

# Water Quality Impact from the Discharge of Coal Mine Wastes to Receiving Streams: Comparison of Impacts from an Active Mine with a Closed Mine

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Abstract This study examined two underground coal mines in the Sydney basin and investigated the water chemistry impact from their wastewater discharges to surface receiving waters. One mine closed 17 years prior to the study, and the other was still active. The geology of both mine locations shared many similarities and some important differences that influenced their wastewater chemistry and its subsequent impact on receiving waterways. Water quality of wastewater discharges from the two mines and their receiving waterways was investigated over a 6-month period. Both mine discharges caused comprehensive modification to receiving water chemistry. The closed mine increased electrical conductivity (EC) 3.3 times from upstream (33 µS/cm) compared to downstream (108 µS/cm). In comparison, the active mine increased EC by 9.4 times (173 µS/cm) upstream to 1628 µS/cm downstream. Both coal mine wastes increased the concentration of different contaminants to levels that are potentially hazardous for receiving water ecosystems. The active mine increased bicarbonate concentration in the receiving water by more than 60 times to 743 mg/L. The closed mine increased zinc and nickel concentrations in its receiving stream by 70 and 20 times to 318 and 360  $\mu$ g/L. The active coal mine discharge was dominated by sodium and bicarbonate ions compared to magnesium and sulphate ions in the closed mine drainage. Although both receiving

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School of Science and Health, Western Sydney University, Locked Bag 1797, Penrith, NSW 2751, Australia e-mail: i.wright@westernsydney.edu.au waters were sodium and chloride dominated upstream of the mine waste, their ionic composition was strongly modified due to the inflow of coal mine wastes. Results from this study are a reminder that water pollution from coal mines is important for both active mines and for closed mines decades after mining activity ceases.

Keywords Underground mining  $\cdot$  Freshwater ecosystems  $\cdot$  Ionic composition  $\cdot$  Bicarbonate  $\cdot$  Metals  $\cdot$ Zinc  $\cdot$  Nickel  $\cdot$  Salinity  $\cdot$  pH  $\cdot$  Pollution  $\cdot$  Water quality  $\cdot$ Geochemistry

## **1** Introduction

Coal mining can contaminate surface waters while mines are active and for decades after they close (Younger 1993; Banks et al. 1997; Wright and Burgin 2009a, b). One well-known form of water pollution often associated with coal mining is *acid mine drainage* (AMD) (Brake et al. 2001). Coal contains sulphur that is oxidised due to the disturbance associated with mining and exposure to both air and water which results in the formation of sulphuric acid (Johnson 2003). The AMD process leaches and mobilises metals in mine water that varies according to many factors such as the amounts of sulphur, the buffering capacity of water and the surrounding geology (Banks et al. 1997; Brake et al. 2001; Johnson 2003).

Water pollution of rivers and streams from coal mine wastes is a widespread environmental problem caused by active and closed coal mine operations (Johnson 2003). Some of the water quality problems associated with disposal of coal wastes to surface waters include reduced pH (Herlihy et al. 1990; Brake et al. 2001); high levels of electrical conductivity (Jarvis and Younger 1997; Pond et al. 2008); and elevated concentrations of metals such as iron, aluminium, copper, nickel, manganese and zinc (Greenfield and Ireland 1978; Short et al. 1990; Brake et al. 2001; Wright and Burgin 2009a; Gray and Harding 2012; Griffith et al. 2012). Investigations have also identified that coal mine wastes can cause substantial modification of stream ionic composition (Griffith et al. 2012; Wright 2012; Wright et al. 2015). One of the characteristic signs of a heavily AMD-contaminated waterway, along with acidic pH and elevated metals, is the dominance of waterway anions by sulphate (Herlihy et al. 1990; Brake et al. 2001; Griffith et al. 2012). There are many potentially adverse consequences of water pollution from coal mine wastes, but one that is particularly common and widespread is the impairment of river and stream ecosystems (García-Criado et al. 1999; Battaglia et al. 2005; Wright and Burgin 2009a,b; Petty et al. 2010; Wright and Ryan 2016).

Water pollution from coal mining is a complex problem of global proportions. Many of the world's earliest coal mining areas in Europe and in the UK have ceased mining yet water pollution is an ongoing legacy for which only some mines have received treatment to mitigate water pollution (Jarvis and Younger 1997; Johnson, 2003). There is a rich literature on coal mines and water pollution in the USA. This includes some broad regional studies of multiple mines and waterways (e.g. Herlihy et al. 1990; Pond et al. 2008; Petty et al. 2010). Some of the US studies have examined water pollution resulting from open-cut coal mining called 'mountaintop mining', where the mining spoil and overburden from higher peaks overlying coal seams are pushed into the lower elevated areas called 'valley fills' (Palmer et al. 2010; Petty et al. 2010; Lindberg et al. 2011; Griffith et al. 2012). Water pollution from coal mining has also been investigated in other coal mining areas of the world including India (Tiwary 2001), New Zealand (Winterbourn 1998; Gray and Harding 2012) and Australia (Battaglia et al. 2005; Wright and Burgin 2009a,b; Wright 2012; Belmer et al. 2014; Wright et al. 2015).

Mining and export of coal from Australia have accelerated over the last 50 years and now exceeds more than 250 million tonnes per year (Mudd 2009; Minerals Council of Australia 2015). Australia exports 77 % of its coal production, and it is Australia's second highest value export commodity, making it one of the top suppliers to the international coal market (Minerals Council of Australia 2015). The Australian coal mining industry began in the late eighteenth century in the Hunter and Illawarra coal fields of the Sydney basin (Mudd 2009). Underground coal mining has been the predominant form of coal mining in the Sydney basin of southeastern Australia, particularly in the southern and western coal fields (Mudd 2009). Many coal mines in the Sydney basin have ceased production and generally ongoing water pollution from the former mining activities is not treated to ameliorate downstream pollution impacts (e.g. Battaglia et al. 2005; Wright et al. 2011b; Wright and Ryan 2016). The management and remediation of closed, abandoned and derelict mines (coal and other mines) in Australia is regarded as a growing environmental problem with recent estimates that there are 52,543 abandoned mines with only a small proportion receiving effective rehabilitation (Unger et al. 2012). Unger et al. (2012) found that the estimates of identification of abandoned mines in many jurisdictions, including New South Wales (NSW), of Australia is sketchy due to the lack of comprehensive information on the location and detailed assessment of the environmental issues associated with each closed mine. The inadequacy of government funding for mine site remediation in NSW has been raised as a major problem with \$A4.1 million in NSW Government funding in 2014-2015 for rehabilitation of derelict mines contrasting with \$A1.5 billion that the mining sector pays to NSW Government for royalties (Geary 2015).

There have been few water chemistry studies (and none in Australia) comparing water quality impacts from coal mine liquid waste discharges from an active mine to those from a closed coal mine. To collect comparative data, we used an equivalent sampling design and equal effort at both locations to achieve a balanced comparison of the effects of the two waste discharges. In this study, we used a temporally replicated water-quality sampling design to detect and measure any water quality effects from the discharge of wastewater from two coal mines to otherwise unpolluted receiving waterways. At both mines, samples were collected from three sub-locations: the receiving waterway immediately above the mine, the receiving water below the waste outlet and the mine wastes itself. Our key questions posed for this investigation are the following:

- 1. How does receiving stream water chemistry change due to the discharge of coal mine waste water from an active compared to a closed coal mine?
- 2. How does the water chemistry of the two mine wastes compare (active vs closed)?

#### 2 Methods

#### 2.1 Study Area

Samples were collected from liquid wastes from two underground coal mines (Canyon Colliery and West Cliff Mine) and the two corresponding waterways (Dalpura Creek and Georges River) that receive inflows of waste water from each mine (Fig. 1). Both mines are in the Sydney basin (Branagan et al. 1979) in New South Wales in southeastern Australia. Canyon Colliery is located in the western coalfield, in the Blue Mountains, west of Sydney and West Cliff Mine is close to the coast, south of Sydney, in the southern coalfield (Fig. 1; Huleatt 1991). One mine closed in 1997 (Canyon Colliery) and the other is actively mined (West Cliff Mine). Canyon coal mine is located 96 km inland from the southeastern coast of Australia, 50 km west of Sydney metropolitan area (Fig. 1). It is about 86 km northwest of West Cliff Mine. The mine closed in 1997 after more than 50 years of operation (Macqueen 2007). West Cliff coal mine is an underground longwall coal mine that has operated continuously since 1976. It is located on the outskirts of the small township of Appin, about 25 km inland from the coast (Fig. 1).

The receiving water catchments for both mines are predominantly naturally vegetated. Both mines are located in the headwaters of river catchments. West Cliff Mine operates in the upper Georges River catchment and Canyon Mine in Grose River catchment (Fig. 1). The catchment in the vicinity of West Cliff Mine does have some disturbance from mining activity from West Cliff Mine's surface workings, and some of the catchment is also occupied by a small urban township (Appin). Despite such modifications, Georges River, above West Cliff Mine, is recognised as having unpolluted water quality and good ecological health (Tippler et al. 2012). Dalpura Creek is a small tributary flowing within a small sub-catchment of the Grose River catchment (Wright and Burgin 2009a; Wright and Ryan 2016). The Dalpura Creek sub-catchment is protected



Fig. 1 Map showing location of the two coal mines (Canyon Mine and West Cliff Mine) and the rivers that receive wastewater from each in relation to the Sydney metropolitan area. Dalpura Creek flows into Grose River and Brennans Creek flows into Georges River in close proximity to each mine

as part of the Blue Mountains National Park and is almost completely undisturbed with no mine workings located upstream of the mine waste discharge.

The geology of the two mines shares many similarities and some important differences. Sedimentation in the Sydney basin took place between the Late Carboniferous and Middle Triassic (Branagan et al. 1979). The stratigraphy of the western coalfield (Canyon Mine) and the southern coalfield (West Cliff Mine) is very similar with Illawarra coal measures from the Late Permian underlying Wianamatta, Hawkesbury and Narrabeen sediments (sandstone and shale formations) from the Triassic (Branagan et al. 1979). The two mines extracted different coal seams with Canyon Mine working the Katoomba Seam for thermal coal and West Cliff Mine the Bulli Seam, which is highly valued as hard coking coal for steel making (Huleatt 1991). Coal samples from both locations in this study have both been assessed as having very low sulphur levels of 0.38 % (Huleatt 1991) which Cohen (2002) points out is about 5 to 10 times lower than typical levels in international sources of coal.

The depositional history of the western compared to the southern coalfield differed in some respects with only the southern coalfield having extensive marine transgressions, particularly during the Greta and Tomago depositional episodes (Branagan et al. 1979). The combination of the depositional history and the proximity of the southern coalfields to the ocean together contribute to the groundwater in the Bulli Seam having higher electrical conductivity (EC) (Nicol et al. 2014) than that has been recorded from groundwater in the Katoomba Seam in the western coalfield (Cohen 2002). The elevation of the land surface at Canyon Mine is 850 m above sea level (ASL), considerably higher than West Cliff Mine (c. 250 m ASL; Table 1).

The discharge of mine wastewater from both mines in this study was regulated by the New South Wales Environment Protection Authority (NSW EPA) through licences called an 'Environment Protection Licence' (EPL) that specify the permitted concentrations of contaminants that each mine could discharge to receiving waters (NSW EPA 2015). Canyon Mine held an EPL #558 that specified the acceptable concentrations of iron, zinc, total dissolved solids, oil and grease and a range for pH (Wright et al. 2011b). After the mine closure in 1997, the mine lease was surrendered in 2005 and the EPL was surrendered in 2001 (Wright et al. 2011b). Groundwater infiltration to the former mine workings continues to flow, under the influence of gravity, from least two horizontal mine shafts, the largest of which provides about 90 % of the flow in Dalpura Creek (Macqueen 2007). Dalpura Creek is a tributary of Grose River and both flow within the Blue Mountains National Park, and the entire area is part of the Greater Blue Mountains World Heritage Area (Wright et al. 2011b). The elevation of the mine drainage shaft is 805 m ASL (Table 1). Earlier research indicated that mine drainage from Canyon coal mine is contaminated with ecologically damaging levels of zinc (Wright and Burgin 2009a,b; Wright and Ryan 2016). In 2003, about 6 years after the mine ceased operation, a survey of water quality and river invertebrates found that at least 10 km of the high conservation-value Grose River was ecologically impaired due to the inflow of mine drainage from Dalpura Creek into Grose River (Wright and Burgin 2009a,b).

The average depth of West Cliff mine is 490 m below the surface, and the mine constantly pumps groundwater to the surface from multiple points within the mine (Volcich 2007). West Cliff mine also collects, stores and recycles a mixture of surface runoff and mine groundwater in a large storage dam (Volcich 2007). Mine wastewater discharge provides the majority of the flow in the upper Georges River as it contributes approximately 0.9 ML of flow per day compared to an average flow of 0.1 ML a day in Georges River above the inflow of the waste discharge (Ecoengineers 2007). In 2012, the NSW Office of Environment and Heritage (NSW OEH) investigated water pollution from the mine and found the mine wastewater was toxic to some aquatic species, in laboratory ecotoxicology testing, and it also had elevated levels of salt and a range of metals (NSW OEH 2012). The EPA has since modified the mine's EPL licence (#2504) to provide a more comprehensive suite of pollutants with quantitative concentration limits for pollutants that are permitted to be discharged (including nine metals, EC, pH and chemical oxygen demand; NSW EPA 2013). Prior to 2012, the EPL 2504 for the mine only included waste limits for three water quality attributes (oil and grease, suspended solids and pH) in the licence to discharge wastewater to Georges River (Wright 2012).

#### 2.2 Water Quality Sampling

Water sampling was conducted at both localities (Canyon coal mine and West Cliff coal mine) and the corresponding receiving waterways on three occasions (at 6to 8-week intervals) over a 6-month period from June 2014 to December 2014 (Table 1). Our investigation used a triangulated upstream versus downstream design with the mine waste itself the third point of the triangle. At each locality, samples were collected from the receiving waterway about 100 m upstream of each mine waste inflow. Samples were also collected from the mine waste inflow just before they discharged (10 m at Dalpura Creek and 50 m at Georges River) into the receiving waterway. Samples were collected about 100 m downstream of the Georges River inflow and about 20 m downstream of the Dalpura Creek inflow. Precipitous and hazardous topography restricted sampling of Dalpura Creek further downstream, and mine waste inflow was observed to be thoroughly mixed with the receiving water at the downstream sampling site.

On each sampling occasion, field meters were used for in situ measurement of physiochemical water quality attributes of pH, electrical conductivity (EC), and water temperature from all study sites. The meters used included a TPS AQUA-Cond-pH meter (for pH and electrical conductivity) and a YSI ProODO meter (water temperature) and a HACH 2100P turbidimeter for turbidity. The calibration of each meter was checked on each day and adjusted if necessary. Five replicated measurements of each of the above water quality attributes were recorded from each site on each sampling occasion. In addition, duplicate grab samples of water were collected from all study sites in decontaminated sample containers provided by a commercial testing laboratory. Samples were chilled and delivered to the laboratory for

Table 1 Sampling sites location	1 details of the	two coal mines and t	heir respective receiving wa	aters that were investigated fron	n June to November 2014 for this study	/
Site name	Site code	Location	Latitude and longitude	Elevation above sea level	Category	Dates of sampling
Dalpura Creek (upstream)	SUQ	Blue Mountains National Park, Bell	S 33° 32' 51.23" E 150° 18' 50.40"	Elevation 815 m	Receiving waterway upstream of mine waste discharge	20 June 2014 10 September 2014 28 November 2014
Canyon coal mine (wastewater)	CCM	Blue Mountains National Park, Bell	S 33° 32' 55.77" E 150° 18' 46.52"	Elevation 805 m	Closed mine, still releases mine drainage to Dalpura Creek	20 June 2014 10 September 2014 28 November 2014
Dalpura Creek (downstream)	SDDS	Blue Mountains National Park, Bell	S 33° 32' 52.40" E 150° 18' 47.18"	Elevation 785 m	Receiving waterway downstream of mine waste discharge	20 June 2014 10 September 2014 28 November 2014
Georges River (upstream)	GUS	Georges River, Appin	S 34° 12' 18.93" E 150° 47' 56.89"	Elevation 233 m	Receiving waterway upstream of mine waste discharge	20 June 2014 19 September 2014 28 November 2014
West Cliff coal mine (Brennans Creek) (wastewater)	MM	Brennans Creek, Appin	S 34° 12' 15.58" E 150° 47' 59.77"	Elevation 230 m	Active mine waste discharged via Brennans Creek to Georges River	20 June 2014 19 September 2014 28 Nov.2014
Georges River (downstream)	GDS	Georges River, Appin	S 34° 12' 14.05" E 150° 47' 52.71'	Elevation 225 m	Receiving waterway downstream of mine waste discharge	20 June 2014 19 September 2014 28 November 2014
The elevation of each site above	sea level (AS)	L) and the category o	f each sampling site in the s	study is provided		

analysis. All grab samples were analysed using standard methods (APHA 1998) by a National Associations of Testing Authorities (NATA) accredited laboratory for major anions, major cations and a range of total metals (aluminium, arsenic, barium, calcium, copper, iron, lead, manganese, nickel, strontium and zinc).

#### 2.3 Data Analysis

Student's *t* test was used to test for differences between upstream versus downstream water quality for water quality attributes, for each locality. Similarly, Student's *t* test was also used to test for differences between each of the mine wastes for water quality attributes.

#### **3 Results**

Water chemistry results show that each of the coal mine waste discharges caused substantial and different changes to the chemical properties of each waterway below the two waste discharges (Tables 2 and 3). Electrical conductivity (EC) increased highly significantly (p < 0.0001) downstream of both waste discharges (Tables 2 and 3). The lowest EC levels (26.4 to 36.9 µS/cm) were recorded from Dalpura Creek above the mine waste entry point. EC in Dalpura Creek increased to a mean of 108  $\mu$ S/cm below the inflow of the mine drainage (Table 3). Mean EC levels in Georges River, above the mine waste, were 173.2  $\mu$ S/cm, rising to 1628  $\mu$ S/cm below the mine waste inflow (Table 2). EC of the mine wastes also varied highly significantly (p < 0.0001) with mean EC of West Cliff Mine  $(2319 \ \mu\text{S/cm})$  more than 18 times higher than that of Canyon Mine (mean = 116.9  $\mu$ S/cm) (Table 4).

The pH of each mine waste discharge differed highly significantly (p < 0.0001) with consistently acidic levels in Canyon Mine wastewater (5.51–6.33) compared to West Cliff Mine wastewater (mean pH 7.94) that displayed considerable fluctuations during the study with pH varying by more than 3 pH units (6.1–9.11) (Table 4). The two upstream receiving waters had differing pH with mean pH of 5.75 at Dalpura compared to 6.9 at Georges River (Tables 2 and 3). Dalpura Creek pH (Table 3) was essentially unchanged with mean pH above the mine (5.75) almost identical to downstream (5.76) with the Canyon Mine inflow only marginally higher (mean pH 5.83; Table 4). The trend was different in Georges River where pH was approximately one pH

unit higher below the mine inflow compared to above it (Table 2).

Each mine waste strongly modified the ionic composition of each receiving waterway (Figs. 2, 3, 4 and 5). Both receiving waterways were sodium and chloride dominated above each mine discharge (Tables 2 and 3). This changed to magnesium and sulphate domination in Dalpura Creek downstream of Canyon Mine (Table 3) and Georges River was sodium and bicarbonate dominated downstream of West Cliff Mine (Table 3). In both cases, the changes in anion and cation dominance of the receiving waters reflected the influence of dominant ions within each mine waste. The ionic composition of the mine wastes contrasted between the two mines. West Cliff Mine was sodium and bicarbonate dominated compared to magnesium and sulphate domination in Canyon Mine wastewater (Tables 2 and 3). The mean concentrations of the dominant ions were much higher in wastewater from West Cliff Mine (sodium 710 mg/L and bicarbonate 939 mg/L) compared to the Canyon Mine (magnesium 7.8 mg/L and sulphate 29.5 mg/L) (Tables 2 and 3).

The inflow of wastewater from both mines broadly increased the individual and total combined concentration of a suite of metals in the receiving waters downstream of each waste discharge (Figs. 6 and 7). The mean combined concentration of metals increased from 0.34 mg/L in Dalpura Creek and 0.48 mg/L in Georges River (above both mines) to 0.93 mg/L (Dalpura Creek) and 1.49 mg/L (Georges River) below the two mines (Tables 2 and 3, Figs. 6 and 7). The mine inflows had combined metal concentrations of 1.36 mg/L (Canyon Mine) and 1.85 mg/L (West Cliff Mine) (Table 4; Figs. 6 and 7). The receiving waters, upstream of both mines, were dominated by three metals iron, aluminium and manganese. Aluminium had the highest mean concentration (205 µg/L) in Dalpura Creek, and iron was highest in Georges River (240  $\mu$ g/L) (Tables 2 and 3). Each of the mine wastewaters had different types and concentrations of metal in its wastewater. West Cliff Mine drainage was dominated by aluminium (557  $\mu$ g/ L) and then with progressively lesser concentrations, Fe > Ba > Str > Ni > Mn > Zn > Cu > As > Pb > Cd(Table 3). In comparison, manganese (417  $\mu$ g/L) was highest in Canyon Mine drainage, followed by Zn>Fe>Ni>Ba>Str>Al>Cd (Table 3). Arsenic, copper and lead were never above detection limits in Canyon Mine drainage.

Table 2 Comparison of water quality variables from Georges River, collected upstream (GUS) and downstream (GDS) of the coal mine waste discharge

Source of variation	t statistic (p value)	Upstream (GUS)		Downstream (GDS)	
		Range	Mean	Range	Mean
pH (pH units)	3.4 (0.0032)	6.38–7.39	6.90	6.38–9.16	8.046
EC (µS/cm)	9.1 (<0.0001)	191.4-176.4	173.18	854-2451	1628.5
Water temperature (°C)	0.95 (0.349)	9.6–18	12.9	11.2-19.9	14.27
Turbidity NTU	3.97 (0.0009)	2.03-6.09	3.28	4.57-6.53	5.09
Carbonate (mg/L)	3.40 (0.0192)	Bd.	_	9–140	82
Bicarbonate (mg/L)	5.62 (0.0025)	9–17	11.85	330–990	743.3
Chloride (mg/L)	4.22 (0.0056)	35-67	45.6	69–170	128.5
Sulphate (mg/L)	5.23 (0.0034)	3–7	5.43	15-39	29
Calcium (mg/L)	5.17 (0.0004)	2.4-4.4	3.28	4.4-6.2	5.625
Potassium (mg/L)	0.32 (0.7595)	0.8-31	5.97	1.9-6.3	4.34
Magnesium (mg/L)	2.55 (0.03445)	3.2-4.1	3.53	2.8-3.4	3.0625
Sodium (mg/L)	7.58 (0.0001)	24–29	26	250-760	608.75
Aluminium (µg/L)	4.40 (0.0070)	20-40	31.43	160-670	453.3
Arsenic (µg/L)	4.90 (0.0043)	Bd.	_	3-10	7.3
Barium (µg/L)	3.65 (0.0354)	13–26	19.75	140-370	257.5
Cadmium (µg/L)	4.92 (0.0027)	Bd0.1	0.06	0.1-0.2	0.17
Copper (µg/L)	4.35 (0.0074)	Bd.	_	3–13	8.5
Iron (µg/L)	0.48 (0.6398)	10-520	240	20-580	303.5
Lead (µg/L)	3.13 (0.0259)	Bd.	_	2–7	3.5
Manganese (µg/L)	0.09 (1.810)	34–320	159.14	23-410	150.7
Nickel (µg/L)	5.30 (0.0032)	Bd1	0.64	45-150	113.8
Strontium (µg/L)	3.58 (0.0373)	17–24	20.5	88-220	155
Zinc (µg/L)	3.95 (0.0180)	5–9	6.86	13–48	33.17

Results for Student's *t* test and *p* values are provided (significant differences appear in bold), along with the range and mean for each variable *EC* electrical conductivity, *Bd*. below detection

Both mine drainage inflows changed metal concentrations in both receiving waterways (Figs. 6 and 7). Downstream of the Canyon Mine discharge the three metals with the highest mean concentrations in Dalpura Creek was manganese (360 µg/L), then iron (324 µg/L) and thirdly zinc (319 µg/L) (Table 3). The corresponding three highest metals in Georges River below West Cliff coal mine was aluminium (453 µg/L), iron (303 µg/L) and barium (257 µg/L) (Table 2). In both cases, the increased concentrations of metals in the receiving water reflected the same order of metal concentrations in the mine wastes.

Increased concentrations of selected chemical attributes provided an estimate of the contribution of each mine waste discharge to downstream receiving waterway flows. Georges River had no detectable concentrations of carbonate, copper and arsenic and had very trace levels of nickel. The comparison of concentration of these attributes in mine waste compared to downstream indicated that West Cliff mine contributed between 74.6 and 82.4 % of the flow in Georges River, below the waste inflow (Tables 2, 3 and 4). A similar comparison was made for Dalpura Creek (using nickel and bicarbonate) indicating that the Canyon Mine drainage contributed 87.1–90.9% of the flow in Dalpura Creek, below the mine (Tables 2, 3 and 4).

The ratio of major anion (chloride/bicarbonate) results for all samples (compared to TDS) was placed on models that were introduced by Gibbs (1970) to represent the broad diversity of dissolved salts across the world's range of surface freshwaters. The receiving 

 Table 3
 Comparison of water quality variables from Dalpura Creek, collected upstream (DUS) and downstream (DDS) of the coal mine waste discharge

Source of variation	t statistic (p value)	Upstream (DUS)		Downstream (DDS)	
		Range	Mean	Range	Mean
pH (pH units)	0.053 (0.9581)	5.01-6.21	5.75	5.22-6.28	5.76
EC (µS/cm)	18.0 (<0.0001)	26.4-36.9	33.07	87-121.3	108.05
Water temperature (°C)	9.5 (<0.0001)	9.1-12.4	10.67333	13.5-16.1	15.13
Turbidity NTU	1.66 (0.1090)	3.72-0.46	1.746	1.37-5.21	2.51
Carbonate (mg/L)	-	Bd.	-	Bd.	-
Bicarbonate (mg/L)	17.1 (<0.0001)	Bd.	-	13–18	16.14
Chloride (mg/L)	3.3 (0.0163)	4–7	6	3–4	3.86
Sulphate (mg/L)	11.4 (<0.0001)	1–2	1.67	18–29	24
Calcium (mg/L)	14.2 (<0.0001)	Bd.	-	4-5.9	5.1
Potassium (mg/L)	9.40 (0.0002)	Bd.	_	1.4–2.2	1.82
Magnesium (mg/L)	14.2 (< <b>0.0001</b> )	0.6-0.7	0.62	5.5-7.8	6.88
Sodium (mg/L)	4.28 (0.0079)	3.6-5.4	4.57	3–3.3	3.13
Aluminium (µg/L)	5.2 (0.0001)	130-280	205	5-90	41.4
Arsenic (µg/L)	-	Bd.	-	Bd.	-
Barium (µg/L)	7.2 (0.0004)	6–10	7.75	20–28	23
Cadmium (µg/L)	1.38 (0.2103)	Bd0.2	0.08	0.1-0.2	0.13
Copper (µg/L)	1.58 (0.1747)	Bd1.0	0.67	Bd.	-
Iron (µg/L)	5.5 (0.0003)	19–130	88.2	230-460	324.3
Lead (µg/L)	-	Bd.	-	Bd.	-
Manganese (µg/L)	12.6 (<0.0001)	13–28	17.8	290-470	360
Nickel (µg/L)	12.0 (<0.0001)	Bd.	_	140-220	182.9
Strontium (µg/L)	9.6 (0.0002)	4-5.5	4.62	16–22	18.2
Zinc (µg/L)	11.0 (<0.0001)	2-8	4.33	240-400	305

Results for Student's *t* test and *p* values are provided (significant differences appear in bold), along with the range and mean for each variable *EC* electrical conductivity, *Bd*. below detection

water samples from upstream of both mines clustered together in the Gibbs sector associated with 'precipitation dominance'. The samples from the mine wastes and from the receiving waters downstream of the mine wastes diverged and formed separate clusters from the two locations in the 'rock dominance' sector of the Gibbs (1970) model, largely separated by differences in salinity (Fig. 8).

### 4 Discussion

This study found that wastewater discharges from two underground coal mines in the Sydney basin both strongly modified the geochemistry of their respective receiving waters. Although the water chemical attributes of the mine wastes differed, the extent to which each mine discharge changed receiving stream water quality was considerable despite only one mine being active and the other being closed for more than 17 years. Due to the waste discharges, the water chemistry of both receiving waterways contained several contaminants at levels likely to be hazardous for aquatic ecosystems.

Both coal mines increased the electrical conductivity (EC) of their receiving waters. The background (upstream) EC was much higher in Georges River (mean 173  $\mu$ S/cm) than in Dalpura Creek (mean 33  $\mu$ S/cm) which largely reflects West Cliff mine having greater proximity to the coast (c. 16.5 km) compared to Canyon Mine (c. 97.1 km). Natural stream ionic composition, and salinity, in coastal flowing streams in southeastern Australia is strongly influenced by distance

 Table 4
 Comparison of water quality variables from mine wastewater samples collected from the West Cliff Mine and the Canyon coal mine immediately prior to their discharge to local receiving waterways

	t statistic (p value)	West Cliff Mine		Canyon coal mine	
Source of variation		Range	Mean	Range	Mean
pH (pH units)	5.81 (<0.0001)	6.1–9.11	7.94	5.51-6.33	5.83
EC (µS/cm)	30.50 (<0.0001)	1937-2510	2319	111.1-122.5	116.9
Water temperature (°C)	0.79 (0.4427)	12.1-15.7	15.1	15.7-16.1	15.88
Turbidity NTU	11.7 (<0.0001)	4.41-8.21	6.0	1.26-1.96	1.55
Carbonate (mg/L)	4.7 (0.0033)	36-140	92.9	Bd.	-
Bicarbonate (mg/L)	21.4 (<0.0001)	810-1100	938.6	18-17	17.75
Chloride (mg/L)	19.2 (<0.0001)	130-170	151.4	4	4
Sulphate (mg/L)	1.9 (0.1071)	27–42	33.8	29–30	29.5
Calcium (mg/L)	4.90 (0.0012)	6.2–6.7	6.4	5.6-6	5.8
Potassium (mg/L)	5.12 (0.0067)	3.5-6.2	4.6	1.9–2.3	2.02
Magnesium (mg/L)	34.8 (<0.0001)	2.7-3.2	3	7.5–7.7	7.8
Sodium (mg/L)	14.9 (0.0001)	590-810	710	2.9-3.2	3
Aluminium (µg/L)	9.03 (0.0001)	320-710	557.1	5-10	6.25
Arsenic (µg/L)	10.45 (0.0001)	6–11	8.86	Bd.	-
Barium (µg/L)	30.16 (<0.0001)	340-390	365	27–28	27.7
Cadmium (µg/L)	0.67 (0.5410)	0.1-0.3	0.17	0.1-0.4	0.22
Copper (µg/L)	11.85 (<0.0001)	9–14	11.43	Bd.	-
Iron ( $\mu$ g/L)	0.13 (0.2690)	24-700	439.1	250-470	310
Lead (µg/L)	5.60 (0.0014)	2–8	5.14	Bd.	-
Manganese (µg/L)	6.89 (0.0002)	24-360	74.6	380-440	417.5
Nickel (µg/L)	6.99 (0.0001)	110-170	138.6	200-220	210
Strontium (µg/L)	19.4 (<0.0001)	190–240	211.7	21–22	21.7
Zinc (µg/L)	23.0 (<0.0001)	25–50	38.6	350-410	370

Results for Student's *t* test and *p* values are provided (significant differences appear in bold), along with the range and mean for each variable *EC* electrical conductivity, *Bd*. below detection

to the coast due to natural atmospheric fallout of sodium and chloride ions which is proportional to the distance from the ocean (Ayers and Gillett 1984; Hart and McKelvie 1986; Wright et al. 2011a; Tippler et al. 2012). The scale of the EC increase was relatively modest in Dalpura Creek with the mine drainage (mean 117  $\mu$ S/cm) causing a rise in downstream EC by 3.3 times that of upstream (mean 33.1  $\mu$ S/cm) to 108  $\mu$ S/ cm, below the mine. These results indicate that the mine drainage inflow to Dalpura Creek is having a comparatively mild increase in EC and that the increase in EC ranks as comparatively low when compared to other published coal mine studies. These results are very similar to the mean EC (107 µS/cm) reported from Appalachian (USA) streams in areas of low mining intensity (Kittanning) (Petty et al. 2010). Slightly higher EC levels resulting from the input of coal mine wastes to otherwise dilute waters were reported from a study in northeastern Spain where EC in non-mined streams ranged from 18 to 34  $\mu$ S/cm increasing more than fivefolds to 202–472  $\mu$ S/cm below coal mines (García-Criado et al. 1999).

The EC of West Cliff mine waste, and its resulting increase in Georges River EC, was large by local and international comparisons. The Georges River EC increased due to inflow of mine waste (mean 2319  $\mu$ S/cm) lifting EC by 12.4 times that of upstream (mean = 173  $\mu$ S/cm) to 1628  $\mu$ S/cm below the mine. This is well in exceedance of the ANZECC (2000) water quality guide-line for protection of ecosystems. For coastal NSW rivers, the recommended EC level is less than 300  $\mu$ S/cm (ANZECC 2000). The Tahmoor Coal Mine, located

Fig. 2 Mean concentration of major anions collected from Georges River upstream (GUS) and downstream (GDS) and West Cliff Mine (WM)



about 20 km west of West Cliff Mine, increased the EC of the Bargo River by about five times, from 186 µS/cm upstream to 1078 µS/cm downstream, due to the inflow of Tahmoor coal mine wastes (Wright et al. 2015). West Cliff mine increased EC in its receiving waters to higher levels than any waterways that received mine drainage in Durham coalfields (UK) where they recorded a maximum EC of 2039 µS/cm (Jarvis and Younger 1997). The maximum level of EC (2451 µS/cm) recorded from Georges River downstream of West Cliff Mine was slightly lower than the maximum EC recorded (2846 µS/cm) in a coal mine impact study of 91 stream sites in New Zealand (Gray and Harding 2012). EC levels higher than those that were recorded in West Cliff Mine wastewater were reported from four 'pumping' coal mines in Durham and Kent in Britain where EC levels ranged from 2850 to 5050 µS/cm (Banks et al. 1997).

The higher EC in West Cliff Mine wastewater is typical of groundwater from coal seams in the southern coalfield (Volcich 2007). For example, a study of groundwater in the Dendrobium mine, about 15 km southeast of West Cliff Mine, reported EC levels entering the mine with a median EC of 2730 µS/cm (Nicol et al. 2014). Even higher EC was recorded (3170 to 4200 µS/cm) from an 18-month study of West Cliff Mine drainage, at the point, it was pumped to the surface (Volcich 2007). Canyon coal mine drainage recorded a much lower EC, which was also recorded at the Clarence Coal Mine that also mines the same seam (Katoomba Seam) about 15 km northwest of Canyon Mine. The Clarence Coal mine is an operating mine and was subject to a detailed water quality study that investigated the water chemistry impact of the mine operation on natural groundwater inflows (Cohen 2002). Water chemistry data was collected within the Clarence Mine by Judell and Andreson (1988) and was reported by Cohen (2002). Water samples flowing from the roof of the mine were much more dilute (EC 40.4–74.3  $\mu$ S/cm) compared to water samples collected in the goaf area disturbed by mining (EC 127-789 µS/cm).

Fig. 3 Mean concentration of major anions collected from Dalpura Creek upstream (DUS) and downstream (DDS) and Canyon coal mine (CCM)



Fig. 4 Mean concentration of major cations collected from Georges River upstream (GUS) and downstream (GDS) and West Cliff Mine (WM)

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In the current study, the impact of coal mine waste discharges either increased receiving water pH or it was neutral. Large differences in pH were detected in Georges River below the waste discharge. The pH of the river increased from an upstream mean pH of 6.90 to 8.05 downstream of the mine waste inflow (Table 1). The mine wastes from West Cliff Mine had a mean pH of 7.94, but there was a large fluctuation in pH detected (ranging from pH 6.1 to 9.1) over the study. Water from the Westcliff mine was reported to have similar pH in an earlier study by Volcich (2007), who recorded a mean pH of 8.65 over an 18-month study of West Cliff Mine water as it was pumped to the surface. In comparison, other coal mine discharges have had different pH impacts. A larger increase in pH from a mine waste discharge into the Wollangambe River (Blue Mountains, Australia) from the Clarence Coal Mine lifted upstream pH from 5.6 to a pH of 8 below the mine (Belmer et al. 2014). An early study of water quality within that mine operation attributed the high pH of the waste discharge to lime dosing treatment of the mine wastes (Cohen 2002). Cohen (2002) reported pH data from Clarence Coal Mine water (collected by Juddel and Anderson 1988) in disturbed areas of the mine with pH of 3.0 to 4.2. The mean pH of Dalpura Creek remained almost identical upstream (5.75) compared to downstream (5.76). The pH of Canyon Mine and West Cliff could be expected to be much lower if acid mine drainage was a problem, but coal from both the Katoomba Seam and the Bulli Seam has been tested and found to have very low sulphur concentrations (Huleatt 1991). This differs from other global examples where non-operational coal mines have continued to release highly acidic wastes to receiving waters. For example, the Green Valley coal mine in Indiana (USA) ceased mining in 1962 and received extensive site rehabilitation yet mine waste discharges are still highly acidic (pH 2.2 to 4.6) 37 years after the mine closed (Brake et al. 2001). Studies on abandoned coal mines in the UK also report strongly acidic mine drainage discharges of pH 3.6 (R. Hipper





Fig. 6 Mean concentration of metals (>5  $\mu$ g/L) collected from Georges River upstream (GUS) and downstream (GDS) and West Cliff Coal Mine (WM)



Discharge) and 4.2 (Ynysarwed) (Banks et al. 1997). The southern hemisphere also has highly acidic water problems from closed coal mines (with pH levels 2.6 to 4.2) reported in streams in a New Zealand study of abandoned coal mines (Winterbourn 1998).

Waste discharges from the two coal mines in this study had different ionic composition, and each modified the ionic composition of its receiving waters differently. One common factor between the two locations was that background water chemistry in each receiving waterway, above both mine waste discharges, was dominated by sodium cations and chloride anions. This is typical of water chemistry in coastal flowing streams in southeastern Australian (Hart and McKelvie 1986). It also corresponds to the typically acidic and dilute stream water chemistry in naturally vegetated and unpolluted catchments across the Sydney basin which are also broadly dominated by sodium and chloride ions (Hayes and Buckney 1995; Davies et al. 2010; Wright et al. 2011a; Tippler et al. 2014; Belmer et al. 2014). Sodium remained the dominant cation in Georges River, below the mine drainage inflow, but the concentration and degree of dominance over the other cations increased substantially.

The two coal mine discharges changed the ionic composition of both receiving waterways. The order of the cation concentrations in Georges River changed from Na > K > Mg = Ca above the mine to Na>Ca>K>Mg downstream. The mine waste discharge also changed the order of anion dominance in Georges River from Cl>HCO<sub>3</sub>>SO<sub>4</sub> above the mine waste inflow to HCO<sub>3</sub>>Cl>CO<sub>2</sub>>SO<sub>4</sub> downstream. Georges River ionic results were very similar to those recorded in the nearby Bargo River (Wright et al. 2015), from Tahmoor Coal Mine wastes, suggesting the strong influence of mining and local geology on the resulting mine wastewater. Canyon Mine changed the order of ionic dominance in Dalpura Creek from Na>Mg and  $Cl > SO_4$  above the mine to Mg > Ca > Na > K and  $SO_4 > HCO_3 > Cl$  downstream of the mine inflow. The







**Fig. 8** Gibbs (1970) Diagram showing the ratio of Cl/ $(Cl+HCO_3)$  compared to TDS (mg/L) in water samples collected in this study. *Square symbols* represent samples from the receiving waterways upstream of the mines. *Triangles* are from receiving water downstream of the mines. *Yellow* is Dalpura and *green* is Georges. *Diamond symbols* represent the mine drainage. *Red* is Canyon coal mine and *Blue* is West Cliff mine

source of the modified ionic composition in the wastewater from each mine is at least partly attributed to the mining activity disturbing the local geology at each mine. A previous study of West Cliff Mine found that it has mine wastewater dominated by carbonates, which, at the typical pH of mine wastewater, were generally present as bicarbonate (Volcich, 2007). Analysis of coal from the Katoomba Seam reported that it had much higher concentration of calcium oxide (6.27 %) compared to coal from the Bulli Seam (0.55-0.93 %) according to Huleatt (1991). The operation of underground coal mining has also been linked to increasing calcium and magnesium concentrations in Katoomba Seam groundwater at the nearby Clarence mine (Cohen 2002). Both mines strongly differ from a case study in Indiana (USA) where background ionic composition was dominated by Ca>Mg>Na>K cations and  $HCO_3 > SO_4 > Cl$  anions (Brake et al. 2001). In that study, the mine drainage did not have a different order of cation dominance to local unaffected waterways, but anion dominance of mine waters changed with SO<sub>4</sub> becoming the dominant anion with HCO<sub>3</sub> not detected due to very low pH (<4.6; Brake et al. 2001).

The bicarbonate concentrations recorded in Georges River in this study, below the coal mine discharge, were as high as 990 mg/L and averaged 743 mg/L. An earlier investigation (in 2012) of this mine and the effects of its waste discharges to Georges River identified slightly lower concentrations of bicarbonate as being toxic to aquatic ecosystems (NSW OEH 2012). A further laboratory study by NSW OEH scientists was published on this topic that found such concentrations of bicarbonate, in the presence of elevated EC and zinc, were toxic to test species in ecotoxicology assays (Vera et al. 2014). However, currently, there are no water quality guidelines for bicarbonate concentrations in Australia (ANZECC, 2000).

The anions (chloride and bicarbonate) were plotted against total dissolved solids on the model developed by Gibbs (1970) to enable comparison of the mine and receiving waters to the range of total dissolved salts across the world's range of surface freshwaters (Fig. 8). The receiving waters upstream of both mines clustered in the 'precipitation dominance' sector reflecting the natural chloride dominance of both waterways, which is typical of waterways flowing in natural and undisturbed catchments of the Sydney area (Wright 2012; Tippler et al. 2014). Samples downstream of both mines clustered separately from upstream samples indicative of the strongly contrasting salinity and chloride/ bicarbonate ratios provide by each mine waste inflow. The mine water and receiving water samples collected downstream of the mines clustered closer to 'rock dominance' zone in the Gibbs (1970) anion (chloride and bicarbonate) model. Comparing results from this study to the Gibbs model provides a clear interpretation showing that water chemistry disturbed by coal mining can provide an unnatural geological effect on downstream major anion chemistry and salinity.

Drainage from Canyon Mine, and downstream in Dalpura Creek, were the only waters in the current study where anions were dominated by SO<sub>4</sub>. Highly elevated concentrations of SO<sub>4</sub> in coal mine wastewaters are commonplace throughout the world. For example, coal mines in a West Virginia (USA) study increased the mean SO<sub>4</sub> concentration of 695 mg/L in streams and the maximum concentration detected was 1520 mg/L (Pond et al. 2008). High sulphate levels have been reported from coal mine wastewaters in Britain where one 'pumped mine' in Durham had a sulphate concentration of 1170 mg/L (Banks et al. 1997). Extreme concentrations of SO<sub>4</sub> were reported from an Indiana

(USA) coal mine study which had concentrations of up to 63,100 mg/L (Brake et al. 2001). The SO<sub>4</sub> concentrations in mine wastes in the current study are much lower than the levels reported from most international studies. The highest sulphate concentration recorded in the current study of 42 mg/L in the Westcliff mine wastewater is quite low compared to other international studies. The comparatively low SO<sub>4</sub> concentrations in mine waters in this current study help explain why pH in both mine waters was never strongly acidic. The low SO<sub>4</sub> concentrations suggest that the coal ore and tailings in both mine workings at both sites had a relatively low abundance of sulphidic minerals. The sulphur content of coal is regarded as a key factor contributing to the degree of acid mine drainage from coal and other mines (Johnson 2003). Australian coal has an international reputation as having a low sulphur content, and the two mines in this study have both been assessed as having very low sulphur levels of 0.38 % (Huleatt 1991). Testing of coal samples from the Katoomba Seam has also identified extremely low pyritic forms of sulphur of 0.02 %, which is 10 to 100 times lower than the levels typically found in coal internationally (Cohen 2002).

Both of the mine discharges increased the concentration of metals in downstream receiving waters. Of most concern for the health of downstream aquatic ecosystems were the elevated concentrations of nickel and zinc, particularly from the closed Canyon coal mine and its receiving water in Dalpura Creek. Both nickel and zinc can be toxic to aquatic ecosystems at relatively low concentrations, and both were at concentrations (in receiving waters at both sites) greater than levels recommended for protection of riverine species in Australian water quality guidelines for aquatic ecosystems (ANZECC 2000). The ANZECC (2000) guideline for protection of 99 % of freshwater ecosystem species is 2.4 to 21.6  $\mu$ g/L for zinc and 8 to 72  $\mu$ g/L for nickel, depending on the level of water hardness. Inflow of wastewater from Canyon Mine increased the concentrations of zinc and nickel in Dalpura Creek to levels well above the ANZECC (2000) guidelines. Canyon Mine drainage increased the mean zinc concentration in Dalpura Creek by more than 70 times, from a mean of 4.3  $\mu$ g/L (upstream) to 318  $\mu$ g/L below the Canyon Mine waste inflow. Nickel was not detected in Dalpura Creek above the mine and the mean concentration was 182.9 µg/L below the mine. The levels of zinc are particularly high in the Australian context, with only one coal mine-affected waterway (Neubecks Creek, Lithgow, NSW) having about a similar concentration of 300  $\mu$ g/L below a derelict coal mine (Battaglia et al. 2005). International coal mine studies have detected zinc at much higher concentrations. For example, zinc in mine drainage from the closed Green Valley mine (Indiana, USA) had zinc levels of up to 13.3 mg/L (Brake et al. 2001). A higher concentration of zinc (19.7 mg/L) was recorded below coal mines in Newhouse Burn (Durham, UK) by Jarvis and Younger (1997).

An earlier study of Grose River found that the inflow of Dalpura Creek (about 2 km further downstream from the current study sites) into Grose River caused a plume of elevated zinc concentrations, linked to ecological impairment of macroinvertebrate communities, in the high conservation-value Grose River for more than 10 km below the inflow (Wright and Burgin 2009a). That study collected samples less than 6 years after the mine closed and 11.5 years before the current study and recorded zinc levels in Dalpura Creek ranging from 545 to 650 µg/L (Wright and Burgin 2009a). Although weather conditions and river flows could have influenced the results, zinc concentrations in Dalpura Creek in this study are almost half the concentrations recorded 11.5 years earlier. We suggest that the decline in the zinc concentrations is due to the progressive and gradual dissipation of zinc from the mine and its environs.

Unlike many other examples overseas, such as the Green Valley mine (Brake et al. 2001), the mine drainage from the now closed mine in this study (Canyon coal mine) has not received any remediation to treat the contaminated drainage that continues to flow from the underground mine workings (Wright et al. 2011b). The mine was not held responsible for treatment of the mine drainage during operation of the mine due to waste discharge conditions in the EPA EPL 558 that permitted the discharge of untreated waste when the mine was operating. The rehabilitation of the mine site by the former owner has concentrated on surface disturbances rather than treatment of the mine contamination that continues to discharge from the mine (Wright et al. 2011b). The contaminated Dalpura Creek flows into Grose River where an earlier study recorded major damage to aquatic ecosystems for many kilometres of river in an area of high environmental significance. Grose River is within a declared Wilderness Area, a NSW National Park and a part of the Greater Blue Mountains World Heritage Area (Wright et al. 2011b; Graham and Wright, 2012).

Both mines in the current study could have discharged less polluting wastewater with more thorough treatment of the mine drainage. There are numerous coal mine waste treatment technologies available (Johnson and Hallberg 2002; Coulton et al. 2003; Johnson and Hallberg 2005). Mine waste treatment systems that use passive treatment such as constructed wetlands (Mays and Edwards 2001), biochemical reactors (Watten et al. 2005; Neculita et al. 2007) or anoxic limestone drains (Skousen 1991) could be suitable for remote locations, such as the treating the ongoing mine drainage from the Canyon coal mine.

This current study provides detailed information on the water chemistry impact of the discharge of coal mine wastewater to local receiving waterways. In both cases, the two mines discharged their waste to waterways that were typical of natural 'reference' waterways flowing in largely undisturbed naturally vegetated catchments. This study used an 'upstream versus downstream' design and included comparison to the waste discharge itself to enable a three-cornered 'triangulated' assessment of the impact resulting from each mine waste discharge. The two mines in this study shared many similarities. They both strongly modified the water chemistry of downstream waters. Both discharges increased receiving water EC and modified the ionic composition, relative to the unaffected upstream waters. Both mines also increased the concentration of a different suite of metals. The mine waste discharges also modified receiving water chemistry in ways that are potentially hazardous to aquatic ecosystems. Elevated EC, bicarbonate and elevated metals (zinc and nickel in particular) were often recorded at levels likely to be ecologically stressful for aquatic ecosystems. This study also provides a reminder that contamination from coal mines is a major environmental issue for both active and derelict coal mines and that contamination from derelict mines may continue to occur for decades after mining has stopped.

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