachates, Campbell & Devlin (1997) merely allowed coal dust slurries to settle, and noted that the final leachate contained a significant amount of non-settleable solids. Interestingly, Carlson et al. (1979) observed 100% mortality within 96 h when exposing fathead minnows to uncentrifuged leachates. This provides further evidence that particulate coal has a greater likelihood of causing adverse biological effects than leachates, even though it is difficult to state whether these adverse effects are due to toxicity or physical irritation. Evidence from the medical literature suggests that PAHs contained in coal dust are not capable of crossing the skin barrier of mammals and are not eluted by pig lung homogenate or human gastric juice (Foa et al. 1998).

Siboni et al. (2004) found that the presence of coal in coastal sediment increased cadmium concentrations in tissues of predatory gastropods. This effect was accompanied by increased metallothionein levels in tissues, increased paracellular permeability, and impeded lysosomal accumulation of lipophilic substances. These sublethal effects were interpreted as evidence for cryptic

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cellular damage, even though no increased mortalities were evident in the field or in laboratory manipulations with coal-amended sediment. Metallothioneins play an important role in the detoxification of metals in invertebrates by converting dissolved metals into an inert, particulate form. Their induction may have been triggered by cadmium present in the coal. Increased permeability of membranes, measurable as incorporation of fluorescein dye, is thought to reflect injury of epithelia and their intercellular junctions, which may have resulted from contact with Cd from the coal substrate. Paracellular fluorescence was especially enhanced in foot tissue, which would have experienced the highest contact time with the coal. Lower lysosomal accumulation of lipid-soluble cationic markers, such as neutral red, has been used as a non-specific indicator of cellular damage.

Effects at the organism level

Whereas a number of studies, discussed in the physical effects section above, have shown direct adverse effects of coal particulates or leachates on survival, growth and reproduction of fishes and a variety of aquatic invertebrates, there have been only a few studies investigating potential toxic effects of unburnt coal to marine organisms. The majority of these concluded that unburnt coal or its leachates are not acutely or chronically toxic. For example, Bender et al. (1987) reported that 28 days of exposure of eastern oysters (*Crassostrea virginica*) to suspensions containing 1 and 10 mg l⁻¹ coal dust did not affect survival or significantly alter shell growth or pumping activity compared with controls nor did coal exposure lead to significant bioaccumulation of PAHs in oyster tissues. These authors considered that because coal dust was observed in the faeces of the oysters, this indicated that toxic compounds contained in coal were unavailable. This conclusion was also supported by the fact that no differences in PAH concentrations were detected between control water and coal-water slurries. However, while Bender et al. (1987) convincingly demonstrated no acute toxicity, in terms of mortality, there may have been a major bias in their interpretation of the growth and accumulation data. The problem lies with the very high background concentrations of PAHs in control estuarine water (York River, U.S.) and oyster tissues (from Rappahannock River), which may have 'swamped' PAH contributions from coal during the actual experiment. For example, phenanthrene and fluorene concentrations in York River water were 0.6 µg l⁻¹ and 0.3 µg l⁻¹, respectively, and total PAH concentrations in control oyster tissues ranged between 1.38–2.27 µg g⁻¹. In comparison, typical tissue concentrations in oysters from mildly contaminated urban estuaries range between 20–100 ng g⁻¹ (G. Olsen, NIWA, personal communication) and the median tissue ΣPAH concentration of shellfish monitored under the U.S. NOAA National Status and Trends Mussel Watch Program (seven bivalve species) is 230 ng g⁻¹ (O'Connor 2002). This result suggests that the oysters from the Rappahannock River used by Bender et al. (1987) may have been highly contaminated by PAHs prior to exposure to coal. Alternatively, the oysters could have accumulated the majority of their PAH tissue burden from York River water over the 28-day exposure period or during the 3-wk acclimation period. Comparing ratios of individual PAHs to the sum of methylphenanthrenes (the dominant PAHs in the coal used) revealed that tissue from coal-exposed, depurated oysters had similar PAH ratios to coal once the high background PAH of control oysters was subtracted. This comparison indicates, in contrast to the authors' interpretation, that some PAHs may have indeed accumulated from coal. Bender et al. (1987) also compared weekly shell length increments in oysters exposed to 0, 1 and 10 mg l⁻¹ suspensions of coal dust and concluded there was no significant difference in shell growth among treatments on a given sampling date for the three test populations. However, it should be noted that numerical differences in growth were indeed observed and that the lack of statistical difference may have been in part due to large variance within treatments. Furthermore, when their growth data are re-analysed by regressing weekly shell growth vs. days of exposure, the 10 mg l⁻¹ treatment had the lowest slope and the control the

highest, suggestive of a treatment effect. Nonetheless, the conclusion remains valid that neither mortality nor markedly reduced growth was observable upon exposure to coal dust, suggesting limited toxicity of coal dust.

Hillaby's (1981) finding of no mortality in crabs exposed to coal dust in aquaria, mentioned above, suggested the absence of a direct acute toxic effect. However, as in many bioassay studies there was large variability among individuals within treatments, which may have resulted in low statistical power to detect effects. In a study of freshwater fishes, Carlson et al. (1979) observed that rainbow trout and fathead minnows (*Pimephales promelas*) exposed to centrifuged coal leachates for 3–24 wk showed no increased mortality, no diminished growth and no pronounced PAH bioaccumulation. However, a lower spawning success was observed in fathead minnows during 2–4 wk exposures. Furthermore, 100% mortality was observed in fishes that were exposed to uncentrifuged coal leachates containing suspended coal. In a study in Kachemak Bay, Alaska, deposit feeding clams (*Macoma balthica*) accumulated an array of hydrocarbons suggesting coal as a source, whereas suspension-feeding mussels (*Mytilus edulis*) and grazing limpets (*Collisella pelta*) accumulated hydrocarbons in their tissues that suggested petroleum origin (Shaw & Wiggs 1980). It should be noted, however, that the shellfish were not depurated before analysis of hydrocarbons and consequently the contaminants may only have been present in the digestive tract and not assimilated into tissues. Paine et al. (1996) found no sediment toxicity to amphipods (*Rhepoxynius abronius*) and sand dollars (*Dendraster excentricus*) from sediments enriched in coke particles or pitch globules in the vicinity of an aluminium smelting plant. While this study did not focus on unburnt coal but, rather, on thermochemically modified coal products, the absence of observable mortality also acts to rule out any potential toxicity from unburnt coal that may have been present in these industrially impacted sediments.

Some stimulatory effects of coal have been reported. For example, Gerhart et al. (1980) observed increased growth and higher chlorophyll a concentration in freshwater algal periphyton exposed to coal leachates in the laboratory (coal concentration: 3.2% and 17% v/v). The growth stimulation in coal treatments occurred regardless of whether leachates were centrifuged or not and the stimulation was not attributable to differences in total phosphorus concentrations. In the field, however, filtered coal leachates inhibited the growth of test algae, which may have been a result of differences in the algal species used or container effects (field exposures used glass bottles, whereas laboratory experiments used plastic containers). Coal distillates, produced to mimic the effect of spontaneous heating in coal storage piles, inhibited growth of algae in a dose-dependent manner, which was attributed to the presence of volatile organic compounds (alkanes, alkylbenzenes and alkyl-naphthalenes). When distillates were bubbled to remove volatile organic compounds, growth stimulation was frequently observed.

Effects at the levels of populations and assemblages

The study by Gerhart et al. (1980) also demonstrated general stimulation of algal growth and shifts in algal species composition (primarily diatoms) upon exposure of periphyton assemblages to coal leachates in the laboratory. In closed, unaerated containers, algal assemblages showed inhibited growth. Field mesocosms with coal distillates resulted in greater algal growth and chlorophyll a, while reducing zooplankton biomass. The main stressors were believed to be volatile organic compounds present in distillates.

As far as could be ascertained, there have been no manipulative experimental studies of effects of coal on populations or assemblages of marine or estuarine organisms *in situ*. Consequently, evidence of effects has to rely on correlational studies in which abundances and diversities of organisms are compared among areas containing different amounts of coal. In common with all correlational studies, these are subject to confounding by covariation among factors, measured or

unmeasured, that may potentially affect the biota, other than the presence of coal. In the case of coal, covariables are likely to include patterns of distribution of natural sediments that are subject to the same sources and mechanisms of transport as the coal. Coal is often dumped or otherwise enters the marine environment in combination with other materials, notably mine spoil and fly ash, further confounding identification of its effects. Surprisingly, and in contrast with freshwater environments, even these types of studies are scarce and relate to only a few geographical areas.

Zhang et al. (1995) refer in the abstract of their paper to effects of coal dust on marine organisms, including photosynthetic effects and changes in sediment quality but no details are given (the main text is in Mandarin). Sampling of sediments around industrialised, coal-mining settlements in Svalbard found elevated concentrations of a range of organic contaminants (Holte et al. 1996). These included PAH, which were assumed to derive from local coal stores via terrestrial runoff. The faunal diversities of these areas were low (based on values of the Shannon-Weaver index), but much of this effect was attributed to inputs of fine particulates from glaciers. The variety of sources of particulates and chemical contaminants to these environments (coal piles, rubbish dumps, industrial activity, glacial runoff) prevented any clear identification of effects of the presence of coal. Paine et al. (1996) observed no reduced species richness of benthic infauna resulting from the presence of coal, coke and pitch particles in sediments, in contrast to the generally observed reduction in diversity of biological assemblages along contamination gradients (Pearson & Rosenberg 1978, Maurer et al. 1993, Chapman et al. 1996b).

Effects of colliery waste on the northeast coast of England

The most extensive set of studies of effects of coal and colliery waste on benthic assemblages is from the coast of northeast England. Colliery waste, together with fly ash from power stations and dredge spoil from the ports of Blyth, Newcastle and Sunderland, has been tipped onto the shore and dumped off the coast of northeast England since the beginning of the 20th century (Eagle et al. 1979). Much of this dumping began before statutory controls were in place to protect the environment and monitoring of the effects of the dumped material did not begin until 1975 (Eagle et al. 1979). Dumping of colliery waste at sea has now ceased and only one coastal dumpsite is still operating (this operation is licensed as beach nourishment under the coastal defence strategy for the area; Barnes & Frid 1999). The sea bed along this stretch of coast consists of fine sand in depths of <10 m with gravel and sand among outcrops of rock further offshore (Norton 1985).

The colliery waste consists of 'minestone', comprised mainly of sandstones and shales from which coal has been separated by a water-based gravity process, and tailings, which represent the fine fractions of the waste from which coal has been extracted by an oil-water flotation method and which have a particle size <0.5 mm (Limpenny et al. 1992). Nearly 80% of the waste was <4 mm in diameter at the time of dumping, but the material was abraded by water movement to particles <1 mm. Colliery waste contains 25–30% coal (determined by loss on combustion: Norton 1985, Hyslop et al. 1997). Trace metal concentrations in the colliery waste were generally lower than in coal from the same area, suggesting that the coal, rather than the associated minerals, was the more important potential source of these contaminants (Eagle et al. 1979, Hyslop et al. 1997). The previously mentioned observation that colliery waste washed up on local beaches contained much lower concentrations of trace metals than either the coal or the waste prior to dumping suggests that metals from colliery waste may be available to animals and plants living on the sea bed in the area where the waste is dumped, and may exert a toxic effect on them. There do not appear to be any measurements of amounts of toxic organic compounds in the waste but these are presumably also high (Hyslop et al. 1997).

Eagle et al. (1979) examined the effects of colliery and other wastes on the sediments and benthic organisms at the dumpsites and found that seabed sediments contained >2% of coal along

a >11 km stretch of coast. Concentrations of trace metals (particularly Cr, Cu, Ni, Pb and Zn) in sediments were high in some areas and corresponded with concentrations of waste. Concentrations of these metals at some sites exceeded sediment quality guidelines at which biological effects are likely to occur (Table 6). A later study (Limpenny et al. 1992) reported that concentrations of trace metals in sediments at the dumpsites were “within the normal range for the north-east coast”, but this is a region of historical contamination from discharges from heavy industrial and urban areas, in addition to dumping of waste at sea. Concentrations of metals in the sediment offshore from beach dumpsites only rarely exceeded the sediment quality guidelines at which biological effects would be expected to occur (Table 6) in the case of Cu, Hg, Pb, Ni and Zn. Limpenny et al. (1992) found more than 50% of coal in sediments around one of the beach disposal sites, and Hyslop et al. (1997) reported 11–27% coal in beach sediments (measured as combustible material after removal of biologically available organic matter with hydrogen peroxide). It must be pointed out, however, that the generally high levels of sediment contamination in this area make it impossible to say that the concentrations of contaminants reported were due to the dumping of colliery waste. The bioavailability of the contaminants, and the associated probability of toxic effects, is also unknown. There was no evidence for accumulation of trace metals by commercially harvested species (various decapod crustaceans, mussels and finfishes) taken from the inshore zone near the dumpsites (Norton 1985).

Eagle et al. (1979) sampled benthic animals in 1975 and 1976 at two dumpsites, one of which had received only colliery waste and small amounts of dredge spoil (i.e., no fly ash). Overall, the surveys indicated that the effects of dumping on the benthos were limited to the area where the material initially settled on the bed. In this region, the benthos was severely depleted in both abundance and number of species. Effects on surrounding areas could not be shown even though the sediments contained particles of waste as a result of sediment transport by water currents.

Johnson & Frid (1995) and Barnes & Frid (1999) examined recovery of the benthic fauna at sites where dumping colliery waste had ceased. The earlier study compared three former dumpsites and an undumped control (or reference) site and concluded that the benthic communities were still perturbed 7.5–12.5 yr after dumping ceased. The number of species was largest at the control site (39 vs. 33–35) but Shannon-Weaver diversity was highest at two of the former dumpsites. The latter observation suggested that the level of disturbance caused by dumping favoured diversity, perhaps by preventing competitively dominant species from excluding others. Unfortunately, this study suffered from possible weaknesses of design. These included the fact that there was only one control site, which may have differed from the dumpsites in the nature of its fauna for a variety of reasons other than the fact that it had not received waste. There was also very limited replication of sampling because the fauna at each of the study sites was characterised from only three replicate grab samples. Furthermore, sediment transport in the area is in a predominantly north-south direction, which also corresponds to the relative positions of sites of increasing time since dumping ceased. Consequently, these sites may have continued to receive inputs of colliery and other wastes even though direct inputs from dumping had ceased.

Barnes & Frid's (1999) study compared the benthic faunas at six sites, including an active dumpsite, undumped controls and sites that had not been dumped for 3–41 yr. They found differences in the composition of the fauna between dumped and undumped sites, and an apparent degree of recovery at those sites where dumping had ceased, though these differences were not clear-cut. Comparisons of the grain-size distribution and organic-matter content of sediments with the composition of the faunal communities present suggested that “dumping of colliery spoil was an important factor determining community composition, rather than the presence of coal per se”. The authors also stated, however, that “While influenced by particle size distribution, a greater effect results from the proportion of coal waste still present... This implies that the greater mobility of the lighter coal particles — and the resulting lower stability of the substratum — are responsible

for the altered communities". Differences between the benthic communities at dumped and undumped sites were, therefore, attributed to the destabilising effect of coal particles on the sediment, making it physically less suitable as a habitat for animals, rather than exerting a toxic effect or smothering resident organisms. Without experimental studies, it is not possible to separate the first two effects, but the slow recovery of these sites is consistent with the suggestion that deposition of material was not the sole cause of observed differences.

In a study of effects of colliery waste on intertidal habitats (as opposed to the subtidal areas studied by Eagle et al. (1979), Johnson & Frid (1995) and Barnes & Frid (1999)), Hyslop et al. (1997) found that colliery waste had an adverse effect on the number of species present in the low-shore and mid-shore levels of sandy beaches. A maximum of eight species was found on uncontaminated shores, compared with a maximum of two species on heavily contaminated shores. There were no detectable differences at the high-shore level. Similarly, uncontaminated rocky shores contained 12–15 species of macroalgae, whereas contaminated shores contained only 5–8. The diversity of animal communities on rocky shores (measured as the Shannon-Weaver index) was, however, sometimes greater on contaminated shores, although the number of species (usually 7–10) did not differ significantly among shores. Hyslop et al. (1997) concluded that reduced numbers of faunal species on sandy shores were due to decreased stability of the sediment resulting from the presence of colliery waste, which was made up of larger, more angular particles than the natural sediment. Concentrations of metals in waste washed up on the shore were much lower than in the unweathered waste. This finding suggested that a large proportion of the metals were leached out of the waste on the sea bed, making it unlikely that the effects on the intertidal fauna were due to the presence of these toxicants.

One of the species whose abundances correlated inversely with the amount of colliery waste in the sediment was the burrowing, deposit-feeding polychaete *Arenicola marina*. By comparing the sediments and the gut contents and faecal material of worms from a heavily coal-contaminated site with those from a lightly contaminated site, Hyslop & Davies (1999) found that the worms at the heavily contaminated site apparently fed selectively on sand grains and rejected coal particles. Selection may have been made on the basis of grain size, and smaller particles of coal were found in the guts and faeces of worms from both sites. Worms from both sites showed a reluctance to burrow into their native sediment when a 2-mm thick surface layer of colliery waste had been placed on the surface, compared with native sediment without the added layer. Despite the rejection of larger particles (mainly coal) by worms at the more contaminated site, there was no evidence to suggest that lack of suitable sediment for feeding was the cause of the observed reduction in abundance of *A. marina* at heavily contaminated sites. Instability of sediments due to the presence of coal may have been a more important factor.

The species of macroalgae absent from coal-contaminated rocky shores were low-biomass, less robust types. Given the fact that toxic effects from coal appeared unlikely, Hyslop et al. (1997) concluded that effects on these species were, therefore, due to abrasion by the suspended particles of waste in the water. This conclusion was confirmed by laboratory experiments in which the individuals of the green alga *Ulva lactuca* exposed for 8 days to colliery waste in containers that were shaken to simulate abrasion lost weight relative to controls without waste (Hyslop & Davies 1998), as discussed previously. Species with thick, waxy cuticles were not markedly affected by the presence of waste. The laboratory studies also showed that over 60 days, *U. lactuca* grew more in the presence of colliery waste in unshaken conditions than in coal-free controls. Hyslop & Davies (1998) suggested that enhanced uptake of inorganic nutrients in the presence of the waste may have been responsible for this increased growth and perhaps accounted for the relatively large abundance of *U. lactuca* in rock pools at their field sites. No evidence of enhanced uptake was given, however, and possible effects on field populations of algae were not tested experimentally.

In summary, the dumping of colliery waste appeared to reduce the species diversity and abundance of intertidal and subtidal assemblages of animals and plants. Although the waste contained significant amounts of trace metals, the effects on benthic assemblages were generally attributed to physical changes (destabilisation) in the sediments and the abrasive action of suspended particles of waste. These physical effects were generally localised around the area of input of the coal. It is important to note that assemblages at dumpsites were compared with nearby controls that themselves contained relatively high concentrations of contaminants in their sediments. Any effects of this background contamination may have obscured toxic effects of the waste. The fact that the waste contained other materials than coal (sandstones and shales), which also have potential physical or chemical effects on biota, confounds interpretation of the observed biological effects as effects of the coal fraction. Furthermore, the spatial and temporal scales of sampling were limited in all of the studies reviewed above. None, for example, sampled on more than two occasions, and the level of spatial replication was generally limited. The naturally large spatial and temporal variability of benthic assemblages makes it difficult to detect environmental impacts without considerable spatial and temporal replication, and clear attribution of cause may not be possible using the correlative approach in situations where there are multiple sources of impact, such as the coast of northeast England. Consequently, the apparent absence of clear toxic effects associated with coal off the northeast coast of England does not rule out such an effect in areas with less background contamination.

Discussion

Assessment of the marine environmental risks of coal

The general opinion, expressed in many of the studies cited above, is that coal may present a physical hazard in the marine environment when present in sufficient quantities, but not a chemical one. The physical effects are largely common to any form of suspended, particulate matter, principally smothering, abrasion, obstruction and damage to respiratory and feeding organs, and effects due to reduction in the quality and quantity of light (Figure 2). Nevertheless, there are some lines of evidence, detailed in preceding sections, supporting the hypothesis that contaminants present in coal can also exert adverse effects on organisms. As with most assessments of risk of adverse ecological effects at higher levels of biological organisation caused by contaminant toxicity, a weight-of-evidence approach is essential (Chapman et al. 2002).

The approach chosen in this discussion is a tiered assessment, analogous to those used to assess environmental risks associated with, for example, disposal of contaminated sediments at sea (e.g., the dredge-spoil disposal guidelines given by U.S. EPA/U.S. ACE 1991, 1998, Chevrier & Topping 1998, Environment Australia 2002). This type of approach could also be used on a case-by-case basis in environmental risk assessments for particular types of coal. Rigorous evidence for or against toxic effects of the type of coal in question at any tier would obviate the need for testing at subsequent tiers. Where conclusive evidence is not provided by lower tiers, higher-tier testing may be required.

In the first tier, information on the chemical nature of the different types of coal is used to assess the toxic potential of the contaminants present. There is no doubt that such contaminants are present in all types of coal, as shown by the studies summarised in Tables 2 and 3. For different types of coal, however, the types and combinations of contaminants may vary, together with other chemical and physical properties, and potential biological effects will therefore also vary. Taking the coal dumped off the northeast coast of England as an example, where the concentration of coal in the sediments at the dump sites ranged from 2–27% by dry weight, and using the concentrations of contaminants in the coal cited by Hyslop et al. (1997), the resulting total concentrations of selected trace metals would be Cr 4–50 $\mu\text{g g}^{-1}$, Cu 5–62 $\mu\text{g g}^{-1}$, Ni 3.5–44 $\mu\text{g g}^{-1}$ and Pb 2.4–30 $\mu\text{g g}^{-1}$.

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Except for Ni, these values are all below sediment quality guidelines (ANZECC 2000) but in conditions where the proportion of coal in the sediment is larger (say, 50%), other metals would also exceed guideline values. Under these conditions, guidelines for some organic toxicants would also be exceeded for at least some types of coal (see Tables 2 and 3). The critical question, of course, is how bioavailable these contaminants are and under what environmental conditions they are present. Furthermore, at these concentrations of coal in sediments, physical effects may have more immediate impact on the biota than any potential toxic effect.

An indication of bioavailability may be provided by information on the quality of runoff from coal stockpiles or laboratory-produced leachates, which show that dissolution of potentially toxic contaminants may occur (see Tables 4 and 5). In an environmental risk assessment for a particular type of coal, this could be determined using elutriate tests (e.g., U.S. EPA/U.S. ACE 1991) or porewater extracts from coal-contaminated sediments. In risk assessment terms, environmental concentrations of the contaminants may be compared with water or sediment quality guidelines, such as those of ANZECC (2000). As a quick-screening test for potentially toxic leachates, it may be worthwhile to determine leachate pH prior to any comprehensive chemical analysis, because low pH, indicative of sulphide oxidation, generally correlates with high concentrations of sulphate and trace metals (Table 4). There is currently little evidence that dissolved PAH concentrations in coal leachates reach levels that exceed water quality guidelines. Coal therefore does not appear to be a significant source of dissolved PAHs. A critical area of uncertainty in terms of bioavailability of contaminants from coal is the degree to which they may be mobilised from particles during passage through the guts of animals. This uncertainty applies especially to deposit-feeders but because, for example, coal particles were found in the guts of fishes exposed to coal in suspension (Gerhart et al. 1981), other animals might also accumulate contaminants by this route.

The second tier involves assessment of evidence that inorganic and organic contaminants derived from coal may be bioaccumulated. While bioaccumulation does not necessarily indicate an adverse biological effect (Chapman 1997), it does provide unequivocal evidence that the accumulated contaminants are present in a bioavailable form. That bioaccumulation of contaminants from coal can occur is demonstrated by the uptake of Cd by the predatory gastropod *Hexaplex trunculus* in the presence of coal particles in the laboratory and field (Siboni et al. 2004). Shaw & Wiggs (1980) suggested that the bivalve *Macoma balthica* had accumulated hydrocarbons from coal-contaminated sediments in Alaska (although they did not clearly distinguish ingestion from assimilation). In a risk-assessment context, this tier might involve laboratory studies of animals exposed to coal under environmentally realistic conditions and routes of exposure (e.g., deposited or suspended coal particles), and incorporate comparisons with appropriate reference sediments. Alternatively, it might follow defined protocols, such as those in U.S. EPA/U.S. ACE (1991), which include factors to assess when interpreting bioaccumulation values in excess of those in reference sediments. These factors include the phylogenetic diversity of the species in which bioaccumulation occurs, and the range of contaminants that are bioaccumulated. Acute and chronic (steady state) bioaccumulation tests might be included, proceeding from the former to the latter as indicated.

The third tier reviews evidence of biological effects of accumulated contaminants derived from coal at the range of levels of biological organisation from the cell to the individual. At one extreme of this continuum, there is evidence that contaminants from coal can have genotoxic effects in fishes (Campbell & Devlin 1997). The severity of pneumoconiosis, a respiratory disease prevalent among coal mine workers caused by quartz particles in coal mine dust, appears to depend not only on inorganic coal constituents but also on organic, cytotoxic coal components, such as phenols and cresols, which are readily leachable by biomimetic physiological fluids (Schulz, 1997). At this level of organisation, it may be reasonable to extrapolate evidence from a range of non-marine to marine organisms, including the range of toxicological data for effects of coal on humans, if there is reason to believe that the mechanisms involved are common to both groups.

Whereas there are a few documented subcellular effects to coal exposure, current evidence suggests that coal poses only a subtle toxic hazard, if at all, to biota in the marine environment. The few studies that have observed pronounced toxic effects of coal and coal leachates were carried out in freshwater environments, where effects related to low pH and high metal concentrations seemed to dominate. Furthermore, acidity and high metal content affect only a subset of coals, namely sulphur-rich types under oxidising conditions. In the marine environment, pH-related effects from coal are likely to be negligible due to the large buffering capacity of sea water, and elevated dissolved metals concentrations are unlikely to occur due to the much greater dilution and mixing potential. Dissolved concentrations of organic compounds, such as PAHs, need to be quite high, typically well beyond $10 \mu\text{g l}^{-1}$, to exceed toxic thresholds in current water-quality guidelines. Such concentrations are unlikely to occur on large scales in the pelagic environment. Thus, toxic effects to planktonic and nektonic organisms, resulting from dissolved toxicants leached from coal, are not very likely in the ocean.

There is, however, a greater probability for metals and PAHs to attain toxic threshold concentrations in pore waters of sediment. The lower exchange rates and limited water volume could allow dissolved compounds to concentrate to high levels before being flushed out, but no measurements of porewater PAH concentrations of coal-containing sediments have been made. Furthermore, wherever coal occurs in small particle size, there is the potential for ingestion by deposit-feeding biota. So far, only a few studies have examined toxicity or bioaccumulation of chemical components of coal and, from the few cases where ingestion was observed, there is little evidence for pronounced dietary uptake of contaminants. This suggests that the majority of metals and hydrocarbons contained in coal are, indeed, present in forms unable to pass through the gut membranes or other epithelia. Digestive bioavailability is likely to be related to the digestive chemistry of the ingesting organism and will depend on factors such as enzyme activity, surfactant concentration and dissolved organic carbon concentration. However, even organisms with relatively aggressive gut conditions, such as mammals or deposit-feeding polychaetes, seem to leach only very small amounts of toxic compounds from coal.

The fourth tier of evidence concerns alterations to populations and assemblages of organisms *in situ*. Here, however, there is very little suitable evidence available at present. Evidence of chemical effects from existing studies, such as those of the northeast coast of England, are confounded by the presence of stressors other than unburnt coal, and by physical effects of the coal itself.

In summary, while the presence of a wide variety of contaminants in most types of coal gives cause for concern, in many situations their low aqueous extractability and bioavailability appears to safeguard against toxic effects. Nevertheless, under certain circumstances and with some types of coal, leaching of toxic components has been demonstrated and these may be of greater biological significance. In addition, very little is known about the effects of ingestion and digestion of particulate coal on the solubility of contaminants. This uncertainty concerning the toxicological effects of coal is in contrast to the demonstrable physical effects. In many cases, the toxic effects may affect biota at lower environmental concentrations of coal than those at which physical effects might become apparent. In many situations requiring an assessment of potential effects of coal, all of these factors may need to be considered in a weight-of-evidence approach.

Mitigating environmental risks from coal

This brief overview is intended only to provide a context for the discussion of marine environmental risks from coal. It is not intended to be exhaustive and certainly does not pretend to any specialist knowledge of the environmental management of coal handling facilities.

The principal sources of unburnt coal to marine environments are the preparation of coal to remove impurities (involving various washing and flotation techniques), disposal of mine wastes

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at sea and the storage and transport of coal, particularly at ports (Figure 1). This coal contamination may enter the marine environment directly, as in the case of coastal coal preparation plants or loading and unloading at sea ports, or via rivers and estuaries. The efficiency of separation and recovery of coal from mine wastes is likely to have been optimised for economic reasons (i.e., minimising wastage of coal). In terms of potentially large, direct inputs to marine environments, losses during storage and transport probably offer the greatest scope for cost-effective mitigation.

Mechanisms of contamination during storage and loading/unloading include runoff from storage and handling areas, airborne transport of dust from stockpiles, slumping and collapse of stockpiles during rainfall, spillage from conveyor belts carrying coal to and from storage areas and spillage at the point of transfer from conveyor to ship or vice versa (Sydor & Stortz 1980). The scale and relative importance of these inputs will depend on factors such as the design of the loading and storage facilities, local climate and the volume and type of coal handled. Appropriate mitigation measures will, therefore, often be site-specific and many modern coal-handling facilities will have environmental management systems in place to assess and manage these environmental risks. General methods of mitigating inputs of coal and leachates to the marine environment are discussed in the following paragraphs.

Contamination from wind-blown coal dust can be considerable. Sydor & Stortz (1980) estimated that 20 t of dust entered Lake Superior each year from a coal shipment facility, 4 t being deposited after winter ice melted. Mitigation of inputs of dust to the environment is achieved by: reducing the amount of time that coal spends in storage areas; covering storage areas (often impractical for large areas); ensuring adequate water content at unloading areas; damping of stockpiles and transfer points with water; managing the stockpiling machinery to minimise the exposure of coal to winds during operation (e.g., maintaining minimal height of stacking booms); compacting of coal in stockpiles; managing stockpile height to minimise exposure to wind, particularly at times of year when strong winds are prevalent; using weather forecast information to respond to predicted adverse weather conditions; cleaning up spillages regularly; and using real-time dust monitoring data to reactively manage dust controls (Ports Corporation of Queensland 2002, World Coal Institute 2004). Sydor & Stortz (1980) observed that dumping of coal from an overhead conveyor generated less dust and caused less down-wind deposition than did grooming operations on stockpiles by caterpillar tractors. Water is also used to suppress dust at transfer points between conveyors and ships, and elevated conveyors and transfer areas can be enclosed to minimise the exposure of coal to winds. In addition to reducing dust generation, enclosing conveyors will also reduce spillage of coal due to vibration and overflow at points where coal is transferred from one conveyor to another.

Use of water to suppress dust adds to rainfall runoff as a mechanism for the generation of leachates from stockpiles. Stockpile areas can be bunded to control runoff and prevent it from entering natural waterways. Runoff can then be directed through drainage systems fitted with sumps and settling ponds to reduce the amount of particulate coal in suspension (Ports Corporation of Queensland 2002, World Coal Institute 2004). Removal of particulates will help trap the majority of PAHs and trace metals before reaching the marine environment. Design of bunds and drainage systems has to take into account the volume of coal to be stored in the future (which affects the volume of the bunded area available for water retention) and the intensity and duration of local rainfall. The quality of the leachate once particulates have settled out will depend very strongly on chemical and physical characteristics of the coals, as discussed above. This, in turn, will determine the level of treatment required before runoff is allowed to enter local waterways or the sea. Water is likely to be recycled for use in dust suppression for environmental and economic reasons (Ports Corporation of Queensland 2002, World Coal Institute 2004). This may increase concentrations of dissolved contaminants over time and lead to potentially more severe environmental effects if runoff accidentally enters natural waterways due, for example, to overflows during intense rainfall, or increase the level of treatment required before intentional discharge. Because concentrations of

contaminants tend to be highest in the 'first flush' of leachate from coal stockpiles during rainfall events (Davis & Boegly 1981a), it may be possible to divert and treat only this component.

Another option, especially for long-term storage, is to treat the coal pile with surfactants and water-repellent compounds that hinder the entry of water into the pile. Cationic surfactants are generally favoured because of their superior wetting ability (Tien & Kim 1997). While surfactants perform well in reducing dust levels, it is conceivable, albeit speculative, that they negatively affect leachate quality by enhancing solubilisation of particle-bound contaminants. The drawbacks of sealing surfaces of storage piles are that the geotechnical properties of the coal are changed and that the coal becomes more difficult to excavate and process. A third option, aimed at minimising the generation of acid leachates and increased metal concentrations in runoff from sulphur-rich coal piles, is to prevent or counteract the oxidation of pyrite and other sulphur-containing compounds by bacteria. One successful approach is to promote the growth of sulphate-reducing bacteria in coal piles (Kim et al. 1999), which has been shown to neutralise acids and reduce dissolved concentrations by 99% for Cd, Cu and Zn, and 87% for Ni. Addition of caustic soda (NaOH) to leachates to neutralise acids is common practice, and it has been suggested that the deliberate storage of different types of coals conjointly (e.g., sulphur-poor coal downstream of sulphur-rich coal) might help neutralise or ameliorate adverse leachate properties (Coward et al. 1978).

Slumping of stockpiles is caused by saturation of the piles with water, for example during heavy rain. The risk of slumping varies with the type of coal and its existing water content. It can be minimised by covering and compacting the stockpile, minimising its height and providing drainage underneath it. Bunding of stockpile areas is used to contain slumps if they do occur. Location of stockpiles as far as practical from the water line will reduce impacts from wind-blown dust and slumping.

Directions for future research

Bioavailability of contaminants from coal

From the foregoing sections it is clear that coal is a heterogeneous mineral whose chemical properties are highly variable. This means that, unlike studies of its physical effects, which relate primarily to particle size, density and concentrations, it is impossible to define a 'representative' coal sample from a chemical standpoint. Thus, even though some studies have suggested low bioavailability of toxic compounds for a number of coal samples, these data are not sufficient to reject the hypothesis that some coals are toxic to aquatic organisms. If one adopts the precautionary principle, any coal sample that is known to contain high concentrations of metals and organic compounds is potentially capable of releasing these to the environment. Leaching studies and bioassays are required to prove otherwise.

It may be safe to say that wherever particulate coal is present in the aquatic environment, effects from increased suspended solids concentrations and modified benthic substrata are likely to dominate over toxic chemical effects. For this reason, it is recommended that any studies on chemical toxic effects of coal should begin with particle-free (i.e., filtered or centrifuged) leachates of coal. This approach would avoid confounding toxic effects with physical effects. The procedures for preparing a leachate whose properties are representative of those encountered during coal pile storage or upon deposition of coal into the aquatic environment will depend on local conditions and need to be carefully evaluated. For example, if coal dust is ultimately discharged into the marine environment and toxic effects of this discharge are to be evaluated, leaching studies need to be conducted with sea water rather than freshwater or distilled water. To investigate whether toxic compounds in coal particles may be assimilated by ingestion, biomimetic leaching experiments that simulate the conditions in the digestive tract of organisms might be useful. Such experiments