



Grounded or submerged bulk carrier: The potential for leaching of coal trace elements to seawater

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ABSTRACT

This study investigates the potential for leaching of coal trace elements to seawater from a grounded bulk carrier. The coal type and ecological scenario was based on the grounding of the “Shen Neng” (April 2010) at Douglas Shoal located within the Great Barrier Reef (Queensland, Australia). The area is of high ecological value and the Queensland Water Quality Guidelines (2009) provided threshold limits to interpret potential impacts.

Coal contains many trace elements that are of major and moderate concern to human health and the environment although many of these concerns are only realised when coal is combusted. However, “unburnt” coal contains trace elements that may be leached to natural waterways and few studies have investigated the potential ecological impact of such an occurrence. For example, coal maritime transport has increased by almost 35% over the last five reported years (Jaffrenou et al., 2007) and as a result there is an increased inherent risk of bulk carrier accidents.

Upon grounding or becoming submerged, coal within a bulk carrier may become saturated with seawater and potentially leach trace elements to the environment and impact on water quality and ecological resilience. The worst case scenario is the breakup of a bulk carrier and dispersal of cargo to the seafloor.

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1. Introduction

Due to increased coal maritime transport, information is needed to predict coal behaviour and the potential for coal trace elements to be leached into seawater after a bulk carrier accident. This is a fundamental step in interpreting potential ecological impacts from coal in seawater, however, information is relatively sparse. The study by Alcaro et al. (2002) interpreted the consequences of a bulk carrier accident (Eurobulker IV, 17 kt in Italy in 2000) but the focus was primarily on the smothering of benthic flora and fauna and not the leaching behaviour of coal in seawater.

Ohki et al. (2004) investigated the leaching of various metals from coal into aqueous solutions containing an acid or a chelating agent. Twelve coals were subjected to leaching where the susceptibility of each metal in coal was roughly divided into three classes; largely leached (Ca, Mg and Mn), moderately leached (Cu, Fe, Pb and Zn) and little leached (Al, Co, Cr and Ni). However, these results were undertaken at pH 2 and pH 4 in aqueous solutions that differ from the chemical composition and pH of seawater (high salinity with pH ≈ 8.1). Cabon et al. (2007) provided a range of leaching results in seawater for South African reference coal types (SARM) and

found that only a few trace elements were leached from coal to seawater. Cabon et al. (2007) explained the leaching dynamics (for Mn in particular) as a function of the bicarbonate (HCO_3^-) system present in seawater. However the leach tests used a few grams of milled (<212 μm) coal samples in 20 mL of seawater and this is not typically representative of the top-size of exported coal in a bulk carrier.

Australian coal is typically exported at a nominal particle size of $-50 \text{ mm} + 0.063 \text{ mm}$. The ways in which trace elements are bound within different minerals in product coal mean that the surface area available for leaching is dependent on the particle size distribution. Furthermore, coal “fines” are likely to float and/or form “froth” on the surface, therefore the particle size distribution is also likely to indicate the percentage of coal that may initially be entrained in local currents and removed from a bulk carrier accident site.

Table 1 shows the major and minor trace elements of concern found in coal that could potentially be leached into seawater.

Sulphur (S), chlorine (Cl) and fluorine (F) in coal are usually measured due to their harmful properties in air after combustion however these elements are relatively abundant in nature and are in relatively low concentrations in coal. These elements were not tested for in this study due to the low concentrations of S, Cl and F in the Bowen Basin coal type present on the “Shen Neng”.

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Table 1
Trace elements in coal of major and minor concern.

Major concern							
Arsenic (As)	Boron (B)	Cadmium (Cd)	Mercury (Hg)	Molybdenum (Mo)	Lead (Pb)	Selenium (Se)	Sulphur (S) (%)
Minor concern							
Chromium (Cr)	Copper (Cu)	Nickel (Ni)	Vanadium (V) (in mg/kg, except S)	Zinc (Zn)	Fluorine (F)	Chlorine (Cl)	Sn + Mn ^a

^a Tin (Sn) was also added to the list due its potential presence in the anti-fouling agent Copper Tributyl Tin. Manganese (Mn) was included because of its known leaching behaviour in some coals (Jaffrenou et al., 2007).

This study aimed to provide information on the potential ecological impact of coal in seawater as a result of a bulk carrier accident in open-water (as opposed to a closed-water scenario such as coal loading basins). This study investigated the export coal type (Bowen Basin coal type) present on the "Shen Neng" at the time of the grounding in April 2010. However, the particle size distribution, trace element content and associated mineralogy will all influence the leaching behaviour of different coal types and the extent to which they may impact on the environment and is the focus of this study. Furthermore, it is important to note that during the grounding incident of the "Shen Neng", coal in the holds was not saturated with seawater.

2. Methods

2.1. Seawater collection

The seawater used in the following tests was obtained (100 L) from a location approximately 10 nautical miles east of Fingal Island (NSW) (32°44'46.26S–152°23'52.00E). The reason for this was to reduce the likelihood of anthropogenic sources of trace elements sometimes found in coastal seawater (potentially from urban wastewater and stormwater runoff). This sample also represented the "control-sample" used in the leaching tests.

2.2. Particle sizing and sample preparation

An initial coal bulk sample of 150 kg of Bowen Basin coal was sub-sampled using a rotary sample divider to obtain sub-samples of approximately 10 kg (wet mass). One of the 10 kg sub-samples was dried at 40 °C to constant weight before a particle sizing was undertaken. The dried coal was sized at 50 mm, 31.5 mm, 16 mm, 2 mm, 1 mm, 0.5 mm, 0.25 mm, 0.125 mm and 0.063 mm using standard sieves. These size fractions were subsequently used in the flotation tests. A representative sub-sample (all size fractions) and a <0.5 mm sub-sample was dried and crushed to 212 µm for trace element and X-ray Diffraction (XRD) analysis.

2.3. Determination of trace elements in coal and mineralogy

Trace element analysis was undertaken by the Australian Coal Industry Research Laboratories (ACIRL) facilities in Brisbane (QLD) and Maitland (NSW). Trace elements in the Bowen Basin coal (export top-size) were determined using standard methods (AS1038.10, 2002). Trace element analysis was also undertaken on the <0.5 mm size fraction as this proved to have a higher likelihood of being transported from a grounded bulk carrier (see flotation test results in 4.4). XRD techniques (at the University of Newcastle) were utilised to determine the mineralogy of a representative sub-sample (all size fractions) and a <0.5 mm sub-sample of the Bowen Basin coal.

2.4. Flotation tests

A flotation test was undertaken on each size fraction. Coal particle sizes > 2 mm were tested using seawater in a 2 L glass beaker as observations and sample recovery (to air dry samples) was relatively simple. For smaller coal particle sizes, approximately 200 g of each size fraction was placed in a 2 L separation flask filled with 750 mL of seawater and slightly agitated to "wet" the coal. After 1 h the lower 200 mL was allowed to run out of the flask and, after being dried (at 40 °C) and weighed, represented the "sinks". The remaining 550 mL was dried and weighed to constant-weight to determine "floats" (and suspended coal particles).

2.5. Leach tests

One of the 10 kg sub-samples was placed in a large polyethylene bucket and then filled with 20 L of seawater. This step was designed to reflect a bulk mass of export coal in a confined cargo-hold filled with seawater (submerged or grounded bulk carrier). The bucket (and lid) was placed on a large "shaker", similar to that used for the coal particle sizing, and shaken for 2 h to remove air-bubbles and maximise contact between coal particles and seawater. The bucket was allowed to rest for 24 h then the leaching solution (seawater) was filtered for analysis.

This was repeated on two other 10 kg sub-samples to complete the triplicate analysis in determining the potential of trace elements in coal being leached to seawater. A control-sample and the triplicate leaching tests were submitted for analyses which were undertaken by the NATA accredited Australian Laboratory Services (Sydney-ALS). All trace elements were determined using inductively coupled plasma-mass spectrometry (ICP-MS) techniques.

3. Results and discussion

3.1. Seawater analysis

So what trace elements are found naturally in seawater? The Artis Zoo Aquarium in Amsterdam relies on good quality seawater so research undertaken by them provides suitable indicators for potential impacts on marine life. Sondervan (2001) describes how the Artis Zoo Aquarium obtains its seawater from the middle of the Atlantic Ocean on the co-ordinates within small ranges from 45°41'N, 24°42'W. This area lies on the route of the large Hual Benelux b.v. Automobile carriers that travel regularly from Amsterdam to Halifax (NS, Canada) and New York (NY, USA). The seawater is taken in from a depth of 9 m and remains no longer than 4 days in the ballast tanks. In Amsterdam, 160,000 L of that seawater is brought by a vessel through the canals near the aquarium building and pumped into the aquarium reservoirs.

Table 2 shows elements and approximate concentrations of water obtained from the North Atlantic. The transport of seawater to the Artis Zoo is similar, in a maritime sense, to bulk carrier

Table 2
Trace elements in natural seawater (from Sondervan (2001)).

Element	Reference (mg/L)	Average (mg/L)
Ca	400	422
K	383	393
Mg	1319	1312
Na	10590	10731
S	878	945
B	4.53	4
Fe	0.01	1
P	0.067	0
Sr	8	7
Br	60	69
	(µg/L)	(µg/L)
Li	176	119
Al	10	26
Ti	0.67	13
Cr	0.6	1
Mn	1.4	28
Co	0.1	1
Ni	4.29	9
Cu	2.07	30
Zn	6.68	353
Rb	121	126
Y	0.11	0
Mo	10.33	2
Ru	?	<0.1
Rh	?	0
Pd	?	<0.1
Ag	0.21	0
Cd	0.1	0
In	10	0
Sn	1.27	0
Sb	0.28	0
I	59	50
Cs	0.37	0
Ba	24	6
La	0.0034	<0.1
Ce	0.14	<0.1
Pr	?	<0.1
W	0.11	<0.1
Pt	?	<0.1
Au	0.005	<0.1
Hg	0.05	<0.1
Tl	0.1	0
Pb	0.03	2
Bi	0.02	<0.1

transport. For example, the seawater is transported in a large container ship as ballast, much like coal, and provides a suitable surrogate for evaluating seawater quality with respect to sampling and storage of seawater.

Seawater used in this study was sampled from open-water and the initial water quality is shown in Table 3. While sampled from the other side of the world, the seawater sampled off Fingal Island (NSW) resided within the average range values shown in Table 2, and thus validated as "typical seawater" for use in the leaching tests.

Table 4 gives the pH, temperature and electrical conductivity (EC) of the seawater both before and after the leach tests and indicated negligible variation in these parameters due to the leaching test. This highlights that S, F and Cl did not change pH and the ionic composition of the seawater was not significantly altered by the leached trace elements.

Table 3
Trace elements found in the seawater used in this study (in µg/L).

Se	Cr	Sn	Hg	As	B	V	Cd	Cu	Pb	Mn	Mo	Ni	Zn
<2	<0.5	<5	<0.1	2.3	5100	1.6	0.4	3	0.4	1.8	13.8	0.6	5

Table 4
Seawater pH, temperature and EC before and after the leaching tests indicating negligible variation due to the leaching test.

	pH (No units)	Temp (°C)	EC (µS/cm)
Seawater (control)	8.14	21.8	48,500
After Leach A	8.09	22.2	48,400
After Leach B	8.11	22.1	48,500
After Leach C	8.09	22.1	48,500
Average	8.11	22.1	48,475
Maximum	8.14	22.2	48,500
Minimum	8.09	21.8	48,400
Standard deviation	0.02	0.2	50

3.2. Particle sizing and flotation tests

Fig. 1 shows results from the particle sizing and flotation tests.

The >2 mm size fraction represented approximately 50% of the total sample mass and readily sank in seawater during the flotation tests. Coal particle sizes <0.250 mm formed "froth" on the seawater surface. These size fractions are likely to be entrained by local currents and transported away from a grounded/submerged bulk carrier, particularly with a hull breach. Results also indicate that some of the -2 + 1 mm and -1 + 0.5 mm size fractions, and all coal particles <0.5 mm, are likely to be suspended and would be particularly susceptible to transport by ocean currents. The mass of coal that floats means that approximately 15.5% of the cargo may be potentially lost to ocean currents.

Jaffrenou et al. (2007) found that coal particles (1–10 mm in size) could be displaced 1 km away from the dumping point in about 10 h at current velocity (0.46–0.66 knots) however bottom inclination and roughness could modulate the extrapolation from their "polludrome" results. The leaching tests by Cabon et al. (2007) were undertaken using milled samples (<212 µm) and small analysis masses (<2 g). The bulk transport of product coal occurs at a nominal particle size of 50 mm with an associated particle size distribution down to 0.063 mm and, while the lab tests of Cabon et al. (2007) provide valuable insight into the leaching trace elements in coal into seawater, the nominal top-size tested was not typical of bulk coal export in bulk carriers.

3.3. Determination of trace elements in coal (Bowen Basin) and mineralogy

Trace elements in Bowen Basin coal (export nominal top-size and the <0.5 mm size fraction) are shown in Table 5. The trace element range for Australian coals, International coals, Earth's crust and Earth shales are also shown to highlight their relative abundance in nature and the fact that Bowen Basin coal typically has lower average trace element concentrations than other Australian coals and international coals.

X-ray diffraction (XRD) allowed the mineralogy of the coal to be determined. The dominant minerals found in the composite sample (nominal top-size -50 + 0.063 mm) were quartz, kaolinite and siderite (refer Table 6). The <0.5 mm size fraction contained similar minerals, however included Calcium Sulfate Hydrate (hydrated gypsum) and synthetic Magnetite, both of which were likely additives to "fines" during the coal washing and fines recovery process. Muscovite was also present in the <0.5 mm size fraction (refer Table 7).

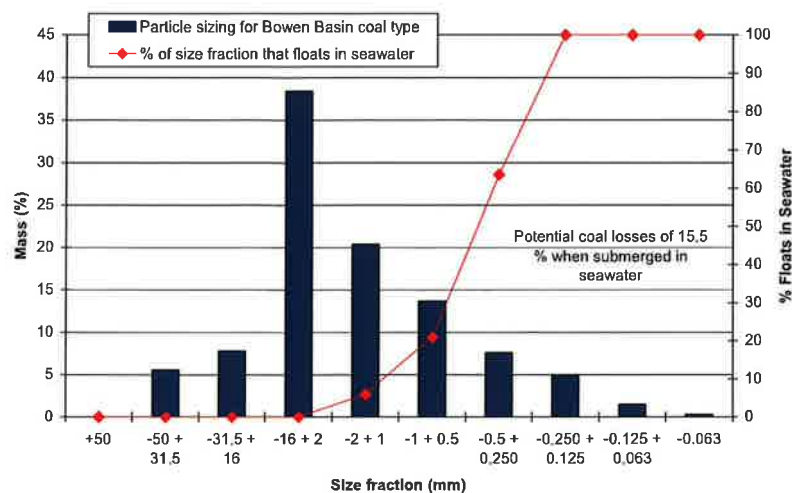


Fig. 1. Particle size distribution and associated percentage that floated in seawater.

Table 5

Trace element analysis for Bowen Basin coal (export nominal top-size and the <0.5 mm size fraction) where the trace element range for Australian coals, International coals, the Earth's crust and Earth shales are also shown.

Element	Symbol	Degree of concern	Units	Bowen Basin (top-size)	Bowen Basin (<0.5 mm)	Range for Australian coals	Range in coal around the world	Earth's crust	Earth shales
Selenium	Se	Major	mg/kg	1	1.1	0.1–1	0.1–5.3	0.05	0.6
Chromium	Cr	Moderate	mg/kg	4	4	2–25	1–35	100	100
Tin	Sn	?	mg/kg	<2	<2	nd	nd	nd	nd
Mercury	Hg	Major	mg/kg	0.02	0.02	0.1–0.11	0.1–0.19	0.08	0.5
Arsenic	As	Major	mg/kg	0.7	0.6	0.2–2.2	3–26	1.8	15
Boron	B	Major	mg/kg	16	19	5–70	6–146	10	100
Vanadium	V	Moderate	mg/kg	41	40	7–75	1–60	135	130
Cadmium	Cd	Major	mg/kg	0.05	0.05	0.01–0.32	0.1–0.31	0.2	0.2
Copper	Cu	Moderate	mg/kg	15	15	6–27	<1–28	55	50
Lead	Pb	Major	mg/kg	5.3	6	2–14	<1–22	12	20
Manganese	Mn	?	mg/kg	42	45	nd	nd	nd	nd
Molybdenum	Mo	Major	mg/kg	<2	<2	0.1–2.6	0.1–4	1.5	3
Nickel	Ni	Moderate	mg/kg	2	2	4–23	2–21	75	70
Zinc	Zn	Moderate	mg/kg	18	18	3–26	1–55	70	100

? Not currently considered as minor, moderate or major concern in the Australian coals.

Table 6

XRD analysis from nominal top-size sample (–50 + 0.063 mm).

Ref. code	Compound name	Chemical formula
01-085-0798	Quartz	SiO ₂
01-080-0885	Kaolinite-1A	Al ₂ (Si ₂ O ₅)(OH) ₄
01-083-1764	Siderite	Fe(CO ₃)

Table 7

XRD analysis from <0.5 mm size fraction.

Ref. code	Compound name	Chemical formula
00-001-0385	Calcium Sulfate Hydrate	CaSO ₄ · 2H ₂ O
01-070-3755	Quartz	SiO ₂
01-080-0885	Kaolinite-1A	Al ₂ (Si ₂ O ₅)(OH) ₄
01-083-1764	Siderite	Fe(CO ₃)
01-087-0245	Magnetite, syn	Fe ₂ · 93O ₄
01-072-1503	Muscovite	K Al ₂ (Si ₃ Al)O ₁₀ (OH) ₂

Table 8
Leaching results–coal in seawater.

	Units	LOR	CONTROL	SW1A	SW1B	SW1C	(Difference)			Trace elements leached from coal to seawater?
							SW1A	SW1B	SW1C	
Selenium (Se)	µg/L	2	<2	<2	<2	<2	0	0	0	No leaching
Chromium (Cr)	µg/L	0.5	<0.5	<0.5	<0.5	<0.5	0	0	0	No leaching
Tin (Sn)	µg/L	5	<5	<5	<5	<5	0	0	0	No leaching
Mercury (Hg)	µg/L	0.1	<0.1	<0.1	<0.1	<0.1	0	0	0	No leaching
Arsenic (As)	µg/L	0.5	2.3	1.2	1.2	1.4	-1.1	-1.1	-0.9	Net removal from seawater
Boron (B)	µg/L	100	5100	4500	4700	4700	-600	-400	-400	Net removal from seawater
Vanadium (V)	µg/L	0.5	1.6	0.5	0.5	0.5	-1.1	-1.1	-1.1	Net removal from seawater
Cadmium (Cd)	µg/L	0.2	0.4	0.6	0.8	0.6	0.2	0.4	0.2	Leaching
Copper (Cu)	µg/L	1	3	12	11	11	9	8	8	Leaching
Lead (Pb)	µg/L	0.2	0.4	0.9	0.8	0.8	0.5	0.4	0.4	Leaching
Manganese (Mn)	µg/L	0.5	1.8	37.1	36.5	37	35.3	34.7	35.2	Leaching
Molybdenum (Mo)	µg/L	0.1	13.8	17	16.8	16.7	3.2	3	2.9	Leaching
Nickel (Ni)	µg/L	0.5	0.6	5.6	5.9	6.2	5	5.3	5.6	Leaching
Zinc (Zn)	µg/L	5	5	23	27	54	18	22	49	Leaching

LOR = limit of reading.

3.4. Leach tests

Table 8 shows results from the leaching tests. The "Control" sample represents the seawater used in the leaching tests and SW1A, SW1B and SW1C represent the triplicate leaching results. The difference between pre and post leaching to seawater is also shown.

No leaching from coal to seawater was observed for Se, Hg, Sn and Cr. There was net removal of As, B and V from seawater, that is, these elements were adsorbed to the coal resulting in a lower concentration in seawater. Leaching from coal to seawater was observed for Cd, Cu, Pb, Mn, Mo, Ni and Zn. How much is leached from coal? A mass balance approach was used to quantify the difference between trace elements in Bowen Basin coal and trace elements found in the seawater leachate. For example, if 10 kg of coal contains 0.05 mg/kg Cd then there was 0.5 mg (500 µg) in the test coal sample. If the initial seawater concentration was 0.4 µg/L (8 µg in 20 L), and it increased by 0.3 µg/L (6 µg in 20 L), then the % leached represents approximately 1.2% of the total Cd content in coal. Table 9 summarises the mass balance for all trace elements that leached from coal to seawater.

The mass balance approach showed that the leaching of trace elements in coal to seawater ranged from 0.03% to 1.2% of the total trace element content in coal. The relatively low amount of trace elements leached from unburnt coal suggests that many trace elements are strongly bound to mineral matrices, particularly when submerged in seawater with a high electrical conductivity and relatively high pH (~8.1). Elemental substitution may occur, for example, such as processes that result in net removal of some elements (As, B and V) and leaching of others (Cd, Cu, Mn, Ni and Zn) from coal in seawater and further research is warranted into such processes.

But what is the ecological threat of such leaching to seawater? Table 10 shows a comparison of water quality guidelines (recommended range) (DERM, 2009) and leachate results, highlighting elements of concern in aquatic environments.

Cu and Mn were the only elements that exceeded guideline values. Fortunately, dilution would be expected in open-waters (like Douglas Shoal on the Great Barrier Reef) that are likely to negate any ecological impact due to leaching of Cu and Mn in coal to seawater. When a semi-enclosed coal loading basin is considered there is likely to be a cumulative impact, however this was outside the scope of this preliminary study. The accumulative ecological impact in such locations also warrants further research.

It is important to note that Mn and Sn are not considered trace elements of major/minor concern in the coal industry. These

Table 10
Comparison of water quality guidelines (recommended range) and leachate results (showing elements of concern in aquatic environments).

Element	Recommended range QLD WQ Guidelines 2009 µg/L	Leach test Average µg/L	Less than Guideline?
As	<50	Net removal	Y
Cd	<3	0.7	Y
Cr	<100	No leaching	Y
Cu	<6	11	N
Mn	<10	37	N
Hg	<0.05	No leaching	Y
Ni	<40	5.9	Y
Sn	<1	<5 ^a	? ^a
Zn	<200	35	Y

^a Likely to be <1 but could not achieve desired limit of reading during analysis.**Table 9**
Leaching results–coal in seawater.

Element	Symbol	Degree of concern	Bowen Basin (top-size) (mg/kg)	In 10 kg of coal (µg)	Initial seawater concentration in 20 L (µg)	Increase after leach test (µg in 20 L)	% leached from total mass in coal
Cadmium	Cd	Major	0.05	500	8	6	1.20
Copper	Cu	Moderate	15	150,000	60	160	0.11
Lead	Pb	Major	5.3	53,000	8	16	0.03
Manganese	Mn	?	42	420,000	36	720	0.17
Molybdenum	Mo	Major	<2	20,000	276	62	0.31
Nickel	Ni	Moderate	2	20,000	12	106	0.53
Zinc	Zn	Moderate	18	180,000	100	620	0.34

? = "No concern". Mn is not considered an element of concern in the coal industry.

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elements were added to the analysis in this study because of their inclusion in water quality guidelines and the leaching behaviour of Mn as noted in the literature (Cabon et al., 2007; Jaffrennou et al., 2007; Ohki et al., 2004). Cabon et al. (2007) found that few trace elements of concern were leached into seawater, however noted the relationship between the calcite and gypsum content of coal and the leaching of Mn. In this respect, both Mn and Sn should be added to existing Australian Coal Association Research Projects (ACARP), particularly the trace element database (CSIRO and ACARP, 2007). Furthermore, their inclusion would assist in interpreting the impact of coal immersion in seawater as highlighted in the literature and in this study.

4. Conclusion

This study was commissioned by Xstrata Coal to provide preliminary information on the potential ecological impact of coal in seawater after an open-water incident. In context of the Bowen Basin coal type on the “Shen Neng” and the open-water incident on Douglas Shoal, leaching results indicated negligible impact to water quality and ecological resilience as a result of trace elements in this coal type being leached to seawater. Ocean currents are highly likely to disperse and dilute leached trace elements in an open-water incident.

This study also highlighted that particle size distribution, trace element content and mineralogy will all influence the leaching behaviour of different coal types and the extent to which they may impact on the environment and warrants further research. Since this study investigated just one export coal type there may be a need for further research on other coal types as all Australian export coals could have certification concerning their ecological impact in case of a bulk carrier accident. Further research is also

required on the potential cumulative impacts of coal “spills” in closed-water systems.

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