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Assessment, regulation and management of water pollution from underground coal mines in the Sydney Basin.

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Summary

This research surveys water quality and chemistry, river sediment chemistry and aquatic macroinvertebrates in eight streams that are the recipient of coal mine wastewaters from seven collieries across the Sydney Basin. The study also surveyed water quality and chemistry at five of the seven collieries wastewater discharge streams (effluent streams). Most of the coal mine wastewater discharges are licensed and regulated by the New South Wales Environment Protection Authority (NSW EPA) through a series of Environmental Protection Licences (EPL's). During this study four of the collieries were actively mining ore, while three were inactive. One of these had permanently ceased mining more than 20 years previously. Results revealed that all seven mines caused mild to significant modifications to a plethora of water quality and water chemistry parameters, river sediment chemical parameters and macroinvertebrate community structure. Many of the pollutant concentrations recorded in this survey were above, or outside of their respective ANZECC 2000 guidelines for protection of aquatic ecosystems. pH and salinity (electrical conductivity) were often above the recommended guidelines and substantially differing from their background conditions.

Of widespread concern are the elevated concentrations of the metals nickel and zinc, with the majority of samples collected from coal mine wastewater streams and downstream of their inflow to rivers well above the recommended ANZECC 2000 guidelines. Iron and manganese were also found to be of elevated ecological concern with aluminium, cadmium, chromium, copper and lead also often above the recommended ANZECC 2000 guidelines for protecting ecosystems. Nickel and zinc were both recorded at their highest concentrations in the discharge waste streams and downstream sample locations at both inactively mined Canyon Colliery and Berrima (Medway) Colliery. Such levels of nickel and zinc are of particular concern, especially as the highest results of this study are from inactive, untreated and uncontrolled mine wastes one of which (Canyon Colliery) ceased mining some 20 years prior to this study.

Macroinvertebrates were collected from upstream and downstream of all eight mine wastewater discharges to measure the impact of the coal mine wastewaters on aquatic ecosystems. Results of the study show that the coal mine wastewaters being discharged were having varying negative impacts to the receiving waterways aquatic ecosystem that this research quantified through assessment of aquatic macroinvertebrates. The macroinvertebrate results showed that impacts were detected whether mining was active or inactive and this research developed a biotic index to enable comparison of the severity of the ecological impact. Macroinvertebrate biotic indices measured at active and inactive coal mines show that actively mined wastewaters are most likely causing lesser impacts to the receiving waterways aquatic ecosystem than inactively mined wastewaters. All of the inactively mined locations recorded statistically significant differences for all macroinvertebrate biotic indices used when compared between upstream and downstream sample locations.

This contrasted with the actively mined locations with only three of the five streams sampled recording negative statistical differences for all macroinvertebrate biotic indices. Results suggest that once mining ceases and the treatment of the coal mine wastewaters subsequently ceases, the receiving waterways can become more degraded and in turn negatively impact the receiving waterways ecosystem to a larger degree. This is of concern as once

mining ceases so does the treatment of their wastewaters. It is recommended that the NSW EPA further investigate measures of treatment post coal mining to ensure further degradation of the receiving waterways does not occur.

Part of this research assessed the bioaccumulation of many metals within one predacious aquatic beetle (Gyrinidae) and one species of terrestrial (riparian) flora (*Acacia rubida*) from a single coal mine wastewater discharge (Clarence Colliery). The bioaccumulation study was undertaken within the near-pristine Wollangambe River and its surrounding tributaries. The majority of the Wollangambe River flows within the World Heritage Greater Blue Mountains National Park and is protected through many layers of legislation from state to Commonwealth and international (Threatened Species Conservation Act 2005, Environment Protection and Biodiversity Conservation Act 1999, United Nations Educational, Scientific and Cultural Organization 2000). Bioaccumulation results show that ten pollutants (mostly metals) were detected at higher concentrations within aquatic beetles collected from the Wollangambe River in the protected conservation area (National Park and World Heritage Area), downstream of the coal mine waste discharge. The ten metal concentrations were at higher concentrations and were highly significantly different compared to metal concentrations in beetles collected from streams not affected by coal mine wastes. In comparison, nine of the ten metal contaminants tested in the flora samples were found to be at higher concentrations (statistically significantly different) when compared between downstream of waste discharge (coal mine impact) and reference conditions.

The bioaccumulation research has allowed for a better understanding of the broader ecological impacts that heavy metal contamination from Clarence Collieries coal mine wastewaters are having on the biota in the surrounding aquatic and terrestrial ecosystems. This has major implications as the wastewater contaminants are bioaccumulating within the aquatic biota in a highly ‘protected’ conservation area. This finding also validates the IUCN concern that ‘discharge of polluted water from collieries into rivers’ is a considered to be a high threat to the outstanding universal value of the Blue Mountains World Heritage Area (IUCN, 2020). This bioaccumulation research also raises major concerns that coal mine wastes may be causing widespread and unregulated metal bioaccumulation within waterways that receive coal mine wastes. Of particular concern is that coal mine contaminants appear to be moving from the contaminated river and are mobilising into vegetation in the riparian terrestrial environment. This research finding (bioaccumulation of coal mine contaminants in riparian vegetation) has not been previously documented in Australia and raises concerns regarding the impact of terrestrial pollutants from active and closed mines. It is recommended that further research be undertaken by the NSW EPA to better assess the implications of heavy metal bioaccumulation and or biomagnification from coal mining wastewater discharges on waterways throughout the state. The results of such work could be used in future to improve the environmental effectiveness of coal mine regulation processes.

A final outcome of this research was the ability to generate preliminary biotic indices for coal mine impact grading using aquatic macroinvertebrates. Macroinvertebrate biotic indices are a well-developed and robust method to assist in the rapid assessment of anthropogenic impacts on aquatic ecosystems and have been used since the early 1980’s (Washington 1984, Chessman 2003). The SIGNAL method (Stream Invertebrate Grade Number Average Level) is a

widely used methodology employed to assess the level and degree of impacts from anthropogenic influences in South Eastern Australia (Chessman 2003).

In this research, 12886 individual macroinvertebrates, the majority of which were identified to the family level (55 families) across eight different waterways have been used to derive a novel Coal Mine Impact Grade (CMIG) biotic index. Results of the CMIG/SIGNAL comparison show the potential benefits of using such a grading system as a robust, relatively low impact and rapid method for the assessment of coal mine wastewater impacts to the receiving waterways aquatic ecosystem. Coal mine wastewater impacts to receiving waterways are regulated by the New South Wales Environmental Protection Authority to ensure the protection of the receiving waterways aquatic ecosystem, though the regulation and subsequent protection is performed through water quality and chemical concentrations. The EPA regulations do not require the coal industry to routinely assess the impact of coal mines wastes directly on the aquatic ecosystem with the majority of regulation focussed on water quality and a range of water pollutants. The CMIG macroinvertebrate grades are the only grading system that assess the impact of coal mine wastewater discharges to a receiving waterways aquatic ecosystem and would provide a much better monitoring system than generalised water column pollutant concentration limits.

Six of the eight chapters presented in this thesis have been published, five of which are published in peer-reviewed scientific journals and one is published in a peer-reviewed conference proceeding. One chapter is being prepared for submission. Due to each chapter being published individually, consequently sections such as introduction, methods (especially sample locations etc) and discussion sections of each of the published manuscripts are somewhat repeated. Chapters are offered in order of publication date. Cover image shows Berrima (Medway) Colliery wastewater discharge stream (orange) just prior to entering the Wingecarribee River (Nakia Belmer 2017).

Acknowledgements

I would like to acknowledge and pay my respect to the traditional custodians of the land in which this research was performed within. The Dharug, Gundungurra, Tharawal, Wiradjuri and Yuin people and their elder's past and present. I would like to thank my family, especially my parents Nada Perisic and Jon Colin Belmer, my Grandparents Gertraud Juliane Behring, Zivan Perisic, Colin Charles Belmer and Phyllis Florence Samuel, my Brother Jason and my better half Amreen Charania for all your support. The majority of this research was supported through an Australian Government Research Training Program Scholarship. I also acknowledge the field work assistance of Nicholas Szafraniec, Mathew Noel, Ben Green and Paul Hammond. Special thanks to Dr Ian Wright for his patience, mentoring, motivation and friendship through my undergraduate and postgraduate studies and Rani Carrol for her editing and constructive review.

Statement of Authentication

The work presented in this thesis is, to the best of my knowledge and belief, original except as acknowledged in the text. I hereby declare that I have not submitted this material, either in full or in part, for a degree at this or any other institution.

A black rectangular box redacting the signature of the author. A small blue mark is visible to the right of the box.

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(Signature)

Preface

This thesis is presented in nine chapters. The first being an introduction followed by seven chapters that are independent manuscripts published within peer-reviewed, scientific journals (five) and two peer-reviewed conference proceedings. The final chapter is a conclusion chapter.

Journal

Chapter 2, Increased Water Pollution After Closure of Australia's Longest Operating Underground Coal Mine: a 13-Month Study of Mine Drainage, Water Chemistry and River Ecology. *Water, Air, Soil Pollution*, Springer 2018.

Conference proceedings

Chapter 3, Heavy metal contamination of water column from a coal mine waste water discharge resulting in mobilisation of metal contaminants to riparian vegetation. Wollangambe River, Blue Mountains Australia. *Proceedings of the 9th Australian Stream Management Conference*, Hobart Tasmania 2018.

Journal

Chapter 4, The regulation and impact of eight Australian coal mine waste-water discharges on downstream river water quality: a regional comparison of active versus closed mines. *Water and Environment Journal*, Wiley 2019.

Journal

Chapter 5, Regional comparison of impact from seven Australian coal mine wastewater discharges on Downstream River Sediment Chemistry, Sydney Basin, New South Wales Australia. *American Journal of Water Science and Engineering*, Science Publishing Group 2019.

Journal

Chapter 6, Regional comparison of the impact on stream macroinvertebrates from Australian underground coal mine wastewater discharges from active and inactive mines in the Sydney Basin, New South Wales Australia. *American Journal of Water Science and Engineering*, Science Publishing Group 2019.

Journal

Chapter 7, Regulated coal mine wastewater discharge contaminants accumulating in an aquatic predatory beetle (*Macrogyrus rivularis*) Wollangambe River, Blue Mountains New South Wales Australia. *American Journal of Water Science and Engineering*, Science Publishing Group 2019.

Conference proceedings

Chapter 8, Grading Coal Mine Wastewater impacts to Aquatic Ecosystems, measured through Macroinvertebrate Diagnostic Biotic Indices. Accepted for publication (publication in August 2021), *Proceedings of the 10th Australian Stream Management Conference*, Kingscliff New South Wales 2021.



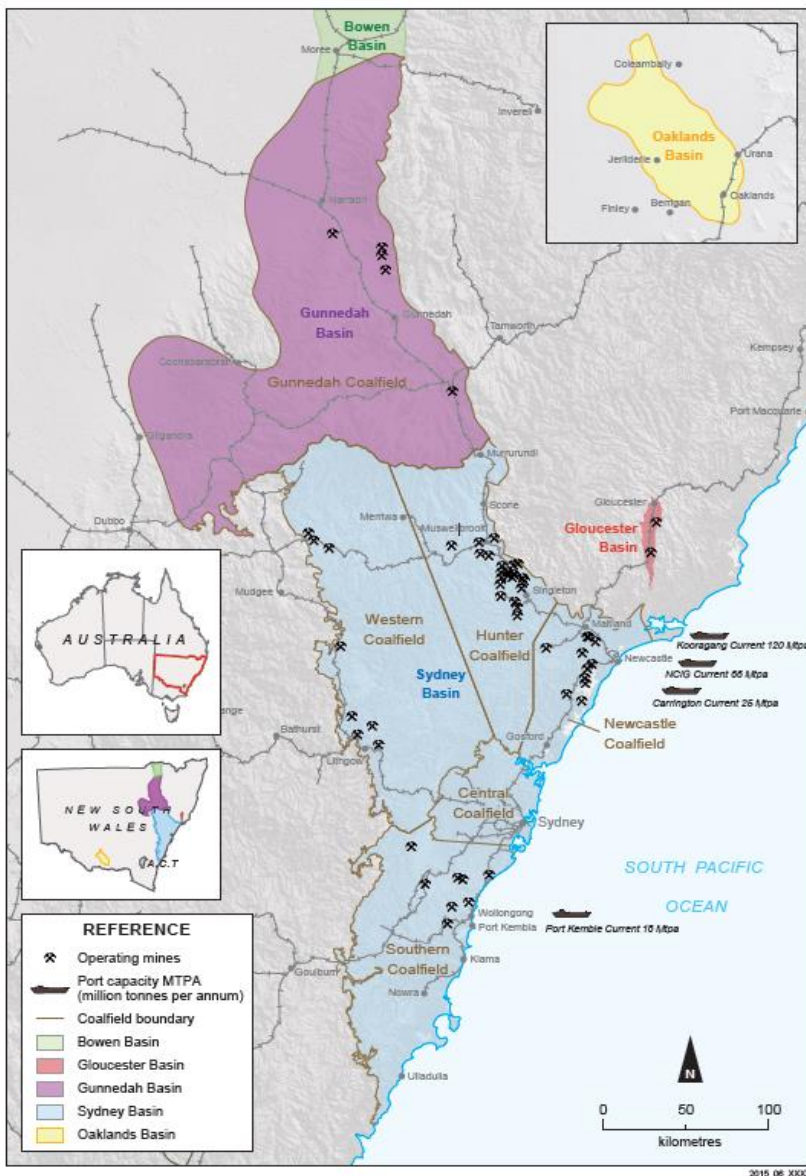
Figure 1 Berrima (Medway) Collieries coal mine wastewater stream just prior to entering the Wingecarribee River (Nakia Belmer 2017).

Chapter 1

Introduction

1. Coal in New South Wales

Coal was first observed in Australia in 1797 by survivors of a shipwreck and subsequently confirmed by Surgeon Bass. These resources were noted to be in the South Coast district and were not used at the time as they were deemed inaccessible. At a similar time, coal reserves were also discovered in the Hunter Valley region by Lieutenant Shortland, this region was referred to as the Northern district. These resources were found to be more easily accessed and mining of this ore started not long after its discovery. By 1881 coal was starting to be extracted at multiple locations and the South Coast district recorded 1,929,236 tons of coal, valued at £570,022 had been extracted in 1908 and the Northern district had extracted 6,511,002 tons, valued at £2,625,446 (Australian Bureau of Statistics 2022).



There are six distinct areas mined for coal ore in New South Wales: Mudgee, Hunter Valley, Newcastle, Gunnedah, Lithgow and Wollongong. The latter two, Lithgow and Wollongong are encompassed in this body of research. It should be noted that these two areas of mining only account for 7.5% of the state’s production pa.

Fast forward to the end of the century and coal production has become New South Wales’s largest natural resource commodity accounting for more than 75% of the total value of NSW mineral production estimated at more than 160 million tonnes of coal, generating more than\$ 23.1 billion in 2018-2019 (NSW Mining 2020). With an estimated Royalty revenue for the NSW state government between some \$169 million and \$181 million.

Figure 1. Map of coal mining districts of New South Wales (New South Wales Government 2022)

Table 1. Raw coal production in NSW 2020 to 2021 financial year. Raw tonnage (pa, per annum) for local areas mined, total tonnage for NSW, total tonnage used in this research and percent of production used in this research (New South Wales Mining 2022, Viewed March 2022).

Area	Raw coal production shown for 12 months ending 30 June 2021
Mudgee	47 tonnes
Hunter Valley	139 tonnes
Newcastle	8.6 tonnes
Gunnedah	28.6 tonnes
Lithgow	5.5 tonnes
Wollongong	12.7 tonnes
Total tonnage pa	241.4 tonnes
Tonnage used in this research pa	18.2 tonnes
Percentage of NSW mined coal used in this research pa	7.5%

2. Overview Coal mining and water pollution impacts

Coal mining practices are well documented to contribute to an array of different environmental problems including air pollution, fire hazards, ground subsidence or deformation, surface and or ground water pollution (Tiwary 2000). Surface water pollution is a major environmental problem associated with coal mining and it occurs through the deliberate or accidental discharge of mine waters that are contaminated by various processes and contaminants associated with mining practices (Jarvis and Younger 1997, Johnson 2003, Pond et al. 2008).

Water pollution from coal mining often occurs as large volumes of surface and groundwater, which are required to be removed from most underground coal mines. This is generally through the pumping of water to the surface, as surface and groundwaters infiltrate the mine shafts from the local geological sub-strata and subsequently accumulates in the underground mine workings. Without this, groundwater would flood most sections of the underground mining operation (Jarvis and Younger 1997, Younger 2004). This practice of coal mine wastewater discharges to the environment is licenced and regulated through the imposition of contaminant limits in New South Wales by the New South Wales Environmental Protection Authority (NSW EPA). The discharge of these contaminants is regulated through Environmental Protection Licences (EPL's) and legislated under the Protection of the Environment Operations Act 1997 (NSW Environment Protection Authority 2013).

Coal mine wastewater is often contaminated due to the disturbance of the local geology and groundwater associated with mining activities. The exact nature of the water contamination will vary depending on local factors such as groundwater geochemistry, hydrology and mineralogy of the local strata (Younger 2004). In addition to the physical activity of the mining operation and the removal of the wastewater, other activities will also often contaminate water used throughout a mining plant which can include; coal washing and the inclusion of other wastes generated by the surface operation at a mine such as sewage wastes (Younger 2004).

3. Water contamination from colliery wastes

Water pollution impacts attributed to treated coal mine wastewaters discharged to surface waters often includes changes to pH, elevated salinity, modified stream ionic composition and elevated heavy metals (Pond et al. 2008, Wright and Burgin 2009, Wright 2012, Belmer et al. 2014, Wright et al. 2015, Wright and Ryan 2016, Price and Wright 2016). Decreased water pH, termed acid mine drainage (AMD) was reported in the UK by Banks et al. (1997) and Younger (2001) after the closure of many coal mines. Other international examples of modified stream pH from closed coal mines was reported from the abandoned Green Valley Coal Mine in Indiana, North America by Brake et al. (2001) with pH as low as 2.2 pH units recorded.

International studies show that waterways receiving coal mine wastewater often have highly elevated concentrations of nickel and zinc (Brake et al. 2001, Pond et al. 2008 and Petty et al. 2010). Closed coal mines commonly produce mine drainage that is contaminated by elevated zinc and nickel for decades after mining ceases. For example, elevated levels of nickel and zinc were detected below the abandoned Green Valley coal mine (Indiana USA) more than 30 years after its closure, with nickel levels above 500 µg/L, and as high as 3780 µg/L and zinc often above 5000 µg/L (Brake et al. 2001). In the West Virginian coal fields Pond et al. (2008) reported salinity increased from 62 µS/cm in unmined streams to 1023 µS/cm downstream of mine discharges. Similar increases in salinity were also reported for the waterways affected by coal mine activity in the Freeport coal seam of the Appalachians (USA) where Petty et al. (2010) reported salinity increased from unmined areas (mean 98 µS/cm) increased at streams exposed to high intensity mining (mean 734 µS/cm). Banks et al. (1997) reported sulphate in the waters of abandoned UK coal mines ranging from 83 to 1554 mg/L. Similarly, Pond et al. (2008) reported much higher sulphate in mined West Virginia streams (mean 696 mg/L) compared to unmined streams (mean 16 mg/L) similar to research performed in the Appalachian Mountains (USA) which found sulfate in unmined streams at (mean 14 mg/L) increasing in intensively mined areas to a mean of 338 mg/L (Petty et al. 2010).

A widespread form of water pollution caused by coal and metalliferous mining is termed ‘acid mine drainage’ (AMD) and often occurs when wastewaters are not treated or when treatment ceases (Brake et al. 2001). This arises when sulphur in coal (or other ores) is oxidised due to the disturbance associated with mining, i.e. its exposure to both air and water, which triggers the formation of sulphuric acid of various strengths (Johnson 2003). The AMD acid leaches and mobilises differing metals within the mine wastewater, depending on the sulphur content of the ore and the characteristics of the surrounding geology (Banks et al. 1997, Brake et al. 2001, Johnson 2003).

Perhaps surprisingly, pollution from coal mines can increase after the commercial operation of a coal mine ceases (Cairney and Frost 1975, Banks et al. 1997 and Younger et al. 2002). Johnson (2003) explains that when the pumping of mine drainage from underground mines ceases, post mining, the groundwater can flood the mine and cause an increase in the concentration of pollutants, such as metals. This process has been termed ‘rebounding’ of mine water as the rising groundwater level increases until it emerges on the surface (Younger 1993 and Younger 2001). The progressive flooding of closed mines, with groundwater, can be a slow process. For example, coal mining ceased in the ore catchment of Scotland in 1967 and the rising level of mine drainage, from various surface locations, emerged between nine and 10 years later (Jackson 1981 reported in Younger 2001).

Coal mine wastewaters are regulated by the NSW EPA to ensure the protection of the receiving waterways aquatic ecosystem. This regulation of coal mine wastewaters focusses on the waste attributes at the 'end-of-pipe' waste discharge. Along with the NSW EPA method of "monthly focus" on a selection of colliery wastewater pollutant concentrations, these approaches do not fully take into account the impact of the waste discharges on the receiving waterways aquatic ecosystems.

4. Contamination of river sediments from colliery wastes

River sediments are also often heavily polluted from mine wastewater discharges as initially the heavy metals become water soluble once oxygenated and discharged through the treatment process, then depending on the nature of the metal, they can become less soluble, eventually falling out of the water column and accumulating in river sediments and contaminating them with many heavy metals (Cohen et al. 1998, Cohen 2002). This accumulation of contaminants in river sediments has broad impacts to the aquatic ecosystem. Battaglia et al. (2005) concluded that increased heavy metals in sediments contributed to the degradation of stream macroinvertebrate assemblages. Wright and Burgin (2009) reported elevated zinc (Zn) levels from drainage flowing from the closed coal mine (Canyon Colliery) impaired the downstream Grose Rivers aquatic ecosystem with reductions in macroinvertebrate taxonomic richness and abundance. A similar study by Belmer et al. (2014) reported that an active coal mine's (Clarence Colliery) wastewater discharge increased the Wollangambe River's salinity, pH, nickel (Ni) and zinc (Zn) levels which were concluded to have reduced macroinvertebrate taxonomic richness and abundance downstream of the mine discharge. These examples show that the regulation of coal mine pollution relies on water chemistry and key pollutants but not the health of aquatic ecosystems that receive the wastewater (Wright and Burgin 2009, Belmer et al. 2014).

5. Biodiversity impacts from colliery wastes

Macroinvertebrates are not the only ecological indicator measuring impacts from coal mine wastewaters as algal diatoms have also been documented to be impacted by treated coal mine wastewaters and untreated acid mine drainage from coal mining activities (Niyogi et al. 1999, Soldo and Behra 2000, Bray 2007, Bray et al. 2008). For example, at a study area in New Zealand where naturally flowing streams were naturally alkaline the process of acid mine drainage modifies the stream pH to be unnaturally acidic (Bray 2007). The decline of pH can change the bioavailability of nutrients and influence the toxicity of some metals (Otter et al. 2012, Durães et al. 2014). In contrast, waterways upstream of the entry of acid mine drainage inflows are found to support many pollution sensitive species along with a much greater richness of species (Kinross et al. 1993, Bray 2007, Bray et al. 2008). Bray (2007) found a range of algae species in his study that were potentially useful as 'sensitive' and 'tolerance' indicators and concluded that algal diatoms may be a useful tool for monitoring these impacts in New Zealand's, West Coast streams.

Hill et al. (2000) also investigated the impacts to algal diatom communities in a Rocky Mountains River, the Eagle River, Colorado USA. It was found that in this river the periphyton community at reference sites recorded two genera increasing to 21 genera downstream of the abandoned mining operations. Two genera were reported to be dominant, *Fragilaria* and *Achnanthes* though *Fragilaria* was dominant at non-impacted sites whilst *Achnanthes* was

dominant in the impacted sites. Finally, Hill et al. (2000) found the greatest taxonomic similarities of the periphyton communities was recorded between samples from the sites with the highest levels of abandoned mine wastewater. An experimental study by Soldo and Behra (2000) was performed in Switzerland investigating the effects of copper (Cu), nickel (Ni), silver (Ag) and zinc (Zn) exposure on periphyton. It was found that exposure to the heavy metals after 12 weeks changed the dominant taxa of the periphyton community along with increasing the relative abundance of one species *Oocystis nephrocystioides* from <1% of the overall community to 56% of the community (Soldo and Behra 2000).

In recent years, concern regarding impacts of toxic metals in global ecosystems has been rising (Allen et al. 1993, Sericano et al. 1995). Exponentially increasing industrial and mining activities and a growing number of closed mines and contaminated sites has led to further environmental pollution through wastes produced by these activities (Sericano et al. 1995, Ashraf et al. 2011). Heavy metals from mining and industrial activities, when released to the environment, have the potential to accumulate within biota at toxic concentrations and cause chronic ecological impacts in ecosystem food chains (Sericano et al. 1995, Ashraf et al. 2011). A study in Australia by Jasonsmith et al. (2008) focussed on selenium (Se) contamination in the water column of an artificial impoundment, Lake Wallace, and reported that concentrations of selenium (Se) in benthic algae were found to be six times higher than concentrations found within epiphytic algae. It was reported that detritus material sampled was found to have higher concentrations when compared to live plant material. This link from live plant material to detritus material is a potential source of selenium (Se) accumulation within Lake Wallace's invertebrates that consume detritus material as a food source (Jasonsmith et al. 2008).

Metal pollution has been a global environmental issue for many decades as waterways have been and continue to be used as a place to discharge industrial and other wastes, and much of this contamination went on relatively unchecked (Allen et al. 1993). In many cases metal pollutants are directly discharged into waterways from anthropogenic activities (Neff 1984). Many of these metal pollutants become absorbed to suspended particulates within the water column. Once absorbed to these suspended particulates the metal contaminated suspended particulates eventually deposit into waterway sediments often remaining indefinitely (Neff 1984, Wang and Rainbow 2008). The pollution is often highly persistent as heavy metal contamination within aquatic environments can persist for much longer than terrestrial organic pollutants. This is due to the lack of a "biodegradation function" of heavy metals in aquatic ecosystems in comparison to a terrestrial ecosystems ability to eliminate metal concentrations through the "biodegradation process" or other chemical means (Ashraf et al. 2011).

6. Bioaccumulation of colliery pollutants in the aquatic environment

Metal bioaccumulation has been reported within many aquatic flora and fauna species worldwide (Hill et al. 2000, Amish and Cowx 2000). For example, Amish and Cowx (2000) reported the persistent legacy pollution of the River Don in South Yorkshire United Kingdom during 1995 and 1997. The river was the recipient of discharges from ceased coal mining operations which commenced during the industrial revolution to current industrial wastes such as ochre and organic pollutants from the current operational paper manufacturing industry (Amish and Cowx 2000). The coal mining operations were all abandoned at the time of the research. It was concluded that the aquatic

macroinvertebrate community was impacted from the legacy pollutants with only pollution tolerant species recorded, it was also observed that two fish species *Salmo trutta* (brown trout) and *Thymallus thymallus* (grayling) appeared to have little recruitment through reproduction and relied heavily on stocking of fingerlings (Amish and Cowx 2000).

A study by Hill et al. (2000) investigated abandoned metal mining operation impacts to stream periphyton communities in the Eagle River, Colorado North America. It was reported that changes in periphyton assemblages between upstream and downstream sample locations were recorded, with different species dominating the different non-impacted and impacted sites (Hill et al. 2000). Another study performed in Ohio, North America found arsenic (As) and selenium (Se) bioaccumulating within caddisflies sampled from coal ash polluted waterways (Reash et al. 2006). The study also used whole fish tissue samples along with gonad tissue samples and results were reported to have been 2 to 3 times higher than proposed for fish, both whole body and ovary (Reash et al. 2006).

Bioaccumulation is a common problem, with increases in concentrations of pollutants being reported for other aquatic biota such as turtles, crayfish, tadpoles and varying fish species in the Tennessee Valley, Tennessee USA by Otter et al. (2012). Whilst pH of water itself can be important for water quality and ecosystems, pH can also exert a strong influence on other water properties. For example, the pH of water is identified as an important factor directly linked to the speciation and bioaccumulation of metals and metalloids (Otter et al. 2012, Durães et al. 2014). pH can affect the bioavailability of metals and metalloids by influencing their solubility and subsequent ability to bioaccumulate to a medium (Otter et al. 2012, Durães et al. 2014). Atkinson et al. (2007) found that lower water pH allowed for greater bioavailability and sequestration of heavy metals in biota and as pH decreased, the precipitation of iron (Fe) and manganese (Mn) increased significantly. This decreasing pH had a secondary effect which allowed a greater sequestration rate of lead (Pb) and zinc (Zn) (Atkinson et al. 2007).

Durães et al. (2014) investigated two differing soil pH levels (acidic and alkaline) impacted by mining activities and their influence on metal and metalloid bioavailability in Portugal and Morocco respectively. Durães et al. (2014) stated that some aquatic and terrestrial flora species are more tolerant than others and can often tolerate high levels of metal concentrations in their tissue. It was found that the mobility of metals and metalloids in acid mine waters (low pH) from Portugal accumulated the metals zinc (Zn), copper (Cu) and lead (Pb) respectively, whilst in contrast the high pH environment of Morocco changed the order of each metals bioavailability to lead (Pb), zinc (Zn) and copper (Cu) respectively. Durães et al. (2014) concludes that the “pH conditions are decisive in the mobility and uptake of metals by plants.” (Durães et al. 2014, pp. 2102). Similar investigations into the bioaccumulation of heavy metals in soil and native plant species, in particular chromium (Cr), cobalt (Co), copper (Cu), iron (Fe), manganese (Mn), nickel (Ni) and zinc (Zn) from mining activities in Northern Pakistan by Nawad et al. (2015) found soil samples to be highly significantly different ($p < 0.01$) when compared to reference soil. Nawad et al. (2015) also found heavy metal accumulation was highly significantly different for all plant species used in their study.

A study assessing the bioaccumulation of heavy metals within plant species growing on land used as a uranium mining dump in the Karkonosze-Izera region of the Sudety Mountains in south-western Poland was undertaken by Wislocka et al. (2006). Heavy metal contamination of the soils was investigated and concentrations of the heavy

metals cadmium (Cd), copper (Cu), iron (Fe), manganese (Mn), nickel (Ni), lead (Pb) and zinc (Zn) were reported within the uranium mining dump sites. Three plant species growing in the area were used to assess the level of bioaccumulation of these contaminants, two being described as trees and one as a shrub. Results showed elevated heavy metal concentrations were accumulating within all three species, with both tree species having higher concentrations than that of the shrub species. These heavy metal concentrations were stated to be “above the average values given for plants in other literature” (Wislocka et al. 2006, pp. 811).

A similar study of the bioaccumulation of the metals iron (Fe), manganese (Mn), zinc (Zn), copper (Cu), cadmium (Cd) and nickel (Ni) within fruit and timber trees grown on grounds contaminated by coal mine spoils in India found concentrations to be higher than that of control trees growing in non-contaminated garden soils (Maiti et al. 2015). The order of bioaccumulation was reported as iron (Fe), manganese (Mn), zinc (Zn), copper (Cu), cadmium (Cd) and nickel (Ni) (ascending from highest to lowest concentrations). pH recorded at the coal mine spoil contaminated site was found to be acidic (pH 4.34 – 4.95) with soil in control areas only slightly higher and reported as mildly acidic (pH 5.4). Significant variations of metal accumulations were recorded for timber tree species whilst high metal accumulations were found for fruit tree species (Maiti et al. 2015). Maiti et al. 2015 concludes that although remediation of the coal mine spoil was undertaken through the addition of topsoils to cover the mine spoil, higher accumulation of metals ($p < 0.05$) was found in trees growing in contaminated soils when compared to trees growing in control soils.

Concentrations of the metals cadmium (Cd), copper (Cu), mercury (Hg), manganese (Mn) and lead (Pb) within water, aquatic sediments and the roots and leaves of aquatic plants was undertaken by Mishra et al. (2007) in lake Govind Ballabh Pant, a man-made lake situated in the southern region of Sonbhadra, Uttar Pradesh, India. The lake receives direct point source wastewaters from a documented group of ten different open cut coal mining operations (Mishra et al. 2007). Eleven flora species in total were used in the study *Eichhornia crassipes*, *Azolla pinnata*, *Lemna minor*, *Spirodela polyrrhiza*, *Potamogeton pectinatus*, *Marsilea quadrifolia*, *Pistia stratiotes*, *Ipomea aquatica*, *Potamogeton crispus*, *Hydrilla verticillata* and *Aponogeton natans*. Results showed that the roots of the aquatic plants sampled had higher concentrations of copper (Cu), manganese (Mn) and lead (Pb) respectively than was recorded within lake sediments, whilst plant leaves had lower concentrations of all metals compared to lake sediments (Mishra et al. 2007).

Research assessing the accumulation of contaminants in fish species have been performed by several studies. One case study is Papagiannis et al. (2004), who investigated copper (Cu) and zinc (Zn) concentrations within fish species from Lake Pamvotis in the Mediterranean region of Europe with high concentrations of copper and zinc within the water column. It was found that high levels of copper (Cu) and zinc (Zn) had bioaccumulated within the fish species organs, notably the liver and gonads ($p < 0.0001$) (Papagiannis et al. 2004). A similar study by Otter et al. (2012) studied the bioaccumulation of arsenic (As) and selenium (Se) in five fish species impacted by a wet coal ash spill after a dyke failed at the Kingston Fossil Plant near Kingston, Tennessee North America in 2008. Otter et al. (2012) found that fish species collected two years after the spill (2010) at impacted sites had higher concentrations (between 25% and 50% greater) of both arsenic (As) and selenium (Se) than control sites. Higher concentrations of selenium (Se) were found in one fish species, the Redear Sunfish (*Lepomis microlophus*) in particular. This was

concluded to potentially be linked to the gut pH of the fish species by influencing the digestion and bioaccumulation of the selenium (Se). Potential changes in trophic dynamics were also linked to the accumulation process at the coal ash spill sites (Otter et al. 2012).

An investigation into the concentrations of arsenic (As), copper (Cd), mercury (Hg) and lead (Pb) within muscle tissue samples and organs of two fish species, *Cyprinus carpio* and *Capoeta sp.*, and water samples from the Kor River in Iran was undertaken by Ebrahimi and Taherianfard (2009). Their study also analysed pathological and hormonal changes due to heavy metal contamination. Three sample sites on the Kor River were used; described as (upper, middle and lower) sampling zones with the middle zone being the most polluted. Results found that both fish muscle tissue samples and organs (ovaries and testes) had significantly higher ($p < 0.05$) heavy metal concentrations than both other sampling zones. Pathological changes in blood cells, the liver and the kidneys of the fish were also recorded as significantly higher in samples collected at the highly polluted middle sampling zone. Ebrahimi and Taherianfard (2009) conclude that concentrations of the heavy metals copper (Cd), mercury (Hg) and lead (Pb) in their study were higher than the permissible levels for human consumption. Similarly, Ashraf et al. (2011) conducted a study on the bioaccumulation of the heavy metals arsenic (As), copper (Cu), lead (Pb), tin (Sn) and zinc (Zn) within eight different fish taxa collected from the Sungai Ayer Hitam River in a former tin mining catchment in Bestari Jaya Peninsular, Malaysia. The order of bioaccumulation contamination within all fish species sampled was recorded as tin (Sn), lead (Pb), zinc (Zn), copper (Cu) and arsenic (As) respectively. Results indicated that elevated levels of tin (Sn), lead (Pb) and zinc (Zn) were found in all fish species collected whilst copper (Cu) and arsenic (As) were found in lower concentrations. Tin (Sn), lead (Pb) and zinc (Zn) in all fish species collected were reported to be higher than the Malaysian Food Acts permitted levels (Ashraf et al. 2011).

A more recent study of the concentrations of cadmium (Cd), copper (Cu), iron (Fe), lead (Pb) and zinc (Zn) in water, river sediment and two fish species, *Pelmatochromis guentheri* and *Pelmatochromis pulcher*, was undertaken by Ajima et al. (2015) in the Mbaa River in south-eastern Nigeria. The section of the Mbaa River used in the study was subject to two predominant pollution sources from the disposal of effluents from domestic uses and the aluminium extrusion industry. Ajima et al. (2015) found that river sediment had the highest concentrations of the heavy metals investigated followed by both fish species then water. The levels of heavy metal concentrations found in both water and fish species were reported to be above the recommended limit set by the World Health Organisation (WHO) (Ajima et al. 2015). The study found the water column metal concentrations were the lowest, followed by both fish species and with sediments containing the largest concentrations. For instance, lead (Pb) was recorded at 2.08 mg/L within the water column, 5.4 and 3.57 mg/kg in the two fish species, and 8.23 mg/kg in river sediment. This trend was the same for the remaining four contaminants. Zinc (Zn) within the water column was 0.65 mg/l, within fish 2.21 and 2.24 mg/kg and sediment 3.18 mg/kg. Cadmium (Cd) in water samples was 0.44 mg/l, both fish species 0.59 and 0.30 mg/kg and sediment 1.09 mg/kg. Copper (Cu) recorded 1.06 mg/l in the water column, 2.61 and 3.65 mg/kg in both fish species and 5.64 mg/kg in sediment and iron (Fe) in water recorded 5.68 mg/l, within both fish species 17.91 and 14.68 mg/kg and within river sediment 56.14 mg/kg (Ajima et al. 2015).

A study by Bharti and Banerjee (2011) investigated concentrations of the heavy metals cadmium (Cd), chromium (Cr), copper (Cu), iron (Fe), manganese (Mn), nickel (Ni), lead (Pb) and zinc (Zn) in the Rihand and Son Rivers,

Singrauli, India, associated with discharged coal mine effluent. The metal concentrations within water were reported to be above the permissible limits as suggested by different “pollution control agencies” (Bharti and Banerjee 2011, pp. 393). Concentrations of cadmium (Cd), chromium (Cr), iron (Fe) and lead (Pb) within edible catfish species caught in the coal mine contaminated zone were reported to be recorded at levels higher than “permissible limits” set by WHO (Bharti and Banerjee 2011, pp. 393).

An experimental study investigating the potential for abandoned coal mine pit lakes contaminated with heavy metals and metalloids to be reclaimed was undertaken by Miller et al. (2013) in Alberta, Canada. Selenium was used as a bioaccumulation indicator to assess heavy metal exposure from the pit lakes water. Two types of coal mine pit lakes were used, one being a thermal coal pit lake (water Se < 2 µg/L) and the other metallurgic coal pit lakes (water Se > 15 µg/L). In-situ invertebrates and two juvenile hatchery fish species, rainbow trout (*Oncorhynchus mykiss*) and brook trout (*Salvelinus fontinalis*) were released into both differing coal mine pit lakes for a 2-year period (Miller et al. 2013). Invertebrates and both fish species *Oncorhynchus mykiss* and *Salvelinus fontinalis* sampled in the metallurgic coal pit lakes had higher concentrations of selenium (Se) than recorded in the thermal coal mine pit lakes. These levels were reported to be higher than the tissue guidelines for selenium (Se) as described by the United States Environmental Protection Agency (USEPA) (Miller et al. 2013). It was concluded that reclamation of coal mine pit lakes under current reclamation practices was not a feasible option as they pose a significant risk to wildlife and or human health (Miller et al. 2013). These findings are similar to that found in coal mine and thermal power station contaminated Lake Wallace, New South Wales Australia. Jasonsmith (2008) investigated an array of biology from algae, macrophytes to aquatic invertebrates and fish similar to Miller et al. (2013)’s research. Jasonsmith (2008) found that the aquatic macroinvertebrates recorded the highest levels of selenium (Se). The two fish species, Eleotridae, a native gudgeon and the rainbow trout (*Oncorhynchus mykiss*) were investigated, and it was reported that the native gudgeon had physical abnormalities and the rainbow trout recorded high concentrations of selenium (Se) within whole tissue and liver tissue (Jasonsmith 2008).

A similar study investigating the health of one fish species (*Squalius vardarensis*) was performed by Jordanova et al. (2016), who used one reference river the Bregalnica River and two rivers impacted by coal mining and agriculture the Zletovska and Kriva Rivers in Macedonia. The rivers were the subject of decades of mining wastewater from lead and zinc mining operations. Health checks were achieved by examination of liver histopathology, mostly through external/internal macroscopic lesions. Results found that hepatic lesions and neoplastic lesions were present in fish from all three rivers sampled, though a higher total prevalence of these lesions were found in fish sampled from the mining polluted rivers (Jordanova et al. 2016). Some pathological differences were found to be more prevalent within fish collected from the Kriva River, which was the less metal contaminated of the two mining impacted rivers. Jordanova et al. (2016) concluded the relationship may be contributed to a higher agricultural impact in the Kriva River and a result of a synergistic effect of metal and organic pollution (Jordanova et al. 2016).

Bioaccumulation of metals from mines can occur long after the operation closes, and the mines become derelict. This was demonstrated in a study that investigated the bioaccumulation of the heavy metals arsenic (As), copper (Cu), chromium (Cr) mercury (Hg), lead (Pb) and zinc (Zn) from abandoned copper and mercury mining areas of

the Nalón River Basin, Asturias, Spain (Méndez-Fernández et al. 2014). In this study, metals in one aquatic oligochaete species *Tubifex tubifex* were assessed (Méndez-Fernández et al. 2014). The highest levels of arsenic (As), chromium (Cr), mercury (Hg), lead (Pb) and zinc (Zn) in the oligochaete samples were recorded at samples collected at mercury mining impact sites, whilst copper (Cu) was recorded at higher levels in samples collected at copper mining impacted sites (Méndez-Fernández et al. 2014). Similar increases in copper (Cu) and zinc (Zn) were reported by Swansburg et al. (2002) who investigated bioaccumulation of cadmium (Cd), cobalt (Co), copper (Cu) and zinc (Zn) within chironomidae tissue samples from five streams receiving mine drainage from a metal mining operation in New Brunswick, Canada. Their study found significantly elevated concentrations ($p < 0.05$) of several metals cadmium (Cd), cobalt (Co), copper (Cu) and zinc (Zn) within the water column and periphyton from all their sample streams. Chironomid tissue samples recorded highly significant increases from mine effected samples to reference samples for copper (Cu), cadmium (Cd) and zinc (Zn). Along with the bioaccumulation of the contaminants within the invertebrates, it was also reported that mine affected chironomids had significantly elevated mentum (mouthpart) deformities. It was also reported that “metal tolerant” chironomid genera increased in abundance within mine effected sample locations. It was concluded that these elevated heavy metal concentrations significantly affected the benthic community and has the potential to highly alter overall stream function (Swansburg et al. 2002).

Another metal mining investigation performed by Maret et al. (2003) studied the bioaccumulation of cadmium (Cd), lead (Pb) and zinc (Zn) in Trichoptera (caddisfly) tissue at eighteen reference and test sites impacted by heavy metal contaminated water and sediment from hard rock mining practices in the Coeur d’Alene and St. Regis River basins, north-western USA. It was reported that concentrations of cadmium (Cd), lead (Pb) and zinc (Zn) in water and river sediment at impacted sites were significantly higher than those recorded at reference streams (Maret et al. 2003). Lead (Pb) and zinc (Zn) were often found to also exceed the ambient water quality criteria limit for river sediment. Metal concentrations within caddisfly tissue was reported to be higher in samples collected downstream of mining activities (mean cadmium (Cd) increased from 0.58 ug/g to 6.68 ug/g, mean lead (Pb) increased from 2.94 ug/g to 142 ug/g and mean zinc (Zn) increased from 181 ug/g to 707 ug/g). Macroinvertebrate community structure was also modified at the mine contaminated sites and was expressed through a loss of sensitive EPT taxa and the loss of know heavy metal sensitive Ephemeroptera (mayfly) species (Maret et al. 2003).

There have been several studies performed on metal pollution of surface waters in the USA. One study in particular was conducted in a stream impacted by uranium mill tailings, the Montezuma Creek in south eastern Utah, North America (Peterson et al. 2002). Invertebrates in this study were collected from upstream and downstream of the uranium mill tailings site as well as a nearby reference stream (Verdure Creek). Three invertebrate trophic status indices were used (detritivores, predators and filter feeders), with sampled taxa analysed for a total of 17 trace metals. Results indicated that invertebrates collected immediately downstream of the uranium mill tailings site had significantly higher arsenic (As), molybdenum (Mo), selenium (Se) and vanadium (V) when compared to all other sites, some two – four times higher. Both samples immediately downstream of the uranium mill tailings site differed to the magnitude of 10 times when compared to the upstream, reference stream samples (Peterson et al. 2002).

Several studies have revealed that some heavy metal pollution can have long term persistent bioaccumulation impacts. One example is the Elbe and Vltava Rivers in the Czech Republic, where Kolaříková et al. (2012) reported elevated arsenic (As), cadmium (Cd) and lead (Pb) from intensive chemical industry activities over decades. These trends were observed over a 12-year period between 1993 and 2005, some years after social-economic changes in the country had led to substantial improvements in the water quality of both the Elbe and Vltava Rivers. The study used four macroinvertebrate taxa *Asellus aquaticus*, *Bythinia tentaculata*, *Erpobdella spp* and an aquatic larva of the Chironomidae family. The heavy metals arsenic (As), cadmium (Cd) and lead (Pb) were found at highest concentrations in samples collected from the industry heavy middle Elbe region of Valy and Obříství. High concentrations of arsenic (As), cadmium (Cd) and lead (Pb) were also observed in samples collected from the upper stretches, whilst cadmium (Cd) and lead (Pb) was recorded in samples from the lower Vltava River, an area known for its former mining activities along with metallurgy and a glass manufacturing industry. Though it was reported by the author that “substantial improvements in the water quality of the Elbe River” occurred during the study period results showed that over the 12-year study no significant reduction in heavy metal levels other than mercury Hg were observed within macroinvertebrates sampled (Kolaříková et al. 2012). The study revealed that metals were bioaccumulated by the chironomid larvae and *Erpobdella spp* more than *Asellus aquaticus* and *Bythinia tentaculata* (Kolaříková et al. 2012).

Further research conducted in North America performed by Cain et al. (2004) investigated the ecotoxicology of metals. Cain et al (2004) used five different aquatic insects and the impact to their distribution patterns, at one mining impacted river, the Clarke Fork River in Idaho. Two mayfly species (*Epeorus albertae* and *Serratella tibialis*) were used as the two species are documented to be sensitive to heavy metal contamination whilst the other three used, two caddisflies (*Hydropsyche spp*) and one mayfly (*Baetis spp*) are well documented to be tolerant to heavy metal contamination. It was reported that three of the known tolerant species were found in high abundance within the impacted sample sites (Cain et al. 2004). Ecotoxicity from organic pollution and heavy metals have been shown to reduce freshwater ecosystem functions such as leaf litter break down, community respiration and primary production worldwide (Peters et al. 2013). Research has revealed losses of up to 20% of the overall ecosystem function for two test biota (algae and an aquatic flea). It was found that the reduction of leaf litter decomposition was reduced more in aquatic ecosystems where the primary leaf litter decomposition process was driven by invertebrates not microorganisms (Peters et al. 2013). Marshal et al. (1983) performed insitu ecotoxicity test in the southern basin of Lake Michigan North America. It was found that zinc concentrations as low as 15 g/L significantly reduced primary productivity, zooplankton populations and species diversity, chlorophyll a and dissolved oxygen levels within two weeks. Several different zooplankton species were reduced severely, one species in particular was reduced to only 1% of its control abundance and some species such as *Bosmina longirostris* and *Keratella cochlearis* significant increases in populations were reported (Marshal et al. 1983). A laboratory-based experiment used freshwater rotifers, specifically *Brachionus calyciflorus*, collected from Gainesville, Florida North America to test toxicity levels of the metals nickel and zinc (Snell et al. 1991). Rotifers were hatched and reared in a rigorous controlled laboratory conditions and subjected to control and increasing metal concentration treatments for 24 hours. It was reported that 24-hour acute toxicity for nickel was 4.0 mg/L and 1.3 mg/L for zinc (Snell et al.

1991). Prenatal toxicity effect characterised by structural or functional defects in the developing embryo or fetus were reported by Jasonsmith et al. (2008). Although bioaccumulation of industrial pollutants is a common problem and well documented worldwide, in Australia there has been relatively few studies of bioaccumulation from industrial pollutants let alone mining and especially coal mining wastewaters other than Jasonsmith et al. (2008).

7. Australian aquatic bioaccumulation studies

Australian aquatic bioaccumulation studies are much rarer. One study, conducted at one stream, Bakers Creek located in Hillgrove New South Wales, investigating the bioaccumulation of antimony (Sb) and arsenic (As) within stream macroinvertebrates from the mining of and subsequent processing of antimony and arsenic ores (Telford et al. 2009). This study found that both contaminants were significantly elevated within aquatic autotrophs except for antimony (Sb) within gastropods. They also found that antimony (Sb) concentrations within riparian vegetation was up to 2 – 3 times greater than recorded in non-impacted samples, although this result was not significantly different (Telford et al. 2009). Increases in arsenic (As) within riparian and terrestrial vegetation have been reported internationally as previously discussed by Ebrahimi and Taherianfard (2008) in Iran, Ashraf et al. (2011) in Malaysia and by Otter et al. (2012) in North America.

Another Australian study investigating bioaccumulation and biomagnification in metal polluted freshwaters was conducted in the Coxs River, near Lithgow in the Blue Mountains (Jasonsmith et al. 2008). The study focussed on selenium (Se) and was undertaken at Lake Wallace, a freshwater coal power station cooling reservoir in the upper Coxs River. The river is the recipient of active and inactive coal mine wastewaters from the Angus Place Colliery and the Springvale Colliery. Selenium (Se) concentrations were measured within water, sediment, phytoplankton, zooplankton, epiphytic and benthic algae, macrophyte roots and leaves, stream detritus material, oligochaetes, gastropods, bivalves, crustaceans, insects, whole fish and fish livers (Jasonsmith et al. 2008). Water concentrations of selenium (Se) were reported to be lower in comparison to other waterways receiving similar coal power station effluent, though these concentrations were at levels considered to be detrimental to overall health and the long-term survival of wildlife (Jasonsmith et al. 2008). Sediment concentrations were found to be nearly double that of the interim Australian sediment quality guidelines provided by ANZECC (2000) and ARMCANZ (2000a).

Jasonsmith et al. (2008)'s research at Lake Wallace is a very important Australian study as it focussed on Selenium (Se) contamination in the water, which is a metal that did not have any ANZECC guidelines at the time of their research. Selenium (Se) has recently been added to the ANZECC guidelines and a trigger value of 5 ug/L has been prescribed to allow for 99% species protection (ANZECC 2000). Results found selenium (Se) concentrations in the water column to be 2.0 ± 0.3 and 1.8 ± 0.3 $\mu\text{g/L}$ these concentrations were reported to be considerably hazardous to the health and long-term survival of wildlife (>2 $\mu\text{g/L}$). It was concluded that contaminants within phytoplankton, zooplankton and epiphytic algae were found to be at concentrations that are unlikely to affect the growth of phytoplankton and epiphytic algae, though concentrations of selenium (Se) in water may promote the growth of some zooplankton. Concentrations of selenium (Se) in benthic algae were found to be six times higher than concentrations found within epiphytic algae sampled (1.3 ± 0.2 $\mu\text{g/g}$ to 8 ± 2 $\mu\text{g/g}$). However, it was concluded that these benthic algae are most likely not an important direct route for selenium (Se) accumulation in the study

(Jasonsmith et al. 2008). Macrophyte roots and leaves were not found to accumulate selenium (Se) at high concentrations though detritus material sampled was found to have higher concentrations of selenium (Se) when compared to live plant material. This link from live plant material to detritus material is considered to be a potential important source of selenium (Se) accumulation within Lake Wallace invertebrates who consume detritus material as a food source (Jasonsmith et al. 2008).

Gastropods, bivalves and crustaceans sampled in the Lake Wallace study were found to have low concentrations of selenium (Se). Selenium (Se) concentrations within oligochaetes was found to be lower than those reported in other literature. Aquatic insects sampled were found to have concentrations 1000 times higher than found in other studies ($5.4 \pm 2 \mu\text{g/g}$) with detritus invertebrates recording the highest concentrations ($8.3 \pm 2 \mu\text{g/g}$) (Jasonsmith et al. 2008). This is concluded to be due to these detritus invertebrates having constant close contact with sediments allowing trace elements such as selenium (Se) to bond through iron oxides to their exoskeleton. Fish tissue samples were found to have selenium (Se) concentrations at levels that have been reported to cause abnormalities in physiological development (teratogenesis), a prenatal toxicity effect characterised by structural or functional defects in the developing embryo or fetus, within one species, the flathead gudgeon. Concentrations of whole tissue and liver tissue showed large increases in *Oncorhynchus mykiss* (Rainbow Trout) sampled (whole body $8.3 \pm 0.3 \mu\text{g/g}$ to liver $250 \pm 64 \mu\text{g/g}$) (Jasonsmith et al. 2008). Although the water column concentrations reported by Jasonsmith et al. (2008) are below the recently derived ANZECC trigger value (Jasonsmith et al. 2008 water column $2 \mu\text{g/L}$, ANZECC $5 \mu\text{g/L}$), the impacts to the environment are clear. This shows that although efforts have been made to introduce a trigger value for selenium, the derived value may not subsequently protect the aquatic ecosystem adequately. It should be noted that the $5 \mu\text{g/L}$ trigger value is for 99% species protection and subsequent values of $11 \mu\text{g/L}$, $18 \mu\text{g/L}$ and $34 \mu\text{g/L}$ have been derived for lower species protection levels of 95%, 90% and 80%.

8. Pollution impacts after closure of coal mines

Many studies across Europe and the United Kingdom have shown that mining pollution from coal mines can continue, and some can increase, after the commercial operation of a coal mine ceases (Cairney and Frost 1975, Banks et al. 1997, Younger et al. 2002). Johnson (2003) explains that when the pumping of mine drainage from underground mines ceases, post mining, the groundwater can flood the mine and cause an increase in the concentration of pollutants, such as metals. This process has been termed 'rebounding' of mine water as the rising groundwater level increases until it emerges on the surface (Younger 1993, Younger 2001). The progressive flooding of closed mines, with groundwater, can be a slow process. For example, coal mining ceased in the Ore River catchment of Scotland in 1967 and the rising level of mine drainage, from various surface locations, emerged between nine and 10 years later (Jackson 1981 reported in Younger 2001). After emergence, this 'rebounding' coal mine water can cause reductions in stream pH (Banks et al. 1997, Younger 2001). Banks et al. (1997) reported elevated sulphate levels in the waters of abandoned UK coal mines. This was similar to findings that Pond et al. (2008) reported in mined West Virginian streams compared to unmined streams. Highly elevated levels of iron (Fe) have also been reported in abandoned UK mines by Cairney and Frost (1975) and Younger (2001). Younger (1993)

reported elevated concentrations of zinc (Zn) in abandoned mines in Durham UK, whilst Banks et al. (1997) reported elevated levels of manganese (Mn).

Despite the importance of the coal mining industry, prior to this research (chapter 2), there have been few Australian research studies published that have undertaken a comprehensive water pollution investigation from an underground coal mine comparing water quality before closure to that after closure. Most published literature on this topic comes from the northern hemisphere, particularly from the UK, where the majority of their coal mines closed in recent decades (e.g. Johnson 2003, Younger 2001). Many studies have revealed that water pollution from the closed coal mines has often increased following the mine closure (Cairney and Frost 1975, Younger 1993, Brake et al. 2001 and Johnson 2003). The lack of Australian literature on this topic is puzzling given that the number of closed and abandoned mines in Australia is currently growing (Unger et al. 2012). Closed mines (coal and other mines) in Australia is an emerging environmental problem with estimates of 52,543 abandoned mines with few receiving rehabilitation (Unger et al. 2012). The Australian Government recognises the growing problem associated with closed mines and is particularly concerned about how to rehabilitate the increasing number of closed and abandoned mines (Noetic, 2016).

9. Conclusion

Many data gaps are present here in Australia, which are showcased above through the more detailed and studied impacts internationally, though this body of research has addressed many. For instance, there are no Australian studies of a comprehensive suite of water quality and other contaminants at a group of active and inactive coal mines until now. Whilst in comparison research on this topic has been performed by Cairney and Frost (1975), Younger 1993 and Banks et al. (1997) in the United Kingdom and the United States by Pond et al. (2008), Brake et al. (2001) and Petty et al. (2010).

No Australian study has investigated macroinvertebrates (a measure of the aquatic ecosystem) above and below a group of coal mines to describe ecological impacts until now. There are very few studies worldwide investigating a large group of inactive and active mines, including some international studies such as Cain et al. (2004) in North America, though this research only used one coal mine, Brake et al. (2001), Pond et al. (2008) and Petty et al. (2010) also performed macroinvertebrate surveys in their research but due to the nature of the mining “Mountain top” mining they were unable to have reference sample locations upstream of the active AMD.

Both internationally and here in Australia there is a lack of research investigating metal and metalloid contaminants comparing upstream water and sediment concentrations from a group of mines until now. For example, as discussed above in the US many coal mining operations are “Mountain top” and no upstream location is available to sample reference stream conditions (Brake et al. 2001 and Petty et al. 2010). In the UK and Europe many of the waterways are often polluted from many different sources and are often degraded prior to coal mining (Jarvis and Younger

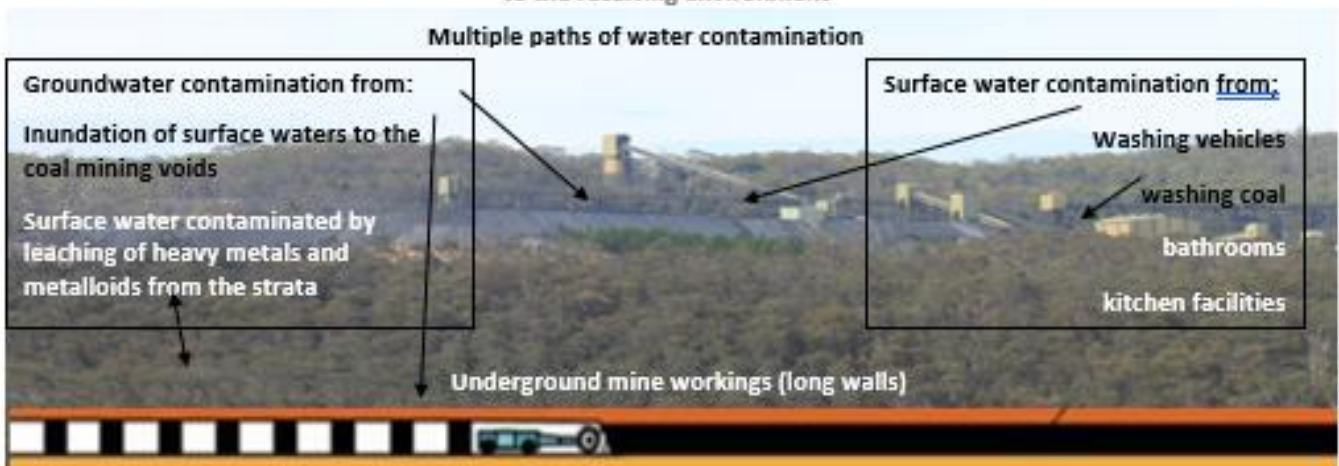
1997, Johnson 2003 and Kolaříková et al. 2012) similar to India and Kenya (Mishra et al. 2007 and Ajima et al. 2015).

The relative lack of Australian research on water pollution and ecological impacts from coal mines is puzzling as coal mining is a major Australian industry, and production has been progressively rising over the last 50 years (Mudd 2009). In 2015, Australian coal production was approximately 440 million t/per year (Australian Government 2016). In addition to its domestic uses for electricity generation and industrial processes, Australia also exports a large proportion of its coal production and it was the largest coal-exporting nation, by volume, in 2015 (Australian Government 2016). The Sydney Basin is an important coal mining region, with most of the mining conducted by underground mines in the southern and western coalfields (Mudd 2009). Although still a very productive region, many coal mines in the Sydney basin have ceased production and water pollution from closed mines has been reported in nearby waterways (e.g. Battaglia et al. 2005, Wright and Burgin 2009a, Price and Wright 2016).

In New South Wales, the regulation of coal mine wastewater discharges are enforced by Environmental Protection Licence's (EPL's) that currently have a water pollution focus on water chemistry pollutants at the 'end of the discharge pipe'. In most cases the regulator uses the default ANZECC (2000) guidelines to set discharge limits which stipulate specific concentrations of pollutants that are permitted to be discharged within the colliery wastes, measured at the 'end of pipe' prior to entering the receiving waterway. This strongly influences the concentration of pollutants that are present in the water column of a receiving waterway (Belmer et al. 2014, Wright et al. 2017 and Belmer and Wright 2019). Setting pollutant limits to EPL's that apply only to pollutant concentration in the 'end of pipe' may well be of limited value if the heavy metals are able to bioaccumulate and cause ecosystem toxicity within the aquatic ecosystem. The regulation generally ignores the instream impact on natural water quality and the health of the aquatic ecosystem. This represents a discord in the EPL process which is unsatisfactorily protecting the receiving aquatic ecosystem from coal mine wastewater discharges. This shows a lack of actual real-world environmental protection when regulation is driven by "end of pipe" water column concentrations.

No data on the ecological impact, using an upstream versus downstream design of a group of coal mines has ever been performed in Australia. There has been no investigation of the impacts to aquatic macroinvertebrates from a group of mines, let alone active and inactive mines. There is also a lack of Australian research investigating the full spectrum of instream pollution concentrations from both the water column and stream sediment from coal mine wastewaters. There is a lack of water quality data on a regional group of mines which use triangulation techniques (upstream, downstream and discharge source), let alone measuring downstream impacts longitudinally for over 15km. There is no research in Australia and limited internationally that investigates the cumulative impacts of coal mine wastewater discharges, from the initial impact of the toxicity of the discharge on the aquatic ecosystem to the eventual bioaccumulation of pollutants within the water column and the mobilisation of pollutants to the terrestrial environment. There is also no coal mine wastewater impacts to aquatic ecosystems research within "high conservation" environments and a general lack of research comparing impacts from actively mined collieries vs inactively mines collieries.

Conceptual model of active and inactive coal mining operations. Wastewater regulation and treatment and impacts to the receiving environment



There are differences in the groundwater removal process determined by if a coal mine is actively mined or inactively mined



Active coal mines are regulated to treat the contaminated wastewaters



Inactive coal mines are not regulated to treat the contaminated

Mine workings (voids) fill with ground water. Water is pumped out of the mine workings to ensure the mining operation can continue. This wastewater is treated to a standard stipulated in the EPL. This wastewater post treatment is then discharged to the receiving waterway

Acid Mine Drainage generates over time as the old mine workings (voids) fill with ground water. This water, once it has filled the mine voids, discharges to the receiving waterway unmitigated and untreated



Wastewater impacts stream water quality and chemistry



Acid mine drainage impacts stream water quality and chemistry



Research significance

There is a rich literature on coal mines and water pollution in some parts of the world, such as the United States which includes many regional studies of active and inactive mines (Brake et al. 2001; Pond et al. 2008; Petty et al. 2010). Many of these studies do not include sampling waterways above the mining operation and, as a result, often do not illustrate the full extent of impact on the receiving waterways and their ecosystems. One major data gap is that there have been very few studies (none in Australia) comparing water quality impacts from a regional group of coal mines that discharge wastes from active (treated) compared to inactive mines (un-treated).

The relative lack of water pollution studies from Australian coal mines is puzzling given the importance of the industry. Despite increased mining of coal in recent decades and coal becoming Australia's second highest value export, there are comparatively fewer studies on water pollution from coal mines in Australia (Mudd, 2009).

Coal mine wastewater discharges in New South Wales are regulated by the New South Wales Environmental Protection Authority (NSW EPA) and environmental protection of receiving waterways is implemented through Environmental Protection Licenses (EPL), under the Protection of the Environment Operations Act 1997 (POEO Act). EPL's set discharge limits for water quality pollutants in coal mine waste waters that are discharged to the environment (Wright, 2012, Belmer et al. 2014).

In many cases the EPL's for coal mine waste discharges are failing to protect the receiving waterways ecosystems by failing to identify ecologically hazardous chemicals in the waste discharges (table 1). EPLs often enable pollution as they impose water quality and pollutant limits that are much higher or significantly different to the receiving waterway or local reference conditions (Belmer et al. 2014, Belmer et al. 2015).

In many cases EPL's often regulate for minor and perhaps insignificant pollutants such as oil and grease and pH, which, as mentioned above, are often much higher than naturally occurring conditions. This can allow the mine wastes to cause pollution of receiving waterways through ineffective regulation (table 1 and table 2). This poor regulation is failing to protect the aquatic environment of these receiving waterways. A major contributing factor is probably the lack of scientific understanding when setting discharge limits (Wright, 2012, Belmer et al. 2014, Price et al. 2015, Wright 2015).

This research builds on the limited research available on coal mine pollution to their receiving waterways within Australia. The research has produced the first detailed chemical and ecological study on the effects of coal mine wastewater discharges from a group of active and inactive coal mines. The inclusion of inactive mines will document the ongoing impacts to waterway health after coal mining has ceased and the continuing degradation of their receiving waterways (chemistry and ecology) which can become a long-term legacy.

The research has filled a major data gap that should help improve the planning and assessment of proposed coal mines. It also will help improve the management of ongoing impacts associated with existing coal mines. In addition, the research provides a better understanding of the legacy of pollutants being left behind when coal mines close. This research reveals that many closed coal mines continue to leak or release contamination within the aquatic

and perhaps also to the terrestrial environment. This knowledge is imperative to better manage current coal mining operations, proposed extensions to existing operating coal mines and also to proposed new coal mines throughout the Sydney Basin and Australia.

The research seeks to improve the rigour of scientific testing of water chemistry and river ecology for Australian underground coal mines. Whilst much of the international literature suggests that coal mining activities heavily degrade aquatic ecosystems, there have been very few Australian studies that have examined this in detail similar to this thesis. Information is particularly scant on the on-going impacts of coal mines after they cease commercial operation. This research shows that water quality and ecological degradation to waterways affected by coal mines continues well beyond the closure of the mine operation. This research directly addresses this important gap in the literature.

Very little research has examined the ongoing food-chain impacts of water pollution from coal mines through metal bioaccumulation. At present in Australia, once coal mining practices cease a coal mine operation undertakes simple remediation practices, often coal mine waste waters will continue to flow into their receiving waterways, unmitigated and untreated. This research directly addresses this gap in the literature.

Current environmental regulation in NSW does not assess the gradual build-up of river sediment contamination that may continue to contaminate waterways long after coal mines cease active operation. This research does address that gap in the literature and correlates water and sediment contamination to the bioaccumulation of contaminants within riparian plants and to an aquatic beetle (river biota).

The research is the most thorough and detailed examination of the impacts of coal mines on the aquatic ecology of local rivers that receive coal mine wastes. Impacts to the aquatic ecosystem through comprehensive macroinvertebrate data will assist in assessing the impacts to the aquatic ecosystem. This research also reveals detailed water quality and river sediment chemistry pollutants, discharged in coal wastes, which contribute to impairment of river water quality and ecological health.

This research produced numerous improvements to environmental regulations. These particular focus on NSW environmental regulations (EPL's) that will help achieve improved environmental outcomes for waterways affected by coal mining operations within the Sydney Basin.

A final outcome of this research is the preliminary development of a "coal mine sensitive and tolerant" rapid assessment tool or guideline for future ecological monitoring of the impacts of coal mine waste waters on their receiving waterways ecosystem. This will be a major improvement on the current environmental regulation that primarily regulates water quality and pollutants in coal mine wastes. The coal-mine biotic index could be used in the future to better ensure that the water quality and chemistry properties specified in coal mine wastewater EPL's are successful to protecting the aquatic ecosystem that they are designed to protect.

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Chapter 2

Increased Water Pollution After Closure of Australia's Longest Operating Underground Coal Mine: a 13-Month Study of Mine Drainage, Water Chemistry and River Ecology

Water, Air, Soil Pollution, Springer 2018.

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Abstract

This study investigated the water pollution impact of mine drainage from an underground colliery that had stopped mining 3 years earlier. After more than a century of operation, the mining stopped, pumping ceased and groundwater accumulated, causing the flooding of the deepest sections (c. 15%) of the mine workings. The mine then began free draining to the adjacent Wingecarribee River. The closure and flooding triggered acid mine drainage that has resulted in mildly acidic pH and higher concentrations of several metals. Of greatest environmental concern were ecologically hazardous concentrations of three metals: nickel (418 µg/L), zinc (1161 µg/L) and manganese (11,909 µg/L) in the mine drainage. Such concentrations are some of the highest concentrations reported for these metals in drainage from an Australian coal mine and are 2.5 to seven times higher than when the mine was operating. The concentration of nickel and manganese were stable, but zinc gradually declined throughout the 13-month study. The inflow of the drainage increased the concentration of the three metals in the river, causing exceedance of water quality guidelines for protection of aquatic species. The ecological impact of the mine drainage was substantial, causing a 63% reduction in family richness and a 90% reduction in proportion of invertebrates from the known pollution-sensitive orders (Ephemeroptera, Plecoptera and Trichoptera). Literature suggests the pollution could continue for decades. Of additional concern is that the mine drainage is currently untreated and pollutes a river in the water catchment of Australia's largest domestic water supply reservoir.

Keywords

underground mining; closed mines; environmental regulation; bicarbonate; pollution; salinity; pH; zinc and nickel.

Introduction

Coal mining operations are a well-known source of water pollution (Tiwary 2000 and Younger 2004). Coal mine drainage can escape, or be discharged, to local waterways causing contamination of surface waters to levels that degrade stream ecosystems (Jarvis and Younger 1997, Pond et al. 2008 and Wright et al. 2017). One well-known form of water pollution that can be triggered by coal mining is acid mine drainage (AMD). Coal mine drainage can be strongly affected by AMD, largely depending on the sulphur content of the coal, which can generate sulphuric acid that leaches and mobilises metals from the surrounding geology (Johnson 2003).

Water pollution, from coal mines, is a major international environmental problem that can have a broad array of symptoms. They include modification to pH (Banks et al. 1997, Brake et al. 2001, Verb and Vis 2000 and Younger 2001), elevated salinity (García-Criado et al. 1999 and Pond et al. 2008), modified stream ionic composition (Younger 2001 and Wright 2012) and elevated heavy metals (Pond et al. 2008, Brake et al. 2001 and Johnson 2003). Water pollution from coal mines can also cause detrimental impacts on river ecosystems (Jarvis and Younger 1997, García-Criado et al. 1999, Pond et al. 2008, Wright and Burgin 2009a, Gray and Harding 2012, Griffith et al. 2012 and Wright et al. 2017). Perhaps surprisingly, pollution from coal mines can increase after the commercial operation of a coal mine ceases (Cairney and Frost 1975, Banks et al. 1997 and Younger et al. 2002). Johnson (2003) explains that when the pumping of mine drainage from underground mines ceases, post mining, the groundwater can flood the mine and cause an increase in the concentration of pollutants, such as metals. This process has been termed 'rebounding' of mine water as the rising groundwater level increases until it emerges on the surface (Younger 1993 and Younger 2001). The progressive flooding of closed mines, with groundwater, can be a slow process. For example, coal mining ceased in the ore catchment of Scotland in 1967 and the rising level of mine drainage, from various surface locations, emerged between nine and 10 years later (Jackson 1981 reported in Younger 2001).

Coal mining is a major Australian industry, and production has been progressively rising over the last 50 years (Mudd 2009). In 2015, Australian coal production was approximately 440 million t per year (Australian Government 2016). In addition to its domestic uses for electricity generation and industrial processes, Australia also exports a large proportion of its coal production, and it was the largest coal-exporting nation, by volume, in 2015 (Australian Government 2016). The Sydney Basin is a very important coal mining region, with most of the mining conducted by underground mines in the southern and western coalfields (Mudd 2009). Although still a very productive region, many coal mines in the Sydney basin have ceased production and water pollution from closed mines has been reported in nearby waterways (e.g. Battaglia et al. 2005, Wright and Burgin 2009a and Price and Wright 2016). Despite the importance of the coal mining industry, there have been no Australian research studies published that have undertaken a comprehensive water pollution investigation from an underground coal mine shortly after its closure when groundwater has flooded its deepest underground coal mine workings. Most published literature on this topic comes from the northern hemisphere, particularly from the UK, where the majority of their coal mines closed in recent decades (e.g. Johnson 2003, Younger 2001). Many studies have revealed that water pollution from closed coal mines has often increased following the mine closure (Cairney and Frost 1975, Younger 1993, Brake et al. 2001 and Johnson 2003). The lack of Australian literature on this topic is puzzling given that the number of closed and abandoned mines in Australia is currently growing (Unger et al. 2012); yet, so many have

ongoing environmental problems (Battaglia et al. 2005, Wright and Burgin 2009a and Wright et al. 2011). Coupled with the lack of literature on the problems associated with coal mine closures and management the of Australian coal mines is that the State Government funding for ongoing management of closed mines in NSW is a relatively paltry Australian \$4.1 million in the NSW Government funding for the 2014–2015 financial year (for rehabilitation of derelict mines); yet, royalty payments of approximately Australian \$1.5 billion per annum are received by the NSW Government from the coal mining industry (Geary 2015).

In this current study, we investigated water pollution changes resulting from coal mine drainage from a recently closed colliery (Berrima Colliery) that operated for more than a century. We conducted a water quality investigation over a 13-month period after it has stopped mining. We also surveyed macroinvertebrates in the Wingecarribee River to detect and measure any adverse ecological and water quality effects from the mine drainage discharged to the river receiving the coal mine drainage. In addition, we also compared our 13 months of post closure mine drainage results with several years of previous collected unpublished data on the water quality of the mine drainage. The data, from the mine owner, was collected in accordance with its environmental license, regulated by the NSW Environment Protection Authority (NSW EPA). The mine's data provided a temporal comparison of water chemistry whilst the mine was in commercial operation, extending to 3.5 years after its cessation of coal extraction, and 6 months after the deepest sections of the underground mine voids were flooded with the partial 'rebound' of the groundwater table submerging sections of the deepest workings. Our study sought to answer three questions:

1. How do river macroinvertebrates respond to coal mine wastewater discharges that continued to emerge from underground colliery workings 39 months after mining has ceased?
2. What are the key water quality changes that the discharge of the mine drainage causes to the receiving river?
3. How has water chemistry of the mine drainage changed since the underground colliery ceased mining and flooded its deepest workings?

Methods

Study Area

Samples of water and invertebrates were collected from the Wingecarribee River (the mine drainage-receiving waterway) and the mine drainage discharges from the Berrima Colliery, in the Hawkesbury-Nepean catchment in the Berrima area, adjacent to the small village of Medway, of New South Wales in south-eastern Australia (34° 15' S, 150° 15' E; Fig. 1). The study area comprised areas of naturally vegetated bushland, rural holdings (mostly livestock grazing), peri-urban areas and urban townships.

The Wingecarribee River has major importance as one of the major rivers supplying water to Lake Burrang. The management and protection of water quality supplied from storages, including Lake Burrang, is the responsibility of the Government agency WaterNSW (WaterNSW 2017a). Lake Burrang is the largest drinking water storage reservoir in Australian and holds 80% of the water supply for 4.6 million people in the Sydney Metropolitan area (WaterNSW 2017b). The Berrima colliery is located about 90 km upstream of Lake Burrang

(Fig. 1). This colliery is one of nine underground coal mines that operate in the Sydney drinking water catchments (GHD 2013). The 2013 independent audit of the catchment recognised that there had been a recent increase in mining activity in the catchment (GHD 2013). The audit identified a number of deficiencies in the regulation of mining in the catchment and made a recommendation that multiple Government agencies should ‘...collaborate to develop a risk assessment methodology to assess the impacts of mining, CSG and industrial developments on water resources in the catchment’ (GHD 2013). An earlier publication, by Krogh (2007), documented many environmental problems associated with coal mining activity in Sydney’s drinking water catchments.

The Berrima Colliery is a small underground mine that has operated since 1872 (Boral 2015). It was one of the longest continually operating underground coal mines in Australia and used a pillar and board mining method, removing ore from lower sections of the Wongawilli seam at depths of 110–190 m below the surface (EMGA 2011; Boral 2015). The Wongawilli coal seam is part of the Illawarra Coal Measures and is highly valued as thermal and metallurgical coal and is classified as semi-hard coking coal (NSW Resources and Energy 2016). The Wongawilli seam has very low (< 0.6%) sulphur content (Huleatt 1991). The seam is overlaid by Hawkesbury Sandstone of the Wianamatta Group, along with basalt flows and dykes from Triassic and Jurassic igneous activity (Boral 2015). The mine was mechanised, and operated continuously, since the late 1920s with the colliery producing an average of 250,000 t of ore per annum, supplying coal to the local cement manufacturing works (Boral 2015).

Wastewater from the mine is generated through the accumulation of groundwater which continuously seeps into the underground coal mining workings, at the rate of approximately 3 ML/day (EMGA 2011). The mine drainage is generated due to the accumulation of groundwater seepage entering the underground workings. The drainage is directed through a single drainage adit to the Wingecarribee River, a distance of about 60 m. This discharge of mine wastewater is regulated by the NSW EPA under the Protection of the Environment Operations (1997) Act (POEO Act 1997). The EPA regulated the Berrima Colliery discharge using an Environment Protection License (EPL) number 608 (NSW EPA 2017b). The EPL 608 provided 100% discharge limits for four pollutants (Oil and grease 10 mg/L; total suspended solids 50 mg/L; pH 6.5–8.5; biochemical oxygen demand 20 mg/L). There are no discharge limits in EPL 608 for any other pollutants, including metals. Although flow data was not recorded from the mine adit, the Wingecarribee River is a large river and the input of the mine adit flow would be considered minimal to the overall average flow of the Wingecarribee River.

The mine ceased extraction of coal in November 2013. The mine owners (Boral) released a plan to close and rehabilitate the mine in October 2015 (Boral 2015). Pumping stopped from sections of the underground works, and areas were partially sealed, between November 2013 to January 2016, with continued groundwater ingress to the mine causing partial flooding of the deepest sections of the mine workings (August 2015 to March 2016). The flooding occupied about 15% of the underground workings (personal communication, Boral).

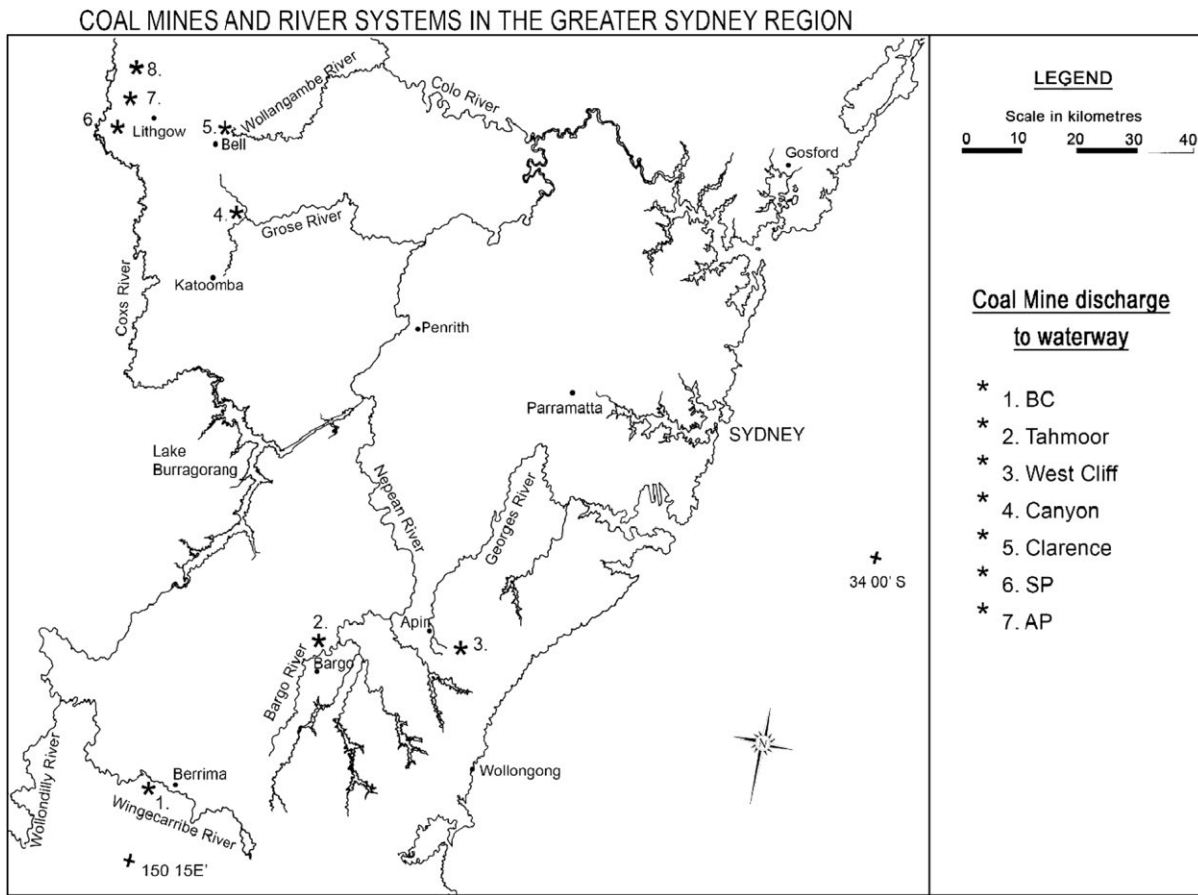


Figure 1. Map of lower Sydney basin, major waterways, settlements and location of various coal mine drainage discharges (marked by * and numbered). Berrima Colliery (BC) is (1) adjacent to the Wingecarribee River, that is recipient of its mine drainage.

At the time of publication, complete sealing of the mine has not occurred. There are concerns that increased groundwater could cause a destructive ‘out- burst’ (e.g. Younger 2002) as some of the historic mine workings are located in close proximity to the steep and fragile Wingecarribee River valley. Mine drainage continues to freely drain from a single drainage adit that directs the drainage to the Wingecarribee River.

Three sampling sites were used in the study (Fig. 1). Two sites were on the Wingecarribee River, the waterway that is the recipient of the coal mine drainage. The upstream site (WiU) was positioned approximately 100 m upstream of the entry of the mine drainage. The second site (WiD) was located approximately 200 m downstream of the mine drainage inflow, in a narrow section of river that ensured the inflowing mine drainage was well mixed. The third sampling site was the mine drainage itself, which was collected approximately 10 m after it emerged from the mine’s drainage adit, prior to its discharge into the Wingecarribee River.

The section of the Wingecarribee River, near the mine drainage adit, flowed within a steeply dissected sandstone and shale canyon. The river ranges from 2 to 8 m wide, is wadable, with average river depths (depending on flow) of approximately 0.5 to 2 m. Several reaches of the river channel are sandstone bedrock, with alternating pool and riffle zones. River discharge is highly variable. The median flow of the Wingecarribee River at Berrima during 2013 to 2016 was reported to be 21.72 ML/day (Alluvium 2017). The Berrima Colliery records the daily flow of mine

drainage that is discharged, in compliance with its EPA environmental license with the NSW EPA (EPL no. 608) which ranged between zero to 7.3 ML/day, with a median discharge of 2.67 ML/day.

Macroinvertebrate Sampling

This study collected river macroinvertebrates to assess and measure the ecological impacts of the mine drainage, if any, to the aquatic ecosystems of the Wingecarribee River. Macroinvertebrates are widely used as ecological indicators of water quality due to well-established methods and ease of sampling (Hellowell 1986).

Macroinvertebrates are regarded as being effective indicators as they have relatively long- life cycles and different taxonomic groups have differing sensitivities to disturbance and water pollution (Hellowell 1986 and Rosenberg and Resh 1993). There is a large and well-established literature on freshwater macroinvertebrates and their applications to assess various anthropogenic disturbances including a broad variety of disturbances and pollution types (Hynes 1960 and Rosenberg and Resh 1993) including coal mine drainage (Jarvis and Younger 1997, Battaglia et al. 2005, Pond et al. 2008 and Wright and Burgin 2009a).

The rapid assessment AUSRIVAS method (Norris and Hawkins 2000 and ANZECC 2000) is widely used for assessment of aquatic ecosystem health in Australia at broad spatial scales but was not considered appropriate for this study where an upstream versus downstream design with multiple quantitative spatial replicates offered an optimum design. Previous water quality studies of mining impacts have shown that the inclusion of abundance data provided important information that assisted in the interpretation of the ecological impact (Clements et al. 2000, Wright and Burgin 2009a, Merriam et al. 2011 and Wright et al. 2017). The level of taxonomic resolution is an important consideration in designing a study that seeks to detect and measure ecological impacts (Ellis 1985). A previous study by the senior author investigated the effect of taxonomic resolution (species, family and order) on detecting ecological impacts on macroinvertebrate communities in stream and rivers that were negatively affected from mining and heavy metal pollution in the Sydney basin (Wright and Ryan 2016). That study established that the family level offered very similar ability to detect impacts to the species level (Wright and Ryan 2016).

Macroinvertebrates were collected from the two sites on the Wingecarribee River, WiU 100-m upstream of the mine discharge and WiD 200-m downstream, in February 2017. Ten quantitative benthic samples were collected from a combination of bedrock and cobble riffle habitat (Resh and Jackson 1993 and Wright et al. 1995). This habitat was widely available at each sampling location. The location of each replicate was randomly selected within a 15-m stream reach. Identical macroinvertebrate sampling techniques were used in similar earlier studies (of coal mine and sewage point sources) by the senior author of the Grose River (Wright and Burgin 2009a) and in the investigation of coal mine waste on the Bargo River (Wright et al. 2015) and Wollangambe River (Wright et al. 2017).

The macroinvertebrate samples were collected using a quantitative 'kick sampling' technique (Rosenberg and Resh 1993). A hand net with a frame of 30 × 30 cm and a robust 250-µm mesh were used (Wright et al. 2017). The sampling net was firmly placed at the random location and dislodging the river invertebrates was done by disturbing the stream bed for a period of 30 s over a 900-cm² area, immediately upstream of the net. The net contents, including stream detritus and macroinvertebrates, were immediately placed into a sealed and labelled storage container and preserved in 70% ethanol. A total of ten replicate samples were collected at the two sites to provide a

strong statistical basis for comparison of any differences in the invertebrate communities between the sites, above and below the mine drainage entry point.

In the laboratory, the entire contents of the sampling containers were examined under a high-resolution dissecting microscope ($\times 10$ to $\times 60$) to extract the macroinvertebrates from stream detritus (e.g. leaves, sticks, rocks, gravel). All insect groups were identified to the family taxonomic level by experienced technicians, using appropriate taxonomic keys and guides (e.g. Hawking 2000 and Gooderham and Tsyrlin 2002) as this level of taxonomic resolution has been demonstrated to provide adequate taxonomic resolution for ecological impact assessment (Wright et al. 1995) including assessment of mining and industrial impacts (Wright and Ryan 2016). Two non-insect groups (Oligochaeta and Hydracarina) were not identified to the family level due to identification difficulties.

Biotic Indices

There are several methodologies and biotic indices available to assist in the interpretation of stream macroinvertebrate data to enable detection and measurement of the ecological condition of waterways. Two of the most commonly and simply calculated indices are taxon richness (often performed with family-level data) and total abundance (see Resh and Jackson 1993). The EPT (Ephemeroptera, Plecoptera and Trichoptera) index is a popular and widely used biotic index, based on the relative abundance or taxonomic richness of three common macroinvertebrate orders that are very sensitive to disturbance and water quality impairment (Lenat and Penrose 1996). The percentage of EPT invertebrates is also widely used for evaluating ecological degradation from coal mine pollution (Clements et al. 2000, Pond et al. 2008, Wright et al. 2015 and Wright 2017).

Water Quality Sampling

Water sampling was conducted at three sites (Wingecarribee River, upstream and downstream of the mine drainage inflow) and thirdly from the mine drainage channel emerging from the Berrima Colliery adit, prior to its dispersion into the Wingecarribee River. Water samples were collected from all three sites on eight occasions over a 13-month period (September 2016 to September 2017). The first four samples (September to December 2016) were collected at approximate month intervals. Due to dangerous weather conditions and access difficulties, sampling was then conducted in February, May, August and September 2017. Historic data collected before 2016 was provided the mine owners.

At each site, on each sampling occasion, physio-chemical water quality attributes of pH, electrical conductivity (EC), dissolved oxygen (DO), turbidity (TU) and water temperature were measured in situ using a TPS WP-82Y meter with a YSI dissolved oxygen probe for dissolved oxygen, TPS WP-88 turbidity meter with a TPS turbidity sensor for turbidity and a TPS WP-81 conductivity, pH and temperature meter with a TPS conductivity and temperature probe and a TPS submersible k407 pH sensor. Each meter was calibrated prior to each sampling day. Five replicated measurements of each of the above water quality attributes were recorded from each site on each sampling occasion, once the meter readings had stabilised. In addition, duplicate or triplicate samples of water were collected in decontaminated sample containers provided by the commercial testing laboratory (Envirolab Services,

Sydney). Water samples for metal determination were collected using unused bottles that had been pre-treated by nitric acid. Samples were chilled and delivered to the laboratory for analysis.

All samples that were analysed in the laboratory used appropriate standard methods (APHA 1998) by a National Association of Testing Authorities (NATA) accredited laboratory (Envirolab Services, Sydney) for determination of major anions, major cations and total metals (aluminium, barium, boron, cadmium, chromium, cobalt, copper, iron, lead, lithium, manganese, molybdenum, nickel, strontium, uranium and zinc). Total metal concentrations were determined by either Inductively Coupled Plasma- Mass Spectrometry (ICP-MS) or Inductively Coupled Plasma-Atomic Emission Spectrometry (ICP-AES). Detection limits for metals were generally 1 µg/L for most metals. Lower detection limits applied for cadmium (0.1 µg/L) and uranium (0.5 µg/L) and higher detection limits applied to manganese (5 µg/L), aluminium and iron (10 µg/L) and boron (20 µg/L).

Data Analysis

Multivariate analyses of family-level macroinvertebrate community data from samples collected from the Wingecarribee River were conducted to investigate the ecological response, if any, of river macroinvertebrates according to differences between the communities collected at sampling sites upstream versus downstream of the Berrima mine drainage inflow. This analysis has been used in many similar studies investigating the influence of coal mine waste waters on stream and river aquatic macroinvertebrate communities (Wright and Burgin 2009a, Wright et al. 2015 and Wright 2017). Non-metric multidimensional scaling (nMDS) has proven very effective in assessing ecological responses to human pressures, including assessing marine and freshwater pollution (Clarke 1993 and Warwick 1993).

In this study, nMDS was performed on the similarity matrix, computed with fourth-root transformed family-level macroinvertebrate taxon abundance data, using the Bray-Curtis dissimilarity measure (Clarke 1993; Warwick 1993). Two-dimensional ordination plots represented the dissimilarity among samples. A total of ten replicates represented for the site upstream, and ten replicates represented the site downstream of the mine drainage discharge. Community differences were evaluated by one-way analysis of similarity (ANOSIM: Clarke 1993). These multivariate analyses were achieved using the software package PRIMER version 6 (Clarke 1993).

Student's t test was used to test for differences between Wingecarribee River water quality and biotic index results (upstream vs. downstream).

Results

Water Chemistry

Water chemistry results show that the entry of the Berrima Colliery drainage caused major changes to the water quality of the Wingecarribee River (Table 1). Wingecarribee River EC increased from a mean of 258 µS/cm above the mine, to 403 µS/cm downstream, an increase of 57% (Table 1). The cause of the EC increase was due to the entry of the Berrima mine drainage, which recorded EC ranging from 848 to 1061 µS/cm during the study. The mean EC of 1000 µS/cm was nearly four times higher than the mean level in the Wingecarribee River, above the

mine. Un-published data supplied by the mine, collected from 2008 to August 2016, showed that the mean EC of the mine drainage was 684 $\mu\text{S}/\text{cm}$ when the mine was operating (up to November 2013; Fig. 2). After the deepest workings were flooded (October 2015 to March 2016) EC increased, peaking at 1150 $\mu\text{S}/\text{cm}$ in August 2016. EC in the mine drainage remained above 1000 $\mu\text{S}/\text{cm}$ between June 2016 and December 2016 and was generally lower from February to September 2017, ranging from 848 to 1060 $\mu\text{S}/\text{cm}$ (Fig. 2). Over the 13 months of this study, the mean pH of the Wingecarribee River marginally declined due to the mine drainage inflow, from 7.48 pH upstream of the mine to 7.24 downstream (Table 1). The pH of the mine drainage itself varied within a narrow range during the study from 6.51 in September 2016 to a low of 6.25 in February 2017 (Fig. 3). Unpublished data from the mine showed that the pH of the Berrima mine drainage was mildly alkaline when the mine was operating. The pH of the drainage slightly increased after the mine stopped commercial operation in late 2013. The pH then progressively fell (after the deepest mine workings were flooded) from 7.74 in June 2016 to 6.25 in February 2017 (Fig. 3).

The concentration of six metals (iron, lithium, manganese, nickel, zinc and strontium) in the Wingecarribee River increased significantly downstream compared to upstream of the Berrima mine drainage inflow (Table 1). The four metals with highest mean concentrations in the Berrima mine drainage were manganese, iron, zinc and nickel. The concentration of the major ions (sulphate, calcium and magnesium) was significantly higher in the Wingecarribee River, below the mine drainage inflow (Table 1).

The sulphate concentration in the Berrima mine drainage increased after the mine closed and its deepest workings were flooded in 2015/6 (Fig. 4). The sulphate concentration was approximately 220 mg/L when the mine operated (2008–2013) rising to just under 300 mg/L in March 2016, at the end of the period when its deepest workings were flooded. The sulphate concentration then progressively increased to a peak of 395 mg/L, recorded in the current study in September 2016, before receding over the following months, ranging from 373 mg/L in December 2016 to 290 mg/L in May 2017 (Fig. 4). The mine drainage inflow increased the mean sulphate concentration of the Wingecarribee River by 4.7 times, from a mean of 13.4 mg/L upstream to 76.4 mg/L downstream (Table 1).

The mean concentration of zinc in the Wingecarribee River increased by more than 100 times from 1.7 $\mu\text{g}/\text{L}$ (upstream) to 178.6 $\mu\text{g}/\text{L}$ downstream, during this study (Table 1). The mine drainage had a mean content of 1161 $\mu\text{g}/\text{L}$ of zinc, during this 13-month study (Table 1). Examination of historic data on zinc concentrations in mine drainage water revealed that the mean zinc concentration in the mine drainage was 254 $\mu\text{g}/\text{L}$ during commercial mining (2008–2013; Fig. 5). When the mine was partially sealed and flooded (October 2015 to March 2016) the concentration of zinc ranged from 115 to 212 $\mu\text{g}/\text{L}$ until increasing by more than ten times in July 2016 (2390 $\mu\text{g}/\text{L}$), reaching a peak in August 2016 (2410 $\mu\text{g}/\text{L}$), immediately prior to the current study. Mine drainage zinc results from the current study showed a steady decline from a peak of 2300 $\mu\text{g}/\text{L}$ in September 2016 to 650 $\mu\text{g}/\text{L}$ in September 2017 (Fig. 5).

The mean concentration of nickel in the Wingecarribee River increased by 90 times from 0.77 $\mu\text{g}/\text{L}$ (upstream) to 73.5 $\mu\text{g}/\text{L}$ (downstream), due to the inflow of the mine drainage, during this 13-month study (Table 1). The Berrima mine drainage had a mean concentration of 418 $\mu\text{g}/\text{L}$ of nickel over this study. Unpublished data from the coal mine showed that the mean nickel concentration in mine drainage was 149 $\mu\text{g}/\text{L}$ when the mine was operating (2008-

2013; Fig. 6). When the mine closed and flooded the deepest workings, the nickel concentrations fell below 100 µg/L, before reaching a low of 36 µg/L in March 2016. Nickel concentrations then increased steeply after the mine flooding (October 2015 to March 2016) to 262 µg/L in June 2016. Nickel then progressively increased each month until it recorded a maximum of 466 µg/L, in December 2016. Nickel has remained at concentrations above 390 µg/L throughout this 13-month study (Fig. 6). The mean concentration of total manganese in the Wingecarribee River increased by more than 80 times from 24.4 µg/L (upstream) to 2040 µg/L (downstream), over the 13 months of the study (Table 1).

Table 1. Water quality and macroinvertebrate biotic index summary statistics for the Wingecarribee River, collected at two river sites (WiU 100m upstream of the mine discharge and WiD 200m downstream) and from Berrima mine drainage adit (September 2016 to September 2017). Results for Students *t* test (and *p* values) are provided for evaluating statistical differences upstream versus downstream of the mine discharge. Other statistics provided are the range, mean and median (med.) for each variable.

Source of variation	<i>t</i> value (<i>p</i>)	WiU (upstream)		WiD (downstream)		Berrima mine drainage	
		Range	Mean (Med.)	Range	Mean (Med.)	Range	Mean (Med.)
Field meter							
pH (pH units)	6.65 (***)	7.27–7.69	7.48 (7.50)	6.98–7.49	7.24 (7.23)	6.25–6.51	6.36 (6.35)
Salinity (µS/cm)	8.39 (***)	87.7–357	254 (259)	258–539	403 (421)	848–1061	1000 (1041)
DO (% Sat.)	2.76 (**)	82.7–101	94.7 (98)	79.2–102	90.6 (87.2)	77.1–85.5	81.6 (81.7)
Turbidity (NTU)	5.30 (***)	0.8–21.4	4.24 (1.95)	3.9–27.6	11.6 (18.9)	2.9–21.1	9.93 (9)
Temperature (°C)	0.01 (ns)	8–21.3	13.9 (12.6)	9.2–20.4	13.9 (12.5)	14.2–16.6	15.4 (15.3)
Major ions (mg/L)							
Bicarbonate	0.12 (ns)	47–69	57.1 (55)	47–64	57.3 (60)	68–94	76.7 (74)
Chloride	1.76 (ns)	25–47	41.3 (41.1)	28–50	44.3 (43.5)	52–66	59.9 (60)
Sulphate	8.76 (***)	7–27	13.4 (12)	19–110	76.4 (74)	280–400	332 (330)
Calcium	7.42 (***)	11–16	13.5 (14)	14–35	24.7 (27)	58–92	70.5 (64.5)
Potassium	1.75 (ns)	3.4–5.8	4.3 (4.21)	3.1–5	3.90 (3.80)	2.7–4.1	3.24 (3.20)
Magnesium	7.92 (***)	6.8–9.0	8.22 (8.60)	8.4–26	18.4 (18)	57–67	60.9 (60)
Sodium	1.17 (ns)	18–45	29.4 (28)	24–46	32.2 (32)	37–51	45.5 (46)
Total metals (µg/L)							
Aluminium	0.63(ns)	30–490	107 (60)	20–470	80.5 (40)	30–50	42.7 (40)
Barium	0.97 (ns)	28–43	34 (34.5)	28–41	33 (33)	35–43	39.4 (39)
Boron	0.68 (ns)	9–20	10.4 (10)	9–20	10.9 (10)	10–25	14.7 (10)
Cadmium	–	Bd.	–	Bd.–0.1	0.055 (0.05)	0.4–0.5	0.42 (0.4)
Chromium	–	Bd.	–	Bd.	–	Bd.	–
Cobalt	–	Bd.	–	4–39	23 (21)	130–160	139.1 (140)
Copper	–	Bd.–2	–	Bd.–2	–	Bd.	–
Iron	8.33 (***)	210–830	398 (360)	950–2900	1829 (1650)	5400–14,000	10,939 (11000)
Lead	–	Bd.	–	Bd.	–	Bd.	–
Lithium	11.4 (***)	Bd.–1	0.74 (0.5)	7–18	12.8 (12)	55–58	57.3 (57.5)
Manganese	9.73 (***)	16–50	24.4 (21)	340–3300	2040 (1850)	11,000–13,000	11,909 (12000)
Molybdenum	–	Bd.	–	Bd.	–	Bd.	–
Nickel	9.93 (***)	Bd.–2	0.77 (0.5)	14–120	73.5 (66.5)	390–470	418 (410)
Strontium	6.74 (***)	63–100	77.3 (78)	73–140	112 (110)	240–270	250 (250)
Zinc	10.2 (***)	Bd.–4	1.7 (1.5)	76–290	178.6 (170)	640–2300	1161 (930)
Uranium	–	Bd.	–	Bd.	–	Bd.	–
Macroinvertebrate biotic indices							
Total abundance	5.1 (**)	18–121	77.5 (86)	4–37	18.8 (17.5)	Nt.	–
Family richness	6.92 (***)	11–26	18 (18.5)	3–11	6.6 (6)	Nt.	–
EPT richness	9.98 (***)	3–8	6.1 (6.5)	0–1	0.6 (1)	Nt.	–
EPT %	6.89 (***)	15.4–72.2	36.6 (33.7)	0–10	3.65 (3.84)	Nt.	–

Mayfly abundance – 3–22 13.4 (13) Nd. – Nt. –

EPT Ephemeroptera (E), Plecoptera (P) and Trichoptera (T), *Bd.* below detection limits, *ns* not significant, *Nd.* not detected, *Nt.* not tested **p* < 0.05; ***p* < 0.001; ****p* < 0.0001

This increase was due to the entry of mine drainage, which had a mean concentration of 11,909 µg/L of manganese (Table 1). In comparison, historic unpublished data showed that the mean manganese concentration in the mine drainage was 2290 µg/L, when the mine was commercially operating (2008–13; Fig. 7). The concentration of manganese initially fell (> 2000 µg/L) during flooding of the mine workings, reaching a minimum of 42 µg/L in June 2016. Concentrations then increased to 6210 µg/L later in June and then progressively increased each month to a peak of 13,000 µg/L in February 2017 (Fig. 7).

The mean concentration of iron in the Wingecarribee River increased from 398 µg/L (upstream) to 1829 µg/L (downstream) due to the mine drainage inflow, during this 13-month study (Table 1). Historic data on the mean iron concentration of the mine drainage showed that it was 127 µg/L when the mine was in active operation (2008–2013; Fig. 8). When the mine closed and flooded the deepest workings the iron concentrations was often less than 50 µg/L (Fig. 8). Three months after the mine workings were flooded, in June 2016, the iron concentration increased to 180 µg/L and then progressively increased on each sampling occasion (Fig. 8). In the 13 months of this study, the mean iron concentration of the drainage on each sampling occasion progressively increased steadily from 5450 µg/L in September 2016 to 14,500 µg/L in September 2017 (Fig. 8).

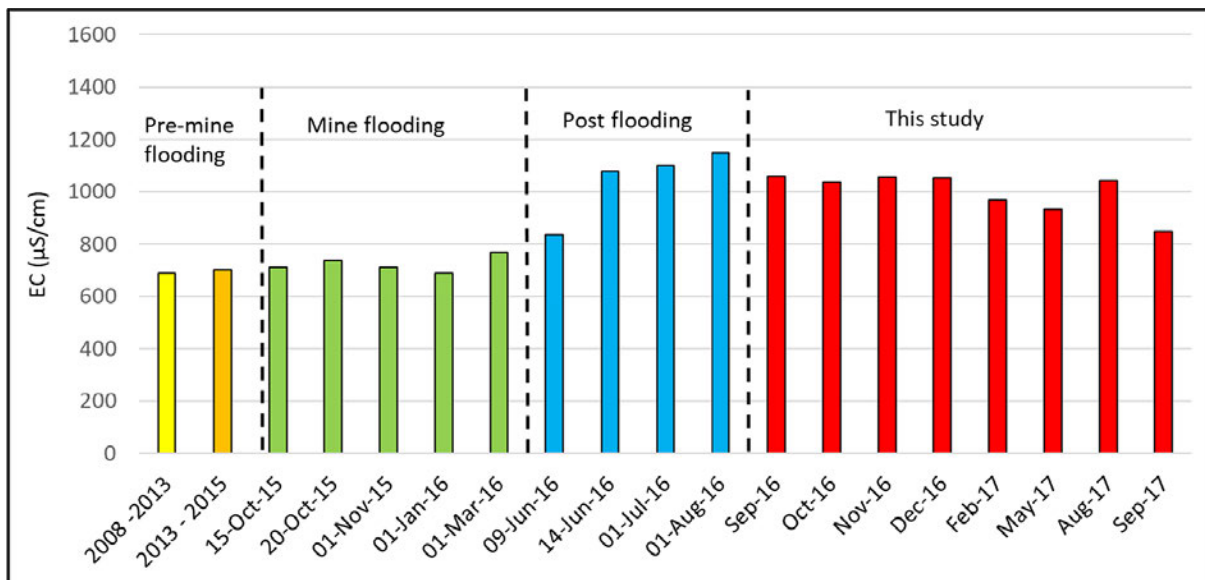


Figure 2. Mean EC (µS/cm) in Berrima mine drainage, including historic data (2008 to August 2016) and data from the current study. The first two bars represent ‘pre-mine flooding’, the first (yellow) bar was during mine operation, the second (orange) bar was during ‘care and maintenance’ with mine drainage pumped. The five green bars ‘Mine Flooding’ was the period when lowest mine working were flooded. The four blue bars were after the flooding when the mine returned to free-draining. The last eight (red) bars were collected during this current 13-month study. The ANZECC guideline for EC is 125 – 2200(µS/cm)

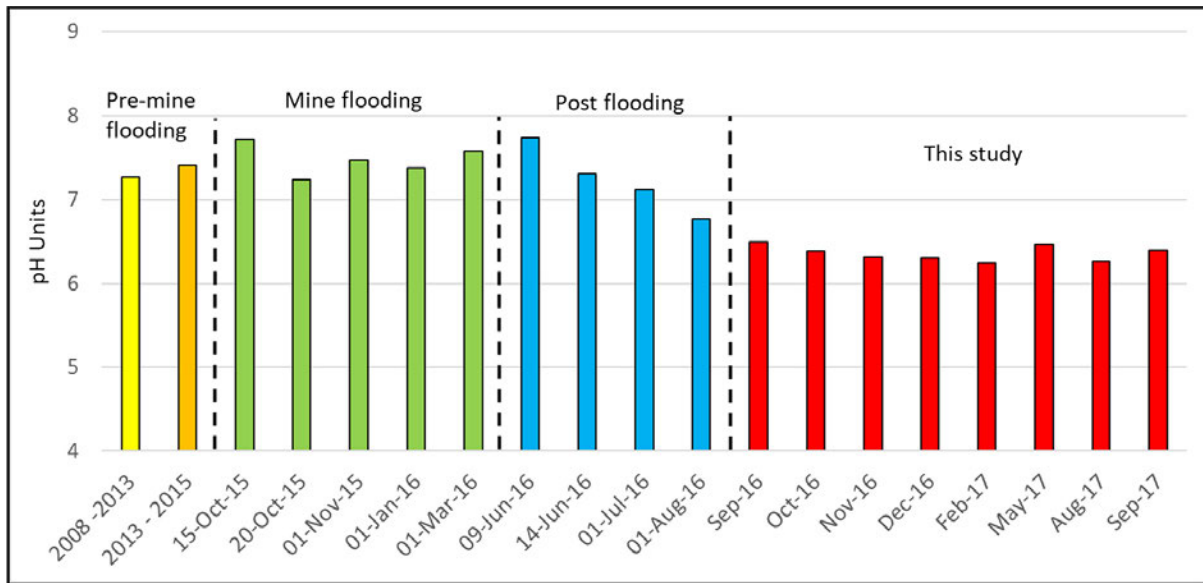


Figure 3. Mean pH (pH units) in Berrima mine drainage, including historic data (2008 to August 2016) and data from the current study. The first two bars represent ‘pre-mine flooding’, the first (yellow) bar was during mine operation, the second (orange) bar was during ‘care and maintenance’ with mine drainage pumped. The five green bars ‘Mine Flooding’ was the period when lowest mine working were flooded. The four blue bars were after the flooding when the mine returned to free-draining. The last eight (red) bars were collected during this current 13-month study. The ANZECC guideline for pH is 6.5 to 8.0 pH units

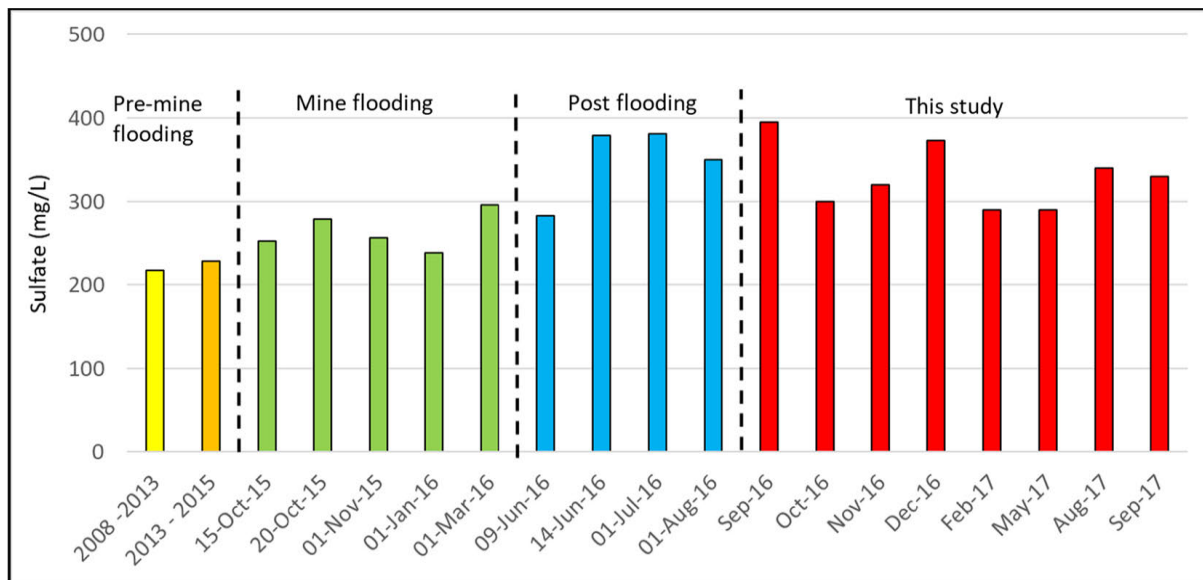


Figure 4. Mean sulphate (mg/L) in Berrima mine drainage, including historic data (2008 to August 2016) and data from the current study. The first two bars represent ‘pre-mine flooding’, the first (yellow) bar was during mine operation, the second (orange) bar was during ‘care and maintenance’ with mine drainage pumped. The five green bars ‘Mine Flooding’ was the period when lowest mine working were flooded. The four blue bars were after the flooding when the mine returned to free-draining. The last eight (red) bars were collected during this current 13-month study. There is no ANZECC guideline for sulphate.

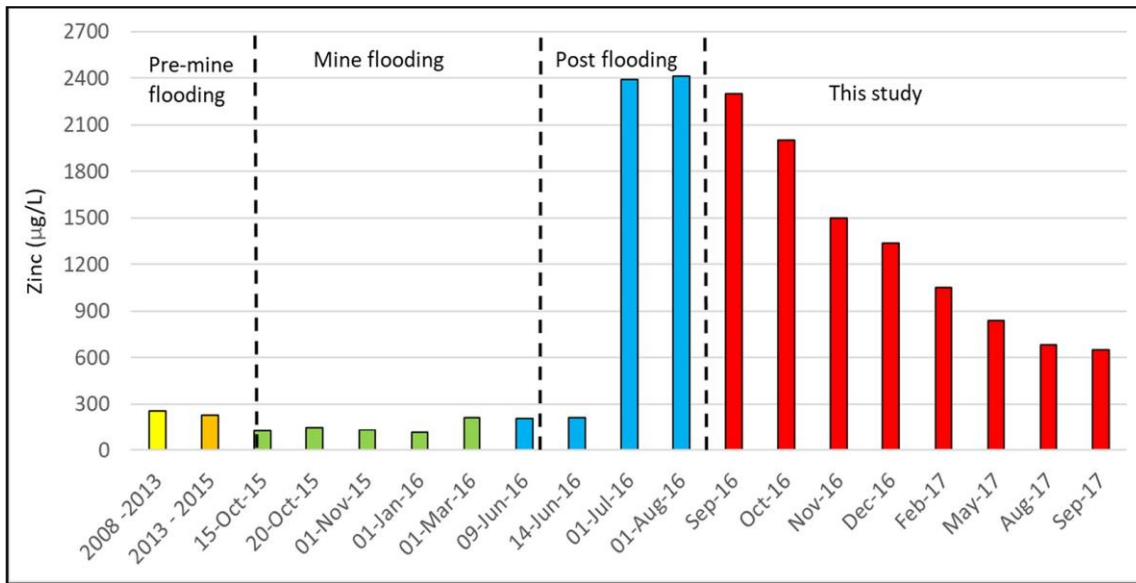


Figure 5. Mean zinc ($\mu\text{g/L}$) in Berrima mine drainage, including historic data (2008 to August 2016) and data from the current study. The first two bars represent ‘pre-mine flooding’, the first (yellow) bar was during mine operation, the second (orange) bar was during ‘care and maintenance’ with mine drainage pumped. The five green bars ‘Mine Flooding’ was the period when lowest mine working were flooded. The four blue bars were after the flooding when the mine returned to free-draining. The last eight (red) bars were collected during this current 13-month study. ANZECC guideline for zinc is $2.4 \mu\text{g/L}$.

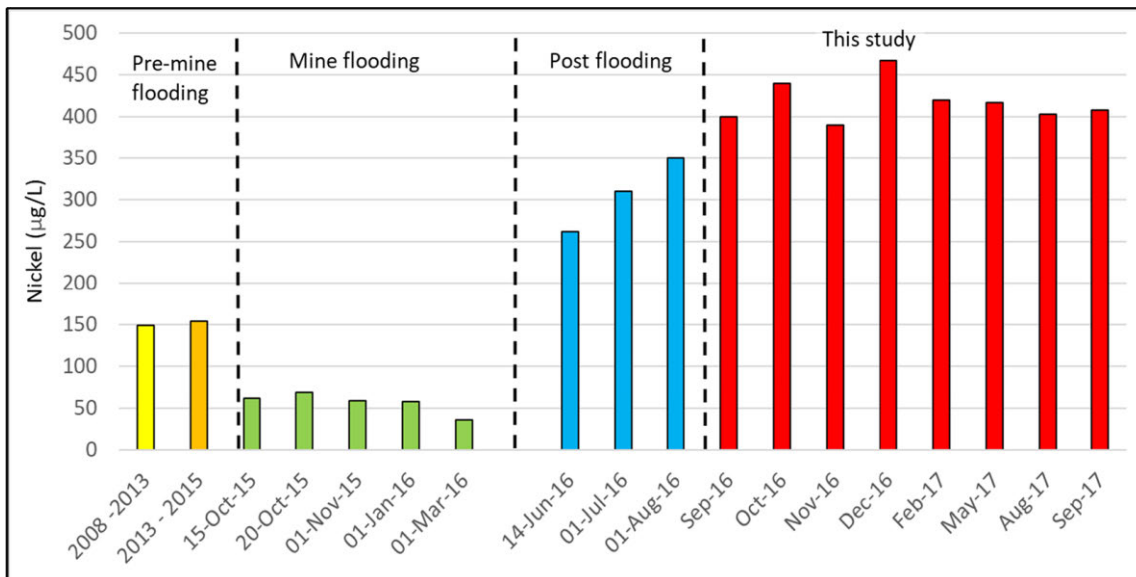


Figure 6. Mean nickel ($\mu\text{g/L}$) in Berrima mine drainage, including historic data (2008 to August 2016) and data from the current study. The first two bars represent ‘pre-mine flooding’, the first (yellow) bar was during mine operation, the second (orange) bar was during ‘care and maintenance’ with mine drainage pumped. The five green bars ‘Mine Flooding’ was the period when lowest mine working were flooded. The four blue bars were after the flooding when the mine returned to free-draining. The last eight (red) bars were collected during this current 13-month study. ANZECC guideline for nickel is $8 \mu\text{g/L}$.

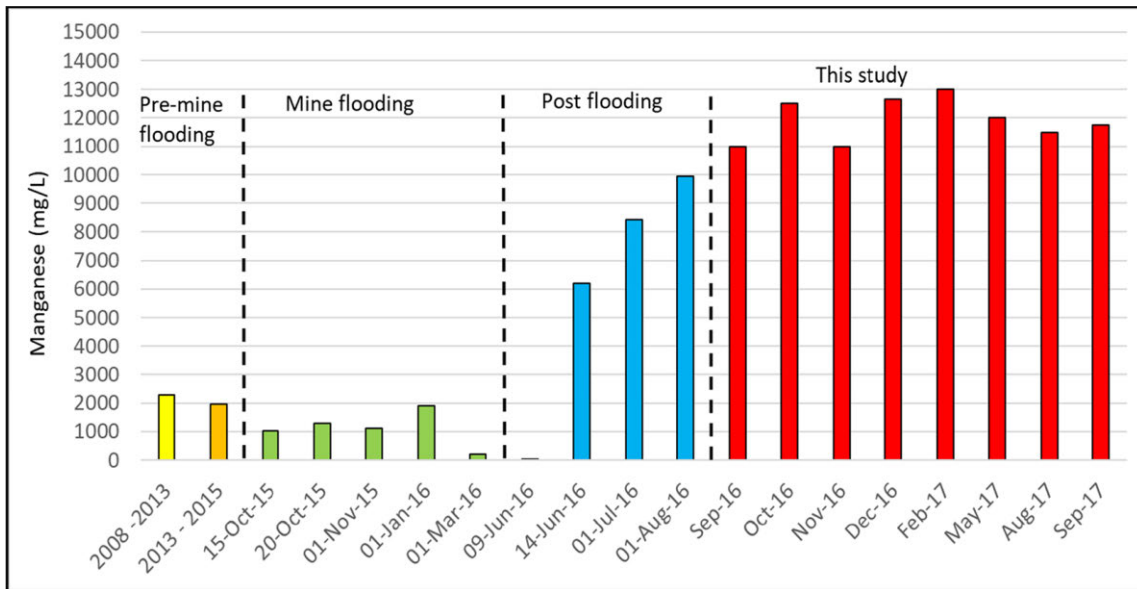


Figure 7. Mean manganese ($\mu\text{g/L}$) in Berrima mine drainage, including historic data (2008 to August 2016) and data from the current study. The first two bars represent ‘pre-mine flooding’, the first (yellow) bar was during mine operation, the second (orange) bar was during ‘care and maintenance’ with mine drainage pumped. The five green bars ‘Mine Flooding’ was the period when lowest mine working were flooded. The four blue bars were after the flooding when the mine returned to free-draining. The last eight (red) bars were collected during this current 13-month study. The Anzecc guideline for manganese is 1200 $\mu\text{g/L}$.

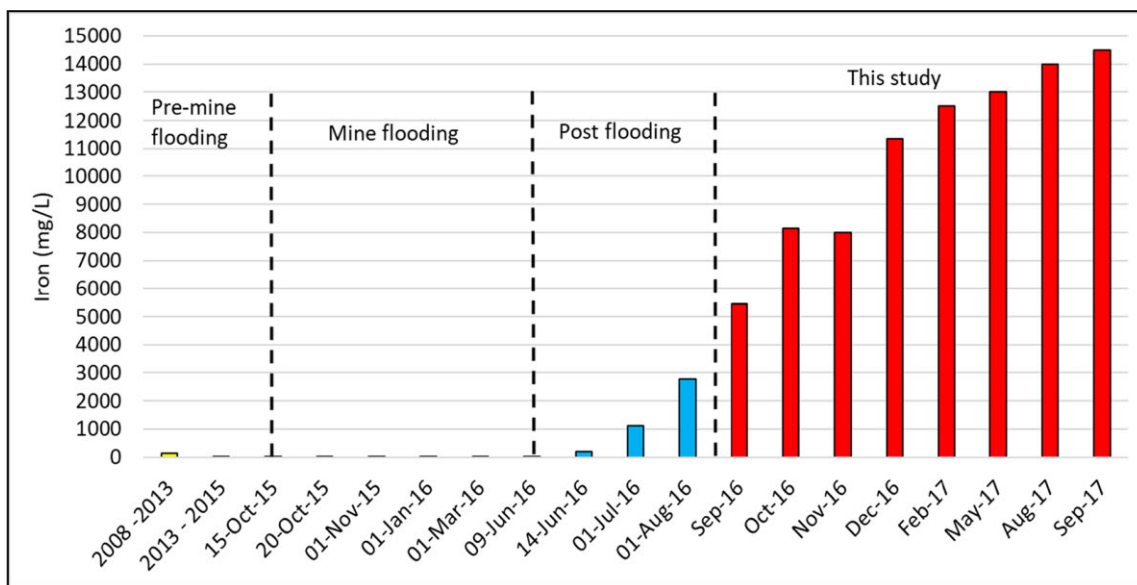


Figure 8. Mean iron ($\mu\text{g/L}$) in Berrima mine drainage, including historic data (2008 to August 2016) and data from the current study. The first two bars represent ‘pre-mine flooding’, the first (yellow) bar was during mine operation, the second (orange) bar was during ‘care and maintenance’ with mine drainage pumped. The five green bars ‘Mine Flooding’ was the period when lowest mine working were flooded. The four blue bars were after the flooding when the mine returned to free-draining. The last eight (red) bars were collected during this current 13-month study. There is no Anzecc guideline for iron.

Macroinvertebrates

A total of 963 aquatic invertebrates were collected and identified from the two sampling sites in this study. The collection comprised of 51 different taxonomic groups, mostly families. Most invertebrates were insects. Whilst the sampling effort was equal at both sites, 775 invertebrates from 45 families were collected from the Wingecarribee River upstream of the mine (mean of 18 families per sample) compared to 188 invertebrates from 25 families downstream (mean of 6.6 families per sample; Table 1). Elmidae beetles were the mostly commonly collected taxonomic group of aquatic invertebrates in this study (228 individuals), more commonly collected upstream (mean of 19.8 per sample) compared to downstream (mean of 3.3 per sample). The EPT orders were represented by 12 families in the study, with EPT taxonomic richness 63.3% lower, downstream of the mine (Table 1). The EPT orders represented a mean (per sample) of 36.6% of all invertebrates in samples collected above the mine, compared to a mean of 3.65% downstream (Table 1).

All 11 EPT families found in the study were detected in at least one sample collected above the mine (mean = 6.1 families per sample). Downstream of the mine, four EPT families were found (mean = 0.6 per sample; Table 1). Downstream of the mine, the four EPT families collected were all in the order Trichoptera (Hydropsychidae, Hydroptilidae, Hydrobiosidae and Philopotamidae). There were no Ephemeroptera (Mayfly) individuals detected in any samples collected downstream of the mine, compared to an average of 13.4 per sample, collected above the mine (Table 1). Multivariate (nMDS) analysis showed that Wingecarribee River macroinvertebrate assemblages were dissimilar, upstream compared to downstream of the entry of the Berrima mine waste (Fig. 9). The macroinvertebrate community structure varied highly significantly according to sampling site (Global R 0.594, $p < 0.001$). The two-dimensional NMDS ordination of macroinvertebrate samples from the Wingecarribee River had two clusters of samples (Fig. 9). The cluster, in the top left of the nMDS, is the group of Wingecarribee River samples collected upstream of the mine drainage. The second cluster is more scattered and is comprised of replicates from the samples collected below the inflow of the mine drainage (Fig. 9).

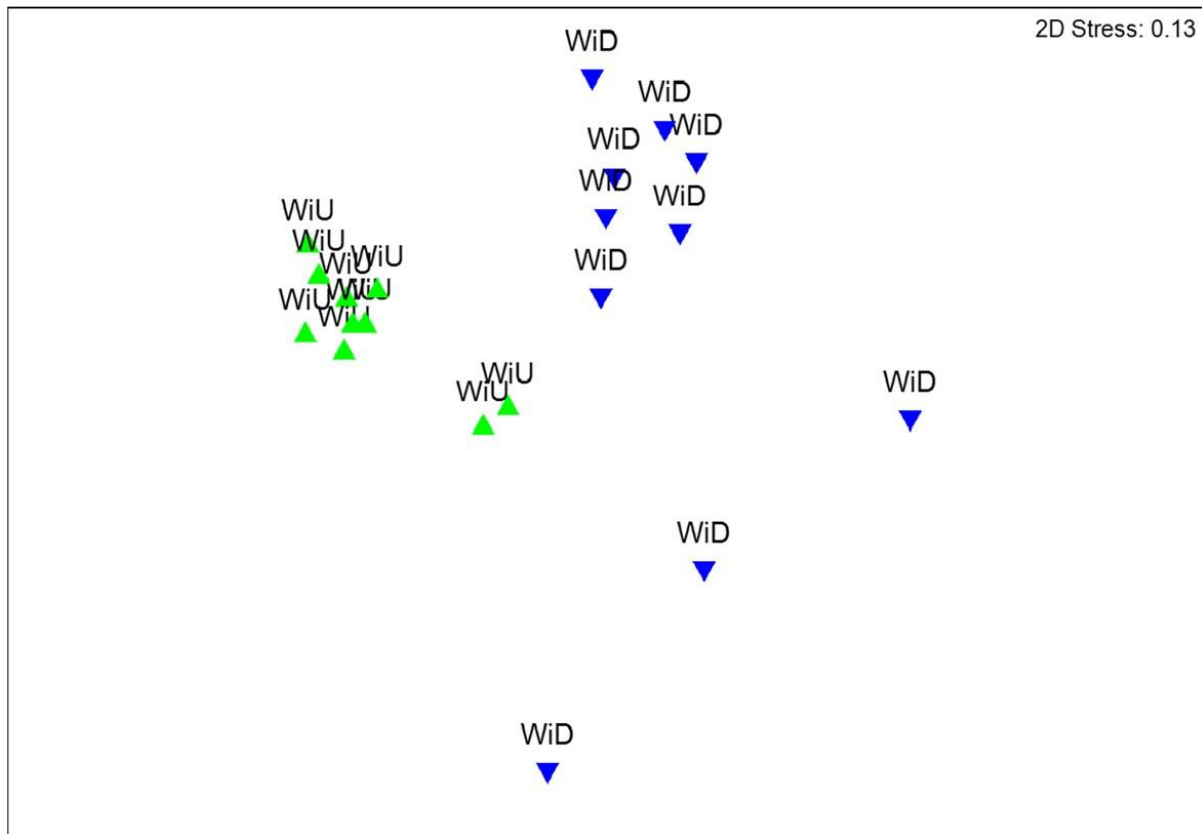


Figure 9. nMDS ordination of macroinvertebrate samples. Green triangles represent samples collected from the Wingecarribee River 100 m upstream (WiU) of the mine drainage. Blue triangles represent samples collected from the Wingecarribee River 200 m downstream (WiD) of the mine drainage.

Discussion

This study found that the inflow of mine drainage, from an underground colliery that had ceased coal extraction 3 years earlier, caused water pollution of a river with many pollutants at concentrations that were ecologically hazardous to its aquatic life. Comparison of data from the current 13-month study with several years of data provided by the mine owner indicated that the concentrations of several pollutants in the mine drainage have increased substantially since the mine stopped operating. The closure of the Berrima mine involved the cessation of underground water pumping and the subsequent flooding of the deepest sections of underground mine workings. Conversations with the mine owner suggest that approximately 15% of the mines underground workings have been flooded. The resulting rising water table in the mine is a process that has been termed rebounding by Johnson (2003) as accumulated groundwater floods some of the underground voids created by the decades of mining activity.

This triggered a substantial deterioration of the mine's drainage water quality, consistent with oxidisation of sulphur in the exposed coal in the previous workings and resulted in the 'first-flush' of contaminants that several authors observed after the widespread closure of many UK collieries in the 1960s to 1980s (Cairney and Frost 1975, Younger 1993 and Banks et al. 1997). The current case study appears to be one of the first Australian studies of an underground colliery that has examined progressive changes in mine drainage chemistry conducted during the mine closure process. Daily flow data from the coal mine owner showed that the discharge of mine drainage from the Berrima Colliery dropped from an average of about 2.6 ML/day, to less than 0.3 ML/day, for a 6-month period

(October 2015 to March 2016). Since March 2016, the drainage emitted from the mine returned to approximately 2.6 ML/day. This 6-month period of very low discharge volumes represents the rebound period of groundwater accumulating and flooding the deepest mine workings after pumping stopped.

It is expected that the rebound of the water table in this mine will not return to pre-mining levels as the mine's drainage adit in a deep gorge alongside the Wingecarribee River is well below the level of the pre-mining water table. Drainage from much of the mine works continues to emerge under gravity and geotechnical stability concerns may prevent it from being fully sealed. The flooding of closed underground coal (and other) mines in the UK has resulted, in some cases, of incidents of mine drainage 'outbursts' that have caused damage to surface structures and sudden floods of contaminated water (Younger 2002). The closure of the Berrima Colliery appears to have triggered an acceleration of AMD within the former underground mine workings. Whilst, the pH of the mine drainage was only mildly acidic (6.25– 6.51) in this study, unpublished data from the mine showed that it was generally circumneutral to mildly alkaline during the mine's operation and remained so until August 2016, following the partial flooding of the workings. Such a reduction of mine water pH after the mine workings was flooded shared similarities to a comparison of (mostly UK) circumneutral pH in 'pumping coal mines' contrasting with flooded coal mines which were generally acidic (Banks et al. 1997 and Younger 2001). The mildly acidic pH of mine drainage in the current study is very mild in comparison to other international examples as the pH of some closed coal mine waters can be highly acidic, with extremely acid waters as low as 2.2 pH in effluent from the abandoned Green Valley Coal Mine (Indiana, USA; Brake et al. 2001).

The sulphate concentration in the Berrima mine drainage was also indicative of AMD activity. After the mine ceased coal production in late 2013, the sulphate concentration steadily increased from c. 220 mg/L rising to 238 mg/L in January 2016 and then progressively increasing to a peak of 395 mg/L in September 2016. Such concentrations of sulphate in mine drainage waters are much higher than are typical for Australian studies. Price and Wright (2016) reported mean sulphate levels from the active Westcliff Colliery (33.8 mg/L) and from drainage from the closed Canyon Colliery (29.5 mg/L) were similar. However, they are much lower than reported in many international studies, with Banks et al. (1997) reporting sulphate in the waters of abandoned UK coal mines ranging from 83 to 1554 mg/L. Similarly, Pond et al. (2008) reported much higher sulphate in mined West Virginia streams (mean 696 mg/L) compared to unmined streams (mean 16 mg/L).

A comparison of data from this study with historic data collected by the mine owners showed that the concentration of four key metals in the Berrima mine drainage (iron, manganese, nickel and zinc) all increased in concentration after the colliery stopped dewatering and the groundwater table partially rebounded, over a 6-month period in 2015/6. The nickel concentration of mine drainage increased 2.5 times over this period, manganese by six times and zinc by 15 times. This was dwarfed by iron, which increased from 127 µg/L, during commercial mining (up to November 2013) with progressive increases after the workings were partially flooded in 2015/6 with a maximum iron concentration of 14,500 µg/L attained in September 2017. Such increased concentrations of metals are symptomatic of the deterioration in water quality of mine drainage as groundwater floods previously worked strata, this process has been referred to as first-flush (Younger 1993). The increased release of iron from a coal mine to local waterways is recognised as a major pollutant that Younger (2001) states: 'In terms of ecology the most

important single influence of the mine waters entering the Ore (River) is the smothering of the benthos with iron hydroxides.

Cairney and Frost (1975) studied the changes in iron concentrations in the Mainsforth Colliery (Durham, UK) before and after mining ceased. When pumping stopped and underground workings were flooded, they documented an increase in iron in the mine drainage, from 8 to 10 mg/L during commercial mining, increasing to 30 to 50 mg/L as the groundwater table in the mine rose by approximately 200 m (Cairney and Frost 1975). In the current case study, the increase in iron concentrations in the mine drainage from the Berrima mine was even steeper, approaching 100 times higher than that of the drainage produced when the mine was operating. The trajectory of the progressive increase in the iron concentrations in this study suggests that the mine drainage was still in the 'first flush' phase at the end of our 13-month study.

Whilst the concentration of iron increased steeply in this study, the concentration is modest by comparison, as it is about 40 times less than was reported from six closed mines (Durham, UK) which Younger (2000) reported mean iron concentrations of 425 mg/L. Despite the lower concentration, in the current study the Berrima mine drainage inflow to the Wingecarribee River increased mean river concentrations from 0.40 mg/L above to, 1.83 mg/L downstream, of the drainage outfall. Such an increase in iron is similar in magnitude to that detected in the River Ore (UK) where coal mine drainage increased iron from 0.1 mg/L upstream to more than 1 mg/L downstream (Younger 2001).

It is likely that the severity of ecological impairment detected in the Wingecarribee River in this study was also strongly influenced by the increased concentration of manganese, nickel and zinc emitted from the mine after it ceased mining. The concentrations of each of these three metals increased in the Wingecarribee River, below the mine drainage inflow, to levels that exceeded Australian guidelines for protection of aquatic species (ANZECC 2000). The ANZECC (2000) guidelines do not have recommended guidelines for iron for protection of aquatic species, unlike manganese, zinc and nickel.

Zinc is one of the three most hazardous pollutants that emerged in this study. The zinc concentrations in the Berrima Colliery mine drainage peaked at 2300 µg/L in September 2016 study, then falling on each of the six sampling occasions over the 8-month study, reaching a minimum of 650 µg/L in September 2017. The mean concentration of zinc in the Wingecarribee River, below the mine, (179 µg/L) was 18 times higher than the hardness corrected zinc guideline of 9.4 µg/L (99% species protection) or 3.6 times higher than 38.3 µg/L (95% species protection: ANZECC 2000). Zinc is a widespread pollutant commonly associated with liquid coal mine wastes in Australia; however, the level of zinc in the Wingecarribee River was higher than that recorded in many other studies across the Sydney basin. For example, a recent study of waste discharges from an active coal mine (Clarence Colliery) in the Wollangambe River caused increased zinc concentrations from < 5 µg/L upstream to a mean of 125 µg/L downstream (Wright et al. 2017). A milder increase in zinc was measured due to the inflow of Westcliff Colliery wastes (from 6.9 µg/L upstream to 33.2 µg/L downstream) in the upper Georges River (Price and Wright 2016). Zinc concentrations in the Wingecarribee River are lower than were recorded in Dalpura Creek in 2003, where

drainage from the Canyon Colliery, 6 years after it had closed, elevated the mean zinc concentration to 595 µg/L (Wright and Burgin 2009a).

Examination of historic drainage data, collected prior to this study, shows whilst the colliery was still extracting coal the mean zinc concentration was 257 µg/L. In January 2016, 8 months before this current study began, the mine was partly flooded with ground-water. Between June and July 2016, the zinc concentration in the drainage spiked, perhaps representing first-flush, from 212 to 2390 µg/L. Our study showed that zinc concentrations remained at levels at, or above, 2000 µg/L for about 4 months. Such elevated concentrations of zinc in coal mine drainage exceed levels published in the drainage from any other Australian colliery. In comparison, the mean zinc concentration of Westcliff Colliery drainage was 38.6 µg/L and the closed Canyon Colliery drainage was 370 µg/L (Price and Wright 2016). Internationally, the zinc content of Berrima Colliery drainage is higher than many other coal mine studies. Banks et al. (1997) reported zinc concentration in 'pumping coal mine' drainages ranged from 34–56 µg/L. The same study reported a wider range of zinc (< 7–221 µg/L) in drainage from abandoned mines. Similar zinc concentrations (3–60 µg/L) were also reported by Younger (1993) from nine Durham colliery 'pumping discharges. However, some of the highest zinc concentrations (540–12,400 µg/L) were reported by Brake et al. (2001) in 'unmixed effluent' associated with coal tailings from the abandoned Green Valley Coal Mine (Indiana, USA).

Nickel was a second ecologically hazardous pollutant associated with the Berrima mine drainage and pollution of the Wingecarribee River. The Berrima mine drainage had an average concentration of 418 µg/L of nickel, over the 13 months of this study. This study found that the mean concentration of nickel in the Wingecarribee River increased by more than 90 times from 0.77 µg/L (upstream) to 73.5 µg/L (downstream), during this 13-month study. Such concentrations confirm that nickel is ecologically hazardous, as most samples collected from the river below the mine drainage exceeded hardness corrected values for nickel (for aquatic ecosystems) of 31.2 µg/L, for 99% species protection, and 42.9 µg/L, for 95% species protection, according to Australian guidelines (ANZECC 2000). Similar to zinc, nickel is a common pollutant associated with coal mine wastes. However, some coal fields do not have similar problems with nickel. For example, waterways affected by coal mining in the central Appalachians (USA) had mean nickel concentrations in mined streams of 14.2 µg/L, compared to unmined streams (< 10 µg/L) according to a study by Griffith et al. (2012).

Nickel concentrations in the Berrima mine drainage increased following cessation of mining. Unpublished data from the mine owner showed that the mean nickel concentration in the drainage was 150 µg/L when the Berrima mine was extracting ore, prior to November 2013. The concentration then progressively increased, particularly after January 2016, when the mine workings were partially flooded, peaking at 466 µg/L in samples collected in this study in December 2016. During the current 13-month study, the nickel concentration in the mine drainage has remained in the range of 390 to 466 µg/L. Unlike zinc, nickel concentrations have not shown any substantial decline over the duration of the current study. It is not known how the nickel concentration will change in future years, but it is possible that the continued release of dangerous concentrations of nickel may remain at ecologically dangerous levels for decades after the coal mine closes. For example, Canyon Colliery, which is located 105 km to the north of the Berrima coal mine, discharged elevated levels of nickel measured at mean concentrations of 210 µg/L into

Dalpura Creek 17 years after its closure (Price and Wright 2016). Contaminated drainage from the Green Valley coal mine (Indiana, USA) was investigated 36 years after its abandonment by Brake et al. (2001) with concentrations of nickel above 500 µg/L. It is plausible that the emissions of elevated concentrations of nickel may continue from the Berrima Colliery for decades.

The third metal in the Berrima Colliery drainage that poses a major hazard to aquatic life in the Wingecarribee River is manganese, which ranged from 11,000 to 13,000 µg/L in the mine's drainage. This study detected concentrations in the river, below the waste discharge point, of 340–3300 µg/L. The mean concentration was 2040 µg/L, which was greater than both the 95% species protection guide-line (1200 µg/L) and the 99% species protection guideline (1900 µg/L: ANZECC 2000). The concentrations of manganese in mine drainage in this current study are unusually high, compared to other Australian studies, which generally do not exceed ANZECC (2000) guidelines. For example, the Westcliff Colliery mine waste had a manganese concentration of 74.6 µg/L, which caused a decrease in Georges River manganese concentrations from 159 µg/L upstream to 151 µg/L downstream (Price and Wright 2016). The Tahmoor Colliery discharge also caused a fall in manganese concentrations from 123 µg/L upstream to 28.2 µg/L downstream (Wright et al. 2015). The highest reported Australian manganese concentrations associated with a coal mine was the closed Canyon Mine which released drainage with a mean 417 µg/L, increasing Dalpura Creek manganese from 18 µg/L (upstream) to 360 µg/L downstream (Price and Wright 2016). Internationally, manganese has often been associated with water contaminated from coal mines. For example, Merriam et al. (2011) reported a mean manganese concentration in a West Virginia (USA) study of coal mine affected waters of 170 µg/L. More similar to the current study, Petty et al. (2010) reported mean manganese concentrations of 2190 µg/L in intensively mined streams in the Freeport areas of the Appalachians (USA).

Abandoned coal mine waters from various UK locations had widely varying manganese concentrations ranging from 138 to 6100 µg/L (Banks et al. 1997). These international comparisons suggest that the manganese concentrations recorded in this mine drainage are very high. The substantial and rapid deterioration in water quality recorded in this study occurred after the Berrima Colliery closed and groundwater flooded sections of previously dry underground workings. The phenomenon of increased concentrations of metal pollutants in coal mine drainage of recently flooded mines has been reported from UK studies (e.g. Younger 2000 and Younger 2002) but has not previously been reported following the closure of an Australian coal mine. It is not known why the concentrations of pollutants increased so quickly and steeply after part of the mine was flooded but appears to be indicative of accelerated AMD. The coal resources of the Wongawilli seam, part of the Illawarra Coal Measures, is understood to have a very low (< 0.6%) sulphur content (NSW Resources and Energy 2016 and Huleatt 1991). The sulphur content of coal is a major driver for AMD (e.g. Brake et al. 2001). However, such low sulphur content values in the Berrima coal seam (Wongawilli; Huleatt 1991) appear inconsistent with the elevated sulphate concentrations in the Berrima Colliery mine drainage.

The sulphate concentration in the flooded Berrima mine drainage, in this study, ranged from 280 to 400 mg/L. This is higher than has been published for sulphate concentrations in undiluted mine drainage in Sydney basin coal mines, with the previous highest concentration of 90 to 225 mg/L recorded from the waste discharge from the active Clarence Colliery (NSW OEH 2015) in the western coalfields of the Sydney basin. Sulphate concentrations were

lower in mine discharges from the Sydney basin southern coalfields, with up to 42 mg/L recorded from the active West Cliff Colliery and 30 mg/L from the closed Canyon Colliery (Price and Wright 2016). It is likely that the sulphur content of coal in the closed workings of the Berrima Colliery is higher than the 0.48–0.59% attributed to the Wongawilli seam (Huleatt 1991). It is recommended that the sulphur content of remaining coal is thoroughly evaluated as part of the planning for mine closure prior to flooding, to predict the future AMD risk.

The damage to aquatic life in this study was assessed by the collection of macroinvertebrate samples from the Wingecarribee River, 39 months after the Berrima Colliery ceased mining. The results illustrate that river invertebrate communities were strongly impaired by the mine discharge. This study found that the mean taxonomic (family) richness in the Wingecarribee River dropped by approximately 63%, from 18 families per sample above the mine waste to 6.6 families below the mine outflow. This impact is similar to that reported from many other Australian and international studies.

In the Wollangambe River, 113 km north of the Berrima mine, the active Clarence Colliery waste discharge caused mean family richness to fall from 8.7 families upstream to 3.2 families downstream (Wright et al. 2017). Wright and Burgin (2009a) also reported a similar level of reduction of taxonomic richness in the Grose River with 14 to 25 families reported at reference sites and 9 to 12 families below the inflow of contamination drainage from the closed Canyon colliery. The reduction of invertebrate family richness in the Wingecarribee River in the current study was larger than was found in a study of waterways affected by mountaintop coal mining of West Virginian streams, which had an average of 20 families in unmined streams compared to an average of 11.7 families at mined sites (Pond et al. 2008).

The current study recorded a 90% decline in the mean proportion of invertebrates in the pollution sensitive EPT orders (Lenat and Penrose 1996) in Wingecarribee River samples collected below the coal mine waste discharge point (EPT 3.65%), compared to upstream (EPT 36.6%). This is one of the largest declines in the proportion of EPT invertebrates reported, due to the discharge of coal mine drainage. This provides an indication that the degree of ecological impairment of aquatic life in the Wingecarribee River, due to the waste discharge, is substantial on a global scale. For example, a study of waterways in West Virginia affected by coal mining (Pond et al. 2008) reported a modest decline in the mean proportion of EPT invertebrates at several mined streams of 51.1% compared to 77.9% at unmined streams. The reduction of EPT invertebrates in the current study was larger than that reported in a recent study of the nearby Wollangambe River. That study reported that the proportion of EPT invertebrates at reference sites was 47% compared to 13% immediately downstream of the Clarence Colliery waste discharge (Wright et al. 2017). Another nearby study of the Bargo River revealed that waste discharges from the Tahmoor Colliery caused a modest reduction in the proportion of EPT invertebrates from 55% upstream of the waste discharge to 37.2% downstream (Wright et al. 2015). Gray and Harding (2012) advocate the use of taxonomic richness of EPT taxa as an indicator of AMD impact after an extensive study of coal mining impacts from 91 locations. In the current case study, the mean taxonomic richness of EPT taxa was 6.1 upstream declining to 0.6 downstream, also indicative of major ecological impairment.

Results of this study question the effectiveness of the environmental regulatory regime imposed on this mine by the NSW EPA which provided inadequate restrictions of the pollutants released by the mine through the mine drainage adit to the Wingecarribee River. The Berrima colliery holds an environmental license (EPL no. 608), enforced by the EPA (NSW EPA 2017a), which permits the discharge of mine drainage to the Wingecarribee River. The license only identified four pollutants (oil and grease, total suspended solids, pH and biochemical oxygen demand) with specific discharge limits for each, in the colliery drainage.

The five most ecologically hazardous pollutants revealed by this study (nickel, zinc, manganese, iron and salinity) were not specified in the license (EPL 608), with specific discharge limits, and thus were effectively unregulated. Such a deficiency in the Berrima mine's environmental license (EPL no. 608) is not isolated to this case, as regulation of specific water pollutants in waste discharges to environmental appropriate levels has been found to be lacking at many coal mine and other industrial discharges in NSW (Wright et al. 2011 and Graham and Wright 2012). Such deficiencies in the regulatory regime have additional implications given that the Berrima Colliery is releasing untreated contamination that is damaging the aquatic ecology in one of the largest rivers in the catchment of Sydney's largest drinking water storage (Lake Burragorang, Warragamba Dam). The pollution and ecological damage caused by the Berrima Colliery, as described in this current study, was not identified in the 2013 drinking water catchment audit (GHD 2013) but was recognised as an issue of community concern in the 2016 catchment audit (Alluvium 2017).

The current study represents the only Australian water quality and stream ecology research that investigates the impact of underground coal mine drainage after an underground colliery has stopped mining after many decades of continuous operation. This study had the benefit of many years of data collected by the mine owner prior to the current study and has revealed that the water quality of the mine drainage became much more hazardous to aquatic life after the mine stopped coal production. The gradual accumulation of groundwater in the underground mine workings that resulted in the partial flooding of sections of the mine appears to have triggered an acceleration of AMD resulting in the higher concentration of iron, nickel, zinc and manganese in the mine drainage. It is unknown how long the dangerous levels of metals will remain present in the mine drainage. Thankfully, the concentration of zinc in the mine drainage has fallen during the study, but nickel and manganese remain at elevated concentrations that are damaging to aquatic life in the Wingecarribee river. The concentration of iron was still increasing at the end of the study. It appears to be very likely that this mine will join the growing list of closed and abandoned mines across the globe that continue to pollute waterways and impair aquatic life for many decades, possibly for centuries.

Research significance

This research builds on the limited research available on coal mine pollution to their receiving waterways within Australia. The research has produced the first detailed chemical and ecological study on the effects of coal mine wastewater discharges from a group of active and inactive coal mines. The inclusion of inactive mines will document the ongoing impacts to waterway health after coal mining has ceased and the degradation of their receiving waterways (chemistry and ecology) which can become a long-term legacy.

The research has filled a major data gap in the planning of proposed coal mines and helps manage the ongoing impacts associated with existing coal mines. In addition, the research provides a better understanding of the legacy of pollutants being left behind and the extent of the contamination within the aquatic and perhaps the terrestrial environment. This knowledge is imperative to better manage current coal mining operations, proposed extensions to existing operating coal mines and newly proposed coal mines throughout the Sydney Basin and Australia.

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Chapter 3

Heavy metal contamination of water column from a coal mine wastewater discharge resulting in mobilisation of metal contaminants to riparian vegetation. Wollangambe River, Blue Mountains Australia.

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Abstract

This study investigates the mobilisation of heavy metal contaminants from the Wollangambe Rivers water column to one species of terrestrial riparian flora (*Acacia rubida*) from one regulated coal mine waste water discharge. The study was conducted at one upland stream (The Wollangambe River) found within the Blue Mountains region of Sydney, New South Wales Australia. Two sample sites were used for this study, one as a reference site upstream of Clarence Collieries waste water inflow to the Wollangambe River (W1). The second was located approximately 200m downstream of Clarence Collieries waste water inflow to the Wollangambe River (W3). Five replicated samples were taken from both sample areas. Plants were selected within a 10m lineal stretch of stream edge. Each replicated sample was delivered to a NATA accredited commercial laboratory (EnviroLab Sydney) and analysed for 10 metals (Aluminium, Arsenic, Beryllium, Cadmium, Cobalt, Molybdenum, Nickel, Strontium, Thallium and Zinc). Results found statistical differences between nine of the ten heavy metals when compared between sample location (upstream and downstream). This study has shown that one coal mine waste water discharge appears to have allowed an avenue for increased heavy metal concentrations within the Wollangambe Rivers water column to mobilise to riparian vegetation found within the terrestrial environment. The implications that the licensed waste water discharges contaminants are mobilising to terrestrial riparian vegetation is of major concern. It is recommended that further research should be undertaken by the NSW EPA to better assess the implications of heavy metal mobilisation to the terrestrial environment from EPL protected waterways. If in fact heavy metal contaminants are leaving the water column of their receiving waterways and mobilising to the terrestrial environment, serious long-term legacy pollutant impacts may persist.

Key words

Bioaccumulation, heavy metal bioaccumulation, flora, native flora, coal mine wastewater, Blue Mountains National Park, Wollangambe River.

Introduction

Coal mining practices are well documented to contribute to an array of differing environmental problems including air pollution, fire hazards, ground subsidence or deformation, surface and or ground water pollution. Surface water pollution is a major environmental problem associated with coal mining and it occurs through the discharge of mine waters that are contaminated by various disturbances associated with mining practices (Jarvis and Younger 1997, Johnson 2003 and Pond et al. 2008).

Water pollution from coal mining occurs as large volumes of surface and groundwater are required to be removed from most underground coal mines. This is generally through the pumping of water to the surface as surface and groundwaters infiltrate the mine shafts from the local geological sub-strata and subsequently accumulates in the underground mine workings. Without this, groundwater would flood most sections of the underground mining operation (Jarvis and Younger 1997 and Younger 2004).

Coal mine wastewater will often be contaminated due to the disturbance of the local geology associated with mining activities. The exact nature of the water contamination will vary depending on local factors such as groundwater geochemistry, hydrology and mineralogy of the local strata. In addition to the physical activity of the mining operation and the removal of the waste water, other activities will also often contaminate water used throughout a mining plant which can include; coal washing and the inclusion of other wastes generated by the surface operation at the mine such as sewage wastes (Younger 2004).

The Wollangambe River flows entirely within the Greater Blue Mountains area some 90 km west of the Sydney CBD and for most of its flow it resides within the World Heritage Greater Blue Mountains National Park and is a protected tributary of the Colo River a declared wild river. In the year 2000 the Greater Blue Mountains National Park was inaugurated into the World Heritage List due to its diverse natural values (Australian Government, Department of the Environment, Water, Heritage and the Arts 2012).

The Wollangambe Rivers headwaters researched in this study lies within the Western section of the Greater Blue Mountains World Heritage National Park bordering the Wollemi National Park and the Gardens of Stone National Park. The Wollangambe River flows from Newnes Junction at an elevation of some 1050m above sea level. It continues through pristine wilderness over a distance of near 60km until it joins the Colo River at an approximate elevation of 50m above sea level (Department of Environment and Climate Change 2009).

In recent years, the concern about impacts of toxic metals in global ecosystems has reached unparalleled heights. Exponentially increasing industrial and mining activities have led to further environmental pollution through wastes produced by these activities. Heavy metals from mining and industrial activities, when released to the environment, have the potential to accumulate within biota at toxic concentrations and cause chronic ecological impacts in ecosystem food chains (Sericano et al. 1995 and Ashraf et al. 2011). Metal pollution is a global environmental issue and has been for many decades and when mobilised metal pollutants can enter waterways indirectly through land runoff. In many cases metal pollutants are directly discharged into waterways from anthropogenic activities. Many of these metal pollutants become absorbed to suspended

particulates within the water column. Once absorbed to these suspended particulates the metal contaminated suspended particulates eventually deposit into waterway sediments often remaining indefinitely (Neff 1984 and Wang and Rainbow 2008).

Aquatic ecosystems are often more highly sensitive to heavy metal contamination from anthropogenic activities, especially as waterways are often used as sources of discharge of many anthropogenic industrial wastes (Allen et al. 1993). Heavy metal contamination within aquatic environments can persist for much longer than terrestrial organic pollutants. This is due to the lack of biodegradation function of heavy metals in aquatic ecosystems in comparison to terrestrial ecosystems (Ashraf et al. 2011). Metal bioaccumulation has been reported within many aquatic flora and fauna species worldwide (Hill et al. 2000 and Amish and Cowx 2000). One study found bioaccumulation of arsenic and selenium within caddisflies Reash et al. (2006) of coal ash polluted sites whilst increases in levels of pollutants were also recorded for turtles, crayfish, tadpoles and varying fish species by Otter et al. (2012).

Durães et al. (2014) investigated two differing soil pH levels (acidic and alkaline) impacted by mining activities and their influence on metal and metalloid bioavailability in Portugal and Morocco respectively. Durães et al. (2014) stated that some aquatic and terrestrial flora species are more tolerant than others and can often tolerate high levels of metal concentrations in their tissue. It was found that the mobility of metals and metalloids in acid mine waters (low pH) from Portugal bio accumulated the metals Zn, Cu and Pb in that order whilst in contrast the (high pH) environment of Morocco changed the order of each metals bioavailability in the order of Pb, Zn and Cu. Durães et al. (2014) concludes that the “local minerology and pH conditions are decisive in the mobility and uptake of metals by plants” (Durães et al. 2014).

Investigations into the bioaccumulation of heavy metals in soil and native plant species, in particular (Cr, Co, Cu, Fe, Mn, Ni and Zn) from mining activities in Northern Pakistan by Nawad et al. (2015) found impacted soil samples to be highly significantly different ($p < 0.01$) when compared to reference soil. Nawad et al. (2015) also found heavy metal accumulation was highly significantly different for all plant species used in their study (Nawad et al. 2015).

A study assessing the bioaccumulation of heavy metals within plant species growing on land used as a uranium mining dump was undertaken in the Karkonosze-Izera region of the Sudety Mountains south western Poland by Wislocka et al. (2006). Heavy metal contamination of soils was investigated and concentrations of the heavy metals Cd, Cu, Fe, Mn, Ni, Pb and Zn were reported within the uranium mining dump sites. Three plant species growing in the area were used to assess the level of bioaccumulation of these contaminants, two being described as trees and one as a shrub. Results showed elevated heavy metal concentrations were accumulating within all three species, with both tree species having higher concentrations than that of the shrub species. These heavy metal concentrations were stated to be “above the average values given for plants in other literature” (Wislocka et al. 2006).

Bioaccumulation of the metals Fe, Mn, Zn, Cu, Cd and Ni within fruit and timber trees grown on grounds contaminated by coal mine spoils was found to be higher than that of control trees Maiti et al. (2015) found. The order of bioaccumulation was reported as Fe, Mn, Zn, Cu, Cd and Ni (ascending from highest to lowest concentrations). pH recorded at the coal mine spoil contaminated site was found to be acidic (pH 4.34 – 4.95). Significant variations of metal accumulations were recorded for timber tree species whilst high metal accumulations were found for fruit tree species (Maiti et al. 2015). Maiti et al. 2015 concludes that although remediation of the coal mine spoil contaminated soil was undertaken via the addition of top soils to cover the mine spoil that no significant improvement was observed for the bioavailability of metal concentrations within the mine contaminated soils.

Methods

This study was conducted at one upland stream (The Wollangambe River) found within the Blue Mountains area of Sydney, New South Wales Australia.

Two sample sites were used for this study, one as a reference site upstream of Clarence Collieries wastewater inflow to the Wollangambe River (W1). The second was located approximately 200 m downstream of Clarence Collieries wastewater inflow to the Wollangambe River (W3). Five samples (whole plants) were taken from both sample areas. Plants were selected within a 10 m lineal stretch of stream edge. All of the foliage was removed from each plant sampled at the Western Sydney University laboratory and stored in separate sample containers. Each replicate sample container was delivered to a NATA accredited commercial laboratory (EnviroLab Sydney) and analysed for 10 metals (Aluminium, Arsenic, Beryllium, Cadmium, Cobalt, Molybdenum, Nickel, Strontium, Thallium and Zinc). One species of Wattle (*Acacia rubida*) was used for this study as it was found to grow in abundance within the riparian zone of each sample location.

The ten metals analysed have been found to be in higher concentrations within the water column and river sediments at the same downstream sample location in previously published and currently submitted manuscripts of the authors of this study, with seven of the metals being statistically different for both water column and river sediment when compared between upstream and downstream sample locations. For instance, Cadmium, Cobalt, Molybdenum and Nickel have been recorded as below laboratory detectable limits within the water column upstream of the coal mine wastewater inflow whilst only Cadmium and Molybdenum remain below laboratory detectable limits downstream of the coal mine wastewater inflow. In contrast, river sediment concentrations upstream record Cadmium, Cobalt and Molybdenum below laboratory detectable limits whilst all of the metals are detectable downstream of the coal mine wastewater inflow.

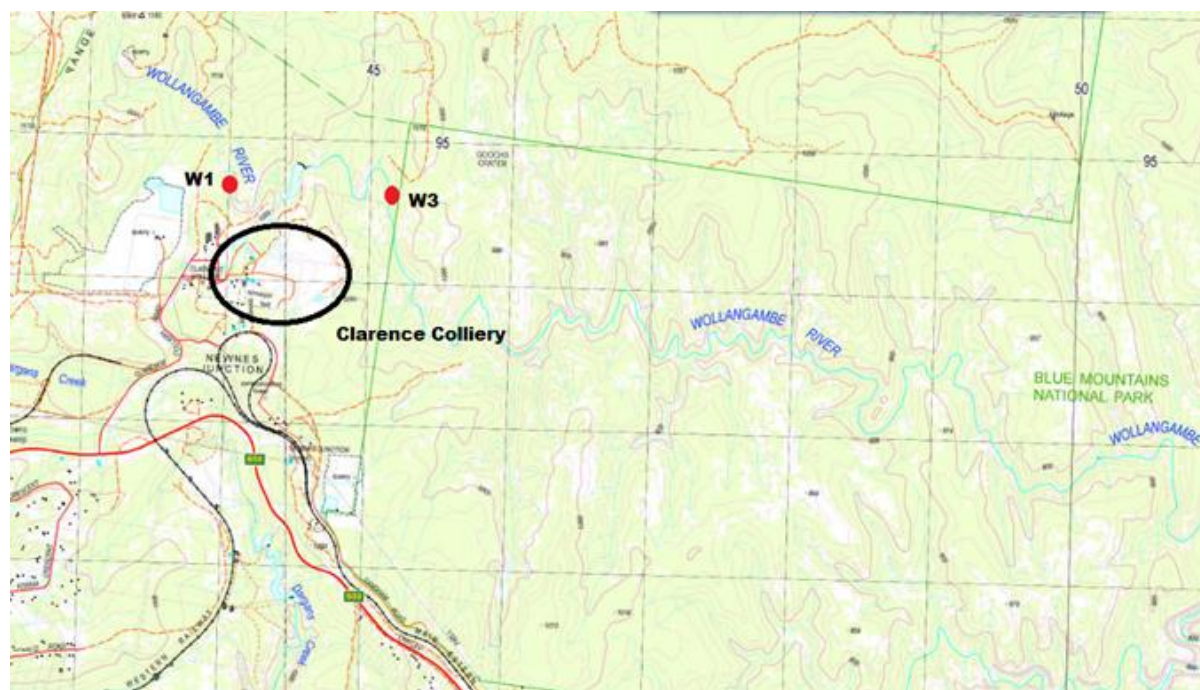


Figure 1. Map of sample locations W1 (upstream) and W3 (downstream). W1 is located approximately 200m upstream of the Clarence Colliery waste water inflow whilst W3 is approximately 200m downstream of the inflow (NSW Six Maps 2017).

Univariate data analysis was used to compare metal concentrations between samples collected upstream of the coal mine wastewater inflow to samples collected downstream. This was performed using Students t-test.

Results

Results show that nine of the ten metals analysed were found to be statistically different between upstream and downstream sample locations with all nine increasing downstream of the coal mine wastewater discharge. Arsenic was not recorded to be statistically different (Table 1).

Aluminium increased from a mean of 12253 ug/kg upstream of the mine wastewater inflow to a mean of 14949 ug/kg downstream ($p=0.009$). Upstream aluminium ranged from 10646 to 13752 ug/kg whilst downstream ranged between 13965 and 16102 ug/kg (Table 1 and Figure 2). Beryllium increased over 4 times from a mean of 6.0 ug/kg upstream of the mine wastewater inflow to a mean of 26.1 ug/kg downstream ($p=3.73E-05$). Upstream beryllium ranged between 5.3 and 6.5ug/kg whilst downstream beryllium ranged between 24.5 and 27.5 ug/kg (Table 1 and Figure 2). Cadmium recorded increases over 4 times downstream of the coal mine waste discharge. Upstream recorded a mean of 7.7 ug/kg in comparison downstream cadmium was recorded at 34.6 ug/kg downstream ($p=0.0001$). Upstream cadmium ranged from 6.4 to 9.7 ug/kg whilst downstream ranged between 26.0 and 41.9 ug/kg (Table 1 and Figure 2).

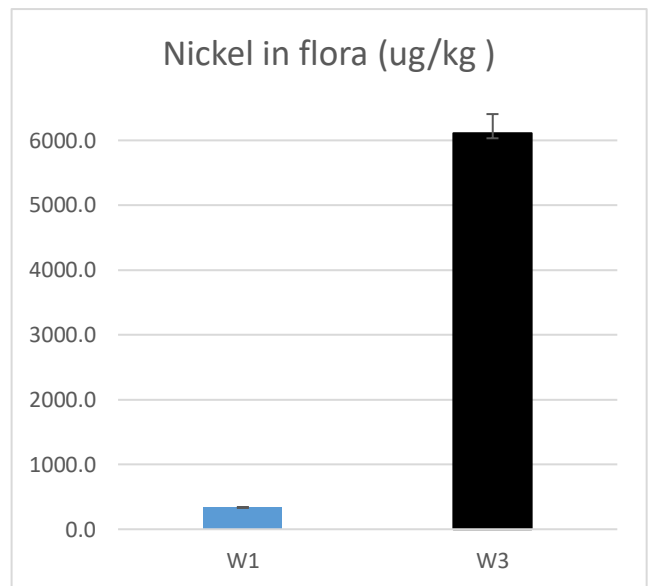
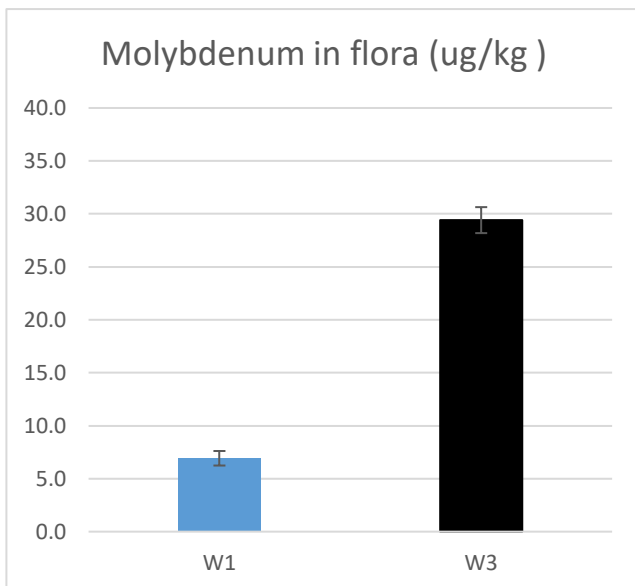
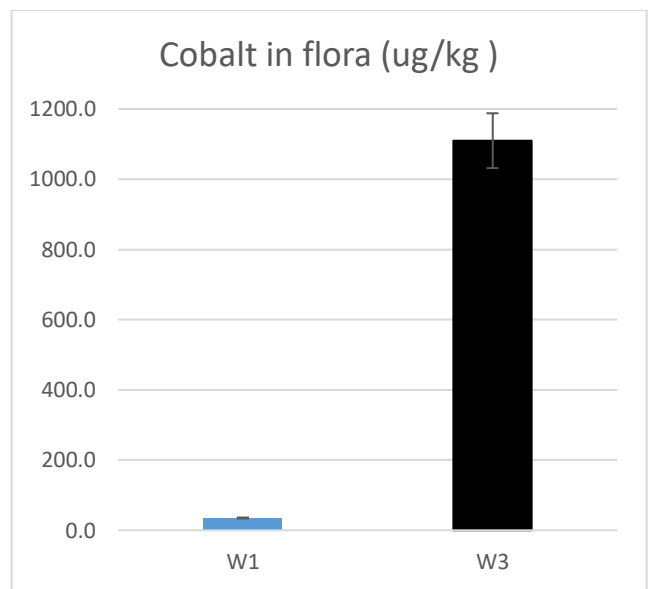
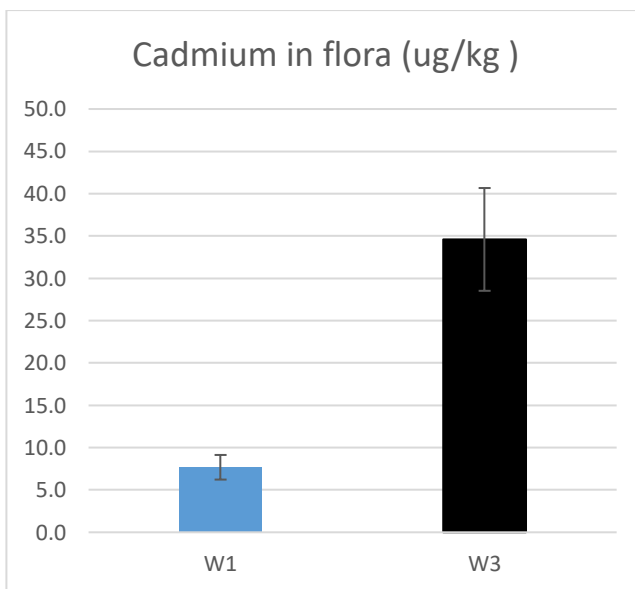
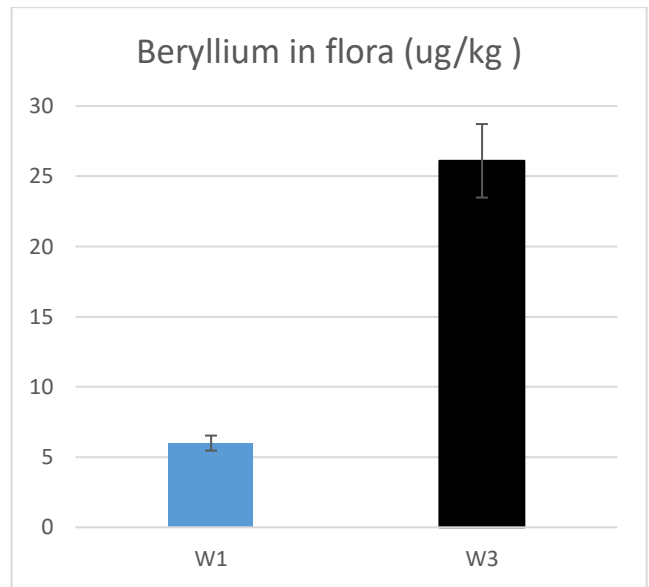
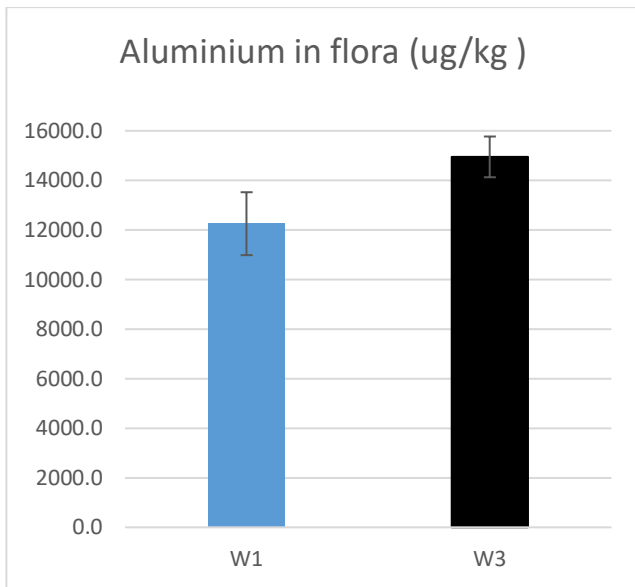
Molybdenum increased over 4 times from a mean of 6.9 ug/kg upstream of the mine wastewater inflow to a mean of 29.4 ug/kg downstream ($p=1.89E-08$). Upstream molybdenum ranged from 6.0 to 7.4ug/kg whilst downstream ranged between 27.8 and 31.2ug/kg (Table 1 and Figure 2). Strontium nearly doubled in concentrations downstream of the coal mine wastewater inflow from a mean of 13917 ug/kg upstream of the mine wastewater inflow to a mean of 21827ug/kg downstream ($p=0.001$). Upstream strontium ranged between 12422 and 15003 ug/kg whilst downstream ranged between 17719 and 24608 ug/kg (Table 1 and Figure 2). Thallium increased over 4 times from an upstream mean of 13.2 ug/kg to a mean of 60.3 ug/kg downstream ($p=0.0001$). Upstream thallium ranged from 10.7 to 16.9 ug/kg whilst in comparison downstream thallium ranged between 44.5 and 74.5ug/kg (Table 1 and Figure 2).

Zinc recorded increases over 7 times downstream of the coal mine waste discharge which is the third greatest increase of this study. Upstream recorded a mean of 6756 ug/kg whilst in comparison downstream zinc was recorded at a mean of 51914 ug/kg downstream ($p=1.64E-05$). Upstream zinc ranged from 6264 to 6152 ug/kg whilst downstream ranged between 46541 and 56932 ug/kg (Table 1 and Figure 2). Nickel increased over 18 times downstream of the coal mine wastewater discharge which is the second greatest increase of metal concentrations in this current study. Nickel increased from a mean of 336 ug/kg upstream of the mine wastewater inflow to a mean of 6110 ug/kg downstream ($p=8.18E-07$). Upstream nickel ranged from 333 to 338 ug/kg whilst downstream ranged between 5799 and 6498 ug/kg (Table 1 and Figure 2).

Cobalt increased over 30 times below the coal mine wastewater discharge which is the greatest increase found in this study. Cobalt increased from a mean of 35.1 ug/kg upstream of the mine wastewater inflow to a mean of 1110 ug/kg downstream ($p=3.33E-06$). Upstream cobalt ranged from 34.0 to 36.8 ug/kg whilst downstream cobalt ranged between 1021 and 1131 16102 ug/kg (Table 1 and Figure 2).

Table 1. Heavy metal parameters, sample locations, T-statistic, p value, range and mean for riparian vegetation. All metals are measured in ug/kg.

Metal/Site	t-stat	p value	Upstream		Downstream	
			Mean	Range	Mean	Range
Aluminium	3.83	0.009	12253	10646-13752	14949	13965-16102
Beryllium	16.7	3.73E-05	6	5.3-6.5	26.1	24.5-27.5
Cadmium	9.57	0.0001	7.7	6.4-9.7	34.6	26.0-41.9
Cobalt	30.7	3.33E-06	35.1	34.0-36.8	1110	1021-1131
Molybdenum	34.7	1.89E-08	6.9	6.0-7.4	29.4	27.8-31.2
Nickel	43.7	8.18E-07	336	333-338	6110	5799-6498
Strontium	5.63	0.001	13917	12422-15003	21827	17719-24608
Thallium	8.99	0.0001	13.2	10.7-16.9	60.3	44.5-74.5
Zinc	20.6	1.64E-05	6756	6264-6152	51914	46541-56932



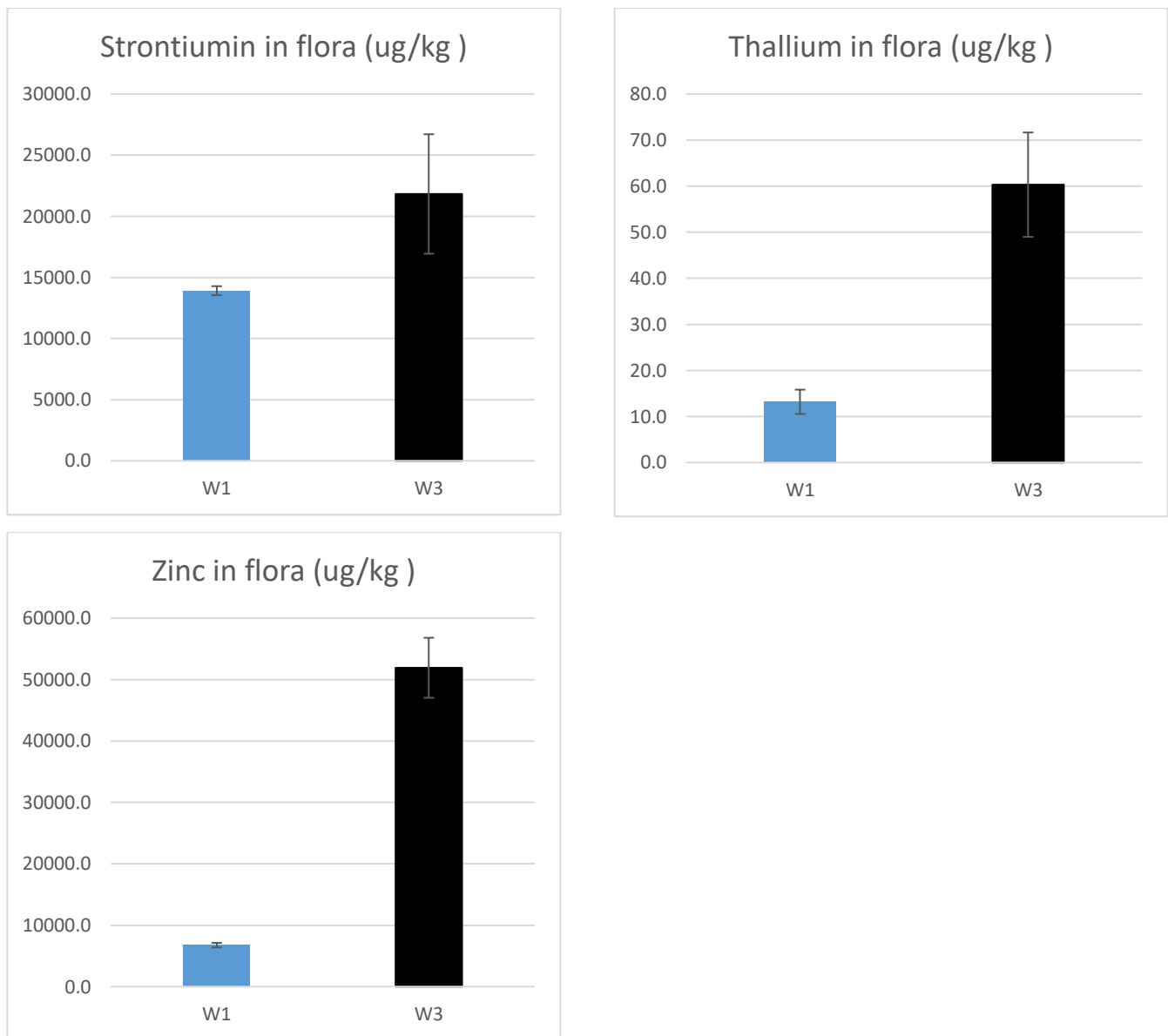


Figure 2. All heavy metal concentrations that were found to be statistically different. Blue is upstream (W1) of the coal mine waste water inflow and black is downstream (W3).

Discussion

This study has shown that one coal mine waste water discharge appears to have created an avenue for increased heavy metal concentrations within the Wollangambe Rivers water column to mobilise to riparian vegetation found within the terrestrial environment. This study could be the first to investigate the mobilisation of metals from a regulated coal mine waste water discharge to terrestrial growing riparian vegetation (*Acacia rubida*).

There are many studies investigating water column pollution from metal contamination from a broad range of mining activities. Many investigate the links between metal mining impacts on water chemistry, river sediments and aquatic flora and fauna. The majority of these studies have found that impacted sample locations, from an array of differing metal mining activities, have increased heavy metal contamination of waters and soils worldwide. These heavily contaminated waters and soils have been reported to have increased the metal concentrations within many differing

terrestrial and aquatic flora and fauna species (Neff 1984, Sericano et al. 1995, Hill et al. 2000; Amish and Cowx 2000, Wang and Rainbow 2008 and Ashraf et al. 2011). This current study supports these findings with increases found in nine of the ten metals analysed.

Studies have been conducted on the impacts of metal bioaccumulation from other coal mining activities on terrestrial and aquatic plant and fish tissue with very few investigating the bioaccumulation of metals from a regulated coal mine waste water discharges. The majority of the studies have provided evidence to suggest that increased bioaccumulation of metals is occurring in areas subject to current or past coal mining surface working activities (Papagiannis et al. 2004, Atkinson et al. 2007, Ebrahimi 2009, Bharti and Banerjee 2011, Otter et al. 2012 and Durães et al. 2014).

Maiti et al. (2015) found the metals Iron, Manganese, Zinc, Copper, Cadmium and Nickel were bioaccumulating in their study of trees growing in coal mine spoils. pH in the impacted soils was recorded between 4.34 and 4.95 and the metal concentrations (highest to lowest) were recorded as Iron, Manganese, Zinc, Copper, Cadmium and Nickel. This is similar to this current study although the order of increase was differing with Nickel and Zinc being two of the three highest recorded increases. Nawad et al. (2015) reported statistically higher concentrations of cobalt in soil and plant material in their study of heavy metal bioaccumulation from chromite mining activities in Pakistan. This is similar to this current study with cobalt found to be the greatest increased metal concentration recorded.

Telford et al. (2009) found that riparian vegetation growing along the banks of Bakers Creek, Hillgrove New South Wales Australia recorded antimony concentrations 2 to 3 times higher than riparian vegetation within non-mine impacted sites. The same study found significant differences between reference stream and impacted stream aquatic autotrophs. These findings suggest that in the right conditions water column pollutants, naturally occurring or not have the ability to be accumulated by terrestrial flora. Telford et al (2009)'s findings can be supported by this current study although the increases recorded are much higher for some of the metals analysed here.

This research has allowed for a better understanding of the broader impacts of the heavy metal contaminants from Clarence Collieries coal mine waste waters being discharged to the Wollangambe River are having on the surrounding terrestrial environment. The implications that the licensed waste water discharge is mobilising to terrestrial riparian vegetation is of major concern. The EPL process is designed to protect the aquatic environment of waterways which receive coal mine waste waters. Discharge limits set by EPL's stipulate levels of pollutants within the water column of a receiving waterway but do not take into account the surrounding terrestrial environment. Setting pollutant limits to EPL's may well be meaningless if the heavy metals are able to mobilise and bioaccumulate within terrestrial riparian vegetation.

This represents a discord in the EPL process which is unsatisfactorily protecting the greater environment from coal mine waste water discharges. The bioaccumulation of zinc, copper and lead in differing concentrations due to differing soil pH was reported by Durães et al. (2014). This study investigated two differing soil pH levels (acidic and alkaline) impacted by mining activities and found that metals bioaccumulated differently depending on soil pH. They

reported that in the lower pH soils (impacted by acid mine waters) Zinc, Copper and lead accumulated in that order of magnitude whilst in higher pH soils the order of magnification was Lead, Zinc and Copper (Durães et al. 2014).

In this study, the reference sites used may have a greater functional ability to sequester and or bioaccumulate heavy metals due to the naturally mildly acid pH of the upstream section of the Wollangambe River. In comparison, the impacted sites are subject to a treated unnatural water pH which is alkaline (Atkinson et al. 2007, Otter et al. 2012 and Durães et al. 2014). This is of major concern as impacted sites in this study have significantly higher concentrations of heavy metals whilst perhaps having a much lower ability to bioaccumulate these heavy metals.

This raises questions as to what will occur after mining activities are no longer undertaken at this colliery as when mining ceases so does the water treatment process. At present pH is increased during the treatment process and water currently being discharged may likely have a much lower rate of bioavailability for flora or fauna (terrestrial or aquatic) as described by Atkinson et al. (2007). Once treatment ceases the Wollangambe Rivers pH will naturally reduce to a background pH which is mildly acidic or as a worse case the rivers pH may decrease through the Acid Mine Drainage process.

This would open a new avenue for increased bioaccumulation of heavy metals from the legacy pollutants within the impacted streams sediments by allowing these legacy heavy metals to become increasingly bioavailable to either the terrestrial or aquatic biota through a pH shift. The results of this current study show that there is a strong likelihood that heavy metal concentrations linked to one coal mine waste water discharge are mobilising and being bioaccumulated by one species of riparian vegetation (*Acacia rubida*). The implications of this regulated water column pollution mobilising out of the aquatic environment shows a major floor in water column pollution licensing.

Recommendations

It is recommended that further research should be undertaken by the NSW EPA to better assess the implications of heavy metal mobilisation to the terrestrial environment from EPL protected waterways at both actively mined (treated waste water) and inactively mined (untreated waste water). If in fact heavy metal contaminants are leaving the water column of their receiving waterways and mobilising to the terrestrial environment, serious long-term legacy pollutant impacts may persist. Of equal concern is if in fact these water column heavy metals are becoming terrestrial pollutants within terrestrial vegetation the bioaccumulation may be wide spread as these terrestrial flora species are food sources for many more terrestrial fauna species.

Research significance

This chapter's research is the first to investigate the link between water column contamination from coal mine wastewaters mobilising and accumulating in plants growing in the terrestrial environment.

Current regulatory focus on coal-mine water quality does not assess the gradual build-up of river sediment contamination that may continue to contaminate long after coal mines cease active operation. This research addresses

this gap in the literature and correlates coal mine river and sediment contamination to the bioaccumulation of contaminants within riparian plants.

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Chapter 4

The regulation and impact of eight Australian coal mine waste-water discharges on downstream river water quality: a regional comparison of active versus closed mines.

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Abstract

Water quality of rivers that received coal mine wastes from active and closed mines were investigated, focussing on pollutants that are hazardous for aquatic ecosystems. Coal mine wastes increased nickel concentrations by an average of 25 times, from 1.5 µg/L upstream to 41.4 µg/L downstream. The average concentration of zinc increased below waste inflows from 8.6 µg/L (upstream) to 83.4 µg/L (downstream). The highest concentrations of zinc and nickel were detected in a stream that continued to receive drainage from a coal mine that closed 20 years earlier. All coal mine discharges increased river salinity. Salinity increased by more than six times (upstream mean 101.4 to 741.7 µS/cm downstream). This study provides a reminder that water pollution from coal mines is a major environmental issue for both active and closed mines. The study highlights the need for more stringent and consistent environmental regulation for all mines in the study.

Keywords

underground mining; closed mines; environmental regulation; bicarbonate; pollution; salinity; pH; zinc and nickel.

Introduction

Water pollution arising from coal mining activities is a major source of water pollution worldwide (Tiwary 2000 and Younger 2004). A broad range of water quality issues has been reported internationally from the release of coal mine wastes into waterways. Commonly reported water quality issues associated with coal mines include increased metal concentrations, elevated salinity and modified stream pH (Banks et al. 1997, García-Criado et al. 1999, Verb and Vis 2000, Brake et al. 2001, Younger 2001 and Pond et al. 2008). A frequently reported impact of coal mine waste discharges into rivers and streams is the degradation of aquatic ecosystems (Jarvis and Younger 1997, Pond et al. 2008, Belmer et al. 2014 and Wright et al. 2017).

A growing environmental water pollution legacy is the continued release of contaminated wastewater from coal mines after they have ceased commercial operation (Robb and Robinson 1995 and Banks et al. 1997). Pumping of accumulated groundwater from closed mines generally stops and, in many cases, this triggers a gradual flooding of the underground voids with groundwater (Younger 1993). The accumulating groundwater is often contaminated with pollutants associated with the oxidation of sulfur and increased mobilisation of minerals and metals (Robb 1994 and Banks et al. 1997). This process termed “rebounding” can occur rapidly as reported by Wright and Belmer (2018) or become a slow protracted process taking up to a decade to become surface water (Jackson (1981) reported in Younger (2001).

Many of the world’s coal mines have ceased mining with environmental problems such as water pollution often becoming a damaging ongoing legacy (Jarvis and Younger 1997, Johnson 2003). There is a rich literature on water pollution problems caused by the closure of coal mines in the United Kingdom (e.g Robb 1994, Robb and Robinson 1995, Younger 1993 and Younger 2001) and from the United States, including a regional comparison of water pollution caused by active and inactive coal mines (Brake et al. 2001, Pond et al. 2008 and Petty et al. 2010). The environmental problems associated with the closure of coal mines is likely to be a large and growing problem in Australia. This may be a surprise considering the increased production of coal from Australian mines in recent decades (Mudd 2009) with the export of coal being one of Australia’s highest value exports (Minerals Council of Australia 2015).

Investigations have reported water pollution from wastes discharged from active Australian coal mines (Belmer et al. 2014, Wright et al. 2015 and Wright et al. 2017). However, fewer studies have examined water pollution from Australian coal mines that have ceased operation whilst continuing to cause water pollution (Battaglia et al. 2005, Wright and Burgin 2009 and Wright and Belmer 2018). One study compared water pollution from an active coal mine (Westcliff Colliery) to that from a closed mine (Canyon Colliery) and showed that wastewater from both caused surface water pollution issues (Price and Wright 2016). Several studies investigated contaminated seepage from the closed Canyon Colliery and the resulting polluting of a high-conservation stream and river within a National Park reserve in the Blue Mountains area (Wright and Burgin, 2009, Wright et al. 2011 and Price and Wright 2016). Closed mines (coal and other mines) in Australia is an emerging environmental problem with estimates of 52,543 abandoned mines with few receiving rehabilitation (Unger et al. 2012). The Australian Government recognises the growing

problem associated with closed mines and is particularly concerned about how to rehabilitate the increasing number of closed and abandoned mines (Noetic, 2016).

Coal mine wastewater discharges in New South Wales (NSW) are regulated by the NSW Environmental Protection Authority (NSW EPA). The EPA issues an individual 'Environmental Protection License' (EPL) agreement to each colliery under the *Protection of the Environment Operations Act 1997* (POEO Act, EPA 2018). Each EPL sets discharge limits for water quality (physical and chemical) properties (usually concentrations) and volumes of the liquid colliery wastes which must be achieved to authorise their discharge to local waterways (Graham and Wright 2012). There is considerable variation in EPL conditions that apply to collieries across the Sydney basin (Table 1). The licences include a few standard pollutants, such as 'oil & grease', 'pH' and 'total suspended solids (TSS)' across the EPLs for most mines. The identification of a specific EPL pollutant concentration limit implies that pollutant is recognised by the EPA as being potentially problematic in that mine waste or in the waste-receiving waterway. However, the absence of identifying a pollutant in an EPL does not mean that it is permitted by the EPA to be discharged. Each EPL has a clause that makes this explicit (EPA 2018), they state:

*'To avoid any doubt, this condition does not authorise the pollution of waters by any pollutant other than those specified in the table*s'

There have been few water quality studies (none in Australia) comparing water quality impacts from a regional group of coal mines that discharge wastes from active and inactive mines. The key question posed for this investigation: does surface water quality change due to the discharge of coal mine wastewater from a regional group of active compared to inactive coal mines? The second question was: how well do the EPA discharge licences ('EPLs') match with the ecologically hazardous pollutants in each mine discharge, or in the waste-receiving river/stream downstream of the mine discharge point?

Materials and methods

This study investigated coal mine waste discharges and the impact of these wastes on water quality of waterways that received colliery wastes from seven underground coal mines in the Sydney Basin (Table 1: Figure 1). The mine wastewater discharges included effluent from actively operating coal mines as well as drainage emerging from closed underground mines. A large component of the mine discharges were from accumulated groundwater that had seeped into each of the underground mines. The geology of all mine locations shared many similarities as they all extracted coal from various seams within the Illawarra coal measures spanning the southern and western coalfields within the larger Sydney Basin (Branagan et al. 1979).

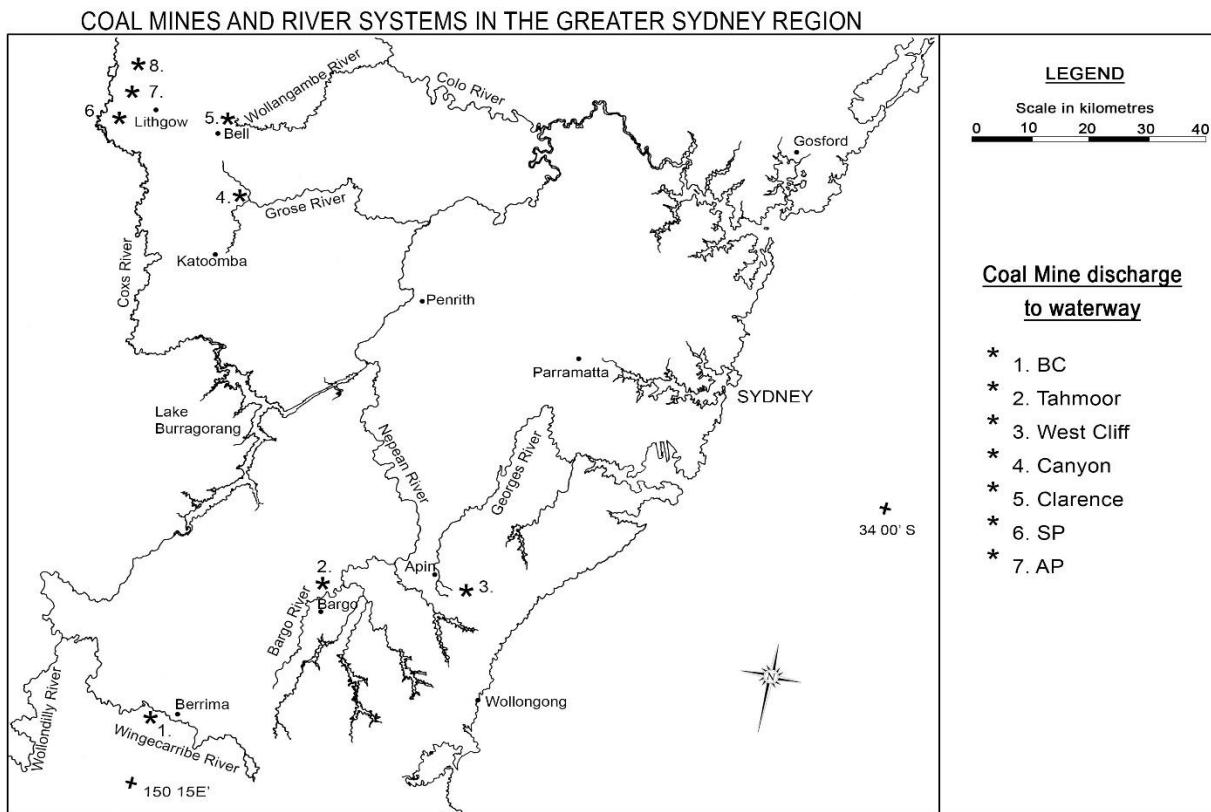


Figure 1. Map of lower Sydney basin, major waterways, settlements and location of the eight coal mine waste discharges (marked by asterisks). Seven mines are numbered (1 ‘BC’ Berrima Colliery, 2. Tahmoor Colliery, 3. Westcliff Colliery, 4. Canyon Colliery, 5. Clarence Colliery, 6. ‘Sp’ Springvale Colliery, 7. ‘AP’ Angus Place Colliery). Springvale and Angus Place Colliery also discharge waste to Sawyers Swamp Creek.

The collieries and waterways in this study were in two broad groups (Figure 1). One group (numbered 4 to 7 in Figure 1) were in the western coalfields, north-west of the Sydney Metropolitan area in the Lithgow / Bell area of the Blue Mountains. These mines are located about 100 to 120 km from the coast in mountainous landscapes at elevations ranging from 790 metres (ASL), Canyon Colliery, to 988 metres (Clarence Colliery) (Table 1, Figure 1). The other two mines in this group are the Angus Place and Springvale Collieries. The other three mines are in the southern coalfields of the Sydney basin, south-west of Sydney (numbered 1 to 3 in Figure 1). They are the Berrima (also called Medway) Colliery which is 57 km from the coast. This closed mine discharges drainage into the Wingecarribee River (530 m ASL). The other two mines are the Tahmoor Colliery (247 m ASL) in the Bargo area 33 km from the coast, and Westcliff Colliery (227 m ASL) near Appin, 16 km from the coast (Figure 1).

Table 1. Study sampling site details. The waterway or colliery name, the description, location (latitude and longitude coordinates) and elevation of sampling sites (above sea level, m ASL). The description includes the distance (in metres) upstream (US) or downstream (DS) of the mine waste discharge point. The month and year of the first and last sampling are provided, as well as the number of sampling visits.

Name of waterway / Colliery	Description relative to waste discharge	Location coordinates	Elevation (M ASL)	Dates of sampling (month/year)	Number of sampling visits
Sampling sites used as reference site upstream (US) of coal mine discharges					
Wingecarribee River (US Berrima)	120 m US drainage adit	34° 29' 15.97" S, 150° 15' 39.61" E	535	8/2016 to 9/2017	8
Dalpura Creek (US Canyon)	75m US drainage adit	33° 32' 32.69" S, 150° 18' 25.97" E	820	12/2016 to 5/2017	3
Coxs River (reference for Angus Place)	4850 m US Kangaroo Ck / Coxs River confluence	33° 18' 19.07" S, 150° 5' 49.83" E	965	9/2015 to 5/2017	6
Georges River (US Westcliff)	150 m US Brennans Creek (waste) discharge	34° 12' 21.24" S, 150° 47' 57.45" E	229	12/2016 to 3/2017	4
Bargo River (US Tahmoor)	1480m US Teatree Hollow (waste) discharge	34° 14' 11.78" S, 150° 34' 46.75" E	255	12/2016 to 3/2017	4
Wollangambe River (US Clarence)	200m US waste inflow	33° 27' 22.35" S, 150° 14' 57.27" E	992	12/2016 to 5/2017	5
Springvale Creek (US Springvale)	1050 m US waste discharge	33° 24' 37.1" S, 150° 6' 38.24" E	905	9/2015 to 5/2017	6
Sawyers Swamp (US Springvale and Angus Place)	3280m US waste discharges	33° 18' 19.07" S, 150° 5' 49.83" E	995	9/2015 to 5/2017	6
Sampling sites downstream of coal mine discharges					
Wingecarribee River (DS Berrima)	140 m DS drainage adit	34° 29' 16.16" S, 150° 15' 40.23" E	530	8/2016 to 9/2017	8
Dalpura Creek (DS Canyon)	50 m DS drainage adit	33° 32' 35.04" S, 150° 18' 25.38" E	770	12/2016 to 5/2017	3
Kangaroo Creek (DS Angus Place)	75 m DS Angus Place Colliery	33° 20' 58.45" S, 150° 5' 56.1" E	905	9/2015 to 5/2017	6
Georges River (DS Westcliff)	90 m DS Brennans Creek (waste) inflow	34° 12' 14.72" S, 150° 47' 53.08" E	227	12/2016 to 3/2017	4
Bargo River (DS Tahmoor)	80 m DS Teatree Hollow (waste) inflow	34° 14' 37.06" S, 150° 35' 20.66" E	247	12/2016 to 3/2017	4
Wollangambe River (DS Clarence)	1250 m DS waste inflow	33° 27' 21.38" S, 150° 15' 26.09" E	979	12/2016 to 5/2017	5
Springvale Creek (DS Springvale)	810 m DS waste discharge	33° 24' 6.8" S, 150° 5' 40.57" E	880	9/2015 to 5/2017	6
Sawyers Swamp Creek (DS Springvale and Angus Place)	900 m DS waste discharge	33° 22' 50.61" S, 150° 5' 10.95" E	890	9/2015 to 5/2017	6

Water samples were collected from rivers or streams upstream of the point where the mine wastes entered. The upstream sampling sites are reference sites and were compared with the water quality results with those collected downstream of the mine waste discharge. The distance upstream ranged from 75 m (Dalpura Creek) to 3.28 km (Sawyers Swamp Creek). For one waterway and mine (Kangaroo Creek / Angus Place Colliery) we were unable to access a sampling site upstream of the mine. In that case we used a nearby (4.3 km upstream) undisturbed waterway (Coxs River) of similar physical characteristics (Table 1). The distance of sampling sites downstream of the mine waste discharge ranged from 30 m (Dalpura Creek) to 1.25 km (Wollangambe River). At all sites the distance downstream was considered adequate to represent complete mixing of the mine drainage with the stream or river that was recipient of the wastes. At Dalpura Creek this mixing occurred in a precipitous and cascading montane stream over a relatively short distance (30 m). This study allocated equal sampling effort to sampling the waterways receiving

mine wastes upstream of (or at a reference waterway) the mine waste outfall to sampling downstream of the mine. In all cases the sampling of a waterway upstream and downstream of the mine inflow was completed on the same day, under the same dry weather conditions.

Two adjoining mines (Angus Place and Springvale) each had two mine discharges included in this study. The waterway 'Sawyers Swamp Creek' received waste discharges from the two mines. Discussions with the NSW EPA indicated that Springvale Colliery provided the greatest volume of wastewater into Sawyers Swamp Creek.

Four of the coal mines in this study were actively mining coal and three were not. For this study we termed 'closed' coal mine as a mine that was not engaged in coal mining during this study. The longest inactive mine closed in 1997 (Canyon Colliery). One closed mine (Angus Place) had not extracted coal for more than two years. It was in a state of 'care and maintenance' where the mine remained ventilated and groundwater in the mine was pumped out, with future mining activity still possible. The third closed mine was Berrima Colliery, that permanently ceased mining two years before this study, after which 15% of the underground workings were flooded (Wright and Belmer 2018).

This study is based on water quality data collected from between three and eight times, from September 2015 to May 2017 (Table 1). It includes data from a previous published study on the Berrima Colliery (Wright and Belmer 2018). On each occasion at each site duplicate water samples were collected, with a minimum of six individual samples collected from each sampling site. Three of the mines were sampled less intensively than other mines (Canyon mine, Westcliff mine and Tahmoor mine) but this was considered acceptable as they had previously been investigated and the results collected in this study were compared and found to be consistent with previous published research (Price and Wright 2016, Wright et al. 2015 and Wright and Ryan 2016).

The coal mines in this study are regulated by the NSW Environment Protection Authority (EPA). The EPA regulate the disposal of mine wastes from each mine using an individual 'environment protection licence' (EPL) for each mine (Wright et al. 2011). The permitted level of pollutants in the liquid wastes from each mine is specified in each EPL, generally as concentrations ($\mu\text{g/L}$ or mg/L) for water quality (physical and chemical) attributes including nitrogen, oil & grease, total suspended sediment (TSS), turbidity (NTU), pH (pH units), salinity (as electrical conductivity in $\mu\text{S/cm}$), chloride, sulfate and 16 metals (EPA 2018). Each EPL also specifies what proportion of time (expressed as percentiles) that the pollutant levels cannot be exceeded. Table 2 summarises the individual EPL discharge limits (often as concentrations) for permitted pollutant levels in each colliery wastewater that the EPA authorises as appropriate to be released into local streams or rivers. This is generally 100% of the time, except for EPL 2504 (Westcliff Colliery) where many of the pollutants must not be exceeded 90% of the time (Table 2).

Table 2. The NSW EPA Environment Protection Licence (EPL) pollutant limits for discharge of colliery wastes to streams/river (EPA, 2018). Each colliery has an individual EPL licence and with a unique licence number. The letter (d) after the EPL pollutant limit refer to the samples requiring filtering, and the pollutant concentration represents the dissolved (d) fraction. The EPL concentration limits apply for different proportions of times, expressed as percentiles: (*) = 100 percentile of the time, (**) 90 percentile of the time. Two waterways receiving waste from Springvale Colliery were investigated, the ‘main’ discharge and ‘minor’ discharge. # Note EPL 558 has been surrendered.

Pollutant attribute (units of measurement)	Angus Place Colliery EPL 467 (*)	Berrima (Medway) Colliery EPL 608 (*)	Canyon Colliery EPL 558 (*) #	Clarence Colliery EPL 726 (*)	Springvale Colliery EPL 3607 Main (*)	Springvale Colliery EPL 3607 Minor (*)	Tahmoor Colliery EPL 1389 (*)	Westcliff Colliery EPL 2504 (**)
pH (pH units)	6.5-9.0	6.5-8.5	-	6.0-8.5	6.5-9.0	6.5-9.0	6.5-9.0	6.5-9.3 (*)
Oil and Grease (mg/L)	10	10	10	10	10	10	10	10 (*)
EC (µS/cm)	-	-	-	-	1200	-	2600	-
TSS (mg/L)	30	50	-	30	50	30	30	50 (*)
Turbidity (NTU)	-	-	-	-	50	-	150	-
Nitrogen (µg/L)	-	-	-	250	-	-	-	-
Aluminium (mg/L)	-	-	-	-	450 (d)	-	-	800 (d)
Arsenic (µg/L)	-	-	-	23 (d)	24 (d)	-	200	19 (d)
Boron (µg/L)	-	-	-	100	370	-	-	-
Cadmium (µg/L)	-	-	-	0.2 (d)	-	-	-	0.5 (d)
Chloride (mg/L)	-	-	-	25	-	-	-	-
Chromium (µg/L)	-	-	-	1 (d)	-	-	-	-
Cobalt (mg/L)	-	-	-	2.5 (d)	-	-	-	20 (d)
Copper (mg/L)	-	-	-	1.4 (d)	7 (d)	-	-	18 (d)
Iron (µg/L)	-	-	1000	300 (d)	400 (d)	-	-	-
Manganese (mg/L)	-	-	-	500 (d)	1700 (d)	-	-	40 (d)
Fluoride (µg/L)	-	-	-	1000	1800	-	-	-
Lead (µg/L)	-	-	-	3.4 (d)	-	-	-	6 (d)
Mercury (µg/L)	-	-	-	0.06 (d)	-	-	-	-
Nickel (µg/L)	-	-	-	11 (d)	47	-	200	200 (d)
Selenium (µg/L)	-	-	-	5	-	-	-	-
Silver (µg/L)	-	-	-	0.05	-	-	-	-
Sulfate (mg/L)	-	-	-	250	-	-	-	-
Zinc (µg/L)	-	-	5000	8	50 (d)	-	300	84 (d)

Water samples were collected on at least three occasions, over a 2-year period, from each sampling site between September 2015 to September 2017 (Table 1, Figure 1). Field meters were tested and calibrated, if required, on each sampling occasion to measure stream and mine discharge pH, salinity (measured as electrical conductivity ‘EC’), and water temperature from all study sites. Five replicate readings were collected from each site on each sampling

occasion. The meters used were a TPS AQUA-Cond-pH meter and TPS WP-81 Conductivity, pH and Temperature meter with TPS Conductivity and Temperature probe and a TPS submersible k407 pH sensor.

Duplicate grab samples were collected using unused bottles and were analysed using standard methods (e.g. USEPA 1998) by Envirolab (Chatswood, NSW) a National Associations of Testing Authorities (NATA) accredited laboratory for 8 metals (aluminium, copper, iron, lead, manganese, nickel, uranium and zinc) and for major anions (chloride, carbonate, sulfate and bicarbonate).

Students *t*-test was used to test for differences in water quality at reference sampling sites (upstream of mine waste discharge) compared to sampling sites downstream of mine waste inflows.

Results and discussion

The physio-chemical properties of all waterways changed substantially due to the influence of coal mine wastewater inflows from seven mines (Tables 3, 4, Figures 3-5). Despite only four of the mines being actively mined during the study, all active and inactive mines continued to release coal mine drainage, or other mine wastes, that caused substantial modification of water quality downstream of their point of entry into one or more local waterways. Of the 15 water quality attributes examined in this study, 12

of them were significantly higher downstream, compared to upstream of the mine discharges (Tables 3, 4).

The coal mine discharges increased the concentration of most metals, with zinc and nickel being of most concern as they were regularly found at hazardous levels for river and stream ecosystems. Nickel was frequently detected in waterways below mines at ecologically dangerous levels that were an average of 25 times higher than upstream (upstream mean 1.55 µg/L, downstream mean 41.4µg/L, Table 4). Similarly, zinc concentrations also increased, rising by nearly 9 times (upstream mean 8.56 µg/L, downstream mean 83.4 µg/L). The downstream nickel and zinc concentrations were generally higher than the Australian guidelines for protection of 99% of freshwater species (nickel: 8 µg/L, zinc: 2.4 µg/L, ANZECC, 2000). The highest nickel and zinc concentrations in waterways receiving mine drainage in this study were detected in Dalpura Creek below the inflow of continuous mine drainage from the drainage adit from the closed Canyon Colliery (mean nickel: 186.3 µg/L, mean zinc: 315.6 µg/L). This colliery ceased mining in 1997 and it continues to cause long-term pollution of high conservation-value waterways within the Blue Mountains World Heritage Area (Wright et al. 2011, Price and Wright 2016).

International studies show that waterways receiving coal mine wastewater often have highly elevated concentrations of nickel and zinc (Brake et al. 2001, Pond et al. 2008 and Petty et al. 2010). Similar to the current study, closed coal mines commonly produce mine drainage that is contaminated by elevated zinc and nickel for decades after mining ceases. For example, elevated levels of nickel and zinc were detected below the abandoned Green Valley coal mine (Indiana USA) more than 30 years after its closure, with nickel levels above 500 µg/L, and as high as 3780 µg/L and zinc often above 5000 µg/L (Brake et al. 2001).

Table 3. Summary statistics (mean and range) for water chemical attributes and major anions (mg/L) collected from waterways upstream (US) and downstream (DS) active and closed coal mines. Summary statistics (mean and range) are given for all upstream sites and for all downstream sites. Downstream sites are listed according to whether mine is ‘active’ or is ‘closed’. The upstream vs. downstream % increase is given. The t-static, degrees of freedom (df), p values are given for upstream versus downstream. (ns=not significant, * p<0.05, ** p<0.001, *** <0.0001) are given for comparison of means US vs. DS. BD = below detection.

US all mines	pH (pH units)	EC (µS/cm)	Water temp (°C)	Chloride	Sulphate	Carbonate	Bicarbonate
Clarence	5.33	19	17.5	41	0.8	BD	BD
	5.18-5.52	17.3-21.7	15.1-19.1	43589	BD-1	BD	BD
Canyon	4.72	26.1	12.7	48	1.8	BD	BD
	4.11-5.21	24.0-27.1	11.5-14.5	43589	43497	BD	BD
Sawyers	4.69	35.7	10.6	52	3.4	BD	4.2
	4.47-4.95	29.2-45.3	6.8-14.1	43620	43618	BD	BD-9
Springvale	5.3	53.6	11.6	8	5.2	BD	12.4
	4.12-6.48	38.3-84.8	9.3-19.0	43804	43712	BD	BD-21
Angus Place	5.13	42.8	12.5	43	0.5	BD	11.6
	4.31-6.07	19.5-88.3	8.6-19.8	43619	BD-1	BD	BD-24
Westcliff	6.3	148.2	20.3	28.8	5.8	BD	18
	6.22-6.39	100-254	20-20.5	17-52	43682	BD	13028
Tahmoor	6.8	206.1	23.2	46	4.4	BD	12.6
	6.39-7.21	159-241	20.5-26.8	25-60	43649	BD	42309
Berrima	7.48	258.4	15.5	44	13	BD	60.8
	7.30-7.68	87.7-357	10.3-21.3	40-47	46569	BD	47-69
All upstream sites							
mean	5.74	101.4	14.02	15.2	4.12	BD	15
range	4.11-7.69	10-357	6.8-26.8	21976	BD-27	-	Bd-69
DS active							
Clarence	7.35	289.5	19.6	38	104	BD	25
	7.24-7.46	286-293	18.3-21.0	43588	88-110	BD	21-31
Sawyers	8.52	1162.4	20.8	59	269	51.2	5977
	8.23-8.81	1113-1226	18.1-24.8	43651	15-35	27-68	540-660
Springvale	7.99	840.2	12.9	13.4	852	8.7	374
	7.53-8.17	471-985	7.8-19.1	44805	63-150	BD-23	110-540
Westcliff	8.92	1256	22	99.8	182	95.9	526
	8.50-9.17	368-2093	21.4-22.7	34-150	46327	BD-160	170-830
Tahmoor	8.47	1011.3	21.8	57.2	9	52	4883
	8.12-8.68	261-1498	20.2-23.0	28-72	43746	BD-87	160-710
DS closed							
Canyon	5.95	129	15.9	38	26	BD	22
	5.58-6.22	113.6-149.8	15.8-16.0	43558	26-26	BD	20-23
Angus Place	8.07	730.2	13.3	6	152	24.1	440
	7.61-8.51	344-994	9.5-20.0	43681	12298	BD-51	150-580
Berrima	7.25	397.6	15.1	45.4	74.7	BD	59
	6.98-7.49	258.4-539	10.4-20.4	41-50	19-110	BD	47-64
All downstream sites							
mean	7.85	741.7	16.5	22.7	469	28.4	342
range	5.58-9.17	113.6-2093	7.8-24.8	3-150	8-150	BD-160	20-830
Mean % increase							
(US vs. DS)	36.8	631.5	17.7	50	1037.6	-	2176.5
t-statistic, df	22.6, 361	20.4, 225	5.3, 408	1.7, 107	9.23, 71	-	10.4, 69
p. value	(***)	(***)	(***)	(ns)	(***)	-	(***)

Table 4. Summary statistics (mean and range) for eight metals (µg/L) collected from waterways upstream (US) and downstream (DS) active and closed coal mines. Summary statistics (mean and range) are given for all upstream sites and for all downstream sites. Downstream sites are listed according to whether mine is ‘active’ or is ‘closed’. The upstream vs. downstream % increase is given. The t-static, degrees of freedom (df), p values are given for upstream versus downstream. (ns=not significant, * p<0.05, ** p<0.001, *** <0.0001) are given for comparison of means US vs. DS. BD = below detection.

US all mines	Aluminium	Copper	Iron	Lead	Manganese	Nickel	Uranium	Zinc
Clarence	93.8	7 6	497.8	BD	25.3	BD	BD	3.1
	70-130	BD-3	290-770	BD	21-29	BD	BD	43618
Canyon	102.9	3 6	20.4	BD	15.3	BD	BD	2.6
	90-110	BD-22	13-27	BD	13-17	BD	BD	43526
Sawyers	277.9	0 5	1613.4	BD	46.6	2.6	BD	20.4
	90-720	BD-1	17-5800	BD	32-62	43557	BD	12359
Springvale	341.8	1 7	1713.1	0.7	124.1	1.7	BD	11.5
	50-740	BD-3	BD-7400	BD-2	13-370	43556	BD	45352
Angus Place	270	0 7	16214.1	BD	434.6	1.3	BD	7.2
	40-840	BD-2	410-45000	BD	20-1500	BD-3	BD	43891
Westcliff	214.3	2 3	497.1	BD	44.6	1.7	BD	7.6
	70-280	BD-9	360-790	BD	20-110	BD-6	BD	43808
Tahmoor	231.7	1	1003.3	BD	119.5	1.5	BD	4
	10-500	BD-2	840-1200	BD	71-180	BD-2	BD	43588
Berrima	184.4	1 1	461.1	BD	31	0.8	BD	2.4
	30-490	BD-2	210-830	BD	19-50	BD-2	BD	43556
All upstream sites								
Mean	231.1	2.05	4239.7	0.53	141.1	1.55	BD	8.56
Range	10-840	BD-22	BD-45000	BD-2	13-1500	BD-6	BD	12055
DS active								
Clarence	22	1 5	61 2	BD	103.3	31.3	BD	36.5
	11232	BD-10	42-86	BD	80-150	23-41	BD	19-56
Sawyers	484.7	0 8	558.3	0.8	69.8	4.8	0.8	12.4
	60-1210	BD-2	47-1500	BD-2	10-220	43679	0.6-0.9	11018
Springvale	515	2 9	2210	2.5	2308.8	10.3	0.7	46.4
	30-2800	BD-15	150-13000	BD-10	35-14000	15373	0.5-0.9	9-240
Westcliff	1420	3 2	456.4	1.3	12	64.8	3.8	13.8
	110-3400	43558	84-1000	43497	43983	14-120	1.7-5.7	43374
Tahmoor	391.7	1 6	611.7	0.9	105.5	28.8	3.6	23.3
	110-740	43497	170-1300	BD-2	22-380	17319	1.8-5.8	16377
DS closed								
Canyon	13.3	9 1	1074.4	BD	457.8	186.3	BD	315.6
	11079	BD-52	670-1900	BD	420-510	180-200	BD	290-340
Angus Place	502.4	0 7	1306.5	0.9	178.9	3.8	1.6	38.2
	30-2000	BD-2	140-5800	BD-3	16-990	43739	1.1-1.8	4-190
Berrima	128.9	0 8	1593.3	BD	1988.9	72.4	BD	228.3
	20-470	BD-2	950-2900	BD	340-3300	14-110	BD	76-290
All downstream sites								
Mean	396.9	2.1	1059.3	1.02	676.4	41.4	1.18	83.4
Range	547986	BD-52	42-13000	BD-10	6-14000	1-200	BD-5.80	3-340
Mean % increase								
(US vs. DS)	71.8	2 4	-75	93.5	379.6	2576	-	874.5
t-statistic, df	2.1, 95	0.5, 141	2.93, 83	2.84, 71	2.53, 78	6.12, 82	-	6.01, 82
p. value	(*)	(NS)	(*)	(*)	(*)	(***)	-	(***)

Although zinc and nickel were detected in waterways downstream of mine discharges at ecologically hazardous concentrations (ANZECC 2000), the NSW EPA regulation of these metals in the colliery discharges show a wide variation (Table 2). Two collieries (Angus Place and Berrima) currently have no limit on the permitted concentration of either nickel or zinc that can be discharged from each mine. Four of the eight EPA licences do specify a discharge limit for nickel concentrations in wastes. The specified concentration varies, ranging from a low of 11 µg/L (Clarence), then 47 µg/L (Springvale discharge to Sawyers Swamp) to the highest permitted concentration of 200 µg/L at both Tahmoor and Westcliff Collieries (Table 2, EPA 2018).

The highest concentrations of nickel, in this study, was detected in Dalpura Creek (ranging from 180 to 200 µg/L) below the closed Canyon mine, which did not have any EPA concentration limit for nickel. Zinc concentration limits were specified in six of the eight EPA licences with a range of discharge limits for zinc concentrations. Again, there were major differences in the permitted zinc concentrations ranging from a high of 5000 µg/L (Canyon), then 300 µg/L (Tahmoor), 84 µg/L (Westcliff), 50 µg/L (Springvale) and 8 µg/L (Clarence) (Table 2, EPA 2018).

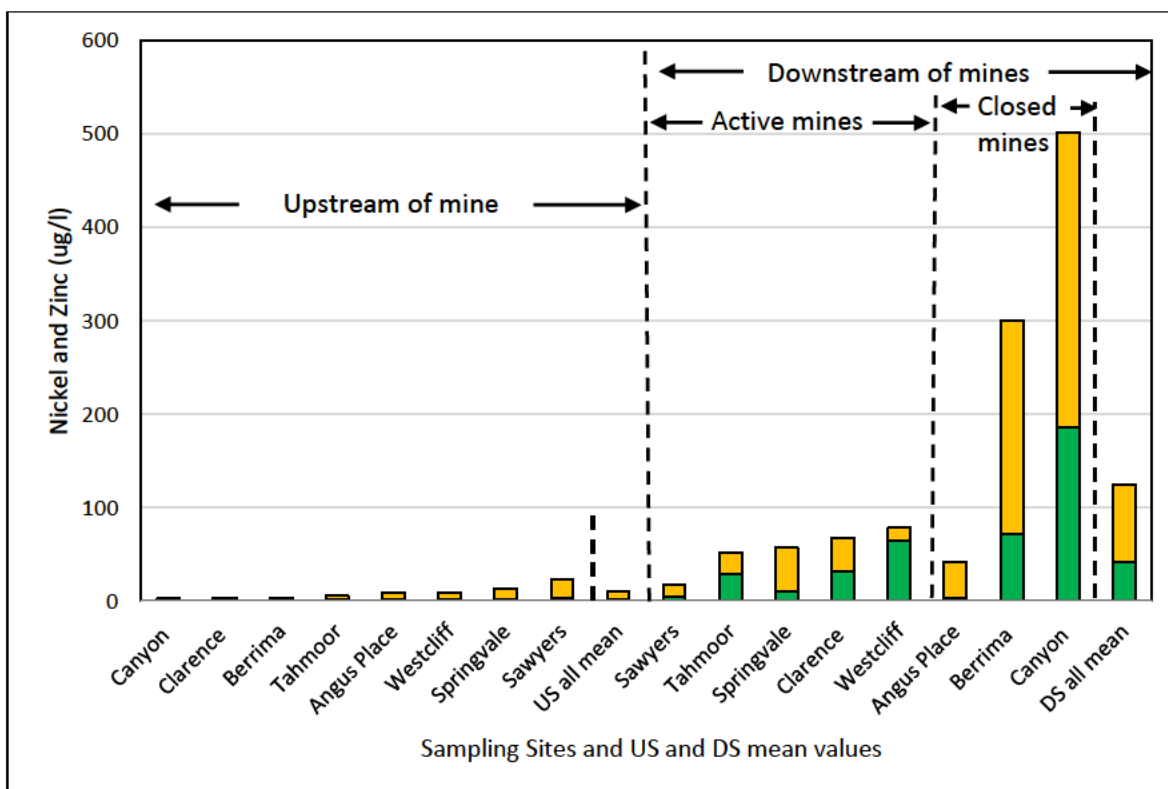


Figure 2. Mean concentration of nickel (bottom, green) and zinc (top, orange) (µg/L) results for sampling sites over this study. Sites upstream of mines are at the left. The overall mean concentrations are represented by ‘US all mean’. Results for sampling sites downstream of waste discharges are on the right, grouped ‘active mines’ and ‘closed mines’. The overall mean concentrations downstream are represented by ‘DS all mean’.

The salinity of waterways upstream of mines ranged from 10-357 µS/cm (mean 101.4 µS/cm, Table 3, Figure 2). The salinity of waterways downstream ranged from 113.6 to 2093 µS/cm (mean 741.7 µS/cm), an increase of 631.5 % (Table 3). The elevated salinity below several mines was at ecologically hazardous levels. The mean salinity level (741.7 µS/cm) downstream of the mine discharges was more than twice the level recommended Australian water quality ecosystem guidelines for upland streams in south eastern Australia (<350 µS/cm, ANZECC 2000). The

increase in salinity appeared modest (mean 129 $\mu\text{S}/\text{cm}$) downstream of one closed mine (Canyon Colliery, Dalpura Creek), yet that was about four times higher than Dalpura Creek upstream of the mine (mean 26.1 $\mu\text{S}/\text{cm}$). At the other end of the scale, the highest salinity (mean 1256 $\mu\text{S}/\text{cm}$) was detected below the Westcliff mine. It was nearly 7.5 times higher than upstream (mean 148.2 $\mu\text{S}/\text{cm}$). It is perhaps surprising that only two of the eight waste discharge licences (EPLs) stipulated any salinity discharge limits. One was 1200 $\mu\text{S}/\text{cm}$ from Springvale Colliery and the other was 2600 $\mu\text{S}/\text{cm}$ from Tahmoor Colliery (Table 2: EPA 2018). In both cases the salinity level of the streams / river downstream exceeded the recommended salinity guideline.

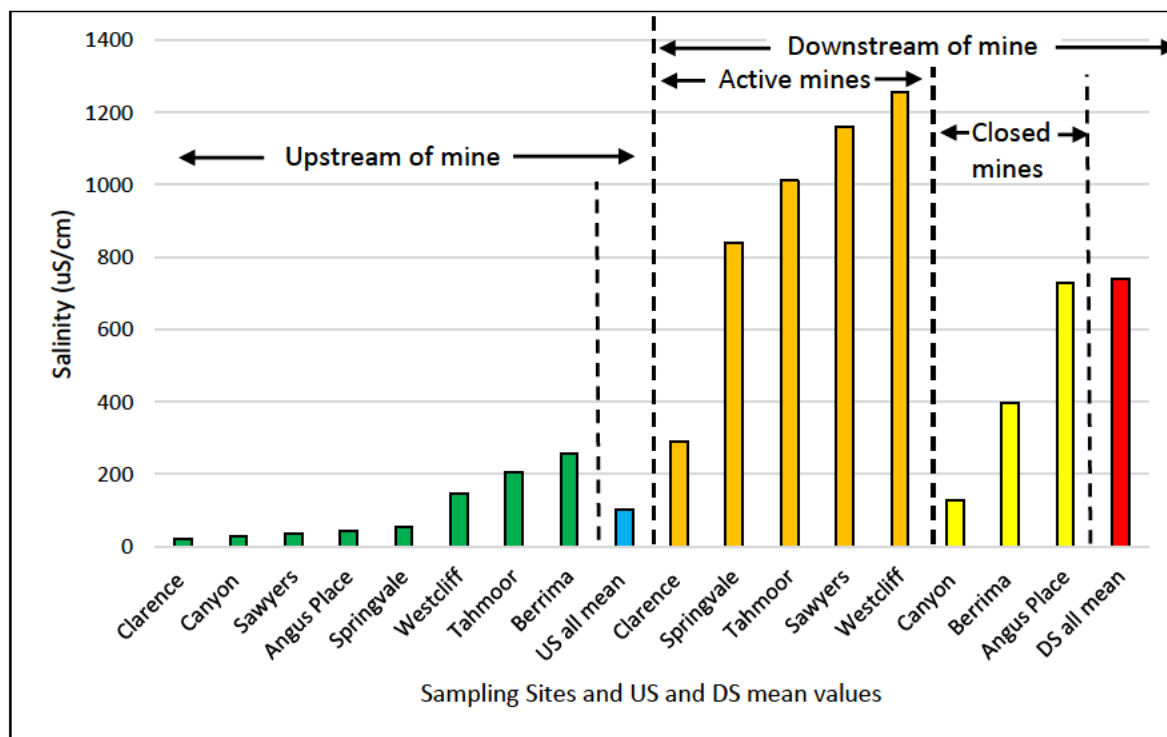


Figure 3. Mean salinity ($\mu\text{S}/\text{cm}$) results for sampling sites over this study. Sites upstream are at left (green bars) of mines. The overall mean upstream salinity value is represented by the blue bar. Results for sampling sites downstream of waste discharges are on the right. Orange bars are sites below active mine and yellow bars are sites below closed mines. The mean downstream salinity value is represented by the red bar. US = upstream and DS = downstream.

In comparison to international studies, the increase in salinity of Sydney basin waterways due to disposal of mine wastewater in this study was comparatively moderate. For example, slightly lesser increases in river salinity, due to coal mine discharges, were detected in the Boeza and Tremor Rivers in north west Spain where García-Criado et al. (1999) reported very low salinity above coal mines (18-58 $\mu\text{S}/\text{cm}$) compared to downstream (144-449 $\mu\text{S}/\text{cm}$). Many waterways in this study had increased salinity comparable to that reported below West Virginian coal mines (Pond et al. 2008) where salinity increased from 62 $\mu\text{S}/\text{cm}$ in unmined streams to 1023 $\mu\text{S}/\text{cm}$ downstream of mine discharges. Similar increases in salinity were also reported for the waterways affected by coal mine activity in the Freeport coal seam of the Appalachians (USA) where Petty et al. (2010) reported salinity increased from unmined areas (mean 98 $\mu\text{S}/\text{cm}$) increased at streams exposed to high intensity mining (mean 734 $\mu\text{S}/\text{cm}$).

The pH of rivers and streams, below mine drainage inflows, was significantly lower than upstream (Table 3, Figure 3). In seven of eight cases the pH of rivers/streams increased due to the influence of the mine drainage (Table 3). On

average, the pH below the mine discharge increased by more than two pH units (mean 5.74 upstream versus 7.85 downstream) below the entry of mine discharges (Table 5). The largest increase in pH was detected in Sawyers Swamp Creek (mean of 4.69 upstream and 8.52 downstream, Table 3). The Wingecarribee River was the only waterway that had lower pH below the mine (mean 7.25), compared to a mean of 7.25 upstream.

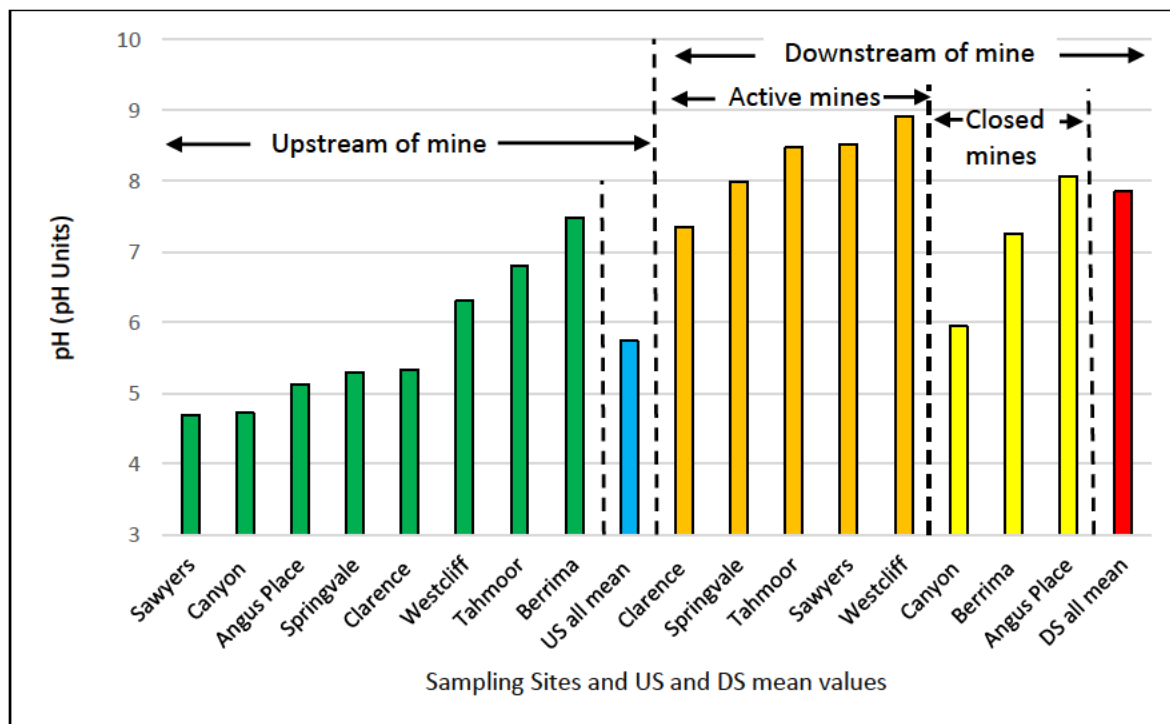


Figure 4. Mean pH (pH units) results for sampling sites over this study. Sites upstream are at left (green bars) of mines. The overall mean upstream pH value is represented by the blue bar. Results for sampling sites downstream of waste discharges are on the right. Orange bars are sites below active mine and yellow bars are sites below closed mines. The mean downstream pH value is represented by the red bar. US = upstream and DS = downstream.

One of the well-known triggers of water pollution from coal mines is the generation of acidic wastewater due to the influence of acid mine drainage (AMD). This is caused by the oxidation of sulfur compounds in the coal, disturbed by the mining process (Robb 1994, Bank et al. 1997 and Tiwary 2000). In contrast to many other studies, our study revealed that the discharge of coal mine wastes alkalized rather than acidified downstream streams or rivers. The acidic pH levels recorded at upstream sites are typical of the naturally acidic and poorly buffered waterways in the Sydney basin in minimally disturbed catchments (Hayes and Buckney 1995, Wright et al. 2011 and Tippler et al. 2014).

An additional factor contributing to the higher pH is that many of the mines provide treatment to their wastes and this often increases the pH of the wastewater to promote precipitation of dissolved metals. For example, an earlier investigation of Clarence Colliery by Cohen (2002) reported that calcium oxide was added in the wastewater treatment process to increase water pH to promote oxidation of the iron and manganese. The largest increase in pH in a waterway was detected in Sawyers Swamp (mean 4.70 upstream and 8.42 downstream). These results contrast with most international coal mine studies which typically report that low pH is often due to coal mines generating acidic wastes (Banks et al. 1997, Winterbourn, 1998, Verb and Vis 2000, Brake et al. 2001, Tiwary 2000, Johnson 2003, and

Petty et al. 2010). There was only one location in this current study where river pH declined due to a coal mine discharge, and that was Berrima Colliery where the pH of the Wingecarribee River declined slightly from a mean of 7.48 upstream, to 7.25 downstream. This data was collected after the Berrima Colliery workings were partially flooded, which caused a drop in the mine drainage pH (Wright and Belmer, 2018).

A contributing factor to the absence of major pH reductions is that coal in the study area (Sydney Basin) has lower sulfur content than many other coal fields. Sydney-basin coal has a reputation for having a lower sulphur content (Huleatt 1991). For example, the sulfur content of coal from the Katoomba Seam (Clarence Colliery and Canyon Colliery) was reported to have low pyritic forms of sulphur of 0.02 %, which is much lower than is generally found internationally (Cohen 2002). The lower sulfur content of coal is reflected in the sulfate concentrations in mine discharges in this study which were generally lower than many international studies. This reinforces the observation that acid mine drainage (AMD) was not a major problem in these coal mines.

However, sulfate was significantly elevated in waters downstream of coal mine discharges, compared to upstream, at all eight discharges. The mean sulfate concentration in waterways receiving mine discharges increased more than ten times from 4.12 mg/L upstream, to 46.9 mg/L downstream. The steepest increase in sulfate concentration was found below the Springvale mine (0.8 mg/L upstream compared to 104 mg/L downstream, Table 4). Comparable to our current study were results from an Appalachian (USA) catchment (Petty et al. 2010) with sulfate in unmined streams (mean 14 mg/L) increasing in intensively mined areas to a mean of 22 mg/L (of Killanning geology) and 338 mg/L in the mined stream in the other area of that study (Freeport geology, Petty et al. 2010).

The concentration of all major anions increased due to the inflow of mine discharges into streams or rivers (Table 3, Figure 4). The anion composition of waterways upstream of mines was co-dominated by chloride and bicarbonate, with sulfate sub-dominant (Table 4, Figure 4). On average, water samples collected downstream of mines revealed that bicarbonate was dominant, with sulfate sub-dominant, followed by carbonate and chloride was lowest. The largest overall increase was measured for bicarbonate concentrations, which increased by nearly 22 times (upstream mean 15.0 mg/L, downstream mean 342.0 mg/L, Table 3). The largest mean bicarbonate increase was recorded in the Georges River where the bicarbonate concentration increased from 18.0 mg/L upstream, rising by 35 times to 526 mg/L downstream, due waste from the Westcliff mine (Table 4).

Bicarbonate has been identified as pollutant that can contribute to ecotoxicology of coal-mine wastewater (Vera et al. 2014). The waterways receiving wastewater from four active and one closed mine had mean bicarbonate concentrations above 225 mg/L. This level of bicarbonate (225 mg/L) has been recommended as a trigger value by NSW OEH (2012) to protect aquatic ecosystems. NSW OEH conducted an ecotoxicology investigation on wastewater discharged from the Westcliff Colliery in 2012 and identified bicarbonate as a key pollutant of concern (in that mine discharge) in 2012 (NSW OEH 2012). The OEH report quoted research by Farag and Harper (2012) on bicarbonate and recommended trigger values for 80% to 95% protection of aquatic species of 225 to 319 mg/L (NSW OEH 2012). The largest increase in the mean bicarbonate concentration in this study was found in Sawyers Swamp Creek, which increased from 4.2 mg/L upstream, to 597.7 mg/L downstream. None of the EPA licence for collieries in this study specify a discharge limit for bicarbonate.

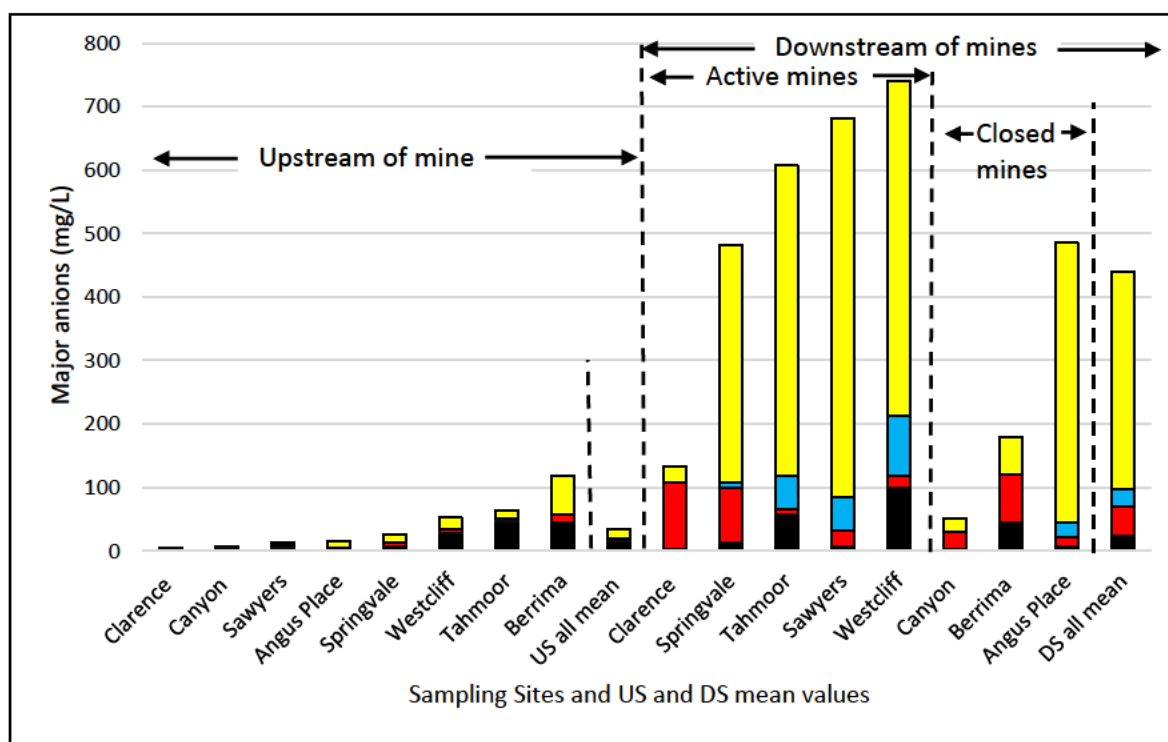


Figure 5. Mean concentration of major anions (mg/L) results for sampling sites over this study. The stacked bars represent the mean concentration of chloride (bottom, black) then sulfate (second, red), then carbonate (third, blue) and bicarbonate (top, yellow). Sites upstream of mines are at the left. The overall mean concentrations are represented by ‘US all mean’. Results for sampling sites downstream of waste discharges are on the right, grouped ‘active mines’ and ‘closed mines’. The overall mean concentrations downstream are represented by ‘DS all mean’.

Thermal water pollution was detected downstream of several coal mines in this investigation (Table 3). Overall, the mean temperature of receiving waterways increased by 2.48° C, with the largest increase measured was a (mean) 10.2° C increase in the water temperature of Sawyers Swamp Creek (Table 3). The second highest increase in water temperature was found in Dalpura Creek, which recorded a mean increase in water temperature of 3.2 ° C downstream of the mine drainage inflow from the closed Canyon Colliery (Table 3). The thermal pollution of streams detected in this study is an important finding. It is the first Australian investigation to link the discharge of wastewater from coal mines to thermal water pollution of waterways receiving the coal mine wastes, although the phenomenon of higher temperatures of flooded mines is internationally well known (Ramos et al. 2015). None of the EPA licence for collieries in this study specify any discharge limit for water temperature (EPA 2018).

The study’s findings are a reminder that active and closed coal mines can both be a major source of water pollution. Berrima and Canyon Collieries both demonstrate that ecologically hazardous water pollution may occur for years after underground coal mines ceased mining. Unlike the UK or US (Robb 1994, Younger, 2004 and Verb and Vis 2000) there has been limited study of the water pollution legacy from closed coal mines in Australia. Battaglia et al. (2005) studied residual pollution and ecological degradation from a waterway affected by a closed coal mine.

Two of the closed mines in this study have previously been investigated. Canyon Colliery ceased mining in 1997 and previous studies have documented aspects of the water pollution caused by this mine (Wright and Burgin 2009 and Price and Wright 2016) and degradation of the ecology of the receiving waterway (Wright and Burgin 2009, Wright et al. 2011 and Wright and Ryan 2016). The Berrima Colliery was still in the closure process when it was investigated in

a 12-month study, by Wright and Belmer (2018). This mine discharged higher concentrations of zinc, nickel, salinity and manganese after it ceased mining and was partially flooded, compared to when it was operating (Wright and Belmer 2018). Whilst water pollution and ecological impairment impacts from closed coal mines has not been commonly studied in Australia, it has been the focus for many studies internationally (e.g. Cairney and Frost 1975, Younger 1993, Brake et al. 2001 and Johnson 2003).

Many of the EPA licences for the collieries in this study have been progressively modified to reduce the pollution of waste-receiving waterways. For example, the Clarence Colliery EPA licence was modified in July 2017 to reduce the concentration of several pollutants permitted to be discharged into the Wollangambe River. An earlier study of this mine and high-conservation value river had documented water pollution and severe ecological damage extending 22 km below the mine (Belmer et al. 2014 and Wright et al. 2017). The new EPA licence reduced the permitted concentration of zinc in the colliery wastewater by 99.5 %, from 1500 µg/L permitted in the previous licence, to 8 µg/L (EPA 2018). The new EPA licence also specified a permitted concentration for nickel of 11 µg/L. The previous licence had not specified any discharge limit for nickel (EPA 2018). It is anticipated that the colliery will upgrade the treatment of their wastewater to conform to the requirements of the new licence.

Conclusion

- 1) Both active and closed coal mines can both modify downstream water quality and generate pollutant concentrations that are hazardous to the receiving river and stream ecosystems.
- 2) The current study provides a reminder that closed coal mines can continue to generate a difficult long-term water pollution legacy.
- 3) Tighter and consistent environmental regulations are needed to reduce pollutants (particularly salinity, zinc and nickel) in wastes from coal mines.
- 4) Long-term rehabilitation strategies to reduce the emission of ecologically hazardous pollutants are needed to avoid legacy pollution problems from closed coal mines.

Research significance

The research from this chapter contributes to the rigour of scientific testing of water chemistry and risk of impairment to river ecology for Australian underground coal mines. Whilst much of the literature suggests that coal mining activities can degrade aquatic ecosystems through changes to river water quality, there have been very few Australian studies that have examined this, and none to a large group of active and inactive mines. Information is particularly scant on the on-going water quality impacts of coal mine impacts to rivers whilst in operation and tentative literature suggests this degradation continues well beyond the time mining practices cease. This chapter's research directly addresses this gap in the literature.

This research helps to demonstrate the need for better environmental regulation through more rigorous and stringent EPL's that reduces the pollution of rivers with harmful contamination and allow for better environmental outcomes for many coal mining operations within the Sydney Basin.

Acknowledgements

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Chapter 5

Regional Comparison of Impacts from Seven Australian Coal Mine Wastewater Discharges on Downstream River Sediment Chemistry, Sydney Basin, New south Wales Australia.

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Abstract

This study investigates the accumulation of licensed and regulated coal mine wastewater pollutants from seven coal mines on each mines respective receiving waterways river sediments. Results from this study shows that the coal mine wastewater pollutants are accumulating within river sediments downstream of the coal mine wastewater inflows at varying levels often greater than the ANZECC guidelines for sediment and often above reference condition sediment concentrations. This is of great concern as these pollutants will likely continue to persist in the river sediment and eventually become legacy pollutants. Coal mine wastewater discharges in New South Wales are regulated by the New South Wales Environmental Protection Authority (NSW EPA) and environmental protection of receiving waterways is implemented through Environmental Protection Licenses. Environmental Protection Licenses set discharge limits for water quality and chemical concentrations within the coal mine waste waters. Though they do not take into account river sediment concentrations. It appears water column pollution regulation at these coal mines is in fact failing to protect the environment whilst still regulated and will continue into the future post mining, licensing and regulation. Water column regulation may well be impractical in protecting the environment as it appears that water column concentrations do not portray the overall environmental impact. It is recommended that the New South Wales Environmental Protection Authority investigate these findings and continue to improve water column pollutant limits as to alleviate the continued accumulation and magnification of the contaminants.

Keywords: Coal mine wastewater, river sediment chemistry, pollutant accumulation, river sediment contamination, environmental regulation, Australia

Introduction

Coal mining practices are well documented to contribute to an array of differing environmental problems including surface and or ground water pollution. Surface water pollution is a major environmental problem associated with coal mining and it occurs through the discharge of mine waters that are contaminated by various disturbances associated with mining practices (Jarvis and Younger 1997, Johnson 2003 and Pond et al. 2008). Water pollution from coal mining occurs as large volumes of surface and groundwater are required to be removed from most underground coal mines. Without this, groundwater would flood most sections of the underground mining operation (Jarvis and Younger 1997 and Younger 2004).

Coal mine wastewater will often be contaminated due to the disturbance of the local geology associated with mining activities. The exact nature of the water contamination will vary depending on local factors such as groundwater geochemistry, hydrology and mineralogy of the local strata (4). Water pollution impacts attributed to treated coal mine waste waters discharged to surface waters often includes changes to pH, elevated salinity, modified stream ionic composition and elevated heavy metals (Wright and Burgin 2009, Wright et al. 2011, Belmer et al. 2014, Wright et al. 2015, Wright and Ryan 2016 and Price and Wright 2016).

Aquatic ecosystems are often more highly sensitive to contamination from anthropogenic activities, especially as waterways are often used as sources of discharge for many industrial wastes (Allen et al. 1993). River sediments are often heavily polluted from the mine wastewater discharges as the heavy metals become water soluble once oxygenated and discharged, often falling out of the water column and accumulating in river sediments often remaining indefinitely (Neff 1984, Cohen et al. 1998, Cohen 2005, Wang and Rainbow 2008, Kolaříková et al. 2012, and NSW OEH 2015). Heavy metal contamination within aquatic environments can persist much longer than terrestrial organic pollutants. This is due to the lack of a “biodegradation function” of heavy metals in aquatic ecosystems in comparison to a terrestrial ecosystem (Twinning et al. 2008 and Ashraf et al. 2011).

The local geology of the study area is described as part of the sedimentary sequence deposited throughout the Sydney Basin. This sedimentary sequence comprises of many layers including the marine dominated Shoalhaven Group, the Illawarra Coal Measures, the Narrabeen and Hawkesbury Sandstone Groups, the Wianamatta Group and small pockets of Basalt flows (Goldbery 1969 and Goldbery and Loughlan 1977). The Illawarra Coal Measure Group is divided into two subgroups, the Nile subgroup and the Charbon subgroup. The Nile subgroup is described as having marine influences whilst the Charbon subgroup is described as being peat influenced and contains large deposits of economically important coal seams (Goldbery 1969 and Goldbery and Loughlan 1977). Mineralogy of most Australian Coals are said to be dominated by phosphates, carbonates, sulphides, silicates and other crystalline mineral groups (Ward 1989).

There is a rich literature on coal mines and water pollution in some parts of the world, such as the United States which includes many regional studies of active and inactive mines (Brake et al. 2001, Johnson 2003 and Petty et al. 2010). One major data gap is that there have been very few studies involving sediment chemistry (none in Australia)

comparing sediment contamination impacts from a regional group of coal mines that discharge waste waters. This relative lack of sediment contamination studies from Australian coal mines is a large scientific gap and requires intensive research. Especially when waterways sediment legacy pollution in many other settings is so well understood.

Coal mine wastewater discharges in New South Wales are regulated by the New South Wales Environmental Protection Authority (NSW EPA) and environmental protection of receiving waterways is implemented through Environmental Protection Licenses (EPL's), under the Protection of the Environment Operations Act 1997 (POEO Act). EPL's set discharge limits for water quality and chemical properties in which coal mine waste waters that are discharged to the environment must adhere to (Wright et al. 2011 and Belmer et al. 2014).

We hypothesize that the heavy metal contamination of the water column is subsequently bioaccumulating and magnifying within the river sediments.

Methods

Study Sites

This study investigates river sediments from waterways receiving wastewater from seven coal mines in the Sydney Basin. Four are located within the Greater Blue Mountains area. They are Angus Place, Canyon, Clarence and Springvale Collieries. Three mines are located in the Greater Southern Highlands area, those being Berrima (Medway), Tahmoor and Westcliff Collieries (Figure 1). The geology of all mine locations share many similarities as they all extract coal from various seams within the Illawarra coal measures spanning the southern and western coalfields within the greater Sydney Basin (Goldbery 1969, Goldbery and Loughlan 1977 and Ward 1989).

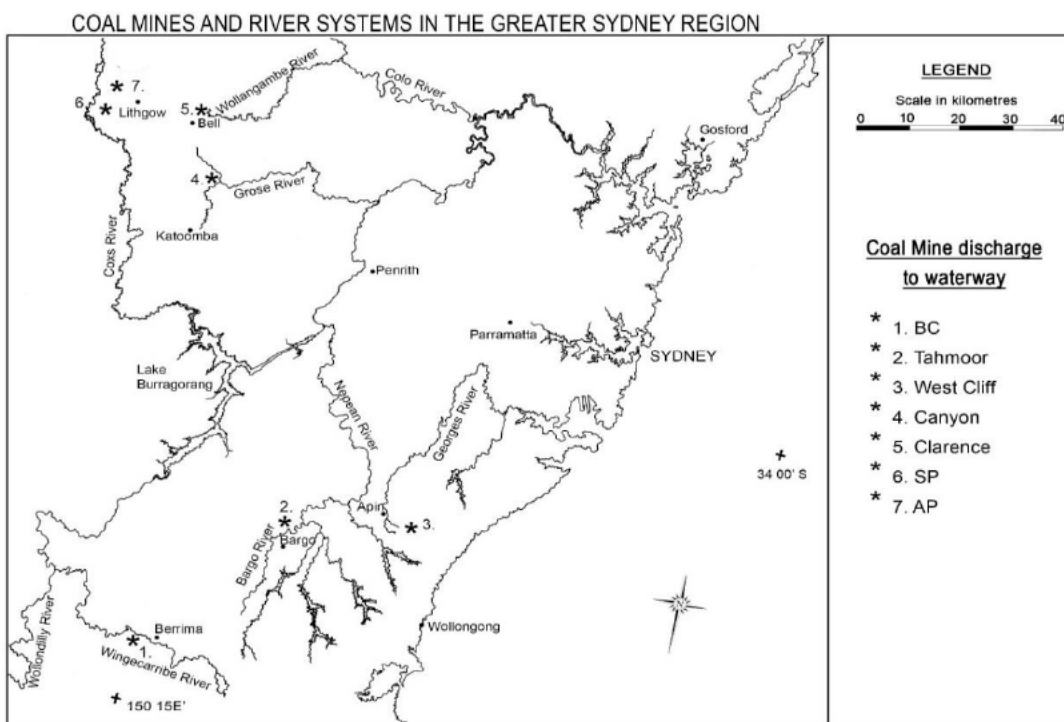


Figure 1. Map of lower Sydney basin, its major waterways and location of the seven coal mines (marked by * and numbered) used in this study. (1 Berrima Colliery, 2. Tahmoor Colliery, 3. West Cliff Colliery, 4. Canyon Colliery, 5. Clarence Colliery, 6. Springvale Colliery, 7. Angus Place Colliery). Some of the smaller tributaries that receive mine waste are not shown (Kangaroo Ck, Sawyers Swamp, Springvale Ck, Dalpura Ck).

Sediment Sampling

Sediment samples were collected on one occasion from upstream and downstream of each coal mines wastewater inflow other than for Angus Place Colliery (Kangaroo Creek). This was due to no upstream location being available. A nearby naturally dilute reference stream was used as its paired upstream sample location (Cox's River). This was achieved by sampling river sediments in a zone of accumulated sediment, following standard methods recommended by the Victorian EPA (25). Samples were taken from approximately 75mm below the surface of the river sediment and placed into sealed glass sampling jars supplied by a commercial laboratory and stored. Three replicated samples were collected and analysed using standard methods (APHA 1998) by Envirolab (Chatswood, NSW) a National Associations of Testing Authorities accredited laboratory for the following pollutants (Barium, Cobalt, Copper, Lithium, Manganese, Nickel, Strontium and Zinc.).

Data Analysis and guideline comparison

For univariate data analysis Students t-test were used to test for differences in river sediment chemistry between samples upstream versus downstream of each mine waste inflows. Results are also compared to Australian sediment guidelines (ANZECC 2000 guideline values for toxicants in sediment) where available (Copper, Nickel and Zinc) and also two international sediment quality guideline values, including a threshold effect concentration (TEC) and a probable effect concentration (PEC) where available (Nickel and Zinc) (Li and Tchounwou 2014). The remaining pollutants which do not have Australian or international guidelines were compared discussed within relation to upstream (reference) sediment values.

Results and Discussion

Barium recorded statistically significant differences between upstream and downstream samples for all mines other than Westcliff Colliery when analysed through Students t-Test (table 1). The ANZECC 2000 guidelines do not stipulate load limits for Barium in sediment. Reference barium at Sawyers Swamp upstream of Angus Place and Springvale Collieries wastewater sources recorded a mean of 10.3 mg/kg whilst downstream barium concentrations were mean 163.3 mg/kg an increase over 15 times. Barium at Cox's River (reference) site was mean 56 mg/kg whilst Kangaroo Creek below Angus Place Collieries wastewater inflow recorded a mean of 173.3 mg/kg an increase greater than three times.

Springvale Creek barium increased over four times from an upstream mean of 115.7 mg/kg to a mean of 490 mg/kg below the coal mine wastewater discharge. Dalpura Creek reference recorded the lowest barium of this study with a mean of 1.33 mg/kg increasing to 2.8 mg/kg downstream. Wollangambe river also recorded low reference barium of 3.0 mg/kg upstream increasing eight times to 25 mg/kg below the discharge. The Wingecarribee River above Berrima (Medway) Collieries wastewater discharge recorded a mean of 41.4 mg/kg increasing over 16 times to a mean of 685 m/kg. The Bargo River reference site recorded low barium concentrations of 5.5 mg/kg similar to Dalpura Creek and the Wollanganbe River. This increased nearly ten times to a mean of 54.5 mg/kg below Tahmoor Collieries wastewater discharge (table 1 and figure 2).

Cobalt was found to be statistically significantly different for Sawyers Swamp, Cox's River/Kangaroo Creek, Dalpura Creek, the Wollangambe River and the Wingecarribee River between upstream and downstream sample locations when

analysed with Students T-test (table 1). There is no ANZECC 2000 guideline for cobalt in sediment. Sawyers Swamp increased from 0.67 mg/kg to 88.5 mg/kg over 100 times higher. Dalpura Creek was below laboratory detection limits upstream of the closed Canyon Coal mines water discharge whilst a mean of 2.1 mg/kg was recorded downstream. The Wollangambe River also recorded below laboratory detectable limits whilst in comparison downstream mean cobalt was 226.7 mg/kg. The greatest concentration in cobalt in sediment was recorded at the Wingecarribee River with upstream reference cobalt measuring a mean of 69.4 mg/kg increasing nearly 80 times downstream of Berrima (Medway) Collieries wastewater inflow to 695 mg/kg. Cox's/Kangaroo Creek recorded cobalt in sediment concentrations upstream of mean 4.8 mg/kg in contrast to Kangaroo Creek downstream of Angus Place Colliery which recorded a mean of 18 mg/kg (table 1 and figure 3).

Copper was found to be statistically significantly different for Sawyers Swamp, Cox's River/Kangaroo Creek, the Wollangambe River, the Wingecarribee River and the Georges River when analysed with Students T-test between upstream and downstream sample locations (table 1). The ANZECC 2000 ISQG-Low guideline for copper in sediment is 65 mg/kg which is above the findings of this study. One sample stream, the Georges River recorded a decrease in copper concentrations in sediment. The Georges River recorded mean copper upstream of Westcliff Collieries wastewater inflow of 3.5 mg/kg decreasing to 1 mg/kg downstream. Sawyers Swamp increased over 10 times from 1.17 mg/kg upstream to 14 mg/kg downstream. Kangaroo Creek below Angus Place Collieries wastewater discharge was 10.1 mg/kg higher (mean 17.3 mg/kg) than the reference stream Cox's River which had mean copper concentrations in sediment of 7.2 mg/kg. The Wollangambe River upstream of Clarence Collieries wastewater discharge recorded mean copper of 0.67 mg/kg in sediment increasing to 2 mg/kg downstream. Berrima (Medway) Collieries wastewater discharge was seen to increase copper in the Wingecarribee Rivers sediments from 3.8 mg/kg upstream to a mean of 13.5 mg/kg downstream (table 1 and figure 4).

Lithium upstream when compared to downstream was seen to be statistically significantly different when analysed with Students T-test for Sawyers Swamp, Springvale Creek, Cox's River/ Kangaroo Creek, the Wollangambe River and the Bargo River (table 1). There is no ANZECC 2000 ISQG-Low guideline for lithium in sediment. All downstream sample locations recorded increases in Lithium other than Dalpura Creek which recorded below laboratory detection limits for both its upstream and downstream sample locations. Lithium concentrations in sediment were found to be below laboratory detectable limits at Sawyers Swamp upstream whilst recording a mean of 29.6 mg/kg downstream of Angus Place and Springvale Collieries. Kangaroo Creek below Angus Place Colliery recorded mean lithium of 13.3 mg/kg in comparison Cox;s Creek upstream recorded below detectable lithium concentrations. The Wollangambe River upstream also recorded below detectable limits of lithium in sediment whilst downstream mean lithium was 3.67 mg/kg. Springvale Creek lithium was found to have a mean of 1.83 mg/kg increasing to 7.75 mg/kg below Springvale Collieries wastewater inflow. The Bargo River increased from below detectable limits of lithium to 35 mg/kg downstream of Tahmoor Collieries wastewater discharge (Table 1 and Figure 5).

Manganese at all sample locations other than the Bargo River and the Georges River recorded statistically significant differences when analysed with Students T-test between upstream and downstream samples (table 1). There is no

ANZECC 2000 ISQG-Low guideline for manganese in sediment. Sawyers Swamp upstream was found to have a mean of 15 mg/kg of manganese whilst downstream manganese increased to 2425 mg/kg an increase over 160 times, the greatest increase in manganese at all sample locations. Springvale Creek increased from a mean of 416.7 mg/kg upstream of Springvale Collieries wastewater discharge to 13300 mg/kg over 30 times higher than reference manganese. Kangaroo Creek below Angus Place Collieries wastewater inflow measured mean manganese of 2233.3 mg/kg whilst its paired reference stream was (mean 90.2 mg/kg). Dalpura Creek upstream of the Canyon Colliery discharge was below laboratory detection limits in comparison downstream was 20.6 mg/kg. The Wollangambe River downstream of Clarence Collieries wastewater inflow recorded mean manganese in sediment of 6520 mg/kg whilst upstream manganese was 48.7 mg/kg this is an increase downstream over 120 the second highest increase recorded for manganese (table 1 and figure 6).

Nickel in sediment recorded statistically significant differences between upstream and downstream sample locations for all sites other than the Bargo and Georges Rivers (table 1). The ANZECC 2000 ISQG-Low guideline for nickel in sediment is 21 mg/kg. Both Dalpura Creek and the Wollangambe River recorded reference upstream nickel concentrations below laboratory detection limits increasing downstream to 6 mg/kg and 180 mg/kg respectively. Sawyers Swamp increased some 140 times from 0.83 mg/kg upstream to 114 mg/kg downstream. Springvale Creek increased from a mean of 18.7 mg/kg to a mean downstream of 60.6 mg/kg. Cox's River recorded mean nickel in sediment of 4.4 mg/kg in contrast its paired impact stream Kangaroo Creek below Angus Place Collieries wastewater discharge was (mean 17.7 mg/kg).

The Wingecarribee River recorded upstream mean nickel of 11 mg/kg whilst downstream a mean of 1030 mg/kg was recorded nearly 95 times higher. This study reveals that the Wollangambe River downstream (near 10 times higher), Sawyers Swamp downstream (over 5 times higher), Springvale Creek downstream (near 3 times higher) and the Wingecarribee River downstream (nearly 50 times higher) all have mean nickel well above the recommended ANZECC guideline (table 1 and figure 7).

Strontium was found to be statistically significantly different when analysed with Students T-test between upstream and downstream for all sample streams other than the Georges River and Dalpura Creek. Dalpura Creek measured below detectable laboratory limits of strontium at both upstream and downstream sample locations (table 1). There is no ANZECC 2000 ISQG-Low guideline for strontium in sediment. Sawyers Swamp increased 10 times downstream from a mean of 1.67 mg/kg to a mean of 16.6 mg/kg. Springvale Creek upstream recorded a mean of 11 mg/kg increasing over three times to a mean of 36.5 mg/kg downstream of Springvale Collieries wastewater discharge. Cox's River recorded mean reference strontium of 6.2 mg/kg whilst its paired downstream site Kangaroo Creek recorded a mean of 41.3 mg/kg an increase over six times. The Wollangambe River upstream of Clarence Collieries wastewater discharge recorded mean strontium in sediment of 0.67 mg/kg increasing slightly to 1.33 mg/kg downstream. The Wingecarribee River upstream of Berrima (Medway) Collieries waste adit recorded mean strontium of 9.4 mg/kg increasing over 6 times to 59 mg/kg downstream. The greatest increase was recorded on the Bargo River with mean strontium downstream

of Tahmoor Collieries wastewater inflow recorded at 25.5 mg/kg an increase over 25 times from reference conditions (mean 0.88 mg/kg) (table 1 and figure 8).

Statistically significant differences were found for zinc when analysed with Students T-test between upstream and downstream samples for all sample streams other than the Bargo River (table 1). The ANZECC 2000 ISQG-Low guideline for zinc in sediment is 200 mg/kg. Sawyers Swamp mean zinc upstream was recorded as 7.33 mg/kg increasing Over 50 times downstream to a mean of 272.5 mg/kg. Springvale Creek recorded the highest upstream zinc concentrations of 87 mg/kg though this still increased downstream of Springvale Collieries wastewater inflow some four times to a mean of 350 mg/kg. Cox's River reference stream measured a mean of 34.6 mg/kg whilst its paired downstream sample recorded mean zinc of 710 mg/kg below Angus Place Collieries wastewater discharge. Dalpura Creek zinc concentrations in sediment were below laboratory detectable limits upstream of Canyon Collieries wastewater discharge whilst downstream recorded a mean of 19.6 mg/kg. The Wollangambe River saw an increase over 80 times from reference (upstream) zinc measuring a mean of 3 mg/kg whilst in comparison downstream of Clarence Collieries wastewater inflow zinc was recorded at a mean of 243.3 mg/kg. The Wingecarribee River recorded the greatest concentrations and increase of this study. Upstream of Berrima (Medway) Collieries wastewater discharge zinc in sediment was recorded at a mean of 31.2 mg/kg increasing nearly 190 times to a mean of 5950 mg/kg below Berrima (Medway) Collieries wastewater adit.

The Georges River in contrast recorded much higher zinc concentrations in sediment upstream (mean 42 mg/kg) than downstream which measured a mean of 5.5 mg/kg. This study reveals that the Wollangambe River downstream (43 mg/kg higher), Sawyers Swamp downstream (72 mg/kg higher), Springvale Creek downstream (150 mg/kg higher), Kangaroo Creek downstream (500 mg/kg higher) and the Wingecarribee River downstream (5750 mg/kg higher) zinc in sediment results are above the recommended ANZECC guideline to varying magnifications (table 1 and figure 9). Similar results were reported by Pavlowsky et al (2017) in the Big River area of the Ozark Highlands Southern Missouri North America. This study investigated sediment legacy pollutants from closed lead mining activities and revealed mean Zinc concentrations of 502 mg/kg ranging between 83 and 1554 mg/kg. This is similar to this study other than downstream of the Berrima (Medway) Colliery.

Table 1 Sediment chemistry results for each coal mine. Mean values and *p values*. BD = Below Laboratory Detection Limits. n/s = not significant. Red font is concentrations above the ANZECC Guideline (28).

Colliery name, p value, sample river and location/ Parameter	Barium	Cobalt	Copper	Lithium	Manganese	Nickel	Strontium	Zinc
Springvale and Angus Place								
p value	0.01	0.002	0.01	0.002	0.0006	0.004	0.02	0.003
Sawyers upstream	10.3	0.67	1.17	BD	15	0.83	1.67	7.33
Sawyers downstream	163.3	88.5	14	29.6	2425	114	16.6	272.5
Springvale								
p value	0.01	0.23	0.1	0.02	0.02	0.01	0.01	0.02
Springvale upstream	115.7	60.7	13	1.83	416.7	18.7	11	87
Springvale downstream	490	51.5	25.6	7.75	13300	60.6	36.5	350
Angus Place								
p value	0.004	0.003	0.01	0.001	0.0001	0.0007	2.54E-05	0.0002
Cox's upstream	56	4.8	7.2	BD	90.2	4.4	6.2	34.6
Kangaroo downstream	173.3	18	17.3	13.3	2233.3	17.7	41.3	710
Canyon								
p value	0.02	0.03	n/a	n/a	0.01	0.0003	n/a	0.0001
Dalpura upstream	1.33	BD	BD	BD	BD	BD	BD	BD
Dalpura downstream	2.8	2.1	BD	BD	20.6	6	BD	19.6
Clarence								
p value	0.007	0.03	0.01	0.03	3.64E-06	0.03	0.006	0.003
Wollangambe upstream	3	BD	0.67	BD	48.7	BD	0.67	3
Wollangambe downstream	25	226.7	2	3.67	6520	180	1.33	243.3
Berrima (Medway)								
p value	0.008	0.02	0.002	0.15	0.25	0.02	5.98E-07	0.003
Wingecarribee upstream	41.4	9.4	3.8	BD	342	11	9.4	31.2
Wingecarribee downstream	685	695	13.5	1.5	26475	1030	59	5950
Tahmoor								
p value	0.01	0.36	n/a	0.009	0.07	0.14	0.02	0.25
Bargo upstream	5.5	1.5	BD	BD	80	1	0.88	6
Bargo downstream	54.5	2	BD	35	25.5	6.5	24.5	9.5
Westcliff								
p value	0.23	0.25	0.06	0.1	0.07	0.09	0.1	0.04
Georges upstream	33.5	0.75	3.5	3.5	130	1	12	42
Georges downstream	24	BD	1	5	36.5	0.75	5.5	5.5

For the three parameters measured in this study (Copper, Nickel and Zinc) which have Australian sediment guidelines (ANZECC 2000 guidelines for toxicants) increased to varying degrees downstream of almost all the mines. Copper recorded increases in downstream sediment concentrations below four of the eight discharges excluding Canyon, Tahmoor and Westcliff collieries ranging from double and ten times the recorded upstream (reference) sediment loads. Although all the downstream samples were below the Australian guidelines such large increases from reference conditions are of concern. Nickel was found to increase in downstream sediments below seven of the eight wastewater inflows from 3 times to 180 times the reference sediment concentrations. Four of the downstream sediment sample concentrations were above the Australian sediment guidelines for nickel in sediment (21 mg/kg) as for both international sediment guidelines TEL (16 mg/kg) and PEL 43 mg/kg). Of most concern is Berrima (Medway) Colliery which recorded an increase from 11 mg/kg upstream (half of the Australian guideline) to 1030 mg/kg

downstream (nearly 50 times the Australian guideline). Clarence Colliery which recorded below laboratory detectable limits upstream (<0.5 mg/kg) increasing to 180 mg/kg downstream (over 7 times the Australian guideline). The combined discharge point from Springvale and Angus Place collieries increased concentrations from 0.83 mg/kg upstream to 114 mg/kg downstream (over 5 times the Australian guideline) whilst the individual discharge from Springvale Colliery recorded 18.7 mg/kg upstream increasing to 60.6 mg/kg downstream nearly three times the Australian guideline.

Zinc is also of major concern at many of the coal mines with large increases measured downstream of six of the eight discharges. Zinc increased downstream of the six coal mines between 6 times and 185 times that of reference sediment concentrations with five of the six recording values above the recommended Australian sediment guidelines (200 mg/kg) and one of the international sediment guideline values TEL (123 mg/kg) whilst three of the five were above the PEL international guideline of 315 mg/kg. Berrima (Medway) Colliery recorded the greatest increase from 31.2 mg/kg upstream increasing to 5950 mg/kg (nearly 30 times the Australian guideline) along with Angus Place Colliery with increased sediment loads from reference conditions (34.6 mg/kg) increasing downstream to 710 mg/kg (3 and a half times the Australian guideline). Sediment concentration downstream of Springvale Colliery recorded increased zinc concentrations from 87 mg/kg to 350 mg/kg. In contrast, downstream of both Clarence Colliery and the combined discharges from Angus Place and Springvale Collieries recorded concentrations just above the Australian and one international guideline (TEL) but still well above that of reference zinc in sediment (Clarence upstream 3 mg/kg increasing to 243.3 mg/kg and Angus Place/Springvale increasing from 7.33 mg/kg upstream to 272.5 mg/kg downstream).

Although five of the eight parameters discussed in this research do not have stipulated Australian guideline values (ANZECC 2000) for load limits in sediment the majority of the five parameters increased drastically within sediments below most coal mine wastewater inflows. Barium downstream of Springvale, Angus Place, Berrima (Medway) and Tahmoor collieries increased from 3 times to 16 times from that of reference conditions with Berrima (Medway) Colliery and Springvale and Angus Place's combined discharge of greatest concern increasing from 15 and 16 times respectively. Cobalt followed a similar trend with Springvale and Angus Place's combined discharge, Clarence and Berrima (Medway) collieries being of most concern. Sediment downstream of the collieries recorded increased concentrations between 60 and 440 times the that of cobalt recorded in reference sediment. Springvale and Angus Place collieries combined discharge increased Cobalt sediment concentration from 0.67 mg/kg upstream to 88.5 mg/kg downstream. Of most concern is Clarence Colliery (increasing from below laboratory detectable limits (<0.5 mg/kg) to 226.7 mg/kg downstream) and Berrima (Medway) Colliery increasing cobalt from 9.4 mg/kg upstream to 695 mg/kg downstream. Lithium did not record great increases other than at Tahmoor colliery and the combined discharge of Springvale and Angus Place collieries increasing from below detectable limits to 35 mg/kg at Tahmoor Colliery and below laboratory detectable limits to 29.6 mg/kg at Springvale and Angus Place Collieries.

Although large increases were not recorded lithium was only measured in laboratory detectable limits (<0.5 mg/kg) at two upstream sites and both at very low concentrations (Springvale Colliery 1.83 mg/kg and Westcliff Colliery 3.5 mg/kg. Magnesium recorded the greatest concentrations per mg/kg of all the pollutants measures, increasing at six of the eight downstream locations between 75 times and 165 times. Of greatest concern is Berrima (Medway) Colliery

which recorded sediment increases downstream of the wastewater inflow some 165 times that of reference sediment manganese. Springvale Colliery is also of concern recording upstream sediment manganese of 15 mg/kg increasing 160 times to 2425 mg/kg along with Clarence Colliery which recorded increases in sediment concentrations in the range of 130 times that of reference conditions (48.76 mg/kg to 6520 mg/kg). Both Springvale and Angus Place Collieries individual wastewater discharges recorded increases greater than 30 times at Springvale Colliery (upstream 416.7 mg/kg and downstream 13300 mg/kg) and nearly 25 times greater downstream of Angus Place Colliery (90.2 mg/kg increasing downstream to 2233.3 mg/kg). Strontium recorded increases downstream of six of the eight wastewater discharges with one of the remaining two recording no difference (Below laboratory detectable limits upstream and downstream). Strontium increased from double to 24 times each waterways respective reference condition.

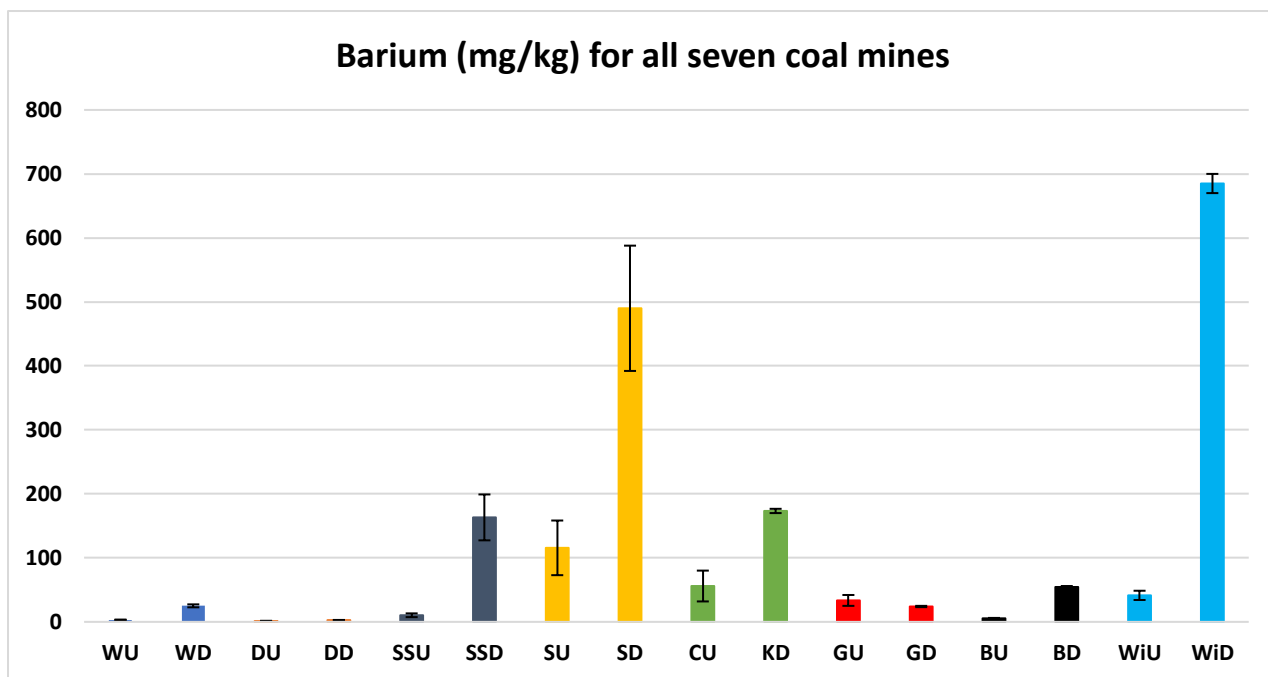


Figure 2 Barium for all seven mines with standard error bars. U = upstream, D = downstream, W = Wollangambe D = Dalpura, SS = Sawyers Swamp, S = Springvale, C = Cox’s, K = Kangaroo, G = Georges, B = Bargo and Wi = Wingecarribee

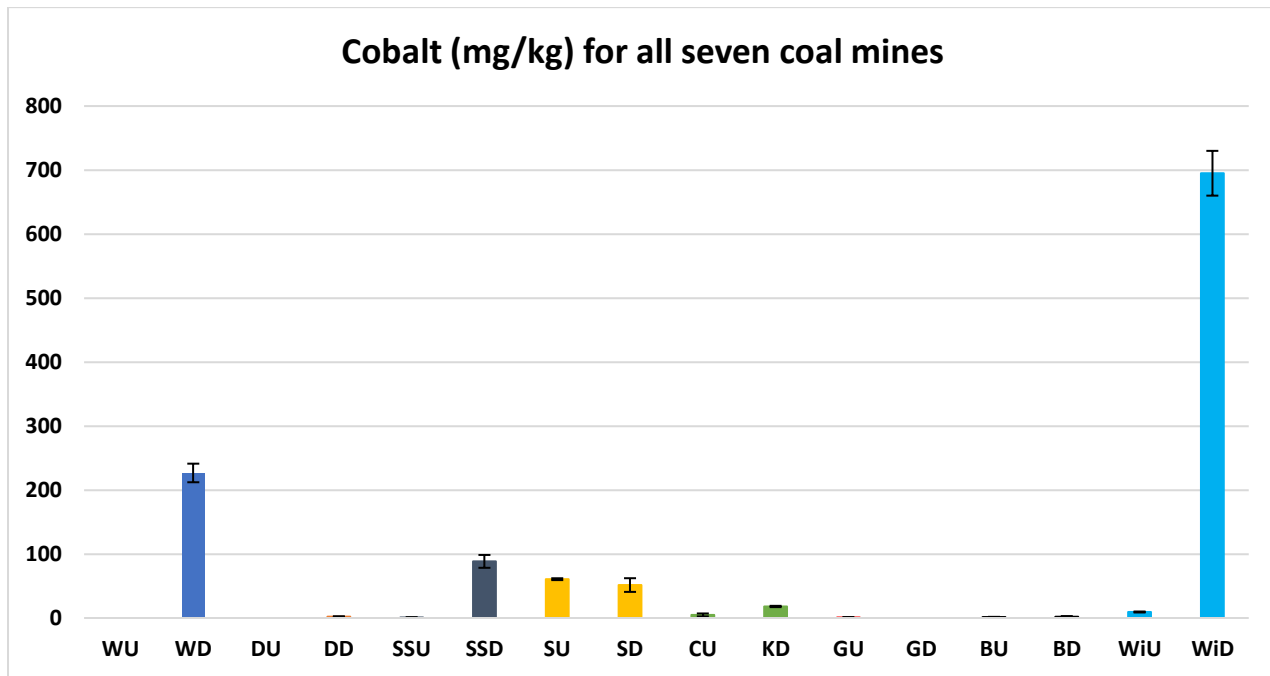


Figure 3 Cobalt for all seven mines with standard error bars. U = upstream, D = downstream, W = Wollangambe D = Dalpura, SS = Sawyers Swamp, S = Springvale, C = Cox’s, K = Kangaroo, G = Georges, B = Bargo and Wi = Wingecarribee

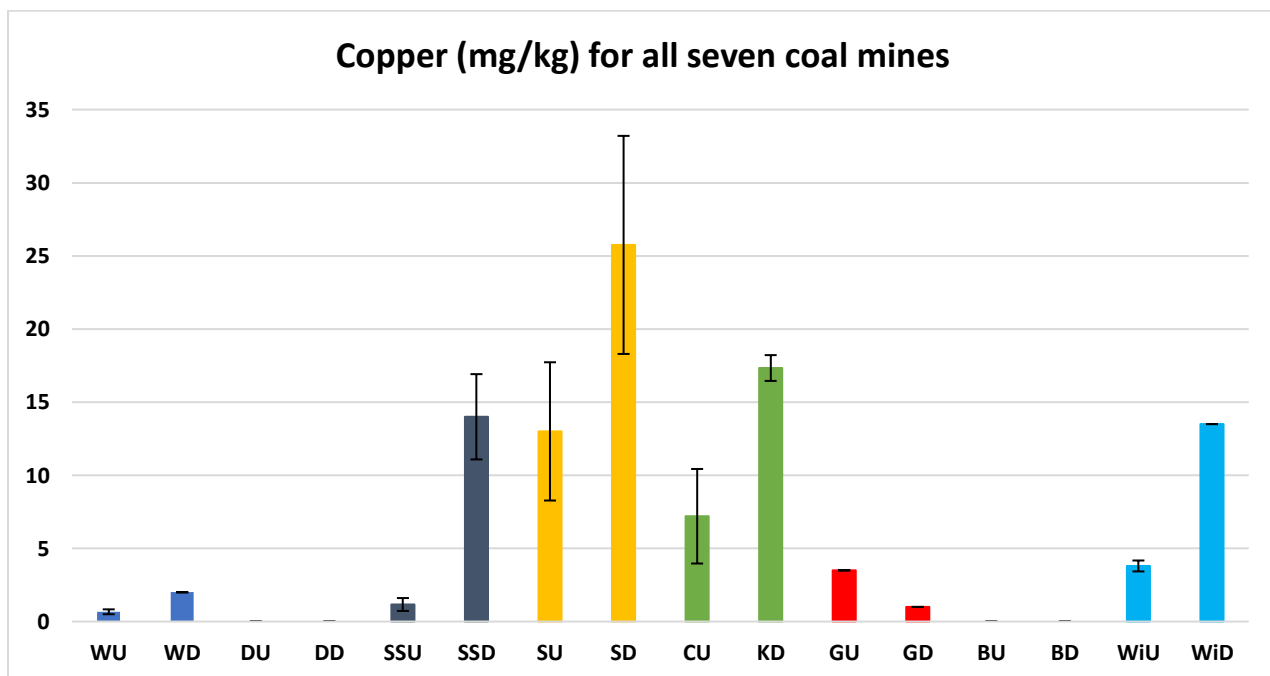


Figure 4 Copper for all seven mines with standard error bars. U = upstream, D = downstream, W = Wollangambe D = Dalpura, SS = Sawyers Swamp, S = Springvale, C = Cox’s, K = Kangaroo, G = Georges, B = Bargo and Wi = Wingecarribee

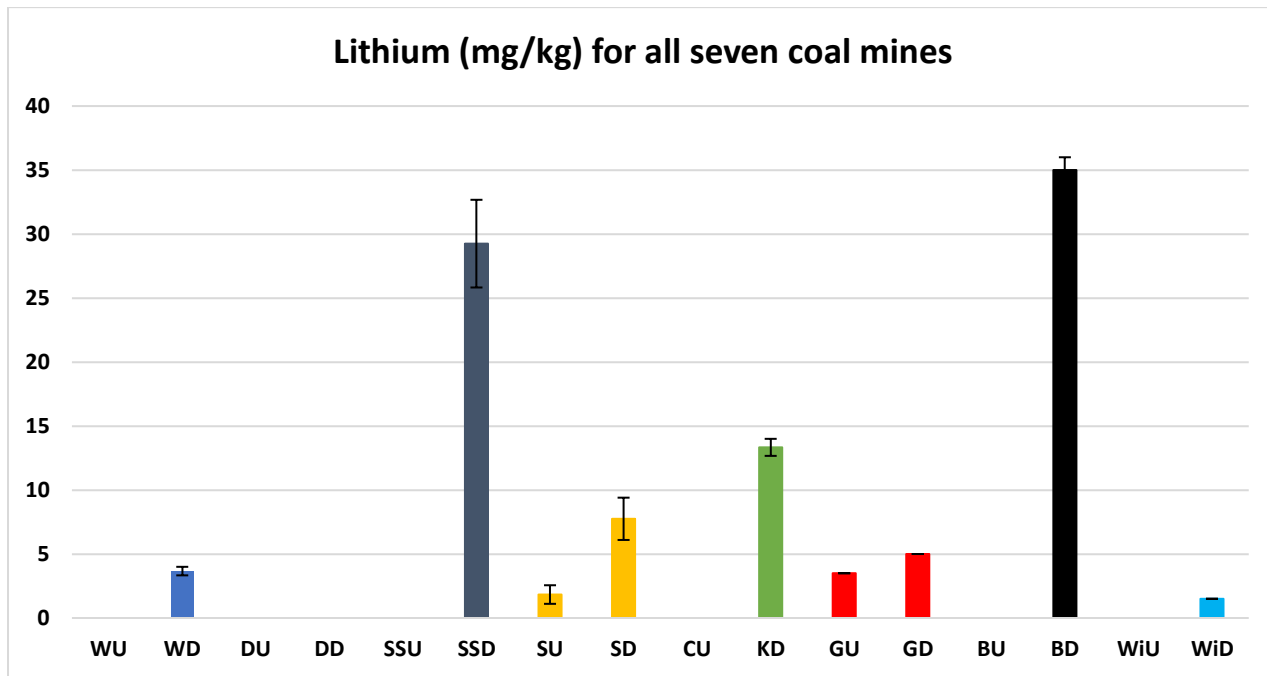


Figure 5 Lithium for all seven mines with standard error bars. U = upstream, D = downstream, W = Wollangambe D = Dalpura, SS = Sawyers Swamp, S = Springvale, C = Cox’s, K = Kangaroo, G = Georges, B = Bargo and Wi = Wingecarribee

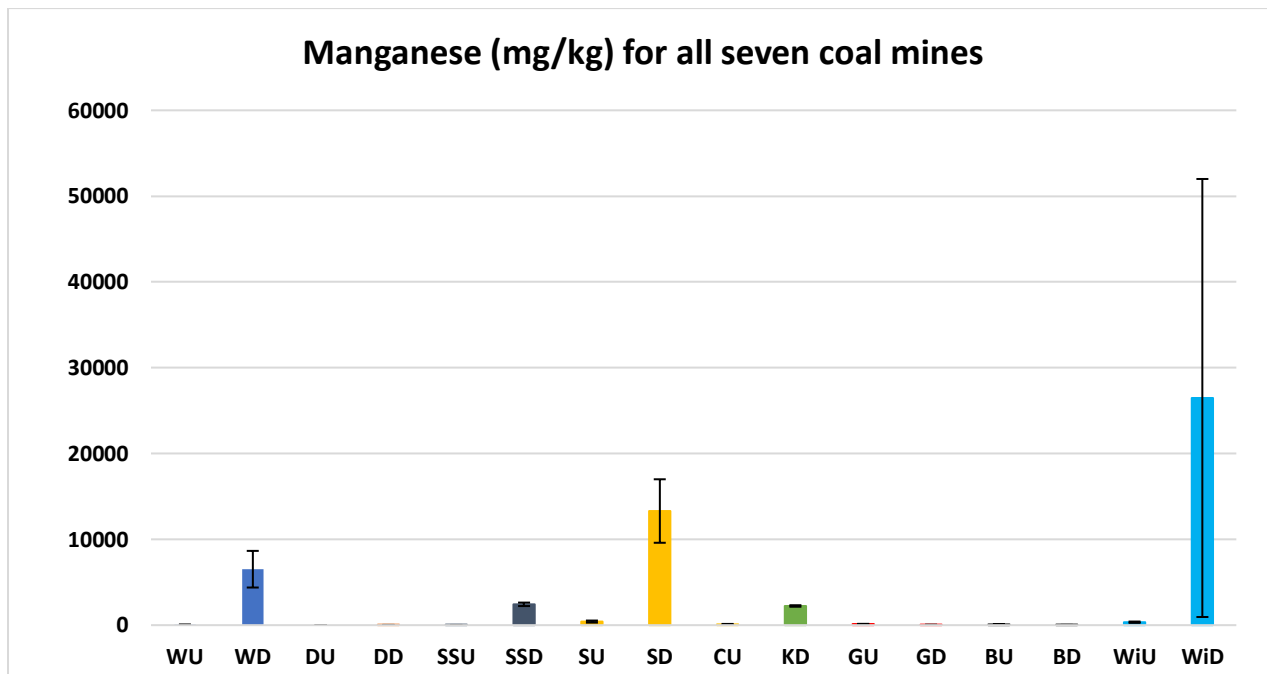


Figure 6 Manganese for all seven mines with standard error bars. U = upstream, D = downstream, W = Wollangambe D = Dalpura, SS = Sawyers Swamp, S = Springvale, C = Cox’s, K = Kangaroo, G = Georges, B = Bargo and Wi = Wingecarribee

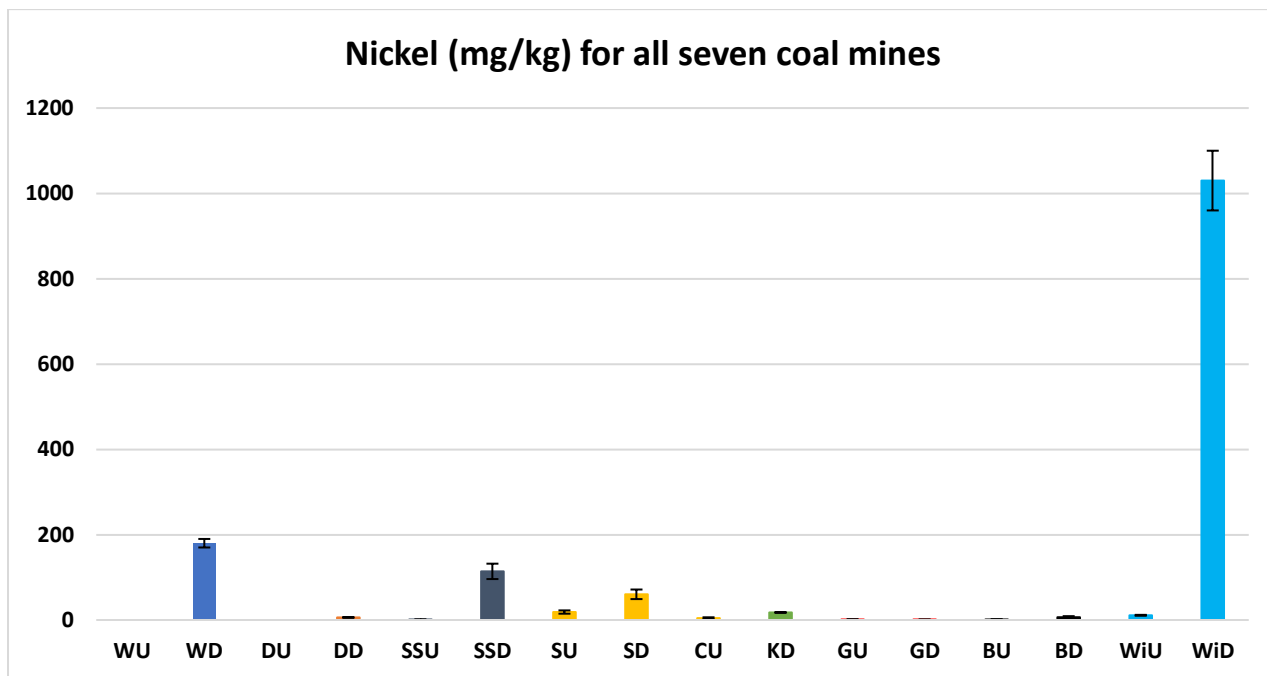


Figure 7 Nickel for all seven mines with standard error bars. U = upstream, D = downstream, W = Wollangambe D = Dalpura, SS = Sawyers Swamp, S = Springvale, C = Cox’s, K = Kangaroo, G = Georges, B = Bargo and Wi = Wingecarribee. The ANZECC 2000 ISQG-Low guideline for nickel in sediment (21 mg/kg).

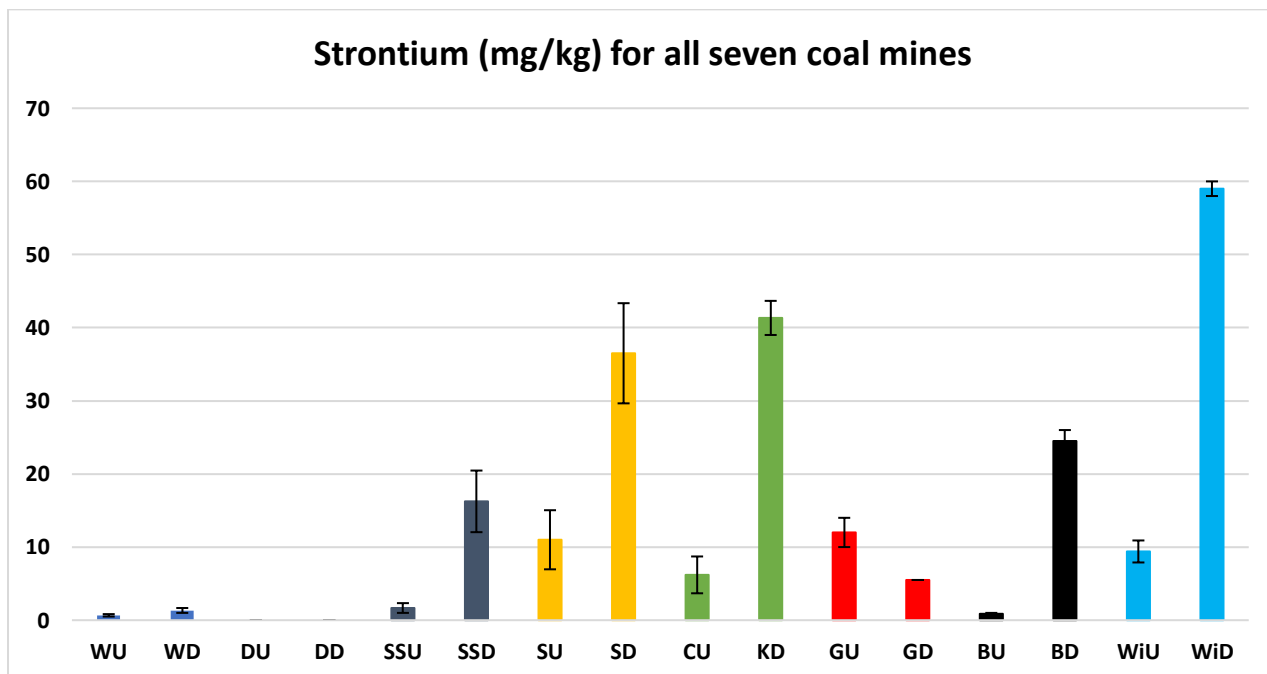


Figure 8 Strontium for all seven mines with standard error bars. U = upstream, D = downstream, W = Wollangambe D = Dalpura, SS = Sawyers Swamp, S = Springvale, C = Cox’s, K = Kangaroo, G = Georges, B = Bargo and Wi = Wingecarribee

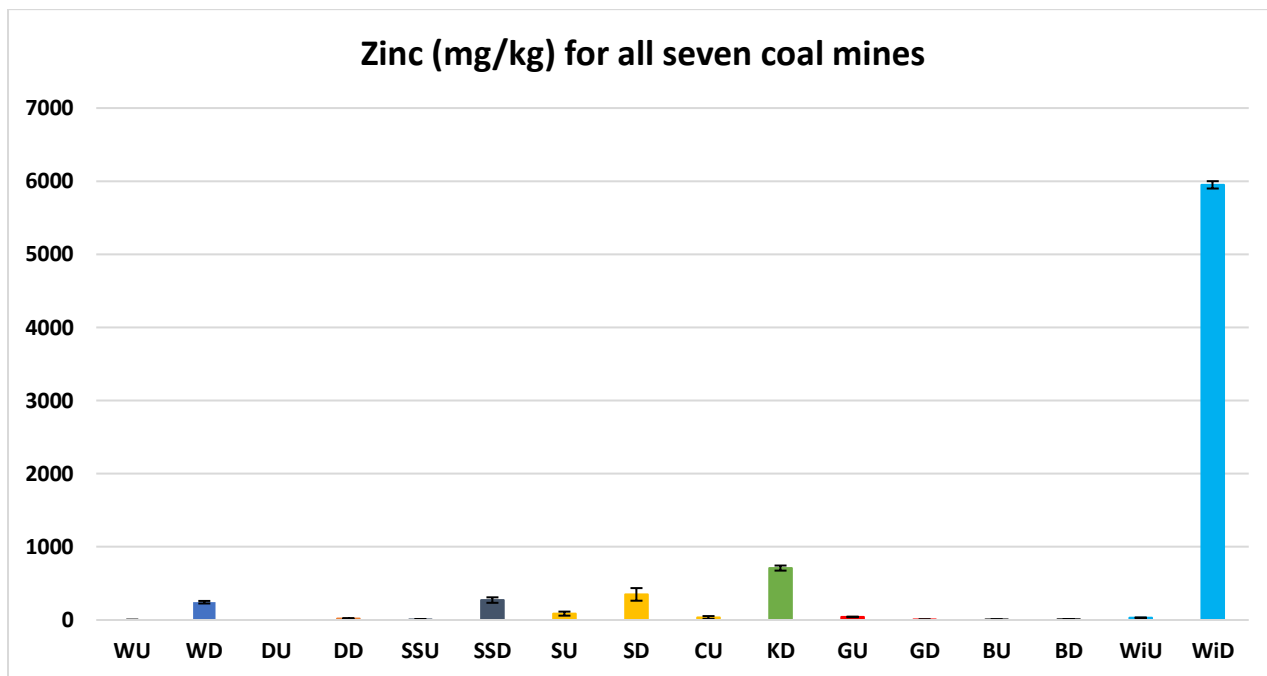


Figure 9 Zinc for all seven mines with standard error bars. U = upstream, D = downstream, W = Wollangambe D = Dalpura, SS = Sawyers Swamp, S = Springvale, C = Cox’s, K = Kangaroo, G = Georges, B = Bargo and Wi = Wingecarribee. The ANZECC 2000 ISQG-Low guideline for nickel in sediment (200 mg/kg).

Conclusions

Results from this study shows that the coal mine wastewater pollutants are accumulating within river sediments downstream of the coal mine wastewater inflows at varying levels often greater than the ANZECC guidelines for sediment and often above reference condition sediment concentrations. This is of great concern as these pollutants will likely continue to persist in the river sediment and eventually become legacy pollutants. Of greatest concern are the levels of Nickel and Zinc in relation to the ANZECC guidelines with four downstream locations recording Zinc levels greater than the recommended levels and three locations for Nickel. Magnesium and strontium were also found the be of concern as they increased statistically significantly downstream of most mines and in large concentrations at times.

It appears water column pollution regulation at these coal mines is in fact failing to protect the environment whilst still regulated and will continue into the future post mining, licensing and regulation. Water column regulation may well be impractical in protecting the environment as it appears that water column concentrations do not portray the overall environmental impact. It is recommended that the New South Wales Environmental Protection Authority investigate these findings and continue to improve water column pollutant limits as to alleviate the continued accumulation and magnification of the contaminants. A limitation of this study is the depth of sampling at 75mm only and further investigation should be undertaken which could include samples from 2 or 3 depths.

Research significance

The research is the most thorough and detailed examination of the impacts of coal mines on the contamination of sediment in rivers that receive coal mine wastes. This research demonstrates the importance of understanding the accumulation of metals in river sediment downstream of coal mine waste discharges. Accumulation of coal mine contaminants in river sediment can contribute to the impacts of the aquatic ecosystem. Research in this chapter

provides one of Australia's first comparisons of river sediment contamination downstream of a group of Australian active and inactive coal mines.

Acknowledgements: We acknowledge and pay our respects to the traditional custodians of the land in which this study was conducted. The Dharug, Gundungurra, Tharawal, Wiradjuri and Yuin people and their elder's past and present. Thanks to Alison Ellis for the preparation of the study area map. Western Sydney University, the Blue Mountains Conservation Society, Sustainable Southern Highlands and the Colong Foundation for Wilderness all provided funding for chemical analysis of water samples. Envirolab (Sydney) generously provided additional testing. We also acknowledge the energetic field work assistance of Nicholas Szafranec, Ben Green and Paul Hammond.

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Chapter 6

Regional comparison of the impact on stream macroinvertebrates from Australian underground coal mine wastewater discharges from active and inactive mines in the Sydney Basin, New South Wales Australia.

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Abstract

This study investigates aquatic macroinvertebrates from waterways receiving wastewater from seven coal mines in the Sydney Basin. Three of the coal mines were inactively mining ore and four actively mining ore during sampling. Macroinvertebrates were collected from each collieries receiving waterway upstream and downstream of all mine wastewater inflow. All the coal mines wastewater discharges are licensed and regulated by the New South Wales Environment Protection Authority (NSW EPA). Results of the study show that the coal mine waste waters being discharged are having varying negative impacts to the receiving waterways aquatic ecosystem through aquatic macroinvertebrate biotic indices, despite whether mining is active or inactive. Biotic indices measured at active and inactive coal mines show that actively mined waste waters are most likely causing less of an impact to the receiving waterways aquatic ecosystem than inactively mined waste waters. All the waterways receiving un-treated (inactively mining) waste waters recorded statistical differences for all biotic indices when analysed between their upstream and downstream sample locations. This was in contrasted to the actively mined (treated waste waters) with only one of the five streams sampled recording statistical differences for all biotic indices. Results suggest that once mining ceases and the treatment of the coal mine waste waters subsequently ceases the receiving waterways aquatic ecosystem are clearly more degraded. This is of great concern as once mining ceases so does the treatment of their waste waters. It is recommended that the NSW EPA further investigate measures of treatment post coal mining at these mines to ensure further degradation of the receiving waterways does not occur.

Keywords

Benthic macroinvertebrates, coal mine wastewater, coal mining, environmental management, coal mine regulation, active mines, inactive mines.

Introduction

Coal mining practices are well documented to contribute to an array of differing environmental problems including air pollution, fire hazards, ground subsidence or deformation, surface and or ground water pollution. Surface water pollution is a major environmental problem associated with coal mining and occurs through the discharge of mine waters that are contaminated by various disturbances associated with mining practices (Jarvis and Younger 1997, Johnson 2003 and Pond et al. 2008).

Water pollution from coal mining occurs as large volumes of surface and groundwater are required to be removed from most underground coal mines. This is generally through the pumping of water to the surface as surface and groundwaters infiltrate the mine shafts through the local geological sub-strata and subsequently accumulates in the underground mine workings. Without this, groundwater would flood most sections of the underground mining operation (Jarvis and Younger 1997, Younger 2004). This practice of mine and wastewater discharge is licensed and regulated through contaminant limits in New South Wales by the New South Wales Environmental Protection Authority (NSW EPA).

Coal mine wastewater will often be contaminated due to the disturbance of the local geology associated with mining activities. The exact nature of the water contamination will vary depending on local factors such as groundwater geochemistry, hydrology and mineralogy of the local strata. In addition to the physical activity of the mining operation and the removal of the waste water, other activities will also often contaminate water used throughout a mining plant which can include; coal washing and the inclusion of other wastes generated by the surface operation at the mine such as sewage wastes (Younger 2004).

A widespread form of water pollution caused by coal and metalliferous mining is termed 'acid mine drainage' (AMD) and often occurs when waste waters are not treated or when treatment ceases (Brake et al. 2001). This arises when sulfur in coal (or other ores) is oxidised due to the disturbance associated with mining and its exposure to both air and water which triggers the formation of sulfuric acid of various strengths (Johnson 2003). The AMD acid leaches and mobilises metals in mine water, depending on the sulfur content of the ore and the characteristics of the surrounding geology (Banks et al. 1997, Brake et al. 2001 and Johnson 2003).

Water pollution impacts attributed to treated coal mine waste waters discharged to surface waters often includes changes to pH, elevated salinity, modified stream ionic composition and elevated heavy-metals (Pond et al. 2008, Wright and Burgin 2009, Wright 2012, Belmer et al. 2014, Wright et al. 2015, Wright and Ryan, 2016 and Price and Wright, 2016).

River sediments are also often heavily polluted from the mine wastewater discharges as the heavy metals become water soluble once oxygenated and discharged, often falling out of the water column and accumulating in river sediments contaminating them with many heavy metals (Cohen et al. 1998, Cohen 2002 and NSW OEH 2015).

A compounding effect of coal mining wastewater discharges into streams and rivers coupled with the eventual contamination of the receiving waterway is the impact on the freshwater ecosystems. Battaglia et al. (2005) concluded that increased heavy metals contributed to the degradation of stream macroinvertebrate assemblages. Wright & Burgin

(2009) reported elevated zinc levels from drainage flowing from the closed coal mine (Canyon Colliery) impaired the downstream Grose Rivers stream ecosystems with reductions in macroinvertebrate taxonomic richness and abundance (Wright & Burgin 2009).

Similar studies by (Belmer et al. 2014 and Wright et al. 2017) reported that a coal mine (Clarence Colliery) wastewater discharge increased the Wollangambe Rivers salinity, pH, nickel and zinc levels which were concluded to have reduced macroinvertebrate taxonomic richness and abundance downstream of the mine discharge.

There is a rich literature on coal mines and water pollution in some parts of the world, such as the United States which includes many regional studies of active and inactive mines (Brake et al. 2001, Pond et al. 2008 and Petty et al. 2010). Many of these studies do not include sampling above the mining operation and, as a result, often do not illustrate the full extent of impact on the receiving waterways and their ecosystems. One major data gap is that there have been very few studies (none in Australia) comparing impacts to coal mine wastewater receiving waterways aquatic ecosystems from a regional group of coal mines that discharge wastes from active (treated) and inactive mines (un-treated).

The relative lack of studies investigating aquatic ecosystem degradation from Australian coal mines is puzzling given the importance of the industry. Despite increased mining of coal in recent decades and coal becoming Australia's second highest value export, there are comparatively fewer studies on the impacts to aquatic ecosystems from coal mines in Australia (Mudd 2009).

Coal mine wastewater discharges in New South Wales are regulated by the New South Wales Environmental Protection Authority (NSW EPA) and environmental protection of receiving waterways is implemented through Environmental Protection Licenses (EPL), under the Protection of the Environment Operations Act 1997 (POEO Act). EPL's set discharge limits for water quality and chemical properties in which coal mine waste waters discharge to the environment must adhere to (Wright, 2012 and Belmer et al. 2014).

In many cases the EPL's for coal mine waste discharges are failing to protect the receiving waterways ecosystems by failing to identify ecologically hazardous chemicals in the waste discharges and often imposing water quality and chemical limits much higher or significantly different to the receiving waterway or local reference conditions (Belmer et al. 2014, Belmer et al. 2015, Wright et al 2017 and Wright et al 2018). The research questioned posed for this research is; how does the receiving aquatic ecosystem (measured via aquatic macroinvertebrates) differ from a regional group of active (treated waste waters) coal mines compared to inactive coal mines (un-treated wastewater)?

Methods

Sample locations

This study investigates eight waterways receiving wastewater from seven coal mines in the Sydney Basin with three inactively mining coal and four actively mining coal during sampling. Four mines are located within the Greater Blue Mountains area. These include Angus Place Colliery (inactively mined), Canyon Colliery (inactively mined), Clarence Colliery (actively mined) and Springvale Colliery (actively mined) (Figure 1). Three mines are located in the Greater Southern Highlands area, those being Berrima (Medway) Colliery (inactively mined) Tahmoor Colliery (actively mined) and Westcliff Colliery (actively mined) (Figure 1). The geology of all mine locations share many similarities

as they all extract coal from various seams within the Illawarra coal measures spanning the southern and western coalfields within the greater Sydney Basin (Goldbery 1969, Goldbey 1977 and Ward 1989).

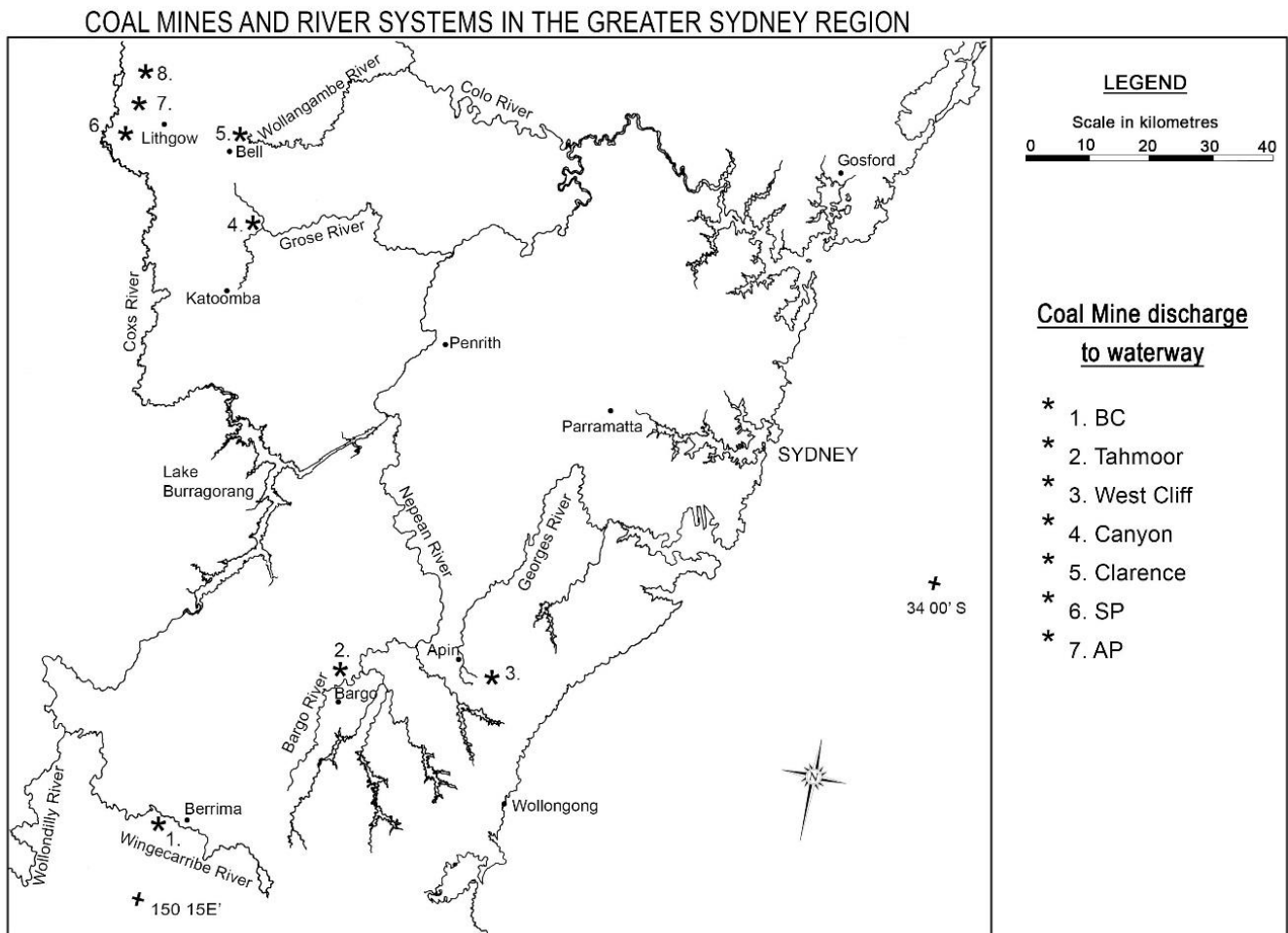


Figure 1 Map of lower Sydney basin, its major waterways and location of the seven coal mines (marked by * and numbered) investigated in this study that discharge wastewater to nearby streams or rivers. (1 Berrima (Medway) Colliery, 2. Tahmoor Colliery, 3. Westcliff Colliery, 4. Canyon Colliery, 5. Clarence Colliery, 6. Springvale Colliery, 7. Angus Place Colliery).

Table 1 Colliery name, waterway name, approximate longitude and latitude and altitude (Metres above sea level) of collieries and waterways used in this study. Stream order is derived from the Strahler method (Strahler 1952).

Colliery name	Waterway name	Sample location	longitude	latitude	Altitude (ASL)	Stream order
Inactive mines						
Angus Place Colliery	Sawyers Swamp	Upstream	-33.396377 S	150.133510 E	1000 m	1
	Kangaroo Creek	Downstream	-33.349507 S	150.098834 E	915 m	1
Berrima (Medway) Colliery	Wingecarribee River	Upstream	-34.489611 S	150.261454 E	590 m	3
	Wingecarribee River	Downstream	-34.488328 S	150.255918 E	530 m	3
Canyon Colliery	Dalpura Creek	Upstream	-33.539753 S	150.308879 E	910 m	1
	Dalpura Creek	Downstream	-33.540910 S	150.308116 E	890 m	1
Active mines						
Clarence Colliery	Wollangambe River	Upstream	-33.455964 S	150.249101 E	1025 m	1
	Wollangambe River	Downstream	-33.455673 S	150.257359 E	960 m	2
Springvale Colliery	Springvale Creek	Upstream	-33.405991 S	150.125420 E	1020 m	1
	Springvale Creek	Downstream	-33.401727 S	150.094156 E	890 m	1
	Sawyers Swamp	Downstream	-33.380748 S	150.086568 E	895 m	1
Tahmoor Colliery	Bargo River	Upstream	-34.236946 S	150.579127 E	260 m	2
	Bargo River	Downstream	-34.244479 S	150.590146 E	250 m	2
Westcliff Colliery	Georges River	Upstream	-34.205055 S	150.798932 E	230 m	1
	Georges River	Downstream	-34.203947 S	150.798088 E	225 m	1

Macroinvertebrates

Aquatic macroinvertebrates were collected on one occasion. All paired upstream and downstream samples were collected on the same day from the respective receiving waterway upstream and downstream of each mines waste inflow (Table 1). A total of ten randomly selected, quantitative benthic macroinvertebrate samples were collected at five receiving waterways and 5 from the remaining three receiving waterways. Samples were collected from flowing sections of each waterway. A 'kick' net (frame of 30 x 30 cm and 250 µm mesh) was used to collect invertebrates and sampling was achieved by disturbing stream substrate in a 30 cm by 30 cm quadrat upstream of the sample net for 30 seconds and collecting all the benthic materials that flow into net (Chessman 1995). Net contents were then placed into individual sample containers and preserved in 70% ethanol.

Aquatic macroinvertebrates were counted and identified to the family level (for the majority of taxa) at the School of Science and Health laboratory facilities at the Western Sydney Universities Hawkesbury Campus using a Nikon stereo microscope 10x magnification and the identification keys (Gooderham and Tsyrlin, 2002 and Hawking and Smith 1997). Family level macroinvertebrate identification has been found to be an adequate taxonomic resolution for coal mine impact assessment (Wright et al. 1995).

Data Analysis

For univariate data analysis (upstream compared to downstream) Students t-test were used to test for differences between aquatic macroinvertebrate community structure. Standard industry biotic indices for aquatic macroinvertebrates were used to infer differences in community structure from upstream and downstream of waste inflows, these include; Total Abundance and Family Richness, Ephemeroptera, Plecoptera and Trichoptera Abundance (EPT abundance), Ephemeroptera, Plecoptera and Trichoptera Family Richness and Ephemeroptera, Plecoptera and Trichoptera Percent (EPT%).

Multivariate data analysis was used to compare community structure of macroinvertebrates with the software package PRIMER 6. PRIMER 6 was used to infer statistical differences in community structure of aquatic macroinvertebrates (Lenat 1988, Plafkin et al. 1989, Chessman 2003 and Metzeling et al. 2006). BIOENV (BEST) was performed to analyse which water quality and chemistry parameters (previously published by the authors) greatly contributed to the change in macroinvertebrate community structure between upstream and downstream sample locations.

Results

A combined total of 12866 individual macroinvertebrates were collected and identified from 8 waterways from 15 individual sample locations (7 upstream and 8 downstream). Of the total aquatic macroinvertebrates collected and identified 5853 were sampled from upstream locations and 7013 from downstream sample locations. Some 58% of the downstream macroinvertebrates were collected at one downstream sample location (Westcliff Colliery) which is some 85% of the total collected macroinvertebrates at Westcliff Colliery. A similar trend was found for upstream samples with some 41% of all upstream macroinvertebrates collected from Tahmoor Colliery.

SIMPER results show combined inactive mines recorded the greatest dissimilarity between their paired upstream and downstream sample locations in comparison to combined actively mined results, inactive mines significance level of 0.5% (Global R): 0.274 and active mines significance level of 0.5% and (Global R) 0.122. Each individual inactive mine; Angus Place Colliery significance level of 0.1% (Global R) 0.949, Canyon Colliery significance level of 0.1% (Global R) 0.689 and Berrima (Medway) Colliery significance level of 0.1% (Global R) 0.594. Whilst each individual active mine recorded less dissimilarity; Springvale Colliery significance level of 0.1% (Global R) 0.940, Westcliff Colliery significance level of 0.1% (Global R) 0.394, Clarence Colliery significance level of 0.1% (Global R) 0.359, significance level of 0.1% (Global R) 0.394, Tahmoor Colliery significance level of 0.5% (Global R) 0.162.

Dissimilarities of macroinvertebrate community structure are depicted as two nMDS plot graphs divided into Blue Mountains mines and Southern Highlands mines (Figures 7 and 8). For the Blue Mountains mines nMDS the majority of the reference samples at both active and inactive mines show similarity to each other with the majority of replicates clustering together in the centre. Whilst in comparison downstream sample replicates are scattered from top to bottom and right, with a few *Dalpura* replicates (D) shifting far left. As for the Southern Highlands mines nMDS the Bargo river replicates are showing some similarity to each with both other collieries (Berrima (Wi) and Westcliff (G)) showing less similarity to their paired sampled replicates (Figures 2 and 3).

Macroinvertebrate community structure was found to be statistically dissimilar when analysed for similarity through ANOSIM at all streams when compared between their upstream and downstream sample locations with a significance level of 0.5% (Global R) 0.033, between active mining upstream and active mining downstream with a significance level of 0.1% (Global R) 0.122, between inactive mining upstream and inactive mining downstream with a significance level of 0.1% (Global R) 0.274 and when compared between active mining downstream and inactive mining downstream with a significance level of 0.1% (Global R) 0.259.

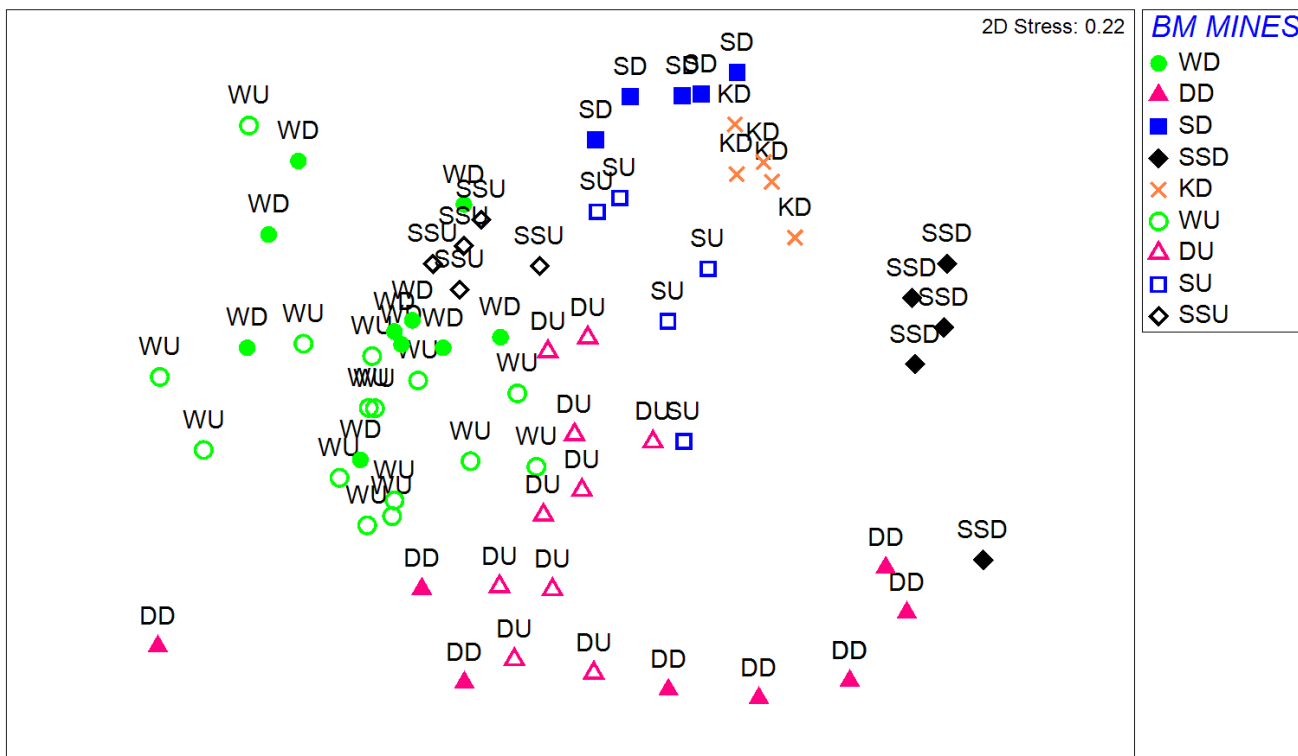


Figure. 2 nMDS plot graph depicting Blue Mountains Collieries macroinvertebrate community structure. Solid shapes are downstream samples and outlined shapes are upstream samples. D = downstream, U = upstream. W (Circles) = Wollangambe River (Clarence Colliery), D (Triangle) = Dalpura Creek (Canyon Colliery), S (Square) = Springvale Creek (Springvale Colliery), SS (Diamond) = Sawyers Swamp (Springvale and Angus Place Colliery) and K (Cross) = Kangaroo Creek (Angus Place Colliery).

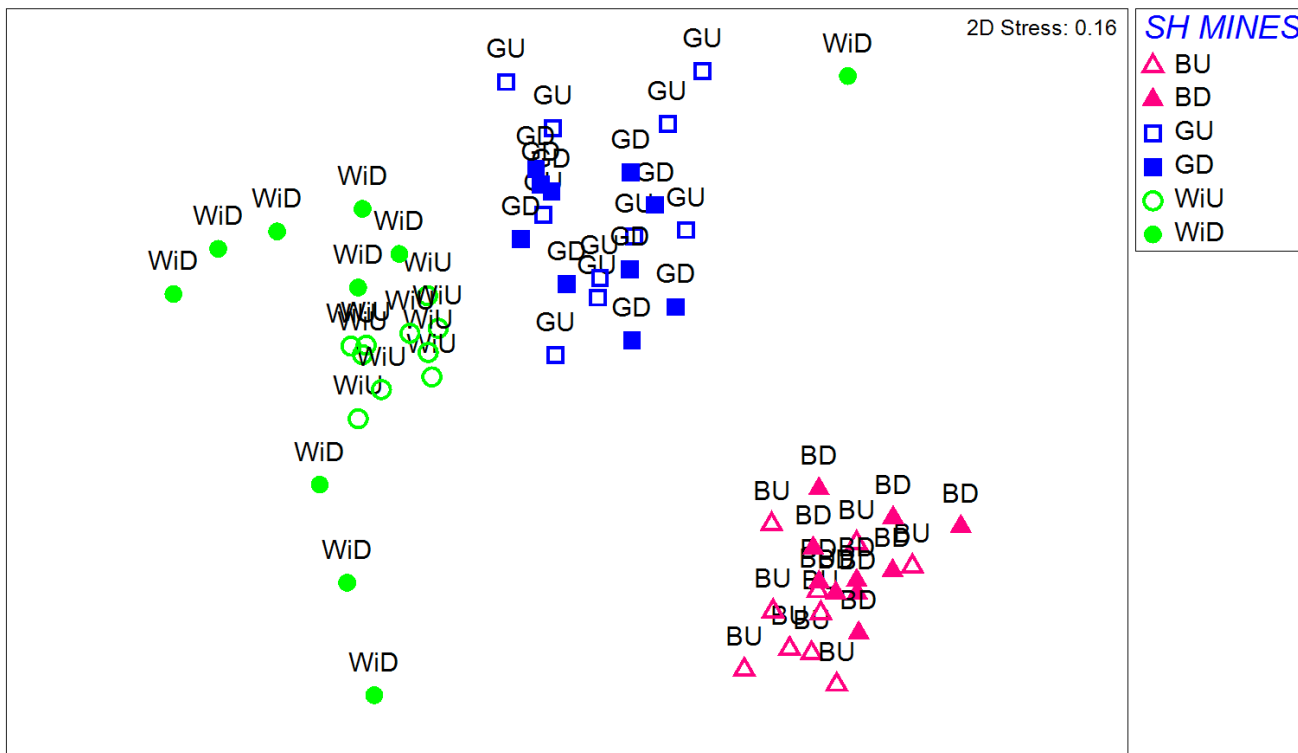


Figure. 3 nMDS plot graph depicting Southern Highland Collieries macroinvertebrate community structure. Solid shapes are downstream samples and outlined shapes are upstream samples. D = downstream, U = upstream. B (Triangle) = Bargo River (Tahmoor Colliery), G (Squares) = Georges River (Westcliff Colliery) and WiD (Circles) = Wingecarribee River (Berrima (Medway) Colliery).

Table 2 Macroinvertebrate total individual abundance, Family richness, EPT abundance and EPT percent (%) range, total counts and means for all mines inactive and active. * = p < 0.05; ** = p < 0.001; *** = p < 0.0001; ns = not significant.

Colliery	Biotic indices	Individual Abundance		Family Richness		EPT Abundance		EPT %		EPT Family Richness	
		Range (Total)	Mean	Range	Mean	Range (Total)	Mean	Range	Mean	Range	Mean
Angus Place Colliery (Inactive)	p value	**		*		**		**		*	
	Sawyers Swamp Upstream (reference)	37 - 125 (363)	72.6	11-16	14	13 - 43 (121)	24.2	27.4 - 44.4	34.4	1 - 4	2.6
Berrima Colliery (Inactive)	p value	***		***		***		***		***	
	Wingecarribee Upstream (reference)	18 - 121 (796)	79.6	11-27	18.5	4 - 5 (299)	29.9	15.4 - 60.5	35.9	3 - 8	6.1
Canyon Colliery (Inactive)	p value	***		***		***		**		***	
	Dalpura Creek Upstream (reference)	22 - 100 (544)	54.4	4-14	9.1	4 - 72 (344)	34.4	8.9 - 84.1	62.3	3 - 5	3.4
Clarence Colliery (Active)	p value	*		***		***		*		n/s	
	Wollangambe River Upstream (reference)	52 - 166 (614)	97.4	10-13	11.4	0 - 87 (284)	23.9	23.1 - 62.7	39.7	1 - 8	4.4
Springvale Colliery (Active)	p value	***		*		***		***		*	
	Springvale Creek Upstream (reference)	54 - 92 (348)	69.6	6-15	10.6	3 - 12 (36)	7.2	4.3 - 16	10.8	1 - 3	1.8
Tahmoor Colliery (Active)	p value	n/s		*		n/s		n/s		n/s	
	Bargo River Upstream (reference)	68 - 933 (2406)	240.6	5-14	8.8	11 - 64 (346)	34.6	6.7 - 4	21.2	3 - 6	4.2
Westcliff Colliery (Active)	p value	***		*		n/s		*		n/s	
	Georges River Upstream (reference)	41 - 155 (782)	78.2	4-18	11.4	0 - 53 (208)	20.8	0 - 64.6	28	0 - 5	2.8
	Georges River Downstream (impact)	130 - 889 (4065)	406.5	10-17	14.4	2 - 64 (345)	34.5	0.7 - 43.8	11.4	1 - 5	2.6

Reference Angus Place macroinvertebrate biotic indices were all found to be statistically different when compared to their paired impacted (downstream) sample. Total abundance for Angus Place Colliery reference site recorded a mean of 72.6 individuals per replicate and ranged between 37 and 125. In comparison, the total abundance downstream of Angus Place Collieries wastewater inflow was (mean of 36.6) nearly half of its paired reference sample. (Table 2 and Figure 4). Family richness for Angus Place Collieries reference site recorded a mean of 14 families per replicate and ranged between 11 and 16. In comparison, family richness downstream recorded a mean of 10 ranging between 6 and 15 families (Table 2 and Figure 5).

EPT abundance for the reference site recorded a mean of 24.2 EPT individuals per replicate and ranged between 13 and 43. In comparison, EPT abundance downstream of Angus Place Collieries wastewater discharge recorded a mean of 5.6 EPT individuals per replicate and ranged between 3 and 7 (Table 2 and Figure 6). %EPT for Angus Place Collieries reference site recorded a mean of 34.4 %EPT individuals and ranged between 27 and 44 %EPT. In comparison, %EPT downstream of Angus Place Collieries wastewater discharge recorded a mean of 15.9 %EPT individuals per replicate and ranged between 11 and 21 (Table 2 and Figure 7). EPT Family Richness upstream of Angus Place Colliery recorded a mean of 2.6 EPT families ranging between 1- 4 families whilst downstream only 1 family was recorded in each replicate (Table 2 and Figure 8).

Reference Springvale Colliery macroinvertebrate biotic indices were all found to be statistically different when compared to their paired impacted (downstream) sample. Total abundance for Springvale Colliery reference site recorded a mean of 69.6 individuals per replicate and ranged between 54 and 92. In comparison, the total abundance downstream of Springvale Collieries wastewater inflow was (mean 38.2 and 12.8 for both receiving waterways) and ranged between 2 and 50, a 50% loss in abundance (Table 2 and Figure 4). Family richness for Springvale Collieries reference site recorded a mean of 10.6 families per replicate and ranged between 6 and 15. In comparison, family richness downstream recorded means of 5.6 and 13 ranging between 6 and 17 families (Table 2 and Figure 5)

EPT abundance for the reference site recorded a mean of 7.2 EPT individuals per replicate and ranged between 3 and 12. In comparison, EPT abundance downstream of Springvale Collieries wastewater discharge recorded means of 2.4 and 2.6 EPT individuals per replicate and ranged between 0 and 4 individuals per replicate (Table 2 and Figure 6). %EPT for Springvale Collieries reference site recorded a mean of 10.8 %EPT individuals and ranged between 4.3 and 16 %EPT. In comparison, %EPT downstream of Springvale Collieries wastewater discharge recorded means of 6 and 25.6 %EPT individuals per replicate and ranged between 0 and 50 %EPT (Table 2 and Figure 7). Although this is a higher percent of EPT taxa, it should be noted that this is represented by a less sensitive EPT community downstream. In contrast to the upstream sensitive taxa such as leptophlebiidae, Hydraboisidae and Philoptomidae the downstream community recorded none of these sensitive EPT taxa and appear to have been replaced with much less sensitive EPT taxa such as baetidae and hydroptilidae (Table 2). EPT Family Richness upstream of Springvale Colliery recorded a mean of 1.8 EPT families ranging between 1- 3 families in contrast downstream means of 0.6 and 0.2 families were recorded ranging between 0 – 1 at both downstream locations (Table 2 and Figure 8).

Macroinvertebrate biotic indices results for Clarence Colliery show statistically significant differences between upstream and downstream samples for Abundance, Family Richness, EPT abundance and %EPT (Table 2). Total abundance for the Wollangambe River reference site recorded a mean of 97.4 individuals per replicate and ranged between 56 and 166. In comparison, mean total abundance for the paired impacted site was 7.9 and ranged between 3 and 34 individuals per replicate showing a decrease some 12 times from reference condition abundance (Table 2 and Figure 4). Family richness for the reference site recorded a mean of 11.4 families per replicate and ranged between 10 and 14. In comparison, the family richness at the paired impacted site was (mean 3.6) and ranged from 2 to 10 this is a loss over 3 times the family richness of reference streams (Table 2 and Figure 5).

EPT abundance for the reference site recorded a mean of 23.9 EPT taxa per replicate and ranged between 0 and 87. In comparison, the EPT abundance at the paired impacted site was (mean 0.8) and ranged from 0 to 3. This shows a decrease of nearly 30 times the abundance from reference conditions (Table 2 and Figure 6). EPT % at Wollangambe River reference site recorded a mean of 39.7% EPT taxa per replicate and ranged between 23.1 and 62.7. In comparison, the EPT % at the downstream impacted site was (mean 6.6%) and ranged from 0 to 26% (Table 2 and Figure 7). EPT Family Richness upstream of Clarence Colliery recorded a mean of 4.4 EPT families ranging between 1- 8 families whilst downstream a mean of 3.1 was recorded ranging between 0 – 6 (Table 2 and Figure 8).

Macroinvertebrate biotic indices results for Dalpura Creek show statistically significant differences between upstream and downstream samples for all biotic indices (Table 2). Total abundance for the reference site recorded a mean of 54.4 individuals per replicate and ranged between 22 - 100. In comparison, total abundance for the impacted site was (mean 4.8) some eleven time lower (Table 2 and Figure 4). Family richness for the reference site recorded a mean of 9.1 families per replicate and ranged between 4 and 14 families. In comparison family richness for the impacted site was 3.1 families per replicated sample and ranged from 0 to 6 families. This shows a decrease of family richness of nearly three times (Table 2 and Figure 5).

EPT abundance for the reference site recorded a mean of 34.4 EPT individuals per replicate and ranged between 4 and 72. In comparison, the EPT abundance for the impacted site was mean 1.1 ranging between 0 and 4 EPT individuals per replicate. This is a loss on average over 30 times (Table 2 and Figure 6). EPT % for the reference site recorded a mean of 62.3% EPT taxa per replicate and ranged between 8.9 and 84.1% EPT taxa per replicate. In comparison, EPT % for the impacted site (mean 19.7) ranging from 0 to 66.7% EPT taxa. On average this is a loss of over three times of EPT% between reference (upstream) and impacted (downstream) samples (Table 2 and Figure 7). EPT Family Richness upstream of Canyon Colliery recorded a mean of 3.4 EPT families ranging between 3 - 5 families in contrast downstream of Canyon Collieries wastewater inflow only 1 family was recorded (mean 0.6 EPT Families) (Table 2 and Figure 8).

Macroinvertebrate biotic indices results for Wingecarribee River show statistically significant differences between upstream and downstream samples for all biotic indices (Table 2). Total abundance for the reference site recorded a mean of 79.6 individuals per replicate and ranged between 18 - 121. In comparison, total abundance for the impacted site was (mean 18.8) ranging from 4 -37 some four time lower (Table 2 and Figure 24. Family richness for the

reference site recorded a mean of 18.5 families per replicate and ranged between 11 and 27 families. In comparison family richness for the impacted site was 6.6 families per replicated sample and ranged from 3 -11 families. This shows a decrease of family richness of nearly three times (Table 2 and Figure 5).

EPT abundance for the reference site recorded a mean of 29.9 EPT individuals per replicate and ranged between 4 and 52. In comparison, the EPT abundance for the impacted site recorded a mean of 0.7 ranging between 0 and 2 EPT individuals per replicate. This is a loss on average over 40 times (Table 2 and Figure 6). EPT % for the reference site recorded a mean of 35.9% EPT taxa per replicate and ranged between 15.4 – 60.5% EPT taxa per replicate. In comparison, EPT % for the impacted site recorded a mean of 3.9 ranging from 0 to 11.1% EPT taxa. On average this is a loss of over nine times of EPT% between reference (upstream) and impacted (downstream) samples (Table 2 and Figure 7). Family Richness upstream of Berrima (Medway) Colliery recorded a mean of 6.1 EPT families ranging between 3 - 8 families in contrast downstream of Canyon Collieries wastewater inflow only 1 family was recorded (mean 0.6 EPT Families) (Table 2 and Figure 8).

Macroinvertebrate biotic indices results for Bargo River show statistically significant differences between upstream and downstream samples for Family Richness only (Table 2). Total abundance for reference site recorded a mean of 240.6 individuals per replicate and ranged between 68 and 933. In comparison, mean total abundance for the paired impacted site was 196.5 and ranged between 11 and 718 (Table 2 and Figure 4). Family richness for the reference site recorded a mean of 8.8 families per replicate and ranged between 5 and 14. In comparison, the family richness at the paired impacted site was (mean 6.9) and ranged from 4 to 10 (Table 2 and Figure 5).

EPT abundance for the reference site recorded a mean of 34.6 EPT taxa per replicate and ranged between 11 - 64. In comparison, the EPT abundance at the paired impacted site was (mean 26.6) and ranged from 5 to 67. (Table 2 and Figure 6). EPT % for the reference site recorded a mean of 21.2% EPT taxa per replicate and ranged between 6.7 and 46. In comparison, the EPT % at the paired impacted site was (mean 18.7%) and ranged from 6.1 to 45.5% (Table 2 and Figure 7). Family Richness upstream of Tahmoor Colliery recorded a mean of 4.2 EPT families ranging between 3 - 6 families in contrast downstream 3.6 EPT families were recorded (range 2 – 5) (Table 2 and Figure 8).

Macroinvertebrate biotic indices results for Georges River show statistically significant differences between upstream and downstream samples for Abundance, Family Richness and EPT % (Table 2). Total abundance for reference site recorded a mean of 78.2 individuals per replicate and ranged between 41 and 155. In comparison, mean total abundance for the paired impacted site was 406.5 and ranged between 130 and 889 individuals per replicate of which the majority (nearly 50%) were chironomidae and simuliidae (1797 of 4065 total impacted macroinvertebrates sampled). Showing an increase over five times from reference condition abundance to impacted sampled abundance though nearly 50% of this sample was comprised of two Diptera (fly) families (chironomidae and simuliidae) (Table 2 and Figure 4).

Family richness for the reference site recorded a mean of 11.4 families per replicate and ranged between 4 and 18. In comparison, the family richness at the paired impacted site was (mean 14.4) and ranged from 10 to 17 an increase of

three families from reference to impacted samples (Table 2 and Figure 5). EPT abundance for the reference site was not found to be statistically different and recorded a mean of 20.8 EPT taxa per replicate and ranged between 0 and 53. In comparison, the EPT abundance at the paired impacted site was (mean 34.5) and ranged from 2 to 64 (Table 2 and Figure 6). EPT % for the reference site recorded a mean of 28% EPT taxa per replicate and ranged between 0 and 64.6. In comparison, the EPT % at the paired impacted site was (mean 11.4%) and ranged between 0.7 and 43.8% (Table 2 and Figure 7) on average a loss of 2.5 times the EPT % of replicates. EPT Family Richness upstream of Westcliff Colliery recorded a mean of 2.8 EPT families ranging between 0- 5 families whilst downstream a mean of 2.6 was recorded ranging between 1 – 5 (Table 2 and Figure 8).

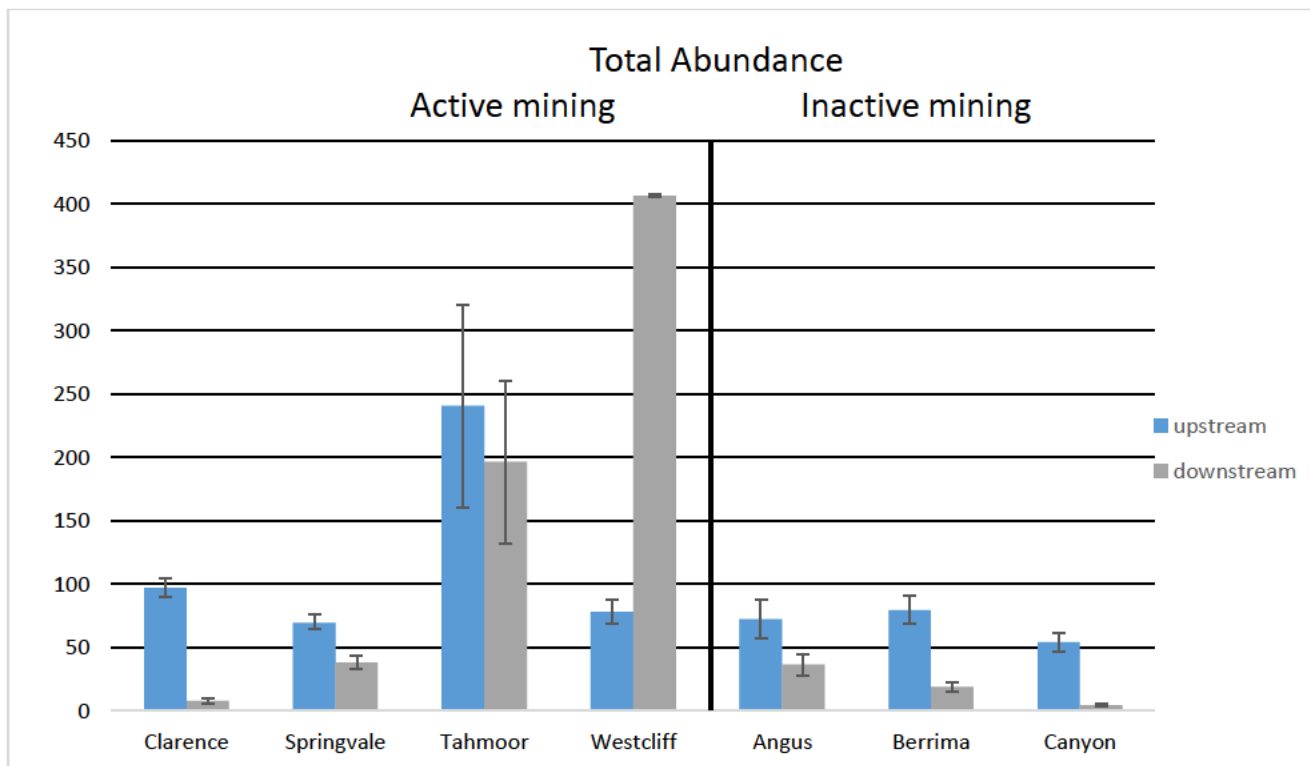


Figure. 4 Macroinvertebrate total abundance. Left columns (Blue) are reference and right columns (Grey) are downstream of each respective coal mines wastewater inflow. Left collieries are actively mining coal (treated wastewater) whilst the right collieries are inactively mining coal (un-treated wastewaters).

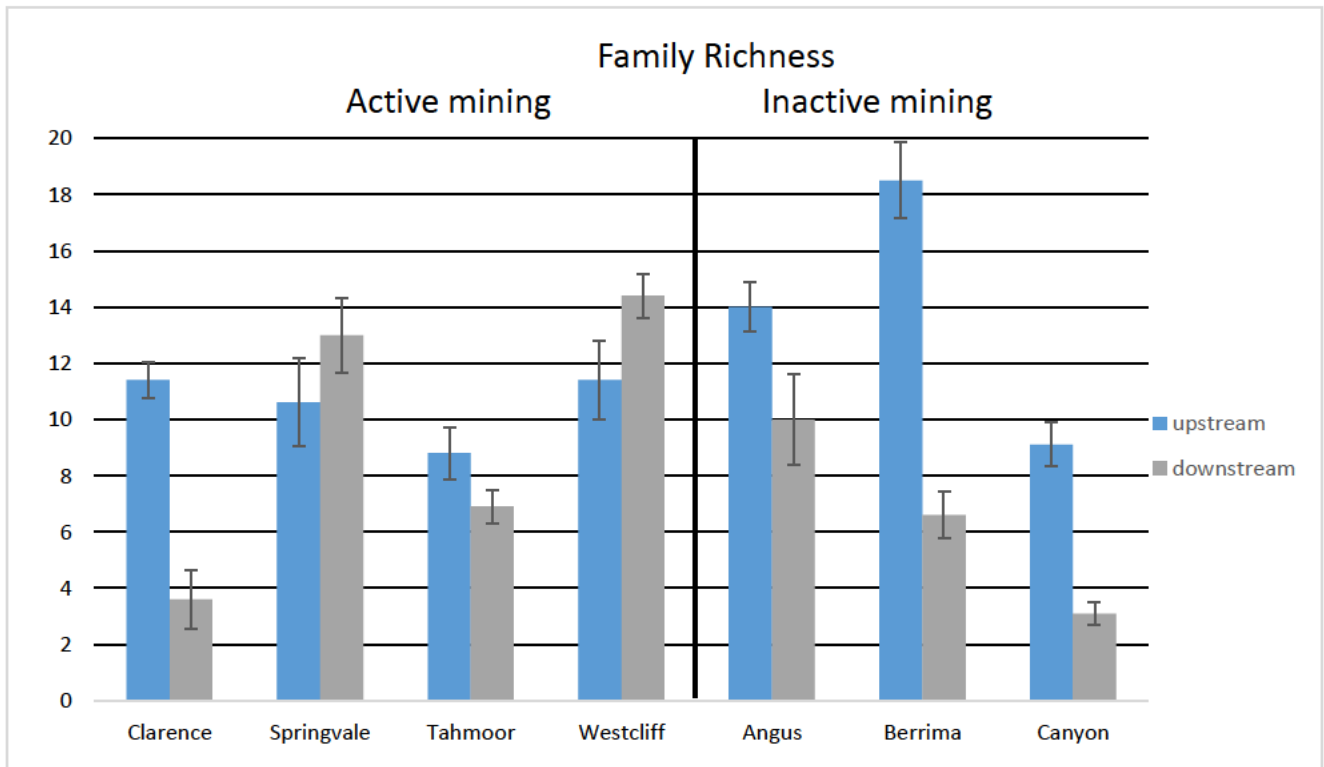


Figure. 5 Macroinvertebrate family richness. Left columns (Blue) are reference and right columns (Grey) are downstream of each respective coal mines wastewater inflow. Left collieries are actively mining coal (treated wastewater) whilst the right collieries are inactively mining coal (un-treated wastewaters).

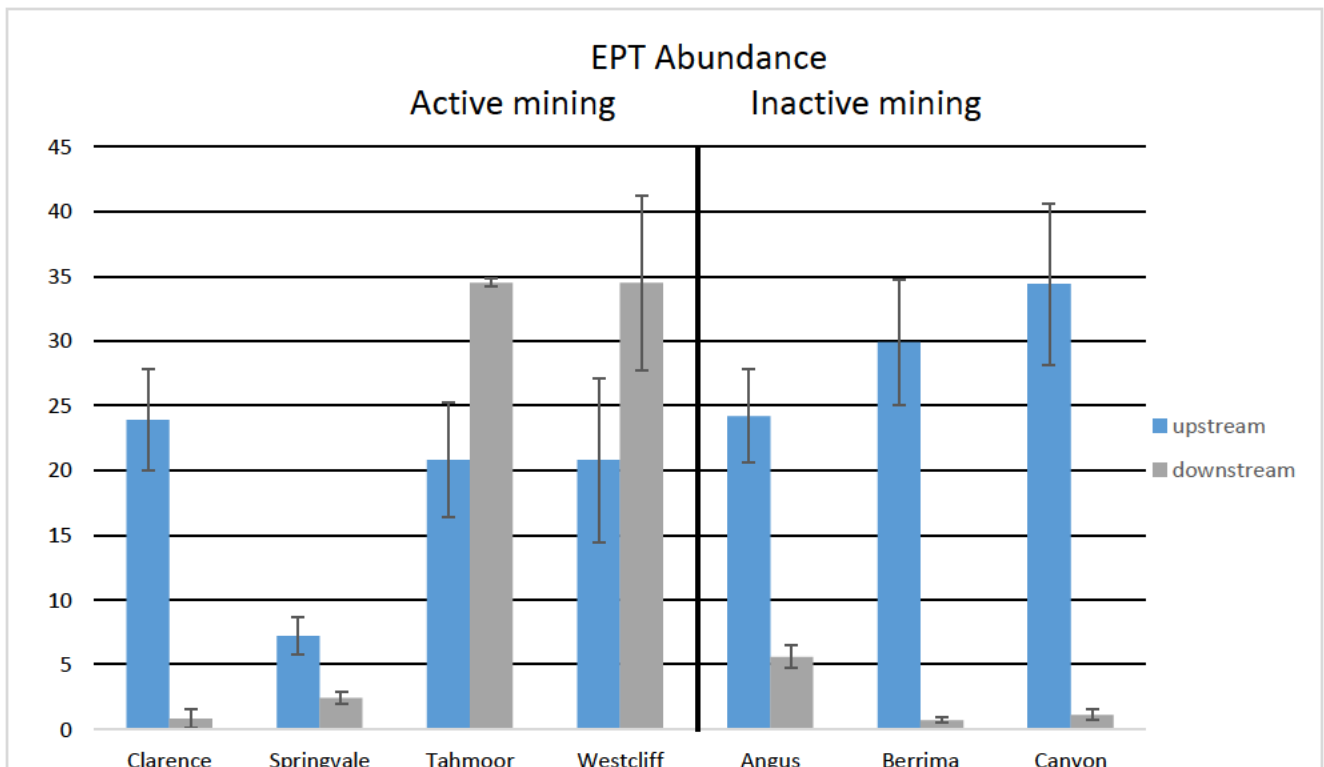


Figure. 6 Macroinvertebrate EPT abundance. Left columns (Blue) are reference and right columns (Grey) are downstream of each respective coal mines wastewater inflow. Left collieries are actively mining coal (treated wastewater) whilst the right collieries are inactively mining coal (un-treated wastewaters).

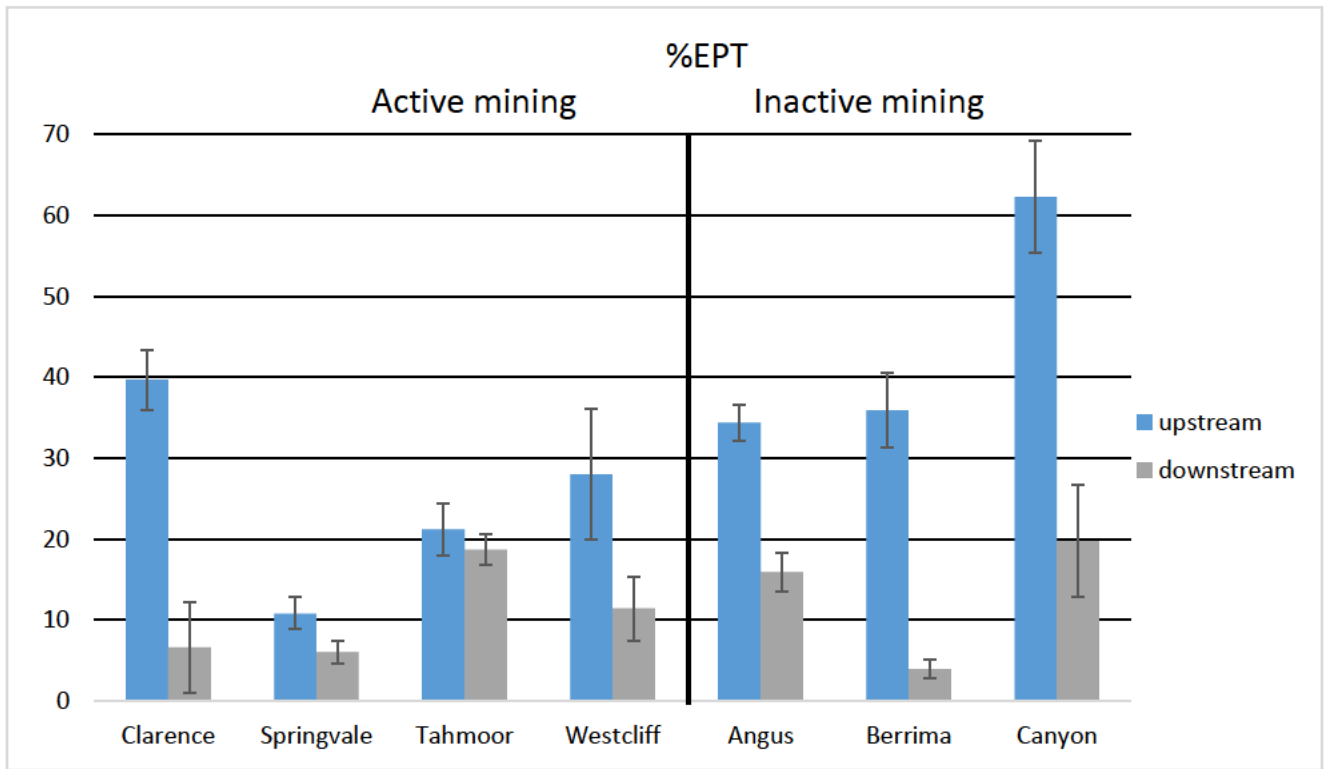


Figure. 7 Macroinvertebrate %EPT. Left columns (Blue) are reference and right columns (Grey) are downstream of each respective coal mines wastewater inflow. Left collieries are actively mining coal (treated wastewater) whilst the right collieries are inactively mining coal (un-treated wastewaters).

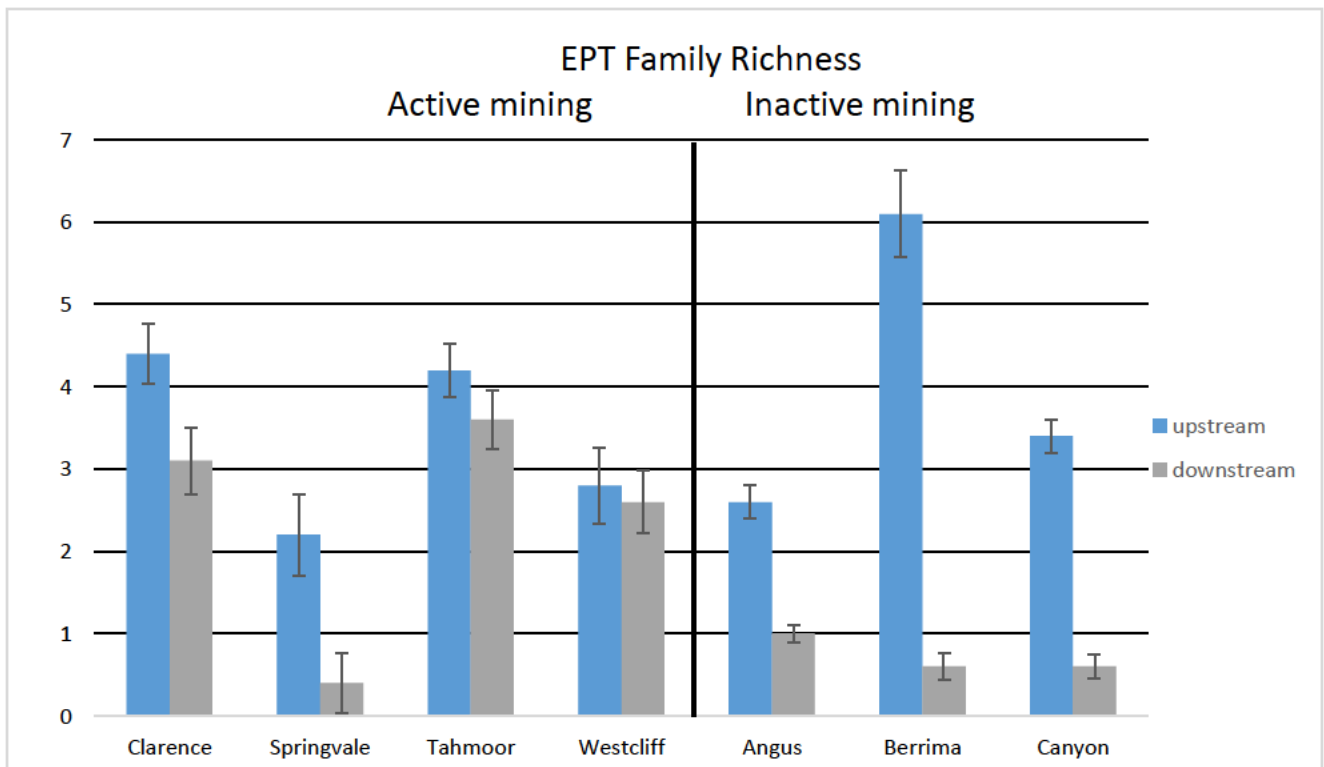


Figure. 8 Macroinvertebrate EPT family richness. Left columns (Blue) are reference and right columns (Grey) are downstream of each respective coal mines wastewater inflow. Left collieries are actively mining coal (treated wastewater) whilst the right collieries are inactively mining coal (un-treated wastewaters).

Table 3 List off macroinvertebrate taxa (order and family).

Order	Family	Impact	Reference
Ephemeroptera	Baetidae	X	X
	Coloburiscidae		X
	Leptophlebiidae	X	X
	Caenidae	X	X
Plecoptera	Eustheniidae		X
	Gripopterygidae	X	X
	Notonemouridae	X	X
Trichoptera	Hydrobiosidae	X	X
	Glossomatidae		X
	Hydroptilidae	X	X
	Hydropsychidae	X	X
	Ecnomidae	X	X
	Conoesucidae		X
	Calocidae		X
	Leptoceridae	X	X
	Philopotamidae	X	X
	Limnephilidae		X
	Helicopsychidae		X
	Philorheithridae		X
	Calamoceratidae		X
	Atriplectididae		X
Coleoptera	Hydraenidae	X	X
	Elmidae	X	X
	Scirtidae	X	X
	Hydrophilidae	X	X
	Curculionidae	X	X
	Gyrinidae	X	X
	Haliplidae		X
	Hydraenidae	X	
	Psephenidae	X	X
Dytiscidae	X		
Hemiptera	Corixidae	X	X
	Gerridae	X	X
	Velidae	X	X
	Notonectidae		X
Diptera	Tipulidae	X	X
	Athericidae		X
	Ceratopogonidae	X	X
	Simuliidae	X	X
	Empididae	X	X
	Chironomidae	X	X
	Dolichopodidae	X	X
	Culicidae	X	
Basommatophora	Bithyniidae	X	X
	Hydrobiidae	X	X
	Planorbidae	X	X
	Physidae	X	X
	Lymnaeidae		X
	Viviparidae		X
Odonata	Aeshnidae	X	X
	Libellulidae	X	
	Gomphidae	X	X
	Diphlebiidae		X

	Corduliidae	X	
Veneroida	Corbiculidae	X	X
	Sphaeriidae	X	X
Decapoda	Atyidae	X	
Megaloptera	Corydalidae	X	X
Neuroptera	Neurorthidae		X
Oligochaeta		X	X
Lepidoptera	Pyralidae	X	X
Collembola		X	X
Tricladida	Dugesidae	X	X
Acarina		X	X
Cladocera			X
Isopoda		X	X

Discussion

Comparisons of the impacts between active and inactive mined coal mines from their coal mine wastewater discharges to the receiving waterways through the use of benthic macroinvertebrates is not well studied. Results of this study show that both active and inactive upstream sample locations showed similarity between each other, other than the Bargo River whilst showing little similarity to active or inactive downstream sample locations. In comparison, active or inactive downstream sample locations shared little similarity with each other. Results suggest that the coal mine wastewaters being discharged are having varying negative impacts to the receiving waterways aquatic ecosystem whether mining of coal is active (treated wastewaters) or inactive (un-treated wastewaters).

The majority of biotic indices recorded at active and inactive mines shows that inactively mined wastewaters are causing a greater impact to the receiving waterways aquatic ecosystem than actively mined wastewaters. With all the inactively mined locations recording statistical differences for all biotic indices when compared between their upstream and downstream sample location (Table 2). This contrasted to the actively mined locations with only one of the five streams sampled recording statistical differences for all biotic indices. Community structure of EPT taxa was also modified with known highly sensitive taxa of the EPT families often being replaced with a less sensitive EPT families at downstream locations (Table 3).

A loss of 18 potential “coal mine wastewater” sensitive taxa was observed from all seven mines. The loss of such individual taxa could lead to the implementation of a coal mine sensitive macroinvertebrate taxa list which could be used as rapid assessment tools for the assessment of coal mine wastewater impacts to their respective receiving waterways (Table 3).

A total of 66 different taxa were recorded at all sample locations (upstream and downstream) the majority of taxa were family level with the remaining order level. 48 total taxa were recorded at all downstream sample locations with a total of 60 being recorded at all upstream sample locations. Of the taxa recorded downstream a total of 6 taxa were not recorded upstream. Taxa were not recorded downstream included; Coloburiscidae, Eustheniidae, Glossomatidae, Conoesucidae, Calocidae, Limnephilidae, Philorheithridae, Calamoceratidae, Atriplectididae, Haliplidae, Notonectidae, Athericidae, Lymnaeidae, Viviparidae, Diphebiidae, Neurorthidae and Cladocera (Table 3 and 4).

BIOENV results (BEST) revealed that the greatest contributing water quality and chemistry parameters influencing the changes in macroinvertebrate community structure between upstream and downstream sample locations were electrical conductivity, lithium, nickel, sulfate and zinc in that respective order. Other contributing parameters though less influential included pH, temperature, calcium, chloride and cobalt.

Total abundance decreased downstream of each respective coal mines wastewater discharge at all sample streams other than one waterway, the Georges River (Westcliff Colliery). The decrease in abundance ranged between 18% and 90% across all the mines. Similar losses in abundance were recorded in the Wollangambe River in a previous study by Belmer et al (2014) with reductions of approximately 90%. Clements et al (2000) recorded similar decreases in total abundance in their study of mining impacts to rivers in the Colorado area of the USA. Georges River (Westcliff Colliery) abundance increased approximately 80% downstream of Westcliff Collieries wastewater inflow. The majority (nearly 50%) of the families that contributes to the increased abundance downstream were Chironomidae and Simuliidae (1797 of the 4065 total impacted sample location macroinvertebrates collected). Showing an increase over five times from reference condition abundance of the two Diptera (fly) families. Average decreases in abundance were reported by Giam et al. (2018) whom used results from eight different studies assessing the impacts from coal mining on stream ecosystem in North America. It was reported that abundance across the mines decreased by 53% (Giam et al 2018).

Family richness decreased below all the coal mines wastewater discharges other than the Georges River (Westcliff Colliery). Declines in family richness for inactive mines was (30, 65 and 65%) whilst active mines recorded smaller declines in family richness (10, 20 and 60%). Similar decreases in family richness have been recorded in the USA by Pond et al (2008) with decreases in the order of 50% recorded in actively mined streams as well as the Colorado Rockies where decreases were significantly lower at mine impacted streams (Clements et al. 2007). The Georges River in contrast recorded an increase from 11.4 families per replicate to 14.4 an increase in family richness of 25%. Giam et al. (2018) reported a 32% decrease in invertebrate richness across eight mines used in their study.

EPT abundance increased downstream of the actively mined Westcliff Colliery which recorded a 70% increase in EPT abundance. This increase was driven by the abundance of the less sensitive mayfly and caddisflies families Canidae, Hydroptillidae and Ecnomidae with downstream abundance recorded as (122, 79 and 79) respectively whilst upstream samples recorded three Canidae, eight Ecnomidae and 9 Hydroptilidae. This is in contrast to the abundance of the more sensitive mayfly Leptophlebiidae with only 42 recorded downstream of Westcliff collieries wastewater inflow compared to 157 collected upstream (Chessman 2003 and Clements et al. 2007).

All other downstream sample locations recorded decreases in EPT abundance with the inactively mined downstream sites showing greater decreases. Inactively mined downstream sample locations recorded 55, 70 and 85% decreases whilst downstream sample locations of actively mined coal mines recorded 10, 50 and 80% decreases in EPT abundance. The greater loss of EPT abundance in this study is similar to those decreases in EPT taxa found in Colorado (Metzeling et al. 2006) and New Zealand (Clements et al. 2007). Decreases in EPT abundance were reported by Clements et al. (2000) in the Rocky Mountains, North America to have decreased by 68% in streams polluted by heavy metals (Metzeling et al. 2006). A comparatively smaller decline in EPT abundance was found in a study of

West Virginia streams impacted by coal mines (Pond et al. 2008) with the proportion of macroinvertebrates in EPT groups at unmined streams of 77.9% compared to 51.1% at mined streams.

EPT% decreased at all downstream sample locations other than Sawyers Swamp downstream of the waste inflow from the active Springvale Colliery. This increase of nearly 50% was mostly driven by the abundance of only two taxa the mayfly Baetidae and the caddisfly Hydroptillidae, whilst in comparison neither of the two EPT taxa were recorded at the paired upstream site. Mayflies and caddisflies were recorded upstream though the family structure was dominated by the much more sensitive mayfly Leptophlebiidae and the more sensitive caddisflies, Hydroboisidae, Philopotamidae and Calamoceratidae (Chessman 2003 and Clements et al. 2007). Decreases in Ephemeroptera% from mined sites to unmined sites were recorded by Pond et al. (2008) and Pond (2010). Pond et al. (2008) found decreases in the taxa Ephemeroptera from 45.6% of samples to 7.4% of samples from West Virginian mines which was similar to mines in the Kentucky region which found 51.5% in comparison to <6% downstream (Pond 2010). All the inactive mines recorded greater decreases in EPT% in comparison to actively mined downstream sample locations. Inactive mines recorded decreases of 55, 70 and 80% whilst downstream of the actively mining operations decreases of 10, 50 and 80% were recorded. Similar reductions in Ephemeroptera were recorded in Acid Mine Drainage effected streams in the River Avoca (Ireland) with upstream Ephemeroptera recording 43.8% of samples whilst downstream of the Acid Mine Drainage inflow only 5.1% of samples recorded Ephemeroptera taxa (Clements 2000).

EPT Family Richness decreased at all downstream sample locations though this was only statistically significantly different at one of the four active mines whilst all three inactive mines downstream samples were statistically significantly different. Active mines recorded decreases from upstream to downstream samples of 7%, 14%, 30%, 66% and 88% Tahmoor, Westcliff, Clarence, Springvale Collieries respectively (Springvale Creek) (Sawyers Swamp), whilst in comparison inactive mines recorded decreases of 61%, 90% and 82% (Angus Place, Berrima (Medway) and Canyon Collieries respectively).

Conclusions

Results of this study show that the coal mine wastewaters discharged by all of the seven mines used in this study are having varying degrading impacts on their respective receiving waterways ecosystem. Whilst coal is still being actively mined water treatment processes of varying degrees to remove or reduce pollutants within the discharged wastewaters is occurring. This is not the case for the mines inactively mining ore in this study. At the stage of mine closure and the subsequent relinquishment of the wastewater discharge licence the water treatment process ceases. This allows for groundwater to accumulate in the underground workings, eventually making its way through adits or discharge points back to the surface and into the original receiving waterway.

This untreated mine wastewater has higher concentrations of heavy metals and other contaminants, due to the Acid Mine Drainage process, than that of the actively mined treated wastewaters. This is of major concern as the impacts to the receiving waterways ecosystem at actively mined, licenced and regulated waterways is significant let alone once the mining operation is completed and water treatment ceases. Giam et al. (2018) found similar results for their study using eight mines in North America. Giam et al. (2018) also found slightly greater decreases in reclaimed mine sites in comparison to actively mined sites. Results show a slight decrease of 32% in taxa richness from actively mined sites

in comparison to a decrease of 34% in reclaimed mine sites. Abundance recorded greater decreases from actively mined sites (53% decrease) whilst reclaimed mine sites recorded a decrease of 68% (Giam et al. 2018).

This research has allowed for a greater understanding of the failings of the NSW EPA to protect the aquatic environment through legislation and the regulation of contaminants within coal mine wastewaters in the Sydney Basin. At the active mines, large losses of biota have been observed, whilst the environmental protection licence is still in place, to ensure the receiving waterways ecosystem is protected. Measures to better protect waterways which receive untreated coal mine wastewaters must be undertaken by the NSW EPA to ensure that once coal is no longer mined the receiving aquatic ecosystem is still protected.

Research significance

The chapter's research contributes to the rigour of scientific testing of water chemistry and river ecology for Australian underground coal mines. Whilst much of the literature suggests that coal mining activities heavily degrade aquatic ecosystems, there have been very few Australian studies that have examined this. Information is particularly scant on the on-going impacts of coal mine impacts to rivers whilst in operation and tentative literature suggests this degradation continues well beyond the time mining practices cease. This research directly addresses this gap in the literature.

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Chapter 7

Regulated coal mine wastewater discharge contaminants accumulating in an aquatic predatory beetle (*Macrogyrus rivularis*) Wollangambe River, Blue Mountains New South Wales Australia.

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Abstract

This study investigates contaminants from a single coal mine wastewater discharge released to the Wollangambe River accumulating in an aquatic predatory beetle (*Macrogyrus rivularis*). The study was undertaken within the Wollangambe River and its surrounding tributaries. The coal mine wastewater discharge is regulated by the New South Wales Environment Protection Authority and the regulation of the pollutants only concentrates on water column concentrations. The majority of the Wollangambe River flows within the World Heritage Greater Blue Mountains National Park and is protected through many layers of legislation from state to federal and international (Threatened Species Conservation Act 2005, Environment Protection and Biodiversity Conservation ACT 1999, United Nations Educational, Scientific and Cultural Organization 2000). Results show that many contaminants are at statistically higher concentrations within the water column, stream sediment and beetles sampled when compared between reference and impacted sample locations. Analysis of Similarity (ANOSIM) found significant differences for contaminants in beetles sampled at impacted sites compared to reference sites with no significant difference recorded between reference sites. Biota and/or Environmental matching (Best) found Manganese, Cobalt, Nickel and Zinc as the factors which have the greatest influence in differences. The implications that contaminants from the regulated wastewater being discharged may be accumulating within aquatic biota is of major concern as the regulation of the wastewater only concentrates on water column pollutants and is not taking into account the greater environmental ramifications of the pollution.

Keywords

Bioaccumulation, heavy metal bioaccumulation, aquatic macroinvertebrates, coal mine wastewater, Blue Mountains National Park, Wollangambe River.

Introduction

In recent years anthropogenic activities and the subsequent contamination from pollutants of global ecosystems has reached unparalleled heights. Exponentially increasing industrial and mining activities have led to further environmental pollution through wastes produced by these activities. A host of pollutants from mining and industrial activities, when released to the environment, have the potential to accumulate within biota at toxic concentrations and cause chronic ecological impacts in ecosystem food chains (Sericano et al. 1995, Wisloka et al. 2004, Ashraf et al. 2011 and Nawab et al. 2015). Metal pollution is a global environmental issue and has been for many decades. In many cases metal pollutants are directly discharged into waterways from anthropogenic activities. Many of these metal pollutants become absorbed to suspended particulates within the water column. Once absorbed to these suspended particulates the metal contaminated suspended particulates eventually deposit into waterway sediments often remaining indefinitely (Neff 1981, Wand and Rainbow 2008 and Kolaříková et al. 2012).

Aquatic ecosystems are often more highly sensitive to contamination from anthropogenic activities, especially as waterways are often used as sources of discharge for many industrial wastes (Allen et al. 1993). Heavy metal contamination within aquatic environments can persist much longer than terrestrial organic pollutants. This is due to the lack of a “biodegradation function” of heavy metals in aquatic ecosystems in comparison to a terrestrial ecosystem (Twining et al. 2008 and Ashraf et al. 2011). Bioaccumulation of contaminants within fish species have been recorded worldwide as a result of metal and coal mining activities (Ashraf et al. 2011, Bharati and Banerjee 2011, Miller et al. 2013 and Ajima et al. 2015) along with other aquatic fauna and flora (Hill et al. 2000 and Amisah et al. 2008).

One study found bioaccumulation of arsenic and selenium within caddisflies of coal ash polluted sites (Reash et al. 2006) whilst increases in the levels of pollutants were also recorded for turtles, crayfish, tadpoles and varying fish species by (Otter et al. 2012). Bioaccumulation of wastewater contaminants within macroinvertebrates from metal mining activities have been reported in Canada, Australia and North America (Peterson et al. 2002, Maret et al. 2003, Cain et al. 2004, Telford et al. 2009, Swansburg et al. 2002 and Jasonsmith et al. 2008).

The pH of water is identified as an important factor directly linked to the speciation and bioaccumulation of metals and metalloids. pH will affect the bioavailability of metals and metalloids by influencing their solubility and subsequent ability of bioaccumulate to a medium (Otter et al. 2012 and Durães et al. 2014). The individual constituents of coal can also influence metal bioavailability (Otter et al. 2012). Atkinson et al. (2007) found that lower water pH allowed for greater bioavailability and sequestration of heavy metals in biota. They found that iron and manganese oxidatively precipitated increasingly as pH decreased. This decreasing pH had a secondary effect which allowed a greater sequestration rate of lead and zinc (Atkinson et al. 2007).

Coal mining practices are well documented to contribute to an array of differing environmental problems including air pollution, fire hazards, ground subsidence or deformation, surface and or ground water pollution. Surface water pollution is a major environmental problem associated with coal mining and it occurs through the discharge of mine wastewaters that are contaminated by various disturbances associated with mining practices (Banks et al. 1997, Jarvis and Younger 1997, Brake et al. 2001, Johnson 2003 and Pond et al. 2008). Water pollution from coal mining occurs as large volumes of surface and groundwater are required to be removed from most underground coal mines. This is generally through the pumping of the wastewater to the surface. Without this, groundwater would flood most sections

of the underground mining operation (Jarvis and Younger 1997 and Younger 2004). Coal mine wastewater will often be contaminated due to the disturbance of the local geology associated with mining activities. The exact nature of the water contamination will vary depending on local factors such as groundwater geochemistry, hydrology and mineralogy of the local strata. In addition to the physical activity of the mining operation and the removal of the wastewater, other activities will also often contaminate water used throughout a mining plant which can include; coal washing and the inclusion of other wastes generated by the surface operation at the mine such as sewage wastes (Younger 2004).

Coal mine wastewater discharges in New South Wales, Australia are regulated by the New South Wales Environmental Protection Authority (NSW EPA) and environmental protection of receiving waterways is implemented through Environmental Protection Licences (EPL's), under the Protection of the Environment Operations Act 1997 (POEO Act 1997) (Belmer et al. 2014). EPL's set discharge limits for water quality and chemical properties in which coal mine wastewaters that are discharged to the environment must adhere to (Wright 2012 and Belmer et al. 2014). In many cases the EPL's for coal mine wastewater discharges are failing to protect the receiving waterways ecosystems by failing to identify ecologically hazardous chemicals in the waste discharges that may accumulate or become persistent in the receiving waterways ecosystem. This non-wholistic regulation is failing to protect the aquatic environment of these receiving waterways by not taking into account legacy pollutants. Studies have been conducted on the impacts of metal bioaccumulation from coal mining activities on terrestrial and aquatic plant and fish tissue with none investigating the bioaccumulation of pollutants from actively licenced and regulated coal mine wastewater discharges.

This study investigates if the contaminants from a single licenced and regulated coal mine wastewater discharge is accumulating in the ambient environment. More specifically within stream sediments and a single species of aquatic predatory beetle (*Macrogyrus rivularis*) from the family Gyrinidae within the high conservation Wollangambe River. Macroinvertebrates are widely used as ecological indicators of water quality due to well-established methods and ease of sampling (Hellowell 1986). Macroinvertebrates are regarded as being effective indicators as they have relatively long-life cycles and different taxonomic groups have differing sensitivities to disturbance and water pollution (Hellowell 1986 and Rosenberg and Resh 1993). There is a large amount of well-established literature on freshwater macroinvertebrates and their applications to assess various human-induced anthropogenic disturbances including a broad variety of pollution types (Rosenberg and Resh 1993) including coal mine drainage (Jarvis and Younger 1997, Pond et al. 2008, Battaglia et al. 2005 and Wright, I.A. & Burgin, S. 2009a).

The beetle species (*Macrogyrus rivularis*) was used as it was identified as a reliable source of sampling, its predacious habit allowing it to consume other aquatic fauna which may be contaminated and due to its long-life cycle (2 years) allowing for a prolonged period in which it can accumulate contaminants from the wastewater (Watts and Hamon 2010). This stream also allows for a great test case as other than the coal mine wastewater the stream resides within an untouched pristine catchment with the two reference locations having no anthropogenic influence on water quality or chemistry (Belmer et al. 2014 and Wright et al. 2017).

It is hypothesised that the contaminants from the single coal mine wastewater are accumulating within a single species of predacious aquatic beetle found within the Wollangambe River downstream of the coal mine wastewater discharge.

Methods

This study was conducted at two upland streams found within the Blue Mountains area of Sydney, Australia. One stream being the Wollangambe River, the other Bell Creek which is a tributary of the Wollangambe River. Both of which flow mostly within the Blue Mountains World Heritage National Park Estate and the majority of their catchments are naturally vegetated (Figure 1). Four sample sites were used in total for this study, Wollangambe 1 (W1) and Bell Creek (Bell) both as reference sites and Wollangambe 3 (W3) and Wollanagmbe 5 (W5) as impact sites. The reference site (W1) is located approximately 200m above the discharge point and is a first order stream, whilst (W3) is approximately 500m downstream and is a second order stream. Both sample locations share similar altitudes between 1025m and 960m above sea level (Figure 1 and Table 1). Two sample locations were located approximately 18km downstream of the coal mine. One being the reference site (Bell Creek) and is a naturally vegetated tributary of the Wollangambe River and is a first order stream. Bell Creeks paired impact site (W5) was located on the Wollangambe River and was approximately 200m downstream of the confluence of Bell Creek and the Wollangambe River and is a second order stream. Both lower sample sites share similar altitudes between 760m and 740m above sea level (Figure 1 and Table 1). Stream order level was determined by the Strahler 1952 method (Strahler 1952).

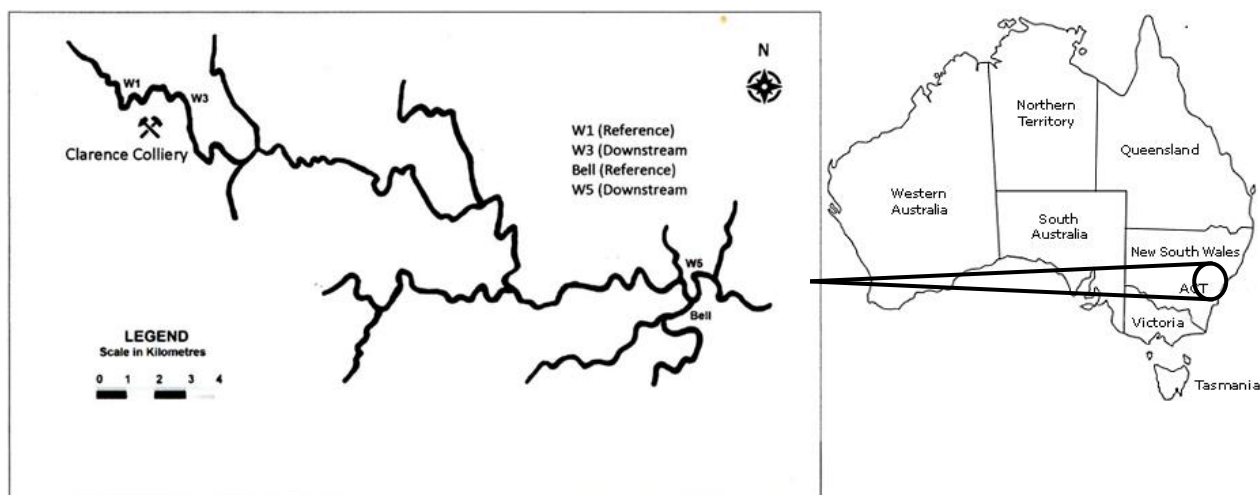


Figure 1 Map of Australia and sample locations and Clarence colliery. W1 (reference) sample location is located approximately 200m above the coal mine wastewater discharge and its paired (impact) sample location W3 is located approximately 200m downstream of the wastewater inflow. W5 (impact) sample location is located on the Wollangambe River approximately 18m downstream of the wastewater inflow and its paired (reference) sample location Bell Creek (Bell) is a clean tributary of the Wollangambe River and is also located approximately 18km downstream of the coal mine wastewater discharge just upstream of W5.

Table 1 Sample location name, longitude and latitude, altitude (Metres above sea level) and stream order of the four sample locations used in this study. Stream order is derived from the Strahler method (Strahler 1952).

Sample location	longitude	latitude	Altitude (ASL)	Stream order
W1 (reference)	150.249101 E	-33.455964 S	1025 m	1
Bell Creek (reference)	150.353770 E	-33.490335 S	760 m	1
W3 (downstream)	150.257359 E	-33.455673 S	960 m	2
W5 (downstream)	150.355208 E	-33.487474 S	740 m	2

Five replicated water samples (grab samples) and insitu field water quality parameters were sampled and measured at the same time as sediment and beetle sampling occurred from the same four sample locations above. Field meters used include a TPS WP-82Y meter with a YSI dissolved oxygen probe for Dissolved Oxygen, TPS WP-88 Turbidity meter with a TPS turbidity sensor for Turbidity and a TPS WP-81 Conductivity, pH and Temperature meter with TPS Conductivity and Temperature probe and a TPS submersible k407 pH sensor. Five field grab samples were collected in commercial preserved sample containers provided by a commercial laboratory (EnviroLab) (to allow for quality control of sampled water) at each site (20 total) and analysed using standard methods (APHA 1998) by EnviroLab (Chatswood, NSW) a National Associations of Testing Authorities accredited laboratory for fifteen metals (Aluminium, Barium, Boron, Cadmium, Chromium, Cobalt, Copper, Iron, Lead, Manganese, Molybdenum, Nickel, Strontium, Uranium and Zinc).

Stream sediments were sampled at the same time as water and beetle samples were collected from the same four sample locations. This was achieved by sampling stream sediments within unused commercial laboratory sample containers provided by (EnviroLab) (to allow for quality control of sampled sediments) in a zone of accumulated sediment, following standard methods recommended by the Victorian Environmental Protection Authority [41]. Samples were placed into sealed glass sampling jars supplied by a commercial laboratory and stored. Five samples were collected at each sample location (20 total) and analysed using standard methods (APHA 1998) by EnviroLab (Chatswood, NSW) a National Associations of Testing Authorities accredited laboratory for the same fifteen metals analysed for water samples (Aluminium, Barium, Boron, Cadmium, Chromium, Cobalt, Copper, Iron, Lead, Manganese, Molybdenum, Nickel, Strontium, Uranium and Zinc).

Six replicated predacious aquatic beetle samples (*Macrogyrus rivularis*) from the family Gyridae were collected on one occasion at all four sample sites (24 total beetles from 4 sites). The beetle species was used as it was identified as a reliable source of sampling, its predacious habit allowing it to consume other aquatic fauna which may be contaminated and due to its long-life cycle (2 years) (Watts and Hamon 2010). Sampling was achieved by identifying and catching beetles from the surface or mid water column as they retreated using a macroinvertebrate sampling 'kick' net with a frame of 30 x 30 cm and 250 µm mesh. Beetles were caught and stored in deionised water within individual sample location sample containers. These containers were sealed and labelled on collection and dispatched to the commercial laboratory (EnviroLab) and analysed for 21 metals (Aluminium, Arsenic, Barium, Beryllium, Boron, Cadmium, Chromium, Cobalt, Copper, Iron, Lead, Manganese, Molybdenum, Nickel, Selenium, Silver, Strontium, Titanium, Uranium, Vanadium and Zinc).

In the laboratory Ultra High Purity (UHP) water was used to wash the beetles and remove any potentially contaminated residues from their outer surface (precipitates). They were then kept in individual sealed plastic containers. The samples were dried in a 60-degree Celsius oven for 48 hours prior to being ground using a clean pestle and mortar. Approximately 0.05 grams of the ground samples were weighed into plastic digestion tubes. High purity Nitric and Hydrochloric acid was added to the tubes and the samples were digested at 98 Degrees Celsius for 1.5

hours. The samples were then made up to 20mL with UHP water. These samples were analysed by Inductively Coupled Mass Spectrometry (ICP-MS). Quality control measures were used to ensure the integrity of the results including blanks and spiked laboratory control samples (LCS).

Multivariate analysis was performed using Single Factor ANOVA and the software package Primer 6 for analysis of similarity (ANOSIM) to test for differences between combined reference sites (W1 and Bell) and both individual impact sites (W3 and W5). Water quality and chemistry, stream sediment chemistry and aquatic predacious beetle chemistry individually. Primer 6 was also used to identify which water quality and chemistry parameters influenced the dissimilarity in beetle contaminants through a BIOENV test. Percentage increases from water column concentrations ($\mu\text{g/L}$) to aquatic beetle concentrations ($\mu\text{g/kg}$) were calculated to assess potential uptake differences between reference water chemistry to impacted water chemistry.

Results

Mean water column concentrations of Barium, Boron, Cobalt, Iron, Manganese, Nickel, Strontium and Zinc were found to be statistically different from reference to impacted sample sites (Table 2). Reference sites recorded mean barium of 9.5 and 8.6 $\mu\text{g/L}$ whilst in comparison downstream sites recorded mean barium of 20.8 and 16.0 $\mu\text{g/L}$. Reference sites recorded mean boron of 8.0 $\mu\text{g/L}$ in comparison downstream sites had mean boron of 9.1 and 9.5 $\mu\text{g/L}$. Reference site Cobalt concentrations were below laboratory detectable limits. Whilst in comparison downstream sites recorded mean cobalt of 26.5 and 1.7 $\mu\text{g/L}$. Mean iron concentrations at reference sites was 300 and 381.7 $\mu\text{g/L}$ whilst in comparison downstream iron was of 134.3 and 66.8 $\mu\text{g/L}$. Reference sites recorded mean manganese concentrations of 22.8 and 35.2 increasing downstream to a mean of 178.5 and 29.2 $\mu\text{g/L}$. Reference sites recorded mean nickel of 0.5 $\mu\text{g/L}$ just above the laboratory threshold. Whilst in comparison downstream mean nickel was 76.7 and 17.7 $\mu\text{g/L}$. Reference strontium concentrations were recorded a mean of 3.6 and 5.2 $\mu\text{g/L}$ increasing downstream to 59 and 36.2 $\mu\text{g/L}$. Reference sites recorded mean zinc of 2.4 and 3.7 $\mu\text{g/L}$ and downstream sites a mean of 105 and 20.8 $\mu\text{g/L}$. Cadmium, chromium, copper, lead, molybdenum and uranium were all at concentrations within water that were below laboratory detectable limits (Table 2).

Mean stream sediment concentrations of Aluminium, Barium, Cadmium, Chromium, Cobalt, Copper, Iron, Manganese, Molybdenum, Nickel, Strontium, Uranium and Zinc were found to be statistically different from reference to impacted sample sites (Table 3). Bell Creek (reference) recorded the lowest mean aluminium in stream sediment of 233.3 mg/kg, whilst in comparison W1 (reference site), W3 and W5 (impacted sites) had mean aluminium of 1248, 5083 and 1034 mg/kg respectively. Reference sites recorded mean barium of 4.75 and 18.0 mg/kg whilst downstream recorded mean barium of 101.6 and 7.5 mg/kg. Both reference sites Bell Creek and W1 as well as the downstream site W5 all recorded cadmium concentrations lower than laboratory detectable limits. W3 recorded mean cadmium of 0.47 mg/kg. Bell Creek, W1 and W5 all recorded replicates which were below laboratory detectable concentrations for chromium. W1 and W5 both recorded mean chromium of 1 mg/kg whilst Bell Creek recorded a mean of 0.25 mg/kg. In comparison W3 recorded chromium concentrations between 2-8 mg/kg and a mean of 4 mg/kg. The reference site Bell Creek recorded below detectable laboratory concentrations of cobalt for all replicates

sampled. W1 ranged between below laboratory concentrations and 5 mg/kg, whilst in comparison W5 was found to have mean cobalt of 6 mg/kg and W3 recorded mean concentrations of 552 mg/kg. The reference sites Bell Creek recorded below detectable laboratory concentrations of copper for all replicates sampled and W1 recorded a mean of 3.5 mg/kg. Whilst the downstream sites W3 and W5 recorded mean copper of 17.0 and 2.4 mg/kg respectively (Table 3).

Bell Creek and W1 recorded mean iron of 1443 and 4000 mg/kg whilst in comparison W3 and W5 recorded mean iron of 9950 and 1982 mg/kg. Manganese recorded at the reference sites (Bell and W1) was mean 13.3 and 127.5 mg/kg in comparison W3 and W5 recorded mean manganese of 5474 and 76 mg/kg respectively. Bell Creek, W1 (reference sites) and W5 (impacted site) samples recorded concentrations of molybdenum within sediment below laboratory detectable limits. Whilst in comparison W3 (downstream site) had mean molybdenum of 4 mg/kg. W3 recorded one replicate below laboratory detectable limits for molybdenum. Bell Creek recorded mean nickel below laboratory detectable limits whilst W1 (reference sites) recorded mean nickel of 2 mg/kg. In comparison W3 and W5 (downstream sites) recorded mean nickel of 606.7 and 19 mg/kg with all samples recording detectable concentrations. Reference site mean strontium was found to be 1.25 and 5 mg/kg and ranged from below laboratory detectable limits and 9 mg/kg across both sites. Whilst in comparison W3 and W5 (downstream sites) recorded mean strontium of 12.3 and 2.8 mg/kg respectively. All sample locations recorded low concentrations of uranium in sediment with the reference stream Bell Creek recording all replicates at concentrations below laboratory detectable limits. W1 was recorded between below laboratory limits and 0.2 mg/kg with a mean of 0.06 mg/kg. W5 was similar to both reference uranium concentrations measuring between below laboratory detectable limits and 0.3 mg/kg with a mean of 0.08 mg/kg. Whilst in comparison W3 mean uranium of 0.53 mg/kg and ranged between 0.1-1.4 mg/kg. The reference site Bell Creek recorded all replicates at concentrations below laboratory detectable limits for Zinc. W1 was found to have a mean of 1.75 mg/kg. Whilst in comparison W3 and W5 (downstream sites) had mean zinc of 734 and 25 mg/kg respectively (Table 3).

Table 2 Water quality and water chemistry parameters, sample locations, Single Factor ANOVA, F value, p value and degrees of freedom (df) between groups indicates statistical difference between merged upstream (reference) and individual downstream (W3 and W5) sample locations, range and mean for water chemistry for all four sample locations. All water chemistry data is measured in µg/L, pH in pH units, Electrical Conductivity in µs/cm, Dissolved Oxygen in % saturation, Turbidity in Nephelometric Turbidity Units and Temperature in degrees Celsius. BD = below laboratory detectable limits, n/a = not any.

Water	Statistics			Reference Sites						Downstream Mine Sites					
	F value	p value	df	W1 Range	W1 Mean	W1 Median	Bell Range	Bell Mean	Bell Median	W3 Range	W3 Mean	W3 Median	W5 Range	W5 Mean	W5 Median
pH	1544	<0.01	2	5.04-5.06	5.05	5.04	5.17-5.20	5.18	5.20	7.27-7.37	7.29	7.35	6.97-6.98	6.97	6.97
Electrical Conductivity	11239	<0.01	2	25.7-25.7	25.7	25.7	27.7-27.7	27.7	27.7	316-316	316	316	217.8-217.9	217.9	217.9
Dissolved Oxygen	0.28	0.75	2	90.4-90.4	90.4	90.4	89.1-89.9	89.6	89.6	92.4-92.5	92.4	91.2	89.6-90.1	89.9	89.8
Turbidity	6.29	<0.01	2	0.1-1.3	0.5	2.2	0.1-1.2	0.6	0.9	0.5-0.7	0.62	3.4	0.4-1.2	0.78	0.9
Water Temperature	10.7	<0.01	2	11.2-11.2	11.2	11.2	10.2-10.2	10.2	10.2	12.4-12.4	12.4	12.4	10.8-10.8	10.8	10.8
Aluminium	4.05	<0.05	2	30-330	178.3	175	40-70	54	50	BD-210	71.7	50	BD-40	21.7	25
Barium	65.3	<0.01	2	8-11	9.5	9.5	8-9	8.6	9	19-23	20.8	20.5	14-19	16	15
Boron	31.6	<0.01	2	7-9	8.0	8	8-8	8.0	8	1-9	9.5	9.5	9-10	9.17	9
Cadmium	n/a	n/a	2	BD	BD	BD	BD	BD	BD	BD	BD	BD	BD	BD	BD
Chromium	n/a	n/a	2	BD	BD	BD	BD	BD	BD	BD	BD	BD	BD	BD	BD
Cobalt	5.57	<0.01	2	BD	BD	BD	BD	BD	BD	3-47	26.5	28	BD-3	1.67	2
Copper	n/a	n/a	2	BD	BD	BD	BD	BD	BD	BD	BD	BD	BD	BD	BD
Iron	18.7	<0.01	2	210-550	381.7	390	140-410	300	375	38-420	134.3	87.5	42-82	66.8	77
Lead	n/a	n/a	2	BD	BD	BD	BD	BD	BD	BD	BD	BD	BD	BD	BD
Manganese	4.24	<0.05	2	14-56	35.2	35	20-24	22.8	23	43-430	178.5	200	22-34	29.2	22
Molybdenum	n/a	n/a	2	BD	BD	BD	BD	BD	BD	BD	BD	BD	BD	BD	BD
Nickel	11.3	<0.01	2	BD-1	0.5	0.5	BD	BD	BD	24-130	76.7	70	8-25	17.7	20
Strontium	88.6	<0.01	2	4.3-6.3	5.22	5.1	3.4-3.8	3.58	3.6	45-68	59	59	28-47	36.2	34
Uranium	n/a	n/a	2	BD	BD	BD	BD	BD	BD	BD	BD	BD	BD	BD	BD
Zinc	7.50	<0.01	2	1-6	3.67	3.5	2-3	2.4	2	21-190	105	99	8-31	20.8	24.5

Table 3 Sediment chemistry parameters, sample locations, Single Factor ANOVA, F value, p value and degrees of freedom (df) between groups indicates statistical difference between merged upstream (reference) and individual downstream (W3 and W5) sample locations range and mean for river sediment for all four sample locations. All river sediment chemistry data is measured in mg/kg. BD = below laboratory detectable limits.

Sediment	Statistics			Reference Sites						Downstream Mine Sites					
	Parameter/Sample location	F value	p value	df	W1 Range	W1 Mean	W1 Median	Bell Range	Bell Mean	Bell Median	W3 Range	W3 Mean	W3 Median	W5 Range	W5 Mean
Aluminium	9.52	<0.01	2	790-2200	1247.5	1000	190-280	233.3	230	1500-8700	5083.3	5200	230-2200	1034	310
Barium	16.1	<0.01	2	10-33	18	14.5	3-8	4.75	4	37-170	101.6	88	2-22	7.5	3
Boron	103.9	<0.01	2	BD	BD	BD	BD	BD	BD	BD-6	2	BD	BD	BD	BD
Cadmium	10.2	<0.01	2	BD	BD	BD	BD	BD	BD	BD-1	0.47	0.5	BD	BD	BD
Chromium	9.99	<0.01	2	BD-2	1.0	1	BD-1	0.25	BD	2-8	4.0	2	BD-3	1.0	BD
Cobalt	18.4	<0.01	2	BD-5	2.0	1.5	BD	BD	BD	61-1000	551.8	635	BD-23	6.0	0.5
Copper	9.59	<0.01	2	2-6	3.5	3	BD	BD	BD	8-29	17.0	15.5	BD-7	2.4	BD
Iron	8.39	<0.01	2	2300-7800	4000	2950	670-2500	1442.5	1300	2400-19000	9950	10350	490-3900	1982	1100
Lead	3.02	0.09	2	3-8	4.5	3.5	BD-2	0.5	BD	5-17	8.4	6	BD-6	2.0	BD
Manganese	36.4	<0.01	2	38-300	127.5	86	7-22	13.3	12	270-9300	5474	6900	3-220	76.0	11
Molybdenum	10.6	<0.01	2	BD	BD	BD	BD	BD	BD	BD-9	4	4	BD	BD	BD
Nickel	20.8	<0.01	2	1-4	2	1.5	BD	BD	BD	160-1100	606.7	630	2-67	19.0	2
Strontium	1.19	0.3	2	3-9	5.0	4	BD-4	1.25	0.5	3-27	12.3	9	BD-8	2.8	0.5
Uranium	19.7	<0.01	2	BD-0.2	0.06	BD	BD	BD	BD	0.1-1.4	0.53	0.4	BD-0.3	0.08	BD
Zinc	27.5	<0.01	2	1-3	1.75	1.5	BD	BD	BD	300-1100	734	760	2-97	25.0	2

Table 4 Beetle chemistry parameters, sample locations, Single Factor ANOVA, F value, p value and degrees of freedom (df) between groups indicates statistical difference between merged upstream (reference) and individual downstream (W3 and W5) sample locations, range, mean for all four sample locations. Percentage increase from W3 water chemistry contaminants to W3 beetle chemistry contaminants are offered in the last column. All beetle data is measured at µg/kg. BD = below laboratory detectable limits, n/a = not any.

Beetle	Statistics			Reference Sites						Downstream Mine Sites						Mean % Increase
	F value	p value	df	W1 Range	W1 Mean	W1 Median	Bell Range	Bell Mean	Bell Median	W3 Range	W3 Mean	W3 Median	W5 Range	W5 Mean	W3 Median	
Aluminium	3.04	0.07	2	3713-8184	6023.3	6496	3963.4-7349.8	4990.8	4578	5624.9-63126	18167.3	11582	5003.3-14091	9227.8	8976	253.4
Arsenic	1.56	0.23	2	177-496.2	501.2	514	260-745.9	318.8	326.1	346.7-584	474.0	486.5	453.2-867.3	541.0	485.3	n/a
Barium	0.99	0.38	2	177.8-908.2	479.9	394.4	205.8-724.2	405.9	339.7	292.7-844.0	623.1	684.3	276.8-1427.9	565.3	399.5	29.9
Beryllium	9.87	<0.01	2	0.3-2.74	1.3	1.36	0.65-2.83	1.4	1.21	5.06-26.8	10.3	5.31	1.83-4.1	2.7	2.39	n/a
Boron	1.23	0.31	2	947.6-1573	1250.9	1309	1039.9-4473.9	1869.2	1494	671.2-1473.5	1065.2	1024	1018.8-14	1230.8	1206	112.1
Cadmium	1.50	0.24	2	6.4-62.1	19.3	12.7	13.0-142.8	59.5	33.7	35.9-373.4	103.5	53.9	14.9-195.9	58.9	33.4	n/a
Chromium	0.16	0.85	2	31.7-258.7	118.1	88.9	42.8-171.4	94.9	70.5	30.2-868.9	124.1	103.4	44.4-156.9	113.2	126	n/a
Cobalt	4.31	<0.01	2	49.45-176.9	87.2	75.2	83.88-492.7	209.6	147.1	2930-8959	6270.5	6310	577.7-2513	1089.8	856.2	236.6
Copper	0.66	0.53	2	13791-29689	23279.8	25755	12563-102888	17147.9	17810	9619-55561	24945.9	18892	1852-44140	25828.3	19838	n/a
Iron	5.21	<0.01	2	32925-59215	45530.3	47658	34133-71457	43373.4	37886	45220-114968	72586.4	64092	40420-109613	68016.2	64954	540.5
Lead	1.16	0.33	2	12.9-48.5	30.8	30.1	18.3-43.4	32.5	34.2	94.1-294.2	42.0	26.8	30.0-257.2	42.9	39.2	n/a
Manganese	19.5	<0.01	2	17998-35040	26850.7	27718	27758-55660	43243.1	43229	79352-184237	117147.9	96872	39751-132119	70644.1	63304	656.3
Molybdenum	10.1	<0.01	2	46.61-64.66	53.4	49.4	40.49-61.75	49.8	48.5	52.42-160.9	100.7	81.2	59.13-123.9	86.2	83.9	2013.7
Nickel	30.2	<0.01	2	16.45-97.61	62.1	73.4	12.27-106.1	40.9	27.5	1918-7780	4491.1	3419.2	585.7-1497	932.9	857	58.6
Selenium	14.8	<0.01	2	918-1855	1376.1	1274	1339-2019	1726.1	1732	2092-3578	2767.4	2864	1673-3178	2106.4	1709	n/a
Silver	1.78	0.19	2	21.5-70.4	47.7	52.3	15.9-50.8	31.0	31.7	20.2-140.8	67.5	53.7	20.4-83.4	46.5	38.8	n/a
Strontium	6.17	<0.01	2	2285-4543	3445.5	3376	3260-6176	4730.3	4642	2232-2973	2490.0	2426	2743-5376	3502.3	3075	42.2
Titanium	0.48	0.62	2	109.9-270.3	189.7	189.6	178.8-203.8	189.0	189.1	128.5-285.3	207.2	206	170.7-284.6	203.8	190.2	n/a
Uranium	6.14	<0.01	2	0.45-1.46	0.9	0.97	0.64-2.25	1.2	1.11	1.98-13.53	4.7	2.88	1.04-3.16	2.3	2.53	94.6
Vanadium	1.79	0.19	2	30.6-85.1	64.8	73.6	55.0-75.6	64.7	63.1	41.8-124.1	71.7	62.1	66.3-126.9	84.5	78.8	n/a
Zinc	5.22	<0.01	2	52037-96949	75489.9	73689	76576-14418	103241.4	99791	67701-121424	96659.4	101064	96772-200559	132707.2	129659	920.6

Mean aquatic beetle concentrations of Arsenic, Beryllium, Cobalt, Iron, Manganese, Molybdenum, Nickel, Selenium, Strontium, Uranium and Zinc were found to be highly statistically different between reference and coal mine impacted samples (Table 4). When analysed by Analysis of Similarity (ANOSIM) significant differences were found between W1 and W3 (R statistic 0.963), W1 and W5 (R statistic 0.639), Bell and W3 (R statistic 0.924) and Bell and W5 (R statistic 0.523) whilst no significant difference was recorded for W1 and Bell (R statistic 0.123). Biota and/or Environmental matching (BEST) results show that Manganese, Cobalt, Nickel and Zinc have the strongest influence in the differences recorded across beetle sample locations (best results, Corr = 0.944), (method, BIOENV).

Reference site beetles recorded mean arsenic concentrations of 318.8 and 501.2 $\mu\text{g}/\text{kg}$ increasing slightly at both downstream sites W3 and W5 (mean 474 and 541 $\mu\text{g}/\text{kg}$). Mean Beryllium at reference sites was recorded at 1.42 and 1.3 $\mu\text{g}/\text{kg}$. The downstream sites W3 and W5 recorded mean Beryllium results of 10.3 and 2.7 $\mu\text{g}/\text{kg}$. Reference sites recorded mean cobalt concentrations of 209.6 and 87.2 $\mu\text{g}/\text{kg}$, in comparison W3 and W5 results showed mean cobalt of 6270.5 and 1089.8 $\mu\text{g}/\text{kg}$. Iron at reference sites was recorded at 43373 and 45530 $\mu\text{g}/\text{kg}$. Whilst in comparison W3 and W5 recorded mean iron results of 72586 and 68016 $\mu\text{g}/\text{kg}$. Reference site results show mean manganese concentrations of 43243 and 45530 $\mu\text{g}/\text{kg}$. The downstream sites W3 and W5 recorded mean manganese of 117148 and 70644 $\mu\text{g}/\text{kg}$. Beetles sampled at both reference sites recorded mean molybdenum of 49.8 and 53.4 $\mu\text{g}/\text{kg}$. In comparison W3 and W5 (downstream sites) recorded mean molybdenum of 100.7 and 86.2 $\mu\text{g}/\text{kg}$. Nickel concentrations from both reference sites was 40.9 and 62.1 $\mu\text{g}/\text{kg}$ (mean), whilst in comparison mean nickel at the downstream sites (W3 and W5) was recorded at 4491.1 and 932.9 $\mu\text{g}/\text{kg}$ (Table 4).

Reference site selenium concentrations were recorded at 1726 and 1376 $\mu\text{g}/\text{kg}$ respectively. In comparison W3 and W5 recorded mean selenium results was 2767 and 2106 $\mu\text{g}/\text{kg}$. Mean strontium concentrations were recorded at reference sites as 4730.3 and 3445.5 $\mu\text{g}/\text{kg}$ with the downstream sites (W3 and W5) recording mean strontium of 2490.0 and 3502.3 $\mu\text{g}/\text{kg}$. Reference sites mean uranium was recorded at 1.2 and 0.9 $\mu\text{g}/\text{kg}$ respectively. In comparison W3 and W5 (downstream sites) recorded mean uranium results of 4.7 and 2.3 $\mu\text{g}/\text{kg}$. Mean zinc results for both reference sites recorded concentrations of 103241 (Bell) and 75490 (W1) $\mu\text{g}/\text{kg}$ respectively, increasing downstream of each impacted sample locations paired reference site to mean zinc concentrations of 132707 (W5) and 96659 (W3) $\mu\text{g}/\text{kg}$ (Table 4).

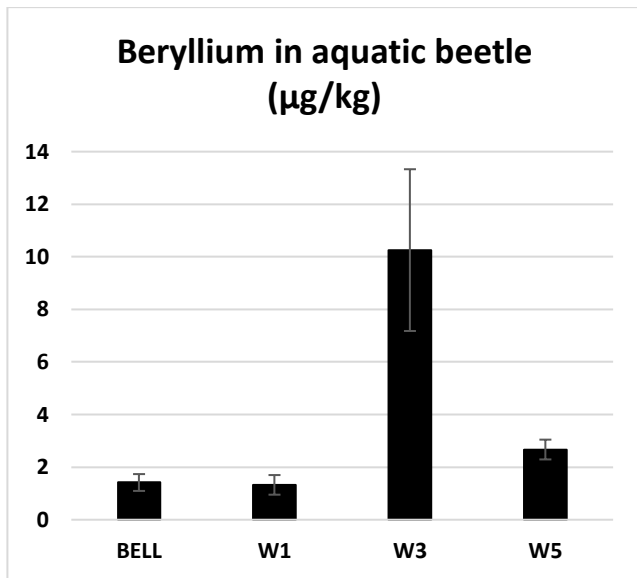


Figure 2 Mean beryllium with standard error bars for aquatic predacious beetles (Gyrinidae) measured in µg/kg. Bell and W1 are reference samples whilst W3 and W5 are coal mine impacted samples.

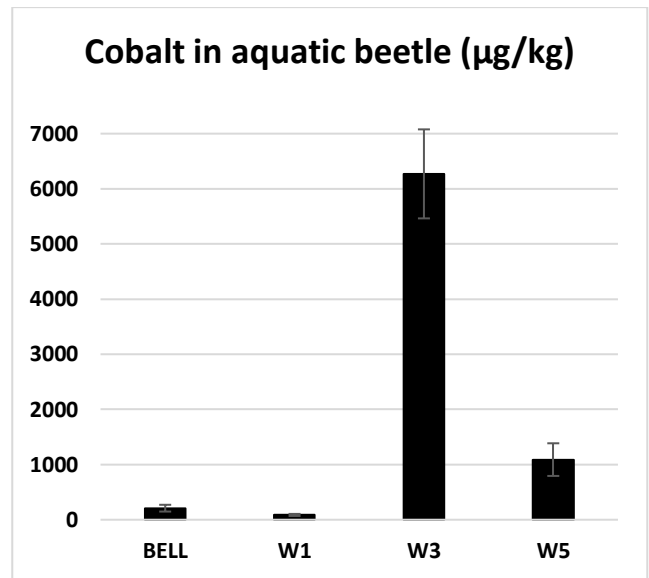


Figure 3 Mean Cobalt with standard error bars in aquatic predacious beetles (Gyrinidae) measured in µg/kg. Bell and W1 are reference samples whilst W3 and W5 are coal mine impacted samples.

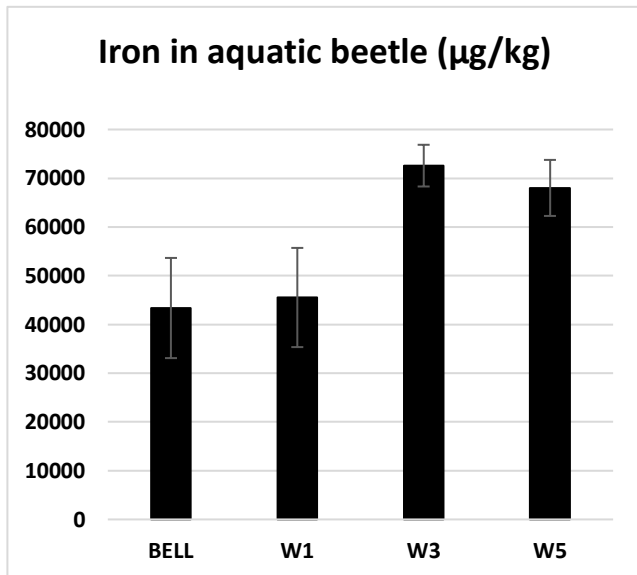


Figure 4 Mean Iron with standard error bars in aquatic predacious beetles (Gyrinidae) measured in µg/kg. Bell and W1 are reference samples whilst W3 and W5 are coal mine impacted samples.

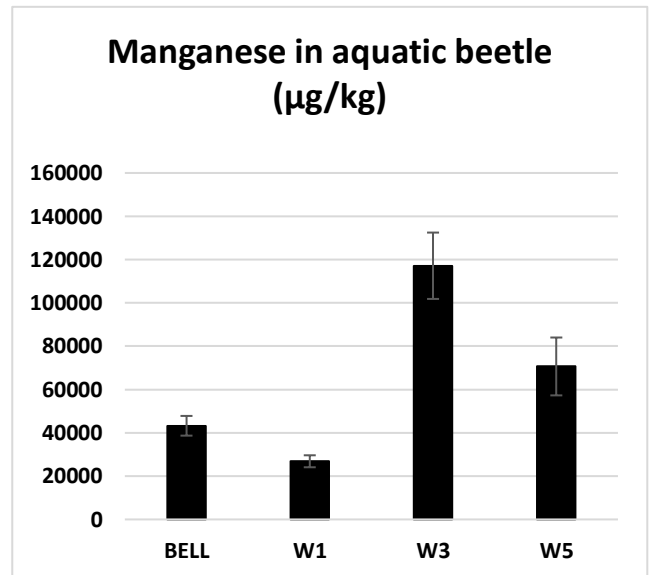


Figure 5 Mean Manganese with standard error bars in aquatic predacious beetles (Gyrinidae) measured in µg/kg. Bell and W1 are reference samples whilst W3 and W5 are coal mine impacted samples.

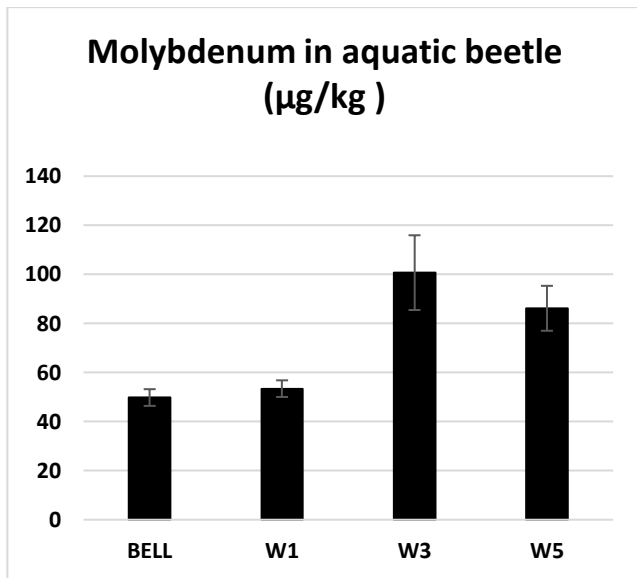


Figure 6 Mean Molybdenum with standard error bars in aquatic predacious beetles (Gyrinidae) measured in µg/kg. Bell and W1 are reference samples whilst W3 and W5 are coal mine impacted samples.

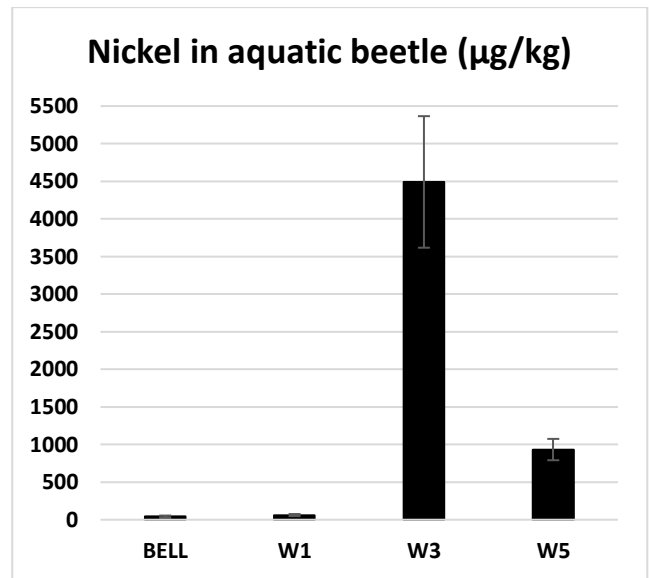


Figure 7 Mean Nickel with standard error bars in aquatic predacious beetles (Gyrinidae) measured in µg/kg. Bell and W1 are reference samples whilst W3 and W5 are coal mine impacted samples.

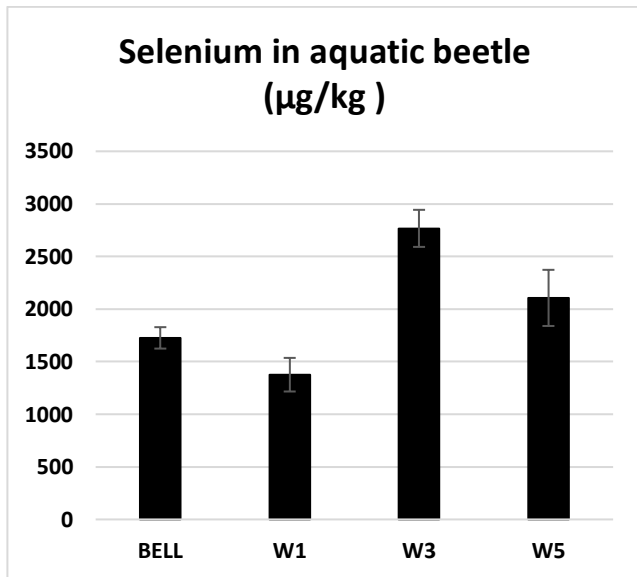


Figure 8 Mean Selenium with standard error bars in aquatic predacious beetles (Gyrinidae) measured in µg/kg. Bell and W1 are reference samples whilst W3 and W5 are coal mine impacted samples.

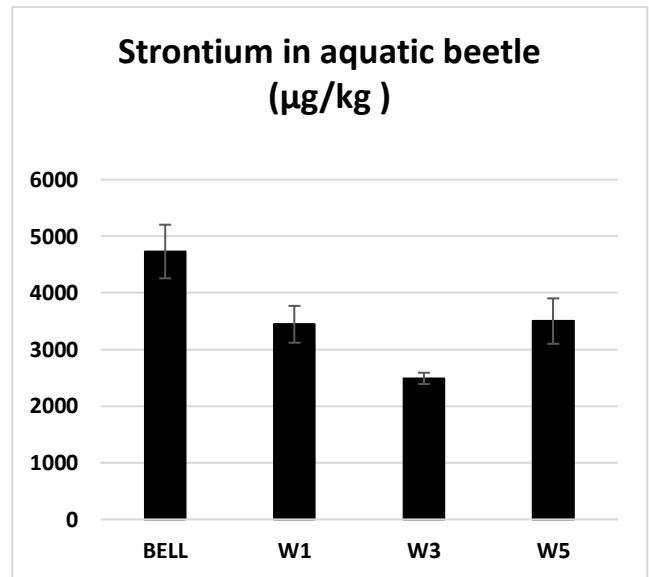


Figure 9 Mean Strontium with standard error bars in aquatic predacious beetles (Gyrinidae) measured in µg/kg. Bell and W1 are reference samples whilst W3 and W5 are coal mine impacted samples.

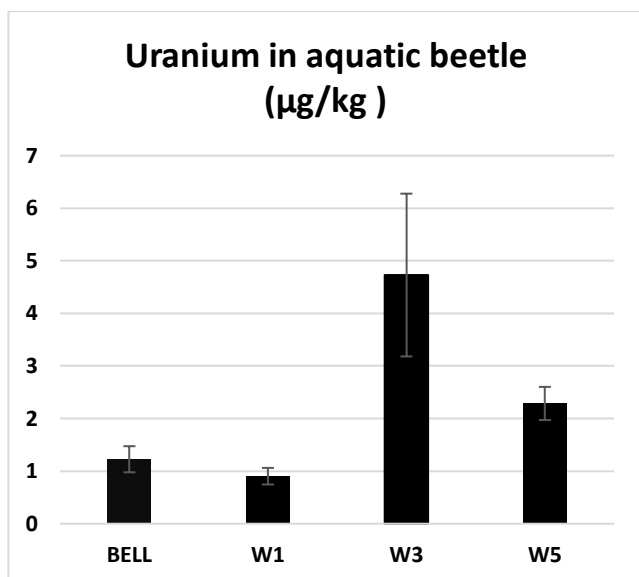


Figure 10 Mean Uranium with standard error bars in aquatic predacious beetles (Gyrinidae) measured in µg/kg. Bell and W1 are reference samples whilst W3 and W5 are coal mine impacted samples.

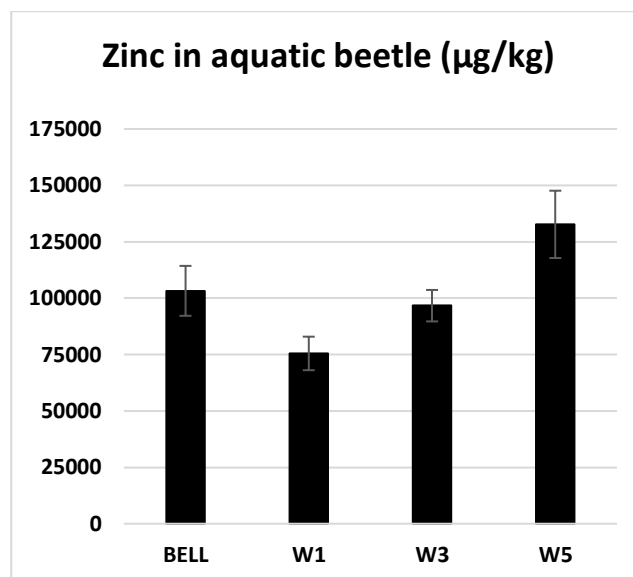


Figure 11 Mean Zinc with standard error bars in aquatic predacious beetles (Gyrinidae) measured in µg/kg. Bell and W1 are reference samples whilst W3 and W5 are coal mine impacted samples.

Discussion

This study may be the first to investigate the bioaccumulation of contaminants from a licenced and regulated coal mine wastewater discharge on an aquatic predacious beetle (*Macrogyrus rivularis*).

Results show that the one species of aquatic predacious beetle has increased concentrations of contaminants at impacted sample locations when compared to non-impacted reference sites. Many of the contaminants can be directly linked to the coal mine wastewater discharge as shown by water column and stream sediment results. In its crux, this one aquatic predacious beetle is at the lower trophic level of the food chain and is a food source for many other aquatic and terrestrial species. The implications of concentrating large amounts of contaminants within lower trophic order species, whom are predated on by aquatic and terrestrial species may allow for a link to continue the biomagnification of the contaminants found within the impacted sampled beetles. The ability for these aquatic beetles to be pray for terrestrial species is also of major concern as this may also allow for the mobilisation of these contaminants from the aquatic ecosystem to the terrestrial environment.

There are many studies investigating water column pollution from metal contamination from a broad range of mining activities. Many investigate the links between metal mining impacts on water chemistry, stream sediments, aquatic flora, aquatic fauna, including; fish, turtles to an array of macroinvertebrates (Neff et al. 1984, Sericano et al. 1995, Wang and Rainbow 2008, Hill et al. 2000, Amisah and Cowx 2008 and Ashraf et al. 2011). Only a few studies have investigated the bioaccumulation of metals from coal mining activities within macroinvertebrates. Most often these impacts are from waste coal ash dams or coal fine spills but not discharged, licenced and regulated coal mining wastewaters.

Miller et al. (2013) studied the bioaccumulation of the metal selenium from abandoned coal mine pit lakes within two fish species. It was reported that selenium was bioaccumulating within the fish tissue to levels above USA EPA tissue guidelines and concluded that the current reclamation practices implemented in the abandoned coal mine pit lakes were failing and that significant risk to wildlife and human health was of concern (Miller et al. 2013). Within

Australia, Telford et al. (2009) and Jasonsmith et al. (2008) have conducted studies assessing the bioaccumulation of metals from mining activities. Telford et al. (2009) investigated bioaccumulation of metals in aquatic gastropods from antimony and arsenic mines and found that concentrations of arsenic were statistically higher in mining impacted samples. Jasonsmith et al. (2008) studied the bioaccumulation of selenium in water, sediment, zooplankton, benthic material, benthic algae, oligochaetes, gastropods, crustaceans, insects and fish residing in a coal power stations cooling reservoir in Lithgow, NSW close (within 15km) to the vicinity of this study. It was found that selenium was found in low concentrations within oligochaetes, gastropods, bivalves and crustaceans sampled and in contrast insects sampled recorded concentrations some 1000 times higher. Jasonsmith et al (2008) concluded that the detritus invertebrates bioaccumulated selenium in much greater concentrations than oligochaetes, gastropods, bivalves and crustaceans they sampled (Telford et al. 2009 and Jasonsmith et al. 2008).

Swansburg et al. (2002) found many metal concentrations below laboratory detectable limits within water samples in their study whilst recording detectable (in some cases statistically different) limits of the same heavy metals in chironomid tissue. Their study investigated metal mining impacts on clean dilute ($EC < 100 \mu\text{s/cm}$), circumneutral (pH 6.5-7.5) streams in New Brunswick, Canada. Cadmium, Chromium, Cobalt, Copper, Lead and Nickel all recording detectable limits within chironomid tissue whilst only Molybdenum still recorded below detectable limits. Significant differences between reference and mine impacted chironomid samples were recorded for copper, cadmium and zinc only (Swansburg et al. 2002). This is similar to this study with many contaminants within the water column recording below laboratory detectable limits at both reference and impacted sample locations, becoming more detectable within stream sediment and in turn sampled beetles at impacted sites.

Jasonsmith et al. (2008).found increases between 3 and 16 times were recorded across the five streams for zinc in chironomid tissue. The greatest increase being the stream impacted by the metal mine facility "Caribou" was reported to increase some 30 times from a reference mean of 113000 mg/kg and an impacted mean of 1813000 mg/kg. This current study found zinc in beetles to be statistically significant recording increases at lower concentrations close to the vicinity of 1.5 times greater at impacted sites and increasing from water column concentrations to beetle concentrations in the magnitude of 77000 times. Cadmium was found by Swansburg et al (2002) to be nearly 65 times higher in impacted chironomid tissue samples at the same mining site increasing from 600 to 37200 mg/kg. This study found cadmium to increase two-fold between the reference site W1 and the impacted site W3 but was not statistically significant. Copper was recorded by Swansburg et al (2002) to increase 11 and 12 times at two impacted sites from 13mg/kg upstream to 153mg/kg downstream and 10mg/kg upstream to 115 mg/kg downstream. Copper in this current study was not statistically significantly different with W1, W3 and W5 all recording similar copper concentrations with Bell Creek recording copper concentrations approximately 0.15 times less.

Jasonsmith et al. (2008).found mine effected chironomids to have deformed mentums. It was also reported increases in cobalt for four of their five sample streams. The highest increase from reference to mine effected cobalt in chironomid tissue was 600 (reference) to 3200 (mine impacted) $\mu\text{g/kg}$ just over five times higher. This study found cobalt increased from 87.2 $\mu\text{g/kg}$ at the reference site W1 to 6271 $\mu\text{g/kg}$ directly below the coal mine wastewater inflow at site W3 which is over 70 times higher than W1. Water column concentrations of cobalt in this current study increased some 50000 times from water column concentrations to beetle concentrations.

Otter et al. (2012) found Selenium and Arsenic bioaccumulated at differing concentrations within two fish species they studied impacted by a wet coal ash spill. Statistical differences were recorded for both metal concentrations when compared between reference and impacted samples. Otter et al. (2012) concluded that the difference in bioaccumulated selenium and arsenic across species in impacted sites was due to the differing stomach pH of the species (Otter et al. 2012). In this current study, the reference sites used may have a greater functional ability to sequester and bioaccumulate heavy metals due to their naturally mildly acid pH in comparison to the impacted sites whom have a “treated” alkaline pH (Otter et al. 2012, Durães et al. 2014 and Atkinson et al. 2007). This is of major concern as impacted sites beetles in this study have significantly higher concentrations of heavy metals whilst having a much lower ability to bioaccumulate these contaminants.

This raises questions as to what will occur after mining activities are no longer undertaken at this colliery as when mining ceases so does the water treatment process. At present pH is increased during the treatment process and water currently being discharged may have a much lower rate of bioavailability of some contaminants for aquatic biota as described by Atkinson et al. (2007). Once treatment ceases and the Wollangambe Rivers pH will naturally reduce to a background pH which is mildly acidic. This decrease in pH may open a new avenue for increased bioaccumulation from the legacy pollutants within the impacted streams sediments. The findings of this research evoke concerns over the validity of water column pollutant limits if they are in fact allowing for legacy pollutants to bioaccumulate and or magnify within the receiving waterways aquatic ecosystem.

Conclusions

Long term legacy pollutants are of great concern worldwide. Sericano et al. (1995) and Ashraf et al. (2011) suggest heavy metals have the potential to accumulate within biota at toxic concentrations and have chronic ecological impacts within ecosystem food chains. Wang and Rainbow (2008) raised concerns over the longevity of heavy metal pollutants in aquatic environments due to their ability to deposit into waterway sediments which can potentially remain indefinitely. Kolaříková et al. (2012) found that bioaccumulation of pollutants within four macroinvertebrate species within the Elbe and Vltava Rivers in the Czech Republic were still persistent after water quality improvements were implemented. It was reported that these heavy metals concentrations were consistent over their 12-year study and they recorded no significant reduction in heavy metal concentrations other than Hg.

The implications that contaminants from the licenced and regulated wastewater being discharged may be accumulating within aquatic biota is of major concern. The implications that this regulated water column pollution is accumulating at magnified rates shows a major floor in water column pollution licensing. It is recommended that further research should be undertaken by the New South Wales Environmental Protection Authority to better assess these broader implications of legacy contaminants from licenced and regulated coal mine wastewater discharges are having on the aquatic ecosystem of EPL protected waterways.

Recommendations

It is recommended that further research should be undertaken by the New South Wales Environmental Protection Authority to better assess the implications of coal mine wastewater contaminants bioaccumulation and or biomagnification in EPL protected waterways stream sediments and their biota. If in fact the contaminants are leaving

the water column and bioaccumulating within the aquatic biota of their receiving waterways, serious long-term legacy pollutant impacts may persist. Of equal concern is if in fact these pollutants are biomagnifying within the aquatic biota there is feasibility that this may transpose to the terrestrial environment and the extent of the contamination may be more far spreading than this study has found.

Research significance

Very little research has examined the ongoing food-chain impacts of water pollution from coal mines through metal bioaccumulation. At present in Australia, once coal mining practices cease a coal mine operation undertakes simple remediation practices, often coal mine waste waters will continue to flow into their receiving waterways, unmitigated and untreated. This research directly addresses this gap in the literature.

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Chapter 8

Grading Coal Mine Wastewater impacts to Aquatic Ecosystems, measured through Macroinvertebrate Diagnostic Biotic Indices.

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Abstract

Macroinvertebrate biotic indices are a well-developed robust method to assess anthropogenic impacts on aquatic ecosystems and have been used since the early 1980's (Washington 1984 and Chessman 2003). The SIGNAL method (Stream Invertebrate Grade Number Average Level) is one method employed to assess the level and degree of impacts from anthropogenic influences in South Eastern Australia (Chessman 2003). This study investigates the impacts from coal mining wastewaters which are often contaminated with high concentrations of heavy metals along with modified ionic composition. 12886 individual macroinvertebrates, the majority of which were identified to the family level (55 families) across eight different waterways have been used here to derive a Coal Mine Impact Grade (CMIG) biotic index. Results of the CMIG/SIGNAL comparison show the great potential of using a grading system as a robust, relatively low impact and rapid method for the assessment of Coal Mine Waste Water impacts to the receiving waterways aquatic ecosystem. Coal Mine Waste Water impacts to receiving waterways are regulated by the New South Wales Environmental Protection Authority to ensure the protection of the receiving waterways aquatic ecosystem, though the regulation and subsequent 'protection' performed through water quality and chemical concentrations never assesses the aquatic ecosystem. Though these grades are only preliminary, they are the only grades which in fact assess the impact of coal mine waste water discharges to a receiving waterways aquatic ecosystem and would provide a much better monitoring system than generalised water column pollutant concentration limits.

Keywords

SIGNAL, biological monitoring, biotic index, macroinvertebrates, water quality, coal mine impact biotic index

Introduction

Macroinvertebrate biotic indices are a well-developed robust method to assess anthropogenic impacts on aquatic ecosystems and have been used since the early 1980's (Washington 1984 and Chessman 2003). The Average Score Per Taxon (ASPT) version of the Biological Monitoring Working Party (BMWP) system is used within Great Britain (Hawkes 1997). The SIGNAL method (Stream Invertebrate Grade Number Average Level) is one method employed to assess the level and degree of impacts from anthropogenic influences in South Eastern Australia and is derived from the afore mentioned methods (Chessman 2003). The SIGNAL method developed in 1995 by Chessman et al (1995) uses family level sensitivity grades for individual taxa ranging from "sensitive taxa" graded as 10 to "tolerant taxa" graded as 1 with many taxa in between varying due to their sensitivity to anthropogenic stressors. The SIGNAL method has evolved over the years and has become a reliable and robust method for the rapid assessment of macroinvertebrate communities impacted by urban development, industrial activities, agricultural impacts and stormwater runoff. The SIGNAL method is currently used as part of multi-layer waterway management practices which include the Victorian Index of Stream Condition (Ladson et al 1999). It was incorporated into the Victorian State Environment Protection Policy proposed by the Victorian Environmental Protection Authority (Anon 2001a). Nationally the SIGNAL method has been incorporated into the Australian River Assessment System (AUSRIVERS) and is a part of Australia's National River Health Program (NRHP) (Davies 2000).

Since its inception in 1995 the SIGNAL method has been modified to ensure the greatest integrity of its assessment outcomes. For instance, it was found by Chessman and McEvoy (1998) that different anthropogenic stressors such as sewage and metal contamination showed differing sensitivities for the same taxa. This later publication divided the anthropogenic stressors into three different categories (Dam, Sewage and Metal) impacts. It was concluded that the derived SIGNAL index's performed to varying degrees. For instance the SIGNAL-DAM method showed less reliability than the SIGNAL-METAL and SIGNAL-SEWAGE index's in that order (Chessman 1998). This current study investigates the impacts from coal mining wastewaters which are often contaminated with high concentrations of heavy metals along with modified ionic composition. 12886 individual macroinvertebrates, the majority of which were identified to the family level (55 families) across eight different waterways have been used here to derive a Coal Mine Impact Grade (CMIG) biotic index. The proposed CMIG biotic index has been derived on presence and absence abundance. In conjunction with Chessman and McEvoy's (1998) SIGNAL-METAL index as a surrogate index to test the rigour of the Coal Mine Impact Grades (CMIG) derived here.

Methods

The Coal Mine Impact Grades (CMIG) derived in this study has been developed from presence and absence data collected at eight waterways within the greater Sydney Basin. All of the eight waterways are or have been subject to licenced and regulated coal mine wastewater discharges. An identical scoring and grading system to that of Chessman et al's SIGNAL method were employed. A total of 12866 individual macroinvertebrates from 55 families were collected from a single upstream and a single downstream sample location at the eight waterways. Ten replicated quantitative macroinvertebrate samples were collected at five of the waterways and 5 identically sampled replicates at each remaining waterways upstream and downstream sample locations. From the entire 12886 macroinvertebrates

collected 7531 were used from 51 families to derive Coal Mine Impact Grade (CMIG) of which 3038 were collected upstream of the coal mine wastewater inflows to the receiving waterways and 4494 downstream. Initially data was analysed for presence and absence of family level taxa between the upstream and downstream samples. Differences were calculated based on how many sample locations (upstream and downstream) recorded each family level taxa. I.e if a family level taxa was observed upstream at all mines (7) and downstream at two mines (2) the difference was recorded as (5), conversely if 5 upstream locations recorded a family level taxa and 3 downstream locations recorded the same family level taxa the difference was 2. If this trend was reversed, i.e greater family level taxa presence downstream than upstream a negative value was achieved, i.e 0 family level taxa were recorded at upstream locations compared to 1 at downstream locations a difference of -1 was recorded. In order to increase the negative number to a positive number each initial differing negative grade of -1 was made 0.5 and -2 0.25 as it was quite obvious that these taxa were very tolerant of the coal mine wastewater pollution hence needed to be graded as such (tolerant = lower grade). This difference in presence and absence calculation was performed for all 45 taxa and gave an initial grade range between -1 (0.5) and 4. These initial grades gave some indication to the degree of spacing between each individual taxa's initial Coal Mine Impact Grade (CMIG). At this stage, the data set was beginning to show differences in each family level taxa's sensitivity to the coal mine pollution. Firstly, it was imperative to adjust the "initial" Coal Mine Impact Grade (CMIG) to encompass a range between 1 and 10, 10 being most sensitive. For the initial grade adjustment, a trial calculation was employed to increase all the grades to fall between 1 and 10, as some taxa were still graded as 0.5. Several multipliers were assessed until the figure of 2.66 was found to suit the entire data set. It should be noted that at this stage the grades had become non-whole numbers and that after the final stage of grade adjustments the grade numbers were rounded either up or down to become whole numbers. After the initial grade adjustment to remove grades below 1 a second multiplier was applied to allow for those taxa that were only collected upstream of the coal mine wastewater discharges to increase to the most sensitive grade of 10. It was found that 1.25 suited the entire data set by allowing for low grades to stay low (for those taxa recorded in great numbers below the coal mine waste inflows) whilst also allowed for those sensitive taxa to be graded as 10 after rounding up.

Results and Discussion

These Coal Mine Impact sensitivity grades were then used with the standard family level taxonomic resolution used for the SIGNAL calculation method and each waterway was assigned a Coal Mine Impact Grade (CMIG). This Coal Mine Impact Grade (CMIG) was assessed along side the standard SIGNAL score and the Chessman and McEvoy (1998) SIGNAL-METAL scores to assess the overall integrity of the newly derived Coal Mine Impact Grades (CMIG) and subsequent scoring system (Table 1).

Table 1. Coal Mine Impact Grade (CMIG) (Belmer and Wright), SIGNAL-METAL (Chessman and McEvoy 1998) and SIGNAL (Chessman et al 1995) grades comparison.

TAXA/SIGNAL grades	Coal Mine Impact Grade (CMIG)	SIGNAL-METAL	SIGNAL
Atriplectididae	5	n/a	8
Baetidae	4	7	5
Caenidae	3	7	4
Calamoceratidae	10	8	7
Calocidae	7	n/a	9
Coloburiscidae	10	10	8
Conoesucidae	10	8	6
Eustheniidae	10	n/a	10
Glossomatidae	10	8	9
Gripopterygidae	8	8	8
Hydrobiosidae	6	6	8
Hydropsychidae	3	10	7
Hydroptilidae	2	6	7
Leptoceridea	5	7	8
Leptophlebiidae	8	8	8
Limnephilidae	8	n/a	8
Notonemouridae	3	1	6
Philopotamidae	4	10	6
Philorheithridae	7	n/a	8
Athericidae	3	n/a	8
Aeshnidae	5	3	4
Bithyniidae	3	n/a	3
Chironomidae	1	6	3
Coenagrionidae	4	3	2
Corbiculidae	4	n/a	4
Corixidae	3	4	2
Corydalidae	4	4	1
Culicidae	4	3	1
Curculionidae	3	n/a	2
Diphlebiidae	5	n/a	6
Dixidae	3	n/a	7
Dolichopodidae	1	n/a	3
Dugesiidae	3	3	2
Dytiscidae	5	4	2
Empididae	1	n/a	5
Gomphidae	3	5	5
Gyrinidae	3	4	4
Haliplidae	3	n/a	2
Hydraenidae	5	7	3
Libellulidae	3	1	4
Lymnaeidae	3	7	1
Notonectidae	3	6	1
Physidae	1	5	1
Planorbidae	4	6	2
Psephenidae	4	8	6
Sciomyzidae	3	n/a	2
Scirtidae	8	8	3
Simuliidae	1	5	10
Sphaeriidae	1	n/a	5
Stratiomyidae	1	n/a	2
Tipulidae	1	n/a	3

After deriving each individual family level taxa grade with the use of family richness data from each site a Coal Mine Impact Grade (CMIG) was calculated and compared to mean SIGNAL Grade Scores (Chessman et al 1995) to assess how each calculation fits within a similar grading system. Results from the above presence/absence calculations show strong relationships between upstream (non-impact) and downstream (impact) samples. By comparing SIGNAL grades and CMIG grades (table 2) a similar pattern of impact was observed, though it should be noted that SIGNAL grades are somewhat more conservative and returned a greater SIGNAL grade in comparison to CMIG grade.

Table 2. Comparison of mean SIGNAL (Chessman et al 1995) and Coal Mine Impact Grade (CMIG) mean scores

Waterway name/Mean derived SIGNAL vs CMIG scores	Mean SIGNAL score (Chessman 1995)	Mean Coal Mine Impact Grade (CMIG) score	Measured Impact (SIGNAL)	Measured Impact (CMIG)
Wollangambe River upstream	6.84	6.05	Wollangambe	Wollangambe
Wollangambe River downstream	4.53	3.38	Mild	Severe
Wingecarribee River upstream	5.06	4.64	Wingecarribee	Wingecarribee
Wingecarribee River downstream	4.69	3.25	Mild	Severe
Bargo River upstream	6.21	6.07	Bargo	Bargo
Bargo River downstream	6.00	5.20	None	Minimal
Dalpura Creek upstream	5.78	4.65	Dalpura	Dalpura
Dalpura Creek downstream	5.10	3.78	None	Moderate
Georges River upstream	4.55	4.14	Georges	Georges
Georges River downstream	4.42	3.92	None	Moderate
Sawyers Swamp upstream	5.05	4.58	Sawyers	Sawyers
Sawyers Swamp downstream	3.89	2.56	Moderate	Severe
Springvale Creek upstream	5.38	5.08	Springvale	Springvale
Springvale Creek downstream	3.79	2.79	Moderate	Severe
Sawyers Swamp upstream	5.05	4.58	Kangaroo	Kangaroo
Kangaroo Creek downstream	4.50	3.00	Mild	Severe

Along with a CMIG grade a colour definition of the grading system has been derived from the Chessman et al (1995) SIGNAL system (table 3).

Table 3. Coal Mine Impact Grade (CMIG) score definitions and original SIGNAL score definitions (Chessman et al 1995).

CMIG		SIGNAL (Chessman et al 1995)	
Healthy	>5.5	Healthy	>6
Minimal Impact	4.5-5.5	Healthy to Minimal Impact	5-6
Mild Impact	4-4.5	Mild Impact	4-5
Moderate Impact	3.5-4	Moderate Impact	3-4
Severe Impact	<3.5	Severe Impact	<3

Comparison between SIGNAL (Chessman et al 1995) and CMIG derived scores (Table 2 and 3) shows a clear depiction of each coal mines wastewater impact on their receiving waterways. Mean SIGNAL scores for five of the mines show that the wastewaters are impacting the macroinvertebrate community but are perhaps under representing the level of impact, especially for the remaining three mines (Bargo River, Dalpura Creek and the Georges River)

which show no impact to the macroinvertebrate community. In comparison, CMIG is depicting differing degrees of impact to the receiving waterways macroinvertebrate community for all eight mine discharges (Table 2). By using the same family level taxa method employed by Chessman et al (1995) along with the derived (CMIG) grades for individual taxa plot graphs depicting each coal mines impact to their respective receiving waterway were created as per SIGNAL method (Chessman et al 1995).

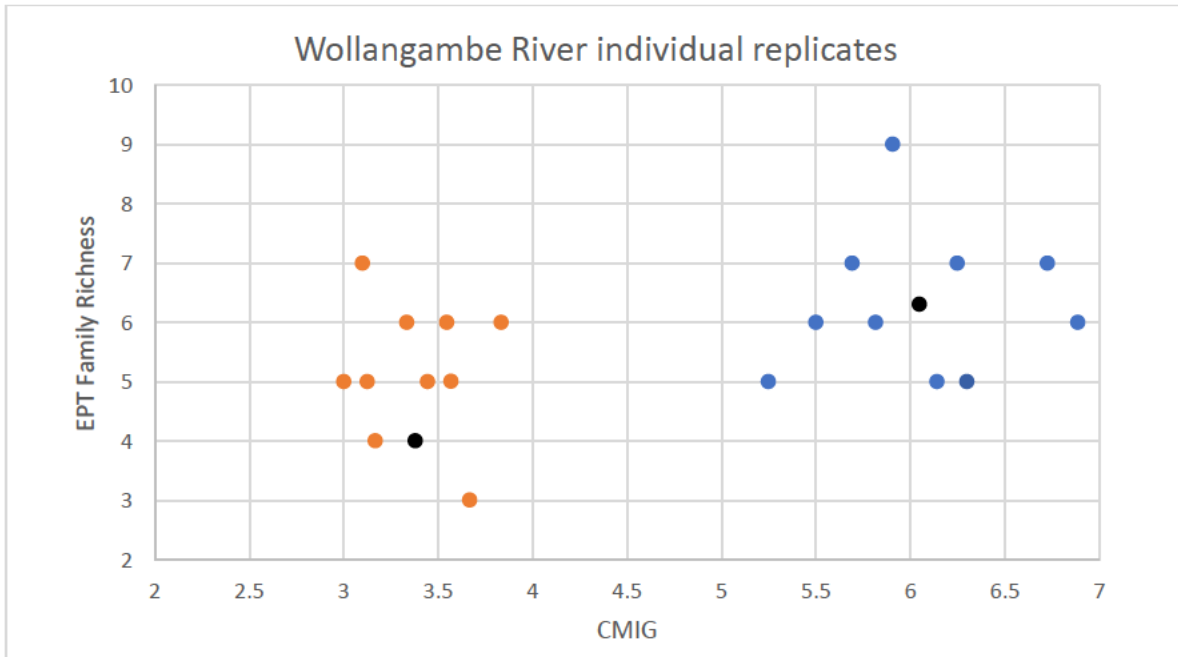


Figure 1. Wollangambe River (Clarence Colliery) CMIG Family Richness plot graph. Orange is downstream of the mine wastewater inflow and blue is upstream. Black is the mean CMIG score for downstream (centre left) and upstream (centre right).

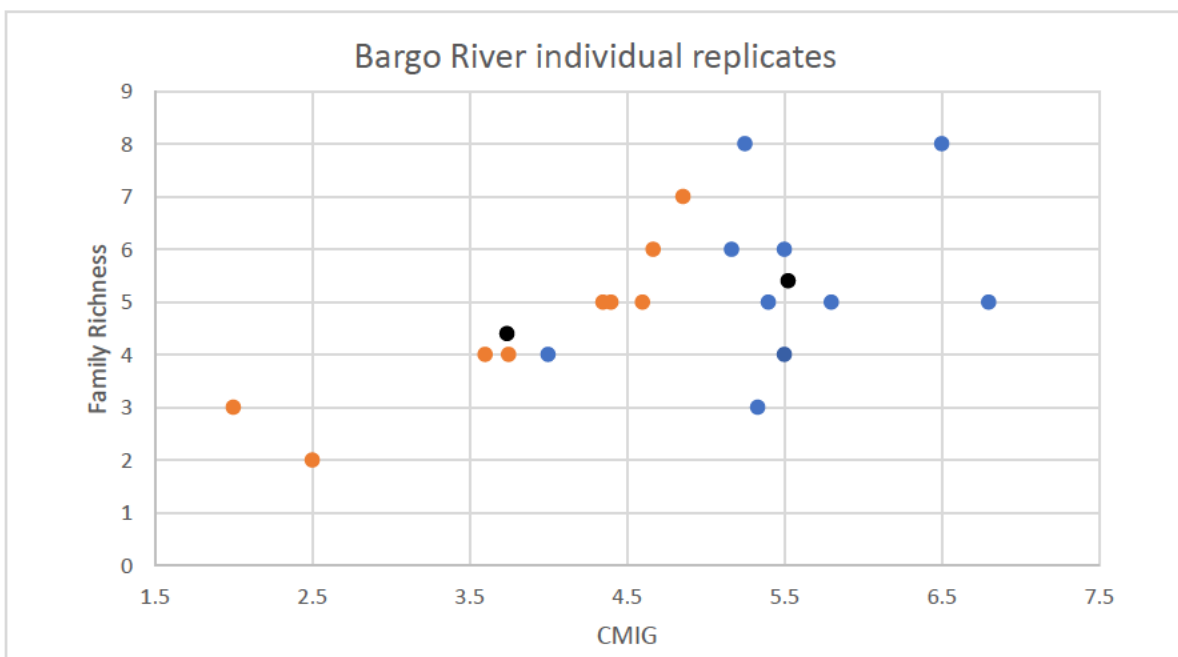


Figure 2. Bargo River (Tahmoor Colliery) CMIG Family Richness plot graph. Orange is downstream of the mine wastewater inflow and blue is upstream. Black is the mean CMIG score for downstream (centre left) and upstream (centre right).

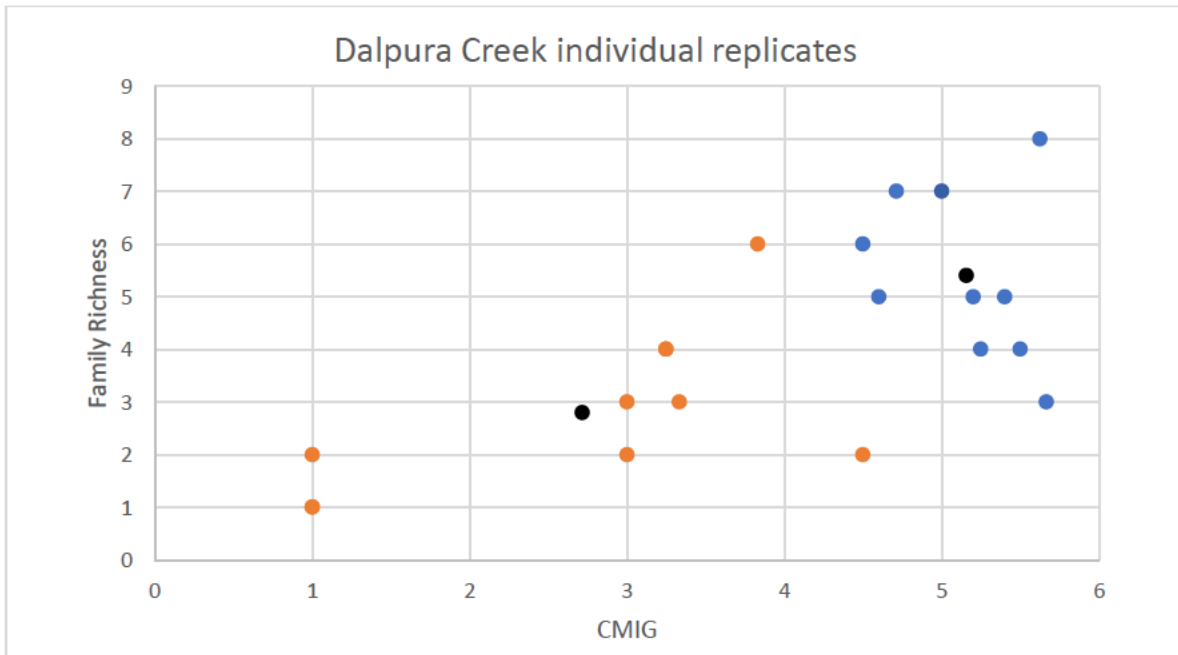


Figure 3. Dalpura Creek (Canyon Colliery) CMIG Family Richness plot graph. Orange is downstream of the mine wastewater inflow and blue is upstream. Black is the mean CMIG score for downstream (centre left) and upstream (centre right).

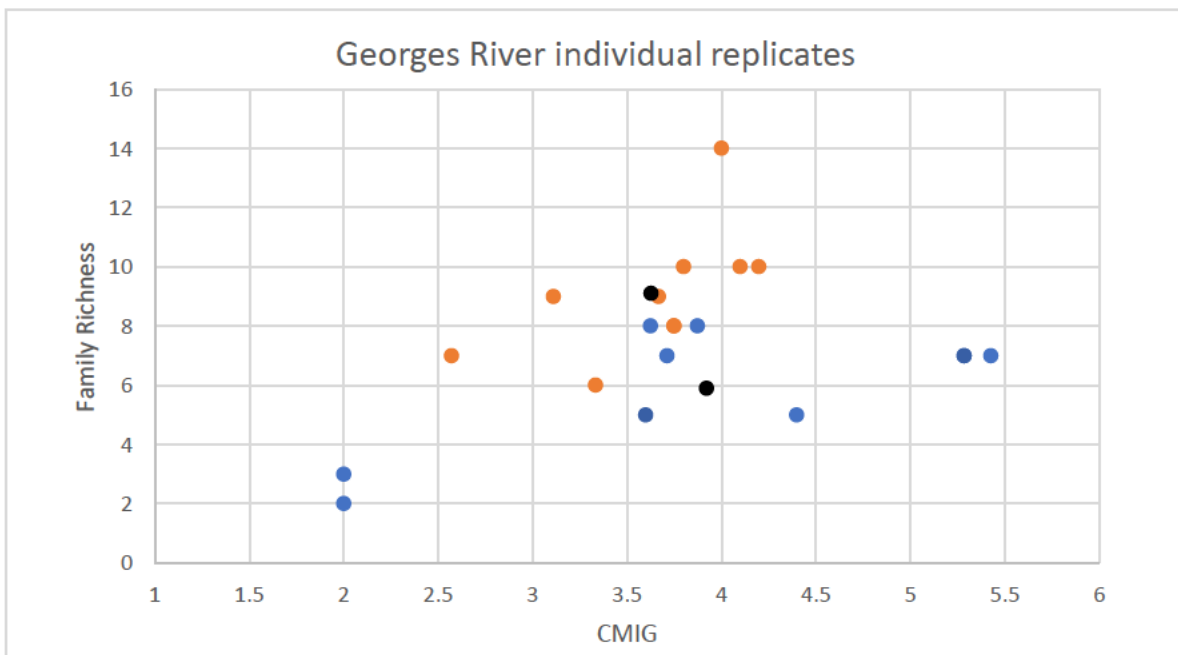


Figure 4. Georges River (West Cliff Colliery) CMIG Family Richness plot graph. Orange is downstream of the mine wastewater inflow and blue is upstream. Black is the mean CMIG score for downstream (centre left) and upstream (centre right).

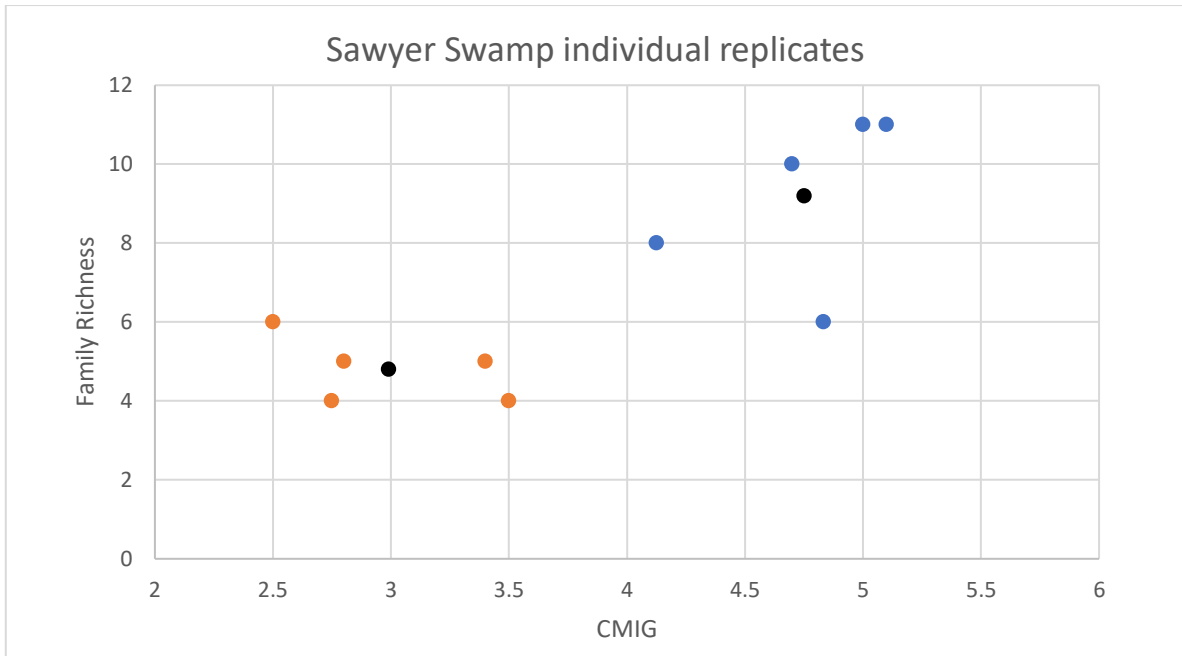


Figure 5. Sawyer Swamp (Angus Place and Springvale Collieries) CMIG Family Richness plot graph. Orange is downstream of the mine wastewater inflow and blue is upstream. Black is the mean CMIG score for downstream (centre left) and upstream (centre right).

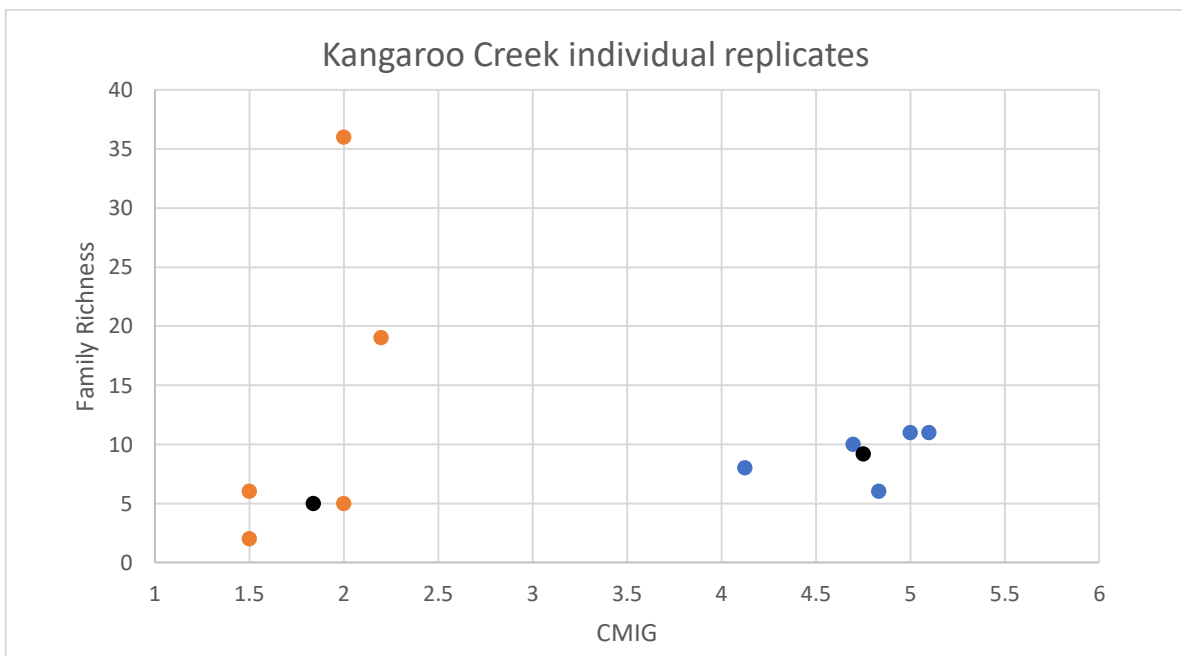


Figure 6. Kangaroo Creek (Angus Place Colliery) CMIG Family Richness plot graph. Orange is downstream of the mine wastewater inflow and blue is upstream. Black is the mean CMIG score for downstream (centre left) and upstream (centre right).

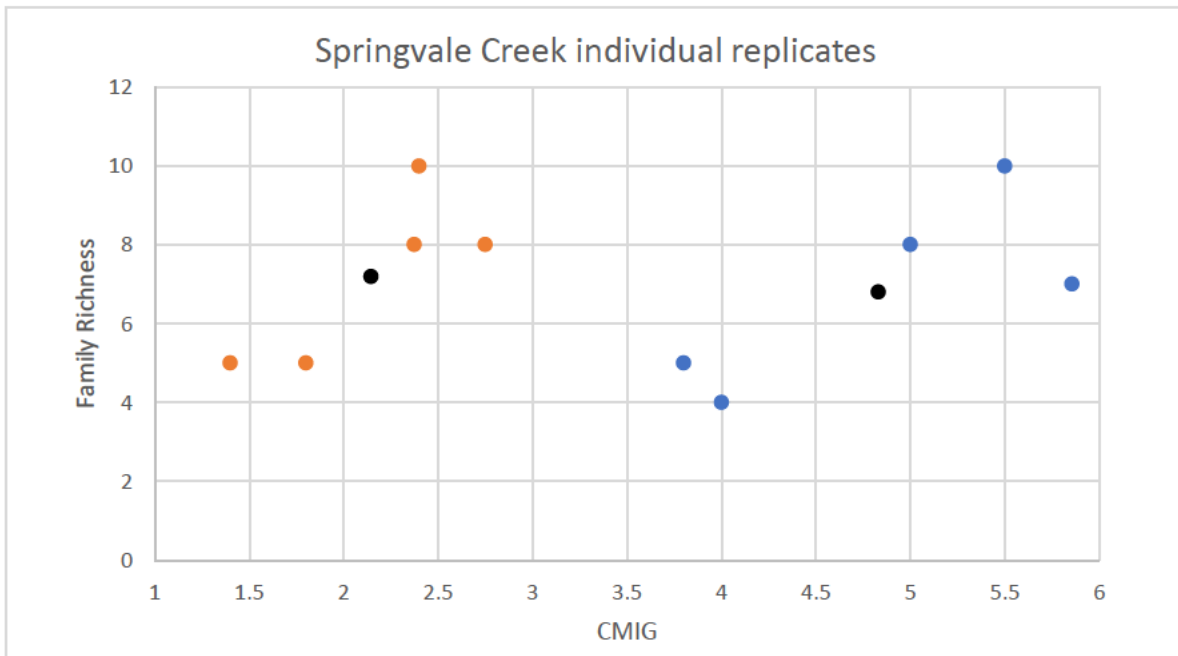


Figure 7. Springvale Creek (Springvale Colliery) CMIG Family Richness plot graph. Orange is downstream of the mine wastewater inflow and blue is upstream. Black is the mean CMIG score for downstream (centre left) and upstream (centre right).

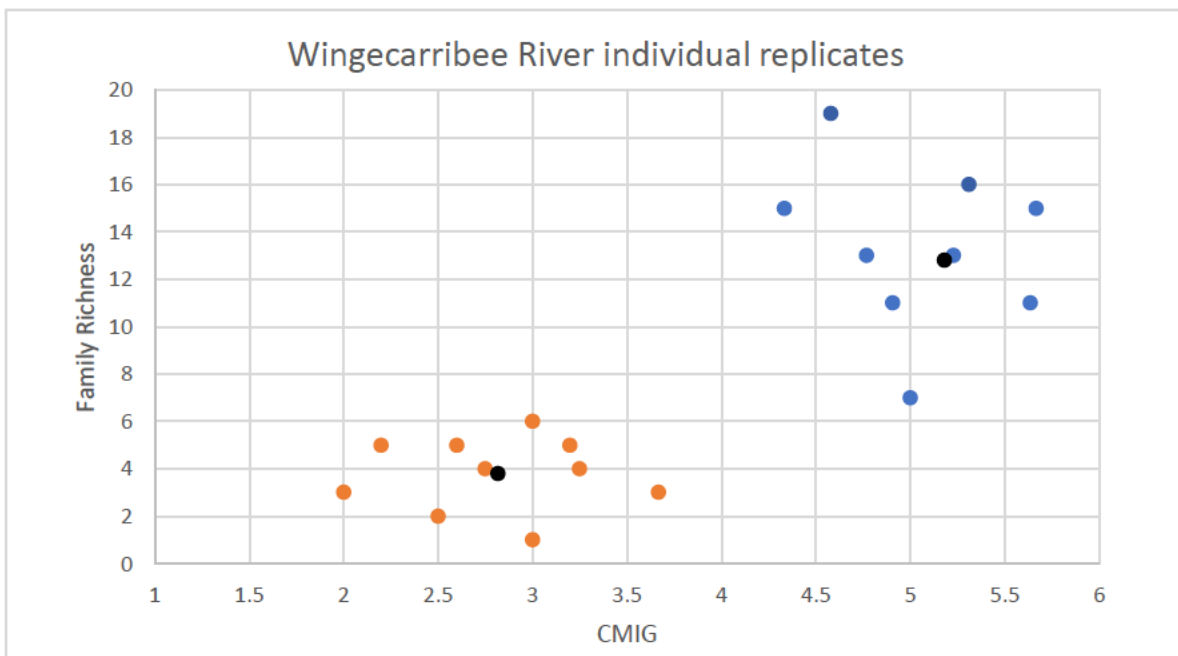


Figure 8. Wingecarribee River (Berrima (Medway) Colliery) CMIG Family Richness plot graph. Orange is downstream of the mine wastewater inflow and blue is upstream. Black is the mean CMIG score for downstream (centre left) and upstream (centre right).

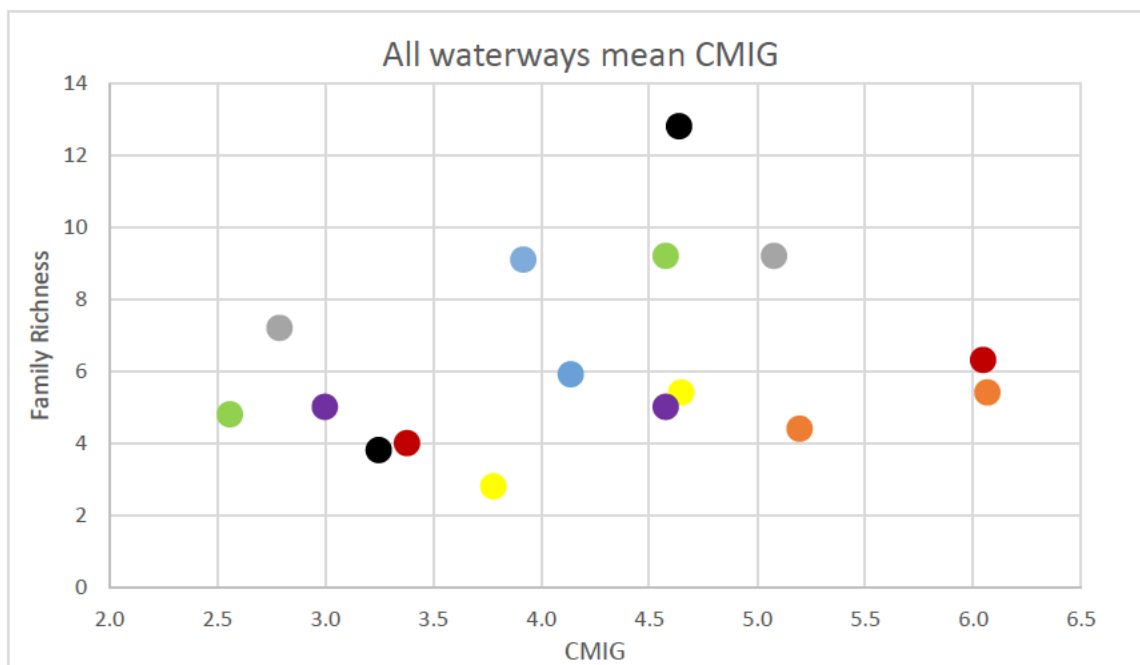


Figure 9. All Waterways and Collieries mean upstream and downstream CMIG Family Richness plot graph. Each respective waterways upstream CMIG grade is positioned to the right and its paired downstream CMIG grade is positioned to the left depicting the difference in CMIG grade and overall waterway health. Red is the Wollangambe River (Clarence Colliery), Orange is Bargo River (Tahmoor Colliery), Grey is Springvale Creek (Springvale Colliery), Black is the Wingecarribee River (Berrima (Medway) Colliery), Green is Sawyers Swamp (Springvale and Angus Place Colliery), Yellow is Dalpura Creek (Canyon Colliery), Purple is Kangaroo Creek (Angus Place Colliery) and Blue is the Georges River (West Cliff Colliery).

Conclusions

Results of the CMIG/SIGNAL grade comparison, CMIG calculated mean scores and the CMIG/Family Richness plot graphs show the great potential of using the grading system as a robust, relatively low impact and rapid method for the assessment of Coal Mine Waste Water impacts to the receiving waterways aquatic ecosystem. Coal Mine Waste Water impacts to receiving waterways are regulated by the New South Wales Environmental Protection Authority to ensure the protection of the receiving waterways aquatic ecosystem, though the regulation and subsequent ‘protection’ performed through water quality and chemical concentrations never actually assess the aquatic ecosystem. It should be noted that the grades derived here may only suit Sydney Basin Coal Mining impacts and in fact perhaps only those which mine ore from identical and or similar coal seam measures. This is certainly a preliminary grading system and will benefit with use, increased data sets and the reworking of each individual taxa’s GMIG grade over time. Though these grades are only preliminary, they are the only grades which in fact assess the impact of coal mine wastewater discharges to a receiving waterways aquatic ecosystem and would provide a much better monitoring system than generalised water column pollutant concentration limits.

Research significance

A final outcome of this research is the preliminary development of a “coal mine sensitive and tolerant” rapid assessment tool or guideline for future ecological monitoring of the impacts of coal mine waste waters on their

receiving waterways ecosystem not just water chemistry. This rapid bioassessment index can be used in the future to rapidly measure the ecological impact of coal mines wastes. This methodology can improve regulation of coal mines. It helps ensure that the water quality and chemistry properties specified in coal mine wastewater environmental regulations can actually measure their success at protecting the aquatic ecosystem they are designed to protect.

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Chapter 9

Conclusions

The findings of this research have revealed major flaws in the current environmental regulation of seven licenced coal mine wastewater discharges within the Sydney Basin. This comprehensive research has shown that at many levels the current NSW EPA water pollution regulation of the seven coal mines fails to protect the downstream waterway from the pollution. The NSW EPA regulation of coal mine wastewaters focus on the waste attributes at the ‘end-of-pipe’ waste discharge. Along with the NSW EPA method of “monthly focus” on a selection of colliery wastewater pollutant concentrations, these approaches do not fully take into account the impact of the waste discharges on the receiving waterways aquatic ecosystems.

Coal mine wastes do not only cause impacts to aquatic ecosystems but are likely causing unknown impacts to the surrounding terrestrial ecosystem. Bioaccumulation of pollutants in fruit and timber trees grown within land contaminated by coal mine spoils was reported by Maiti et al., (2015) along with Wislocka et al., (2006). This research has shown more specifically that pollutants from one coal mine (Clarence Colliery) wastewater discharge appears to be mobilising trace metals which are bioaccumulating in the riparian terrestrial environment (Chapter 3; Belmer et al. 2018). It is very likely that this is not the only coal mine that results in the bioaccumulation of colliery waste pollutants in different organisms and or in the broader ecosystem. It should be noted that this one coal mine (Clarence Colliery) is still in operation and it is possible that undocumented bioaccumulation of colliery pollutants may well be occurring and could be of a greater magnitude at the closed, inactive mines investigated in this research. For example, the closed Canyon Colliery (Chapter 4; Belmer and Wright 2019 and 5; Belmer and Wright 2019) has some of the highest concentrations of metals recorded below the discharge of its waste, when compared to the other six mines in this study. The potential mobilisation and bioaccumulation of pollutants from the Canyon Colliery is highly problematic as the mine wastes flow into a high conservation area (The Blue Mountains National Park a part of the Greater Blue Mountains World Heritage Area; Chapter 4; Belmer and Wright 2019).

The Greater Blue Mountains National Park is perhaps the most sensitive and ‘highly protected’ conservation areas in the world. The impacts of coal mine water pollution in this location reported by (Chapter 4; Belmer and Wright 2019 and 5; Belmer and Wright 2019; Belmer et al 2014 and Wright et al 2017) has recently gained international attention. The. It has been assessed as a ‘high threat’ by IUCN in their 2020 review of the conservation outlook for the Greater Blue Mountains World Heritage Area (IUCN 2020). In 2017 the NSW EPA modified the Clarence Collieries waste discharge limits for Zinc from 1500ug/L to 8ug/L vastly improvinmg the allowable concentrations of zinc being discharged to the Wollangambe River. These licence modifications were the direct result of the recommendations discussed by Belmer et al 2014 and Wright et al 2017 and the subseautent Office of Environment and Heritage investigation (Belmer et al 2015; Wright et al 2017 and OEH 2015).

This research has critically evaluated the failings of the current and historic processes used by the NSW EPA to protect the aquatic environment through legislation and the regulation of contaminants within coal mine wastewaters in the Sydney Basin (*Protection of the Environment Operations Act 1997*). Results show that inactively mined wastewaters (such as drainage waters emerging from Berrima Colliery and Canyon Colliery) are causing a greater

water quality impact to the receiving waterways aquatic ecosystem than actively mined wastewaters (Chapter 4; Belmer and Wright 2019). This research has found that once mining ceases and pollution licences are surrendered (e.g. Canyon Colliery) the contaminated wastewaters from the closed mines continue to pollute unmitigated, and in the case of Canyon Colliery, at higher concentrations than from active mines (Chapter 4; Belmer and Wright 2019, 5; Belmer and Wright 2019 and 6; Belmer and Wright 2019).

Continuing discharge of contamination from closed coal mines, such as Canyon Colliery (closed in 1997), are left unmitigated, unlicensed and can now, 20 years after closing, be considered as unregulated pollution discharges. They are creating legacy pollution that can be more harmful to the receiving aquatic environment than the licenced and regulated water pollution from active mines (Chapter 4; Belmer and Wright 2019, 5; Belmer and Wright 2019 and 6; Belmer and Wright 2019).



Figure 2. Canyon and Berrima (Medway) Collieries coal mine wastewater discharges (Nakia Belmer 2017).

Some similar international case studies are the abandoned coal mines investigated by Cairney and Frost (1975), Younger (1993) and Banks et al. (1997) in the United Kingdom. The coal mines investigated ceased mining during the 1960's and the 1980's yet continue to pollute. Reduced pH generating acid mine drainage was still reported decades later along with increased sulfate concentrations (Younger 1993, Banks et al. 1997), similar to this research. Of great concern is the levels of sulfate reported by Banks et al. (1997) which is much higher than reported at Berrima (Medway) Colliery and Canyon Colliery in this research (Chapter 3; Wright et al. 2019 and 4; Belmer and Wright 2019). It is likely that the sulfate levels within the wastewater at Berrima (Medway) and Canyon Colliery will continue to increase unmitigated for decades. In fact, the findings of this research (e.g Canyon Colliery and Berrima (Medway) Colliery) suggest that if measures are not undertaken to mitigate pollution post mining, it may well be irrelevant to treat the wastewater during mining as the negative fallout post mining could overshadow water pollution and impacts to aquatic life offered "protection" during the mining period. For instance, mean zinc from the four active mines measured 46.4, 36.5, 23.3 and 13.8 $\mu\text{g/L}$, whilst in contrast the inactive mines measured mean zinc concentrations of 315.7, 228.3 and 38.2 $\mu\text{g/L}$ at Berrima (Medway), Canyon and Angus Place Collieries respectively. The ANZECC guideline for zinc at 99% protection is 2.4 $\mu\text{g/L}$, hundreds of times less than the recorded levels

(Chapters 3; Wright et al. and 3; Belmer and Wright 2019). It must be stated that Berrima and Angus Place collieries did not even have discharge limits for zinc and although Canyon colliery did have a limit for zinc, the discharge limits were 5000 ug/L (Wright et al 2011). Perhaps of greatest significance is the steep increase of zinc concentrations at Berrima colliery which increased sharply after mine closure and the subsequent flooding of the mine shafts. This is similar, albeit faster than has been observed in the United Kingdoms closed coal mines (Jackson 1981, Younger 1993 and Younger 2001) supporting the suggestion that post closure impacts to the receiving waterways ecosystem are far worse than whilst mining is still occurring.

This research has conducted some of Australia's most detailed research investigating colliery wastewater contaminants in water, sediment and the subsequent bioaccumulation in waterway biota (Chapters 2; Belmer and Wright 2018, 3; Wright et al. 2019, 4; Belmer and Wright 2019, 5; Belmer and Wright 2019 and 6; Belmer and Wright 2019). This novel research is likely to have many implications as it demonstrates that at least one coal mine (Clarence Colliery) is releasing wastewater contaminants which are bioaccumulating and or biomagnifying in the waterway's sediments, their aquatic biota and terrestrial riparian vegetation (Chapters 5; Belmer et al. 2019 and 7; Belmer et al. 2019). The investigation of coal mine pollution in the GBMWA World Heritage Area's Wollangambe River and the Clarence Colliery indicates that contaminants are moving from the water column, sediment and are bioaccumulating within the aquatic biota of their receiving waterways demonstrates that serious long-term legacy pollutant impacts are likely to persist. The pollutants are also leaving the water column and many are accumulating in the river sediment (Chapter 5; Belmer et al. 2019). Perhaps of additional concern for such a high conservation-value area is the high likelihood that many pollutants are also being transported to the terrestrial environment via the roots of riparian vegetation, of which the extent of the contamination may be more far spreading than this study has found (Chapter 2; Belmer and Wright 2018). This study could be the first to investigate the mobilisation of metals from a regulated coal mine wastewater discharge to terrestrial growing riparian vegetation (*Acacia rubida*) downstream of a mine waste discharge. It is noteworthy that the contamination is occurring within a highly regulated coal mine that discharges its wastewater into a high conservation area (The World Heritage Blue Mountains National Park) (Belmer et al. 2014 and Chapter 2; Belmer and Wright 2018). While the Clarence Colliery is currently actively mined, the Canyon Colliery closed in 1997 and it is likely that similar bioaccumulation impacts are occurring there.

There are many previous studies investigating water column pollution from metal contamination from a broad range of mining activities. Many investigate the links between metal mining impacts on water chemistry, river sediments and aquatic flora and fauna (Hill et al. 2000, Amish and Cowx 2000). The majority of these studies have found that impacted sample locations, from an array of differing metal mining activities, have increased heavy metal contamination of waters (Ashraf et al. 2011, Maiti et al. 2015) and soils (Neff 1984, Wang and Rainbow 2008) worldwide. These investigations have reported contaminated waters and soils with increased metal concentrations within many differing terrestrial and aquatic flora and fauna species (Neff 1984, Sericano et al. 1995, Hill et al. 2000; Amish and Cowx 2000, Wang and Rainbow 2008, Ashraf et al. 2011). This current study (Wright et al. 2017, and Chapters 2; Belmer and Wright 2018, 3; Wright et al. 2018, 4; Belmer and Wright 2019, 5; Belmer and Wright 2019 and 7; Belmer and Wright 2019) supports these findings with increases found in nine of the ten metals analysed. Sericano et al. (1995) and Ashraf et al. (2011) suggest heavy metals have the potential to accumulate within biota at

toxic concentrations and have chronic ecological impacts within ecosystem food chains. Wang and Rainbow (2008) raised concerns over the longevity of heavy metal pollutants in aquatic environments due to their ability to deposit into waterway sediments, which can potentially remain indefinitely. Kolaříková et al. (2012) found that bioaccumulation of pollutants within four macroinvertebrate species within the Elbe and Vltava Rivers in the Czech Republic were still persistent after water quality improvements were implemented. It was reported that these heavy metals concentrations were consistent over their 12-year study and they recorded no significant reduction in heavy metal concentrations other than mercury (Hg) (Kolaříková et al. 2012).

This current body of research only investigated contamination in one species of aquatic invertebrate downstream of one colliery and there is a strong possibility that similar contamination may be occurring in the waterways below the other mines in this study. Results show that metals have biomagnified from the water column, to the river sediment and the aquatic beetle (*Macrogyrus rivularis*). The beetle is long lived (approximately 24 months) and a predator (Watts and Hamon 2010) and is most likely consuming large volumes of contamination through predated on other contaminated macroinvertebrates. For instance, cobalt was below laboratory detection within the water column at reference sites whilst measurable at 26.5 µg/L, just below the mine and 2 µg/L, 18km below the mine. Cobalt was still undetectable in sediment at reference sites whilst measuring 551 mg/kg just below the mine and 6 mg/kg 18km downstream. Concentrations within beetles at the reference sites was measurable (87.2 and 209 mg/kg) and increased at both impacted sites to (6271 and 1089.8 mg/kg). This was also similar for nickel, at reference sites nickel was <0.5 µg/L below laboratory detectable limits within the water column, whilst the impacted sites recorded mean nickel of 76.7 and 17.7 µg/L. Nickel concentrations within river sediment at the reference sites was 2 mg/kg and below laboratory detectable limits, whilst at the impacted sites 606.7 mg/kg and 18km downstream 19.0 mg/kg. Nickel within beetles at reference sites measured 62.1 and 40.9 mg/kg, whilst just below the coal mine discharge nickel in beetles was measured at 4491.1 mg/kg and 18km downstream nickel recorded a mean of 932.9 mg/kg (Chapter 7; Belmer and Wright 2019).



Figure 3. *Macrogyrus Rivularis* (Nakia Belmer 2018).

This research has revealed information that provides a more detailed understanding of the broader impacts that the heavy metal contaminants from Clarence Collieries coal mine wastewaters being discharged to the Wollangambe River are having on the aquatic and terrestrial environment. The implications that the licensed wastewater discharge is mobilising pollutants to terrestrial riparian vegetation is of major concern and appears to be the first Australian research of this nature. The Environmental Protection Licensing (EPL) process used by the NSW EPA are driven by the POEO Act 1997 and are designed to protect the aquatic environment of waterways which receive coal mine wastewaters. The POEO Act 1997 states a hand full of “objects of the Act” which are to protect, restore and enhance the quality of the environment in New South Wales, having regard to the need to maintain ecologically sustainable development, to rationalise, simplify and strengthen the regulatory framework for environment protection, to improve the efficiency of administration of the environment protection legislation and to reduce risks to human health and prevent the degradation of the environment by the use of mechanisms that promote: pollution prevention and cleaner production, the reduction to harmless levels of the discharge of substances likely to cause harm to the environment, the making of progressive environmental improvements, including the reduction of pollution at source and the monitoring and reporting of environmental quality on a regular basis (POEO Act 1997).

Discharge limits set by EPL’s stipulate levels of pollutants that are permitted to be discharged within the colliery wastes at the ‘end of pipe’. This strongly influences the concentration of pollutants that are present in the water column of a receiving waterway, as seen in the Wollangambe River (Belmer et al. 2014, Wright et al. 2017 and Chapter 4; Belmer and Wright 2019), but the EPL does not take into account the surrounding terrestrial environment. Setting pollutant limits to EPL’s that apply only to pollutant concentration in the ‘end of pipe’ may well be of limited value if the heavy metals are able to mobilise and bioaccumulate within the aquatic ecosystem and terrestrial riparian vegetation. In the case of Clarence Colliery the terrestrial environment 1 km below the mine waste discharge is part of the Greater Blue Mountains World Heritage Area (Belmer et al. 2014, Wright et al. 2017). This represents a discord in the EPL process which is unsatisfactorily protecting the greater environment from coal mine wastewater discharges. Currently the regulation focuses on concentrations within the water column whilst disregarding loads within stream sediments. Load limits such as (tonnes/year) or measures of a similar nature should be taken into consideration by the NSW EPA to attempt to mitigate the bioaccumulation of pollutants within stream sediments, biota and or terrestrial vegetation. Especially in the scenario of Clarence Colliery, with such a high level of protection offered at the legislative level (Nationally and Internationally) but failing at the regulatory level. The IUCN Conservation Outlook (IUCN, 2020) has identified that water pollution from coal mines outside of the GBMWhA boundary are a ‘high threat’ to the conservation values of the World Heritage estate. This research shows that regulation of the coal mines wastes by the NSW EPA, prior to 2017, allowed pollutants to enter the food chain of the Wollangambe River within the world heritage area.

Chapters 4; Belmer and Wright (2019), 5; Belmer and Wright (2019), 6; Belmer and Wright (2019) and 7; Belmer and Wright (2019) provide a detailed comparison of the water quality, sediment quality and ecological impacts of active and inactive coal mines from their coal mine wastewater discharges. The ecological impacts of coal mine wastewaters discharged to receiving waterways through the use of benthic macroinvertebrates is not well studied in Australia. This is not the case internationally with many studies performed in North America including in the Indianan, West

Virginian and the Appalachian Mountain coal fields (Brake et al. 2001, Petty et al. 2010, Merriam et al. 2011), as well as the United Kingdom throughout the Durham coal fields and Southern Yorkshire coal fields (Banks et al. 1997, Younger 2000, Younger 2002). The handful of international studies using a regional group of mines in the United States had similar findings to this research with increased sulfate (Petty et al. 2010), salinity (Pond et al. 2008, Petty et al. 2010), nickel and zinc (Brake et al. 2001, Pond et al. 2008), along with modified pH generating acid mine drainage (Banks et al. 1997). The lack of Australian literature on this topic is puzzling given that the number of closed and abandoned mines in Australia, which is currently growing (Unger et al. 2012); yet, so many have ongoing environmental problems (Battaglia et al. 2005).

Results of this study show that all the waterways receiving mine effluent had similar healthy macroinvertebrate communities upstream of both active and inactive coal mines (Chapters 3; Wright et al. 2018 and 6; Belmer and Wright 2019). The major exception was the Bargo River where macroinvertebrate communities in the river were only mildly different downstream, compared to upstream, indicative of a very mild ecological impact (Chapter 6; Belmer and Wright 2019). In comparison, most other coal mines, either active or inactive, showed that the ecological condition (according to macroinvertebrates) downstream of coal mine wastewater discharges were generally moderately to highly degraded, compared to upstream of each respective coal mine (Chapter 6; Belmer and Wright 2019). This research is one of only a few internationally that investigates impacts to aquatic macroinvertebrates from a group of mines, let alone active and inactive mines. It is also the first Australian investigation to study a group of coal mines and investigate their water and sediment quality and ecological impact. Results from this research (Chapter 6; Belmer and Wright 2019) conclude that the discharge of coal mine wastewaters to surface waters are having varying negative impacts to the receiving waterways aquatic ecosystem, whether mining of coal is active or inactive. This research has showcased the lack of actual real world environmental protection when regulation is driven by “end of pipe” water column concentrations and not a measure of the aquatic ecosystem. The research from chapters 3; Wright et al. (2018), 4; Belmer and Wright (2019), 5; Belmer and Wright (2019), 6; Belmer and Wright (2019) and 7; Belmer and Wright (2019) show adverse impacts of varying degrees to the receiving waterways aquatic ecosystem whilst that aquatic ecosystem is fundamentally being protected. To not consider the cumulative impacts, from the initial impact of the toxicity of the discharge on the macroinvertebrate community to the eventual bioaccumulation of the pollutants, is a failure of environmental protection.

The study’s findings are a reminder that active and closed coal mines can both be a major source of water pollution and ecological degradation of waterways receiving the wastes. Berrima (Medway) and Canyon Collieries, (Chapters 3; Wright et al. 2018 and 4; Belmer and Wright 2019) both demonstrate that ecologically hazardous water pollution may occur for years after underground coal mines cease mining. Unlike the United Kingdom or North America (Robb 1994, Younger 2004, Verb and Vis 2000), there has been limited research investigating legacy water pollution from closed coal mines in Australia. Battaglia et al. (2005) is one of the few Australian studies that assessed residual pollution and ecological degradation from a waterway affected by a closed coal mine. This case study highlights that after mine closure, increased untreated and unmanaged acid mine drainage is likely to be triggered driving metal concentrations to increase within the receiving waterway. Canyon Colliery is an important case study for this current research as it ceased mining in 1997 yet continues to release some of the highest concentrations of nickel and zinc of

all the active and inactive mines used in this research (Chapter 4; Belmer and Wright 2019) (Table 1). This builds upon previous studies that have documented some aspects of the water pollution caused by this mine (Wright and Burgin 2009, Price and Wright 2016) and degradation of the ecology of the receiving waterway further downstream (Wright and Burgin 2009, Wright et al. 2011, Wright and Ryan 2016). This current research studied this important case study in more detail than previous studies, and produced the first ecological study of Dalpura Creek above and below the mine adit that continues to release contaminated drainage (Figure 2). Remarkably this coal mine remains an example of the many failings of the coal mine closure process to address residual pollution that continues to emerge from a closed coal mine (Table 1). The governance of coal mine closures is particularly questionable, as the NSW EPA allowed the coal mine owner to ‘surrender’ its EPL when commercial coal mining ceased, whilst contaminated drainage continued to flow into a high conservation value ‘protected’ area such as a World Heritage National Park (Dalpura Creek into the Grose River). The IUCN’s 2020 assessment made reference to coal mine water pollution as a ‘high threat’ to the conservation values of the Greater Blue Mountains World Heritage Area. It also highlighted water pollution of the Grose River from this mine in particular. This research highlights the failure of the coal miner to conduct any remediation of the coal mine to address pollution emerging from this mine in the midst of such a high conservation-value area.

Table 1. Mean wastewater discharge concentrations of nickel and zinc of all seven mines ($\mu\text{g/L}$) showing both Canyon and Berrima (Medway) Collieries elevated nickel and zinc concentrations.

Colliery name	Nickel ($\mu\text{g/L}$)	Zinc ($\mu\text{g/L}$)
Clarence (Active)	31.3	36.5
Springvale (Active)	10.3	46.4
Tahmoor (Active)	28.8	23.3
West Cliff (Active)	64.8	13.8
Angus Place (Inactive)	3.8	38.2
Berrima (Medway) (Closed)	72.4	228.3
Canyon (Closed)	186.3	315.7

This research prompts questions about whether the New South Wales Government could improve its governance of water pollution associated with coal mine closures. For example, the Berrima Colliery was still in the closure process when it was investigated in a 12-month study, by Wright et al. (2018) (Chapter 3). This mine discharged much higher concentrations of zinc, nickel (Table 1), salinity and manganese after it ceased mining and was partially flooded, compared to when it was operating (Chapter 3; Wright et al. 2018). The research triggered action to address some improvement of the water pollution through the implementation of post mining discharge limits imposed on the colliery (NSW EPA 2017). It could be concluded that without this research no action would have been implemented to address the pollution.

Table 2. Timeline of the closure of Berrima (Medway) Colliery and research impact events triggered by research of this thesis.

Year	Event
2013	Coal mine ceases mining activity
2015	Boral release mine closure plan
2016	WSU starts sampling mine effluent and river (Thesis Author and Supervisor)
2017	Media interest in WSU research, subsequent publication demonstrating increasing zinc and nickel concentrations

2018	EPA issue directive to Boral
2019	Boral install water treatment
2020	Boral install barriers to seal contamination within mine

Whilst water pollution and the ecological impacts from closed coal mines is not yet commonly studied in Australia, it has been the focus for many studies internationally (e.g. Cairney and Frost 1975, Younger 1993, Brake et al. 2001, Johnson 2003). Results of the Berrima (Medway) Colliery investigation questions the effectiveness of the environmental regulatory regime imposed on this mine by the NSW EPA which provided inadequate restrictions of the pollutants released by the mine through the mine drainage adit to the Wingecarribee River. The Berrima (Medway) Colliery holds an environmental protection license (EPL no. 608), enforced by the NSW EPA (NSW EPA 2017a), which permits the discharge of mine drainage to the Wingecarribee River. The license only identified four pollutants (oil and grease, total suspended solids, pH and biochemical oxygen demand) with specific discharge limits for each, in the colliery drainage. It is well documented that all the mines used in this research are discharging wastewaters contaminated by a plethora of heavy metals and metaloids. This lack of identification of the calidascope of pollutants being discharged at Berrima (Medway) Colliery and in fact all other mines is a grose lack of stewardship by the NSW EPA. It also should be noted that the Wingecarribee River is one of the largest rivers that flows into the protected Lake Burragorang empounded behind Warragamba Dam, the source of over 80% of the Sydney Basin’s drinking water supply.



Figure 4. Berrima (Medway) Colliery untreated “acid mine drainage” (orange) entering the Wingecarribee River in 2017, four years post closure (Nakia Belmer 2017).

The research at Berrima (Medway) Colliery was performed in conjunction with the miner (Boral) and the NSW EPA and are aiding in the implementation of mitigation measures implemented within the mine workings.

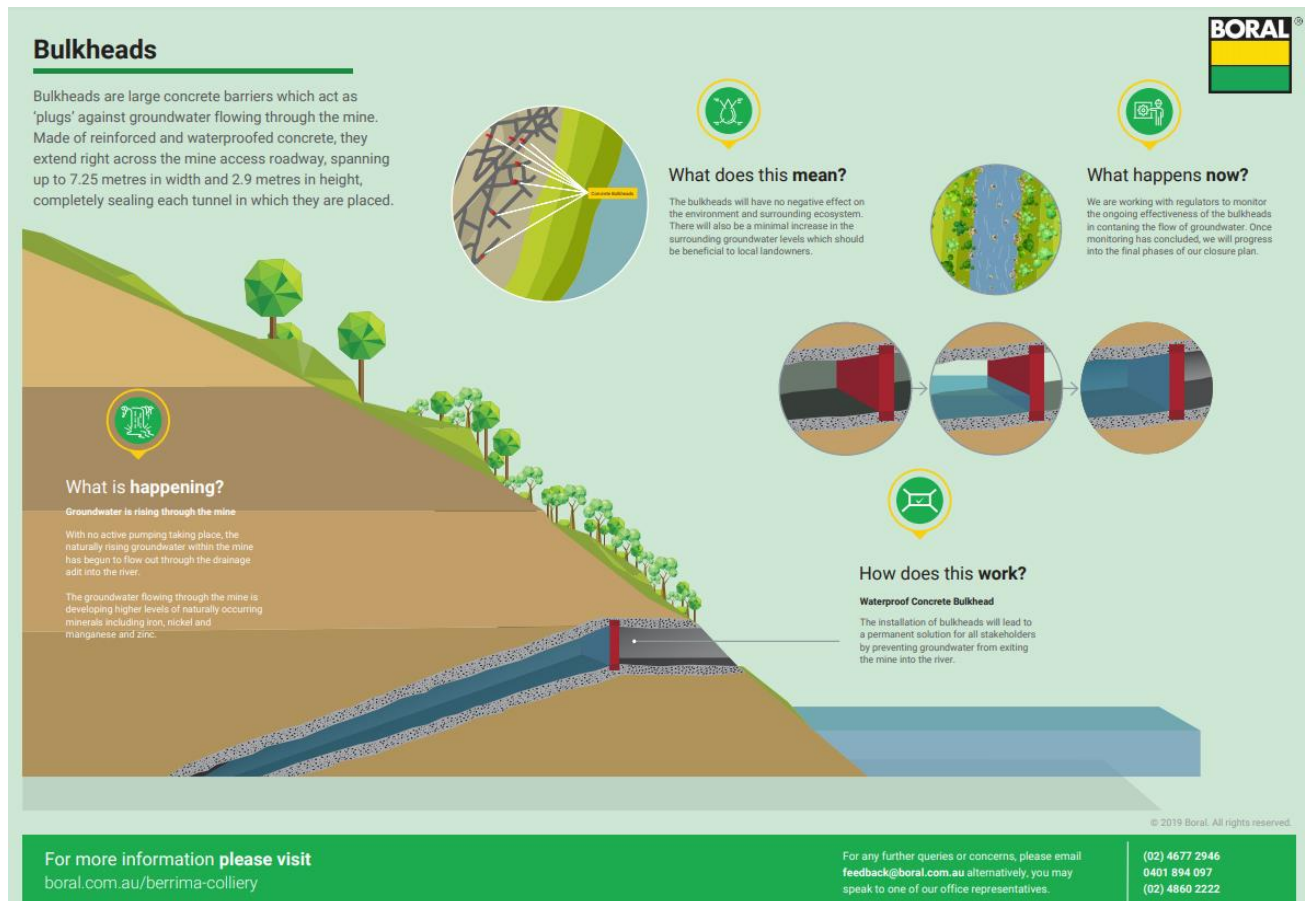


Figure 5. Boral Bullhead diagrams, apart of the ongoing closed mine working mitigation works (Boral 2019).

The outcomes of the Berrima Colliery investigation is perhaps one of the biggest research impacts of this body of work. This research has been touted as “World Leading” and considered an “exemplar of international best practice in environmental regulation” (Nature Research Custom Media 2018). The notice of variation of licence 608 document clearly states that the investigation and subsequent variation was driven by the partnership between the NSW EPA and Western Sydney University (NSW EPA 2017). The research performed for the completion of this thesis (Chapters 2; Wright et al. 2018 and 3; Belmer and Wright 2019) has helped guide the NSW EPA to improve the NSW Governments environmental regulation of coal mines (Table 2). In the case of Berrima (Medway) Colliery, ongoing remediation procedures are underway to treat the “inactive” mine wastewater and the NSW EPA has stipulated water quality and chemistry concentrations as part of a post mining Environmental Protection Licence, another major achievement of this research and the first for post coal mine wastewater discharges (NSW EPA 2017). The five most ecologically hazardous pollutants in Berrima (Medway) Collieries wastewater discharge revealed in Chapter 3; Wright et al. (2018), nickel, zinc, manganese, iron and salinity, were not specified in the active mines license (EPL 608) and

have subsequently being unregulatedly discharged for decades at potentially toxic levels. When this research first investigated the mine drainage that was being released by the Berrima mine it was noted that the mine drainage received no treatment. Such a deficiency in the Berrima mine’s environmental license (EPL no. 608) is not isolated to this case, as regulation of specific water pollutants in waste discharges to environmentally appropriate levels has been found to be lacking at many coal mines (Table 3) and other industrial discharges in NSW (Wright et al. 2011, Graham and Wright 2012). It must be stated that the Wingicarrabee River is one of the largest rivers to flow directly into Lake Burrorang and Warragambah Dam and the river flow makes up a large percentage of Sydney’s Drinking Water supply, well documented by ABC News (Greg Miskelly 2019).

Table 3. The NSW EPA Environment Protection Licence (EPL) pollutant limits for discharge of colliery wastes to streams/river (EPA, 2018). Each colliery has an individual EPL licence and a unique licence number. The letter (d) after the EPL pollutant limit refers to the samples requiring filtering, and the pollutant concentration represents the dissolved (d) fraction. The EPL concentration limits apply for different proportions of times, expressed as percentiles: (*) = 100 percentile of the time, (**) 90 percentile of the time. Two waterways receiving waste from Springvale Colliery were investigated, the ‘main’ discharge and ‘minor’ discharge. # Note EPL 558 has been surrendered. A dash (-) = no concentration limit applied for that pollutant, as you can see many pollutants are not regulated and thus allow the discharge of those pollutants at any concentration (even toxic) to the receiving waterway.

Pollutant attribute (units of measurement)	Angus Place Colliery EPL 467 (*)	Berrima (Medway) Colliery EPL 608 (*)	Canyon Colliery EPL 558 (*) #	Clarence Colliery EPL 726 (*)	Springvale Colliery EPL 3607 Main (*)	Springvale Colliery EPL 3607 Minor (*)	Tahmoor Colliery EPL 1389 (*)	Westcliff Colliery EPL 2504 (**)
pH (pH units)	6.5-9.0	6.5-8.5	-	6.0-8.5	6.5-9.0	6.5-9.0	6.5-9.0	6.5-9.3 (*)
Oil and Grease (mg/L)	10	10	10	10	10	10	10	10 (*)
EC (µS/cm)	-	-	-	-	1200	-	2600	-
TSS (mg/L)	30	50	-	30	50	30	30	50 (*)
Turbidity (NTU)	-	-	-	-	50	-	150	-
Nitrogen (µg/L)	-	-	-	250	-	-	-	-
Aluminium (mg/L)	-	-	-	-	450 (d)	-	-	800 (d)
Arsenic (µg/L)	-	-	-	23 (d)	24 (d)	-	200	19 (d)
Boron (µg/L)	-	-	-	100	370	-	-	-
Cadmium (µg/L)	-	-	-	0.2 (d)	-	-	-	0.5 (d)
Chloride (mg/L)	-	-	-	25	-	-	-	-
Chromium (µg/L)	-	-	-	1 (d)	-	-	-	-
Cobalt (mg/L)	-	-	-	2.5 (d)	-	-	-	20 (d)
Copper (mg/L)	-	-	-	1.4 (d)	7 (d)	-	-	18 (d)
Iron (µg/L)	-	-	1000	300 (d)	400 (d)	-	-	-
Manganese (mg/L)	-	-	-	500 (d)	1700 (d)	-	-	40 (d)
Fluoride (µg/L)	-	-	-	1000	1800	-	-	-
Lead (µg/L)	-	-	-	3.4 (d)	-	-	-	6 (d)
Mercury (µg/L)	-	-	-	0.06 (d)	-	-	-	-
Nickel (µg/L)	-	-	-	11 (d)	47	-	200	200 (d)

Selenium ($\mu\text{g/L}$)	-	-	-	5	-	-	-	-
Silver ($\mu\text{g/L}$)	-	-	-	0.05	-	-	-	-
Sulfate (mg/L)	-	-	-	250	-	-	-	-
Zinc ($\mu\text{g/L}$)	-	-	5000	8	50 (d)		300	84 (d)

Many parts of this research have positive environmental research impacts which continue to help guide the NSW EPA in better regulating the levels of contaminants from these coal mine wastewater discharges. It has gathered evidence that has assisted the NSW Resource Regulator demand that Boral take action to reduce pollution from the closed coal mine for those pollutants of most concern. Many of the NSW EPA licences for the collieries in this study have been progressively modified to reduce the pollution of waste-receiving waterways through direct association of the research findings of this thesis. Perhaps the greatest improvement is the Clarence Colliery (table 3). This EPA licence was modified in July 2017 to reduce the concentration of several pollutants permitted to be discharged into the Wollangambe River as a result of previous research undertaken by the Author of this thesis and his Supervisor (Belmer et al. 2014 and Wright et al. 2017). The research found that this mine, which discharges wastewater to a high-conservation value river, causes significant water pollution and severe ecological damage extending 22 km below the mine, deep within the Blue Mountains National Park (World Heritage Area). This pollution received international prominence in December 2020 when IUCN recognised that water pollution from this mine is a ‘high threat’ as it is causing major contamination of the Wollangambe River within Greater Blue Mountains World Heritage Area (IUCN, 2020). This research (Belmer et al. 2014) was reported to the NSW EPA and this triggered the NSW OEH to investigate the findings and ultimately drove changes in the collieries EPL (NSW OEH 2015). For example, the new EPA licence for Clarence Colliery reduced the permitted concentration of zinc in the colliery wastewater by 99.5 %, from 1500 $\mu\text{g/L}$ permitted in the previous licence, to 8 $\mu\text{g/L}$ (EPA 2018). The new EPA licence also specified a permitted concentration for nickel of 11 $\mu\text{g/L}$. The previous licence had not specified any discharge limit for nickel (EPA 2018). It is anticipated that the colliery will upgrade the treatment of their wastewater to conform to the requirements of the new licence. Along with the outcomes at Clarence Colliery and Berrima (Medway) Colliery, more recently (March 2020) another coal mine wastewater EPL was modified due to the findings of chapter 4’s publication (Belmer and Wright 2019) at the Westcliff Colliery. The Westcliff Colliery released wastewaters to the Georges River which increased salinity concentrations nearly 10 times within the river (148.2 $\mu\text{s/cm}$ upstream and 1256 $\mu\text{s/cm}$, pH increased significantly from 6.3 pH units upstream and 8.92 pH units within the river downstream. Chloride, sulphate, carbonate and bicarbonate all increased in the Georges River downstream of the mine wastewater discharge (28.8 to 99.8 mg/L, 5.8 to 18.2 mg/L, below laboratory detection to 93.9 mg/L and 18 to 52.6 mg/L) respectively. The EPL initially had no limits for salinity, chloride, bicarbonate or carbonate and under “special conditions” of the current EPL salinity is regulated at 400 and 600 $\mu\text{s/cm}$ and bicarbonate alkalinity at 185 mg/L. Along with these two pollutants aluminium, cobalt, copper, nickel, zinc and total nitrogen have been added and given discharge limits (NSW EPA 2021).

One specific avenue that this research investigated was how could routine assessment of aquatic ecosystems, using river macroinvertebrates, be used to measure impacts of coal mine wastewaters on rivers, rather than rudimentary water column concentrations which have been found to offer limited protection for the ecosystem. As part of this research macroinvertebrate biotic indices were calculated seeking a rapid, sensitive and efficient methodology for

routine assessment for surveying actual ecosystem impacts from coal mine wastewaters. The grading system was developed to detect and measure any negative impact to a receiving waterways ecosystem (Chapter 3; Wright and Belmer 2018, 6; Belmer and Wright 2019 and unpublished Chapter 8). A much better focus than the current simple “end of pipe” monitoring focus. Popular indices such as SIGNAL (Chessman 1995, Chessman 2003) and their qualitative measures were found to be relatively insensitive for measuring the degree of ecological impairment associated with coal mine wastes (Chapter 8). This is to be expected as SIGNAL was initially calibrated to measure the macroinvertebrate response to organic and urban pollution on waterways (Chessman, 1995). For instance, community structure of EPT taxa (three orders of known sensitive biota) was impacted, though it was found that the community structure was modified from known sensitive taxa of the EPT order (Ephemeroptera, Plecoptera and Trichoptera) being replaced with a less sensitive EPT taxa. This shows that although a robust method (EPT analysis), the sensitivity of such biotic indices may not completely portray the impact in a coal mine wastewater scenario. The loss of 17 potential “coal mine wastewater” sensitive taxa was observed from waterways affected by mine wastes from all seven mines in this study. The loss of these individual taxa was an important element in the development of a coal mine sensitive macroinvertebrate taxa list which could be used as a rapid assessment tool for the assessment of coal mine wastewaters ecological impacts to their respective receiving waterways (Chapter 8). Preliminary data from (Chapter 8) indicates that this method has encouraging signs. For instance, when compared to the well developed SIGNAL method (Chessman 1995, Chessman 2003), the derived grades in Chapter 8 show similarity for known sensitive species (Table 4). Although similar, the CMIG grades have adjusted the sensitivity of some of the SIGNAL graded taxa slightly higher or lower. This shows the importance of the CMIG grades to better assess direct coal mine wastewater impacts.

Table 4. Coal Mine Impact Grade (CMIG) (Belmer and Wright unpublished and SIGNAL (Chessman et al. 1995) grades comparison for sensitive taxa.

TAXA/SIGNAL grades	Coal Mine Impact Grade (CMIG)	SIGNAL
Calamoceratidae	10	7
Calocidae	7	9
Coloburiscidae	10	8
Conoesucidae	10	6
Eustheniidae	10	10
Glossomatidae	10	9
Gripopterygidae	8	8

Table 5 is a selection of slightly sensitive SIGNAL grade taxa in relation to their CMIG grades and shows how some SIGNAL graded taxa are more tolerant of coal mine wastewater, in turn lowering their derived CMIG grade in line with the impact of the wastewater rather than the urban water quality stressors driving SIGNAL grading (Chapter 8).

Table 5. Coal Mine Impact Grade (CMIG) (Belmer and Wright) and SIGNAL (Chessman et al. 1995) grades comparison.

TAXA/SIGNAL grades	Coal Mine Impact Grade (CMIG)	SIGNAL
Baetidae	4	5
Caenidae	3	4
Hydrobiosidae	6	8
Hydropsychidae	3	7
Hydroptilidae	2	7
Notonemouridae	3	6

Athericidae	3	8
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The grading system was created and assessment of its robustness was performed between comparisons of the SIGNAL grading system (Chessman 1995, Chessman 2003) used to assess impacts to urban waterways and the derived CMIG grading system described in (Chapter 8). Initial results show it is likely that the SIGNAL grading system is underrepresenting the level of impact, especially for three of the coal mines (Bargo River, Dalpura Creek and the Georges River) when compared to the newly derived CMIG grading system (Chapter 8). Although these grades are only preliminary, they are the only macroinvertebrate grades which in fact assess the impact of coal mine wastewater discharges to a receiving waterways aquatic ecosystem and would provide a much better monitoring system than generalised “end of pipe” water column pollutant concentration limits at monthly intervals.

This research has also shown that true “protection” of the colliery impacted waterways investigated in this study is not occurring, and the waterways aquatic ecosystem is unlikely to ever fully recover due to the legacy contamination that is occurring long after coal mines close (Cairney and Frost 1975, Jackson 1981 and Chapters 4, 5 and 6). This research is intended to help improve coal mine wastewater licensing and regulation, particularly to improve the assessment and protection of aquatic or terrestrial environments. This research has shown that environmental contamination may not just be occurring to waterways that are direct recipients of the wastewater (the waterway) but also the surrounding terrestrial environment may also be contaminated. This research shows that whilst coal mining is occurring, and the treatment of wastewaters continues, the ecosystem is generally not effectively protected. The assumption that waterways affected by coal mine wastes may perhaps return to their original pre-mining ecological condition post mining is perhaps expected but remains highly unlikely in reality. It is apparent that once a coal mining operation starts mining ore and discharging coal mine wastewater, the receiving rivers water quality and aquatic ecosystem will most likely be degraded indefinitely and reaching its most degraded once mining and any subsequent water treatment ceases. This research has triggered intervention from the NSW EPA showing that improved environmental regulation can be applied to reduce water pollution impacts.

This research has achieved six major outcomes which are listed below;

1. This research has produced the most comprehensive water chemistry data set (water chemistry including ionics and metals) on the contaminants in a group of coal mine wastes ever conducted from Australian collieries. Although there are other publications investigating similar impacts internationally, none have been conducted in such detail on a group of regional mines in Australia (Chapters 2, 4, 5, 6 and 7).
2. Until now, very few studies compare water quality between upstream and downstream of the entry of coal mine waste point source discharges– and none, ever, on upstream versus downstream water quality and ecological studies of a group of regional mines in Australia (Chapters 2, 3, 4, 5 and 6).
3. NSW regulation of coal mines wastes use Environmental Protection Licence’s that currently have a water pollution focus on water chemistry pollutants at the ‘end of the discharge pipe’. In few cases these regulations use ANZECC (2000) guidelines. The regulations generally ignore the instream impact on natural water quality and the health of the aquatic ecosystem that is exposed to the coal mine waste discharges. No data on the ecological impact,

using an upstream versus downstream design of a group of coal mines, has ever been performed in Australia until this study. It is now one of only a few in the world (Chapters 2, 6 and 7) (Brake et al. 2001, Pond et al. 2008 and Petty et al. 2010).

4. This research will help improve the application of scientific methods using river macroinvertebrates as a sensitive, rapid and effective indicator of the ecological health impact to the receiving aquatic environment from coal mine wastewater pollution. And has already helped in reducing pollution limits at both Berrima and Clarence Collieries (Chapter 8).

5. Until now there was very little data on the nature and magnitude of metal pollutants from coal mine wastewaters to the receiving waterways river sediments within Australia and Internationally. There have been few, if any, studies on river sediments above and below a regional group of coal mines (Chapter 5) (Maiti et al. 2015).

6. Until now there was no literature or understanding of the broader implications of bioaccumulation and or biomagnification of metals from coal mine wastewaters into aquatic invertebrates or riparian plants in Australia. This research is the first assessing the bioaccumulation of pollutants in the aquatic community and the mobilisation of the wastewater pollutants to the surrounding terrestrial vegetation (Chapter 6).

Recommendations

It is apparent that long-term rehabilitation strategies are needed at all the closed mines used in this study, along with future planning for the eventual closure of those coal mines that are still actively operating. This research has shown that the river water column pollution concentrations, although provided some protection from EPA coal mine pollution discharge limits, require more effective environmental regulations. This research reveals that many pollutants are unregulated and others are at hazardous concentrations. EPA regulation of coal mines should ensure that pollutants are not bioaccumulating and or biomagnified in aquatic and riparian foodchains.

It is recommended that at the operational coal mines improved wastewater treatment processes should be implemented to better protect the receiving waterways ecosystem. This should be led by the licencing system that regulates these discharge concentration limits and is driven by the continued improvement of the regulation of pollutants within each coal mines EPL. In addition, the implementation of regulation and treatment post-mining is urgently required if the waterway below the mine drainage is ever to return to a natural water quality and ecological health once mining ceases. Measures to reduce the pollution concentrations within the wastewater whilst mining is still in operation to much lower levels will benefit the receiving waterways. This would entail reductions in the pollutants already identified by the NSW EPA in their regulations. In addition, those pollutants that are being discharged and are not currently regulated within each coal mines environmental protection licence should be added to the licence and regulated at levels that also reduce the impairment to stream ecology and the ongoing bioaccumulation. By reducing the licensed discharge limits the concentration and load of contaminants released from a coal mine to the receiving waterway will be reduced and it is likely will reduce the accumulation of contaminated sediment and the contamination within the aquatic and surrounding terrestrial environment. Ongoing monitoring of this should be undertaken and continued reductions should be implemented through more effective licences. Lower pollution concentrations could be achieved by improving many of the coal mines water treatment processes as most of the

mines used in this research use rudimentary precipitation treatment processes (Cohen 2002). Chemical precipitation is the most widely used form of treatment to remove heavy metals from wastewaters, this process requires the addition of chemicals to buffer the wastewaters pH to allow for the dissolved and suspended solids (metals) to fall out through sedimentation (Gautam et al. 2016). Chemical reduction is another treatment process where as sodium borohydride is used to effectively reduce the metals mercury, cadmium, lead, silver and gold from wastewaters along with the Xanthate Process though this is effective in removing large amounts of heavy metals it in turn replaces them with sodium and magnesium which may lead to a different water pollution problem (Gautam et al. 2016). Most likely the best solution is a membrane treatment process, these processes use fine polymer filtration such as electro dialysis and or reverse osmosis. Although greatly effective at reducing the levels of contaminants in wastewaters, this can be costly and is most likely more feasible whilst mining is still occurring (Gautam et al. 2016).

This research has made it apparent that measures to better protect waterways which receive untreated coal mine wastewaters from closed mines should be investigated by the NSW EPA to ensure that once coal is no longer mined the receiving aquatic ecosystem is still protected. There are two forms that could be used at the abandoned mines used in this research. One being a non-biological treatment process and the other a biological process. The non-biological techniques are simple though costly and often highly variable in their results and are used more so as mitigation controls rather than efficient long term prevention control. This technique simply “plugs” the hole as such by backfilling mine workings with soil, sand, cement and or paste. (Villian et al. 2013, Johnson and Santos 2020). An alternative treatment method worth investigating would be the absorption process. This is documented to be an economically feasible alternative to other treatment processes. Absorption uses materials such as activated carbon, clays, zeolites and or siliceous materials to absorb the heavy metals without releasing any new contaminants. Activated carbon is effective but like all other absorption processes there is a limit in how much heavy metals can be absorbed by the treatment material, eventually leading to a saturation point where the absorption material will no longer absorb the pollutants. At this stage the removal and replacement of the contaminated absorption material is required (Gautam et al. 2016). Another approach to backfilling is the emplacement of anoxic limestone drains or open limestone channels. This technique uses limestone trenches or pits which allows the wastewater to percolate through a layer of limestone which increases the pH of the Acid Mine Drainage and allows for precipitation of the pollutants. This is often used in conjunction with a wetland downstream of the limestone drains where the wetland is used as a biological treatment of the precipitated contaminants, as such allowing the constructed wetland ecosystem to accumulate the pollutants prior to being discharge further downstream to the receiving waterway (Johnson and Santos 2020).

Biochemical techniques such as constructed wetlands have been used effectively to remediate some Acid Mine Drainages, these include oxidative wetland and reductive wetland techniques. Oxidative (aerobic) wetlands are used as “compost bioreactors” and are shallow with large amounts of metal tolerant macrophytes (water plants) which are used to stabilise wastewater flow whilst absorbing precipitated pollutants. Vertical flow wetlands have been used since the 1980s and use a series of perforated pipes buried in a layer of limestone backfilled with a top layer of organic matter. The acid mine drainage flows through the organic and limestone layers in turn reducing sulfate concentrations and eventually neutralizing the acidity of the wastewater (Younger et al. 2002 and Johnson and Santos 2020).

Perhaps more feasible and cost effective are some emerging biological approaches to heavy metal removal, where the use of microorganisms which detoxify the heavy metals are used and or the phytoremediation technique where plants are used to decontaminate the metals from water and or soils through constructed wetlands (Gautam et al. 2016). The phytoremediation technique could be used to reduce contaminants within riparian soils, though perhaps not feasible in cases where the riparian community is within a natural setting such as a National Park. The phytoremediation technique, although it is still an emerging technology has shown to be more cost effective than chemical treatment. The microorganism treatment process uses bacteria, algae, fungi and or yeast that are naturally able to accumulate high levels of metals from the environment. These microorganisms have shown promise in removing the metals lead, cadmium, mercury, zinc, copper and silver (Gautam et al. 2016). A more industrial approach would be the construction of a bioreactor. A benefit of the bioreactor system is that the waste sludge generated is more feasible for re-use and being an engineered industrial pond manipulation of the biological response is more easily achieved than a natural constructed wetland that will have natural variation in its biological responses (Neculita et al. 2007). The bioreactor system has shown promise in a few settings such as the Caribou Mine in New Brunswick Canada, the Copper Queen Mine in Arizona and the Raglan Nickel Mine in Quebec Canada (Bratty et al. 2006, Johnson and Santos 2020).

Although there are many old and emerging techniques for treating coal mine wastewaters, all of the techniques have varying effectiveness in pollutant removal from wastewaters and different operating costs. The downside to using biological processes are that these constructed wetlands naturally vary as they rely mostly on natural processes that can vary from season to season, though this is still seen as the more cost effective method currently (Johnson and Hallberg, 2002 and Johnson and Santos 2020). Although the downsides of the biological process of pollutant removal is variable within a natural setting, i.e. natural or constructed wetland the bioreactor system where the construction of a bioreactor allows for greater control and manipulation of these biological processes is more favourable for the treatment of acid mine drainages (Johnson and Hallberg 2002 and Johnson and Santos 2020).

It is apparent that the chemical treatment of coal mine wastewaters is much more costly and has a much larger ecological footprint (Gautam et al. 2016). The precipitation method often generates large volumes of waste sludge which is less reusable than that generated in the bioreactor process. This treatment option may be the standard whilst coal mining is occurring and is offering some protection to the environment although this thesis shows that all mines investigated were underperforming in regard to protecting the environment. The best practice for operational coal mines would be a membrane treatment process such as electrodialysis and or reverse osmosis (Gautam et al. 2016). Biological treatment methods, especially the bioreactor system has shown great promise internationally in treating post mining Acid Mine Drainage in Canada (Bratty et al. 2006 and Johnson and Santos 2020). Although the bioreactor system requires greater start-up costs than that of natural and or constructed wetlands the contaminant load removal and subsequent ability to re-use the waste sludge make it a much more ecological sustainable treatment process and if cost effective should be implemented at abandoned coal mines (Younger et al. 2002 and Johnson and Santos 2020). Future research should investigate the performance of such treatment processes, especially once mining has ceased. Investigating biological Acid Mine Drainage treatment processes such as bioreactor systems or vertical flow wetlands at abandoned coal mines is a must. Chapters 2, 4, 5 and 6 clearly show the impacts associated with the closed or

inactive mines used in this research and demonstrates the need for post mining wastewater treatment. The example of Canyon Colliery, some 30 years post mining still impacting the Greater World Heritage National Park speaks volumes.

It is recommended that the NSW EPA investigate the findings of this research and continue to advocate improved waste treatment and also improve the effectiveness of EPL coal mine pollutant discharge limits to ensure protection of the receiving water quality, sediment quality, waterways aquatic ecosystem and reduce bioaccumulation of contaminants is achieved.

Of most concern is;

1. The implications that the coal mine wastewater contaminants are continuing to be released, or even increasing (eg Berrima Colliery), in the mine drainage water that is emerging from mines after mining ceases. This should be investigated and regulated by the NSW EPA. The EPA should not accept the surrender of any mine if they still contaminate the waterways or local environment.
2. The implications that the coal mine wastewater contaminants are causing greater degradation to the receiving waterways aquatic ecosystem after mining ceases (measured through aquatic macroinvertebrates) should be investigated by the NSW EPA. Careful preparation and monitoring is required to assess the water quality and ecological impacts after coal mines close.
3. The implications that the coal mine wastewater contaminants are accumulating in river and stream sediments due to coal mine discharges is currently unregulated and should be investigated and regulated by the NSW EPA.
4. The implications that the coal mine wastewater contaminants are bioaccumulating and biomagnifying in stream aquatic biota should be investigated by the NSW EPA.
5. The implications that heavy metal mobilisation to the terrestrial environment from coal mine wastewater emerging from both actively mined (treated wastewater) and inactively mined (untreated wastewater) is of concern and should be investigated by the NSW EPA. Such foodweb contamination in high conservation areas, such as the Greater Blue Mountains World Heritage Area should be investigated and regulated by the EPA.
6. Investigations into water pollution mitigation techniques at both active and closed coal mines is urgently needed.

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