CONTAMINATION OF AN UPLAND STREAM BY HEAVY METALS FROM AN OLD MINE SITE

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ABSTRACT

This paper presents the results of a detailed investigation of heavy metal contamination in an upland stream, Cadiangullong Creek, in New South Wales, Australia. Previous studies suggested enriched copper concentrations from an historical mine site, Big Cadia, were impacting upon organisms within the creek. Samples were obtained from sediments, pools and runoff from within Big Cadia, as well as creek bank soil, water, plant, biofilm and sediment from Cadiangullong Creek and a nearby reference stream, Panuara Rivulet. Samples were analysed for ten heavy metals: arsenic, cadmium, chromium, copper, iron, mercury, manganese, nickel, lead and zinc.

Big Cadia was found to be a major source of copper, iron and nickel in Cadiangullong Creek. Copper concentrations at the sites adjacent and downstream of Big Cadia were found to reach levels which may be toxic to the two fish species present in Cadiangullong Creek.

Key words: heavy metals, runoff, mining, water quality, contamination.

INTRODUCTION

Newcrest Mining Limited is currently mining and processing a large gold bearing copper ore body in the Cadia Valley of Cadia Hill. The Cadia Valley is located in the Central Tablelands region of New South Wales, Australia, approximately twenty-five kilometres southwest of Orange. Since 1850 a variety of mining and refining ventures have been carried out in the valley. The Big Cadia mine was mined for copper early in the twentieth century and was the largest producer of copper in the valley, as well as being mined for iron ore (Ryan 1995).

Cadiangullong Creek is a relatively small first and second order upland stream, which runs through the Cadia Project Mine Lease Area and at the base of the old Big Cadia mine site. Newcrest Mining Limited recently completed the construction of a dam across the creek, upstream of Big Cadia, to provide a reliable water source for the Cadia Hill goldmine. Researchers from the Environmental Studies Unit, Charles Sturt University Bathurst, are presently undertaking research to develop appropriate flow release strategies to optimise biodiversity and maintain or enhance ecological integrity in Cadiangullong Creek and the adjacent riparian system.

Previous research carried out by researchers from the Environmental Studies Unit found that macroinvertebrate communities adjacent to and immediately below Big Cadia had a reduced abundance and diversity compared with other sites along Cadiangullong Creek (Hayman 1998). An examination of the physicochemical parameters of Cadiangullong Creek led to the conclusion that copper was the only parameter with an increase in value which could be attributed to Big Cadia runoff. Copper concentrations in the water column adjacent to and immediately downstream of Big Cadia were significantly higher than at other sites, as were Chironomid whole body copper concentrations (Porter 1998).

Other studies also suggested the possibility of copper contamination from Big Cadia. A study conducted by Dames and Moore (1981) found high concentrations of copper in Cadiangullong Creek downstream of Big Cadia. A water quality monitoring program carried out by Newcrest and the Department of Mineral Resources found that water downstream of Big Cadia had elevated levels of copper, cadmium, iron, and occasionally, lead. The study identified two main anthropogenic sources of heavy metals in Cadiangullong Creek, Big Cadia and another historical mine site, Little Cadia (Ryan 1995). Since the time of the water quality monitoring program, the tributary on which Little Cadia is situated has been diverted away from Cadiangullong Creek, so Little Cadia has ceased to be a heavy metal source to the creek. A baseline soil geochemical survey of areas surrounding the present Cadia Hill goldmine found elevated copper concentrations in soils within the Big Cadia area and elevated arsenic concentrations in soils associated with the historical workings at Big Cadia (AGC Woodward Clyde 1995).

In the development of flow release strategies for Cadiangullong Creek, heavy metal concentrations will need to be considered. Flows can have a considerable influence on the concentrations of heavy metals within the water column of rivers and streams. During low flow periods, leaching of metals from sediments may be increased because of increased sediment-water contact times (Mok and Wai 1990). Heavy metal concentrations are also higher during low flow periods as there is a lower volume of water in the river (Mok and Wai 1990). During high flow periods, dilution by uncontaminated runoff decreases heavy metal concentrations (Grimshaw *et al*. 1976; Norris et al. 1981). However, at the onset of increasing runoff, heavy metal concentrations have been found to rise. This can be explained by a ‘flushing’ effect, where heavy metals may be flushed into rivers by runoff after warm, dry spells (Grimshaw *et al*. 1976).

Since the Big Cadia mine site had been previously identified by Newcrest and the Department of Mineral Resources as the major source of heavy metal contamination (Ryan 1995), the current study focused on Big Cadia as a potential anthropogenic source of heavy metals to Cadiangullong Creek. The project aimed to determine whether Big Cadia was a significant source of heavy metal contamination to Cadiangullong Creek, and to discover the extent of any contamination. Panuara Rivulet, in a nearby catchment, was chosen as a reference stream as it has not been disturbed by previous mining activities and is unlikely to be affected by planned future activities (Porter 1998).

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To determine whether Big Cadia was a significant source of heavy metal contamination to Cadiangullong Creek, this project investigated whether Big Cadia was capable of introducing heavy metals to Cadiangullong Creek; whether heavy metals were enriched above background levels within the Big Cadia site; and whether heavy metal concentrations in Cadiangullong Creek were greater in the vicinity of Big Cadia than at other sites along the creek. The project incorporated three main components:

(i) a study of heavy metals in sediments, pools and runoff within Big Cadia;
(ii) a study of heavy metals in creek bank soils along Cadiangullong Creek and Panuara Rivulet; and
(iii) a study of heavy metals in various instream media (water, plants, biofilm, sediment) in Cadiangullong Creek and Panuara Rivulet.

MATERIALS AND METHODS

Fine soils were collected from along drainage lines within the Big Cadia site in May 1999. Twenty-four sites were selected along the main drainage line, with four sites along a secondary drainage line, using a judgemental sampling pattern (Arakel 1995; Standards Australia 1997). Four samples were taken from each site. Soils were collected with plastic spades to a depth of three centimetres. Water samples from surface pools associated with small ditches and from runoff were also collected at Big Cadia during two separate rainfall events, in June and July 1999. Water samples for heavy metal analyses were acidified and samples for pH analyses were placed in ice and refrigerated in the laboratory.

Soils were sampled from seven sites along the banks of Cadiangullong Creek and three sites along the banks of Panuara Rivulet (Figure 1) in May 1999. A corer was used to sample to a depth of three centimetres. Three samples were taken from each site.

Water, plant, biofilm and sediments were sampled in Cadiangullong Creek and Panuara Rivulet in May, June and July 1999. Study sites for the instream survey were the same as those used in the creek bank soil survey (Figure 1). Six water samples and three plant, biofilm and sediment samples were taken from each site on each sampling occasion. Water samples were collected with polyethylene sample bottles. Water samples for heavy metal analyses were acidified and samples for physicochemical parameters analyses were placed in ice and refrigerated in the laboratory. Measurements of temperature, dissolved oxygen, dissolved oxygen saturation, salinity, turbidity and pH were taken at each site using a Grant YSI 3800 Water Quality Logging System. Sediments were collected using a syringe so that fine, mobile sediments could be analysed. Sediments were dried to constant weight in the laboratory before being ground for heavy metal analyses. *Carex* sp. was the plant chosen for analysis because it was available at most sites. *Carex* sp. leaves were collected by hand and placed in plastic bags on ice. Biofilm samples were collected by scraping the surface of rocks with a plastic spatula and stored in glass jars. *Carex* sp. and biofilm samples were dried to constant weight in the laboratory and cut into pieces for heavy metal analyses.

The sediment, soil, plant and biofilm samples were converted to aqueous solutions before being analysed. The conversions were carried out with a wet ashing digestion technique using concentrated nitric acid (Förstner and Wittman 1979). A Varian Liberty Series II Inductively Coupled Plasma Emission Spectrometer (ICP-AES) was

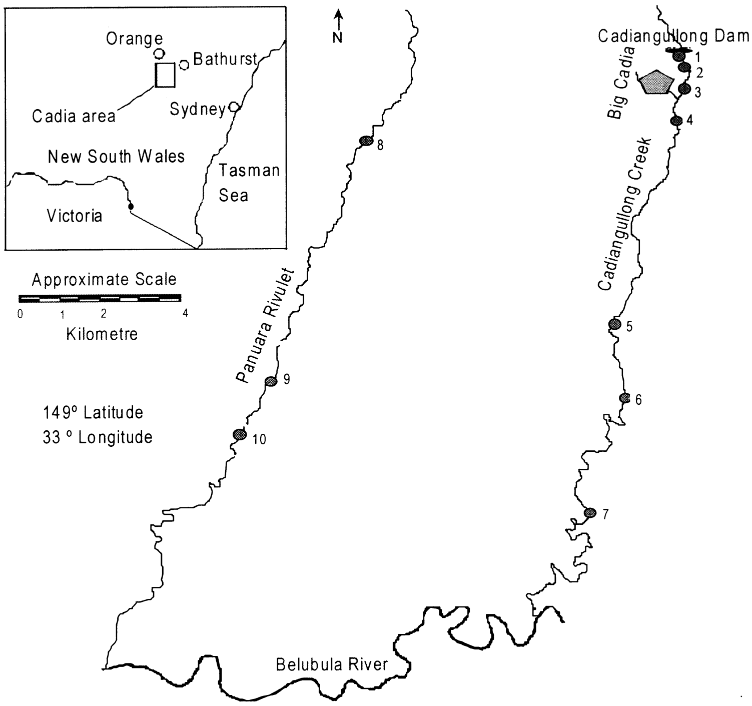


Figure 1. *Cadiangullong Creek and Panuara Rivulet creek bank soil and instream survey site locations.*

Table 1. *Detection limits for ICP-AES determination of ten heavy metals.*

Metal	Detection limit for water samples (mg/L)	Detection limit for digested samples (mg/L)
As	0.1000	1.000
Cd	0.0010	0.010
Cr	0.0040	0.040
Cu	0.0020	0.020
Fe	0.0100	0.100
Hg	0.0100	0.100
Mn	0.0004	0.004
Ni	0.0100	0.100
Pb	0.0100	0.100
Zn	0.0020	0.020

used to determine the concentrations of ten heavy metals in each sample: iron, copper, lead, cadmium, arsenic, nickel, manganese, chromium, zinc and mercury. The detection limits for these analyses are presented in Table 1.

A QC Plus^{+} Quality Control Standard was used for the water samples (SRM 014). The results for the quality control standard showed that copper was always within the acceptance limits, while cadmium, chromium, iron and zinc always deviated less than five percent from the acceptance limits. Manganese, nickel and lead always deviated less than 10%. Arsenic and mercury were observed to deviate more than 10%. The reason for this deviation lies in the fact that the concentrations of arsenic and mercury in the quality control standard were very close to the detection limits for the ICP-AES. The concentration of arsenic in the standard was 0.180 mg/L, only 0.08 mg/L above the arsenic detection limit. The concentration of mercury in the standard was 0.015 mg/L, which is only 0.005 mg/L higher than the detection limit.

A TPS LC80A pH/mV/temp meter was used to measure the pH of the Big Cadia sediment and runoff samples. Calcium chloride was added to a 1:5 soil:water suspension before measurements were taken (White 1969). The following physicochemical parameters were determined in the Cadiangullong Creek and Panuara Rivulet water samples: alkalinity (APHA 2320B); acidity (APHA 2310B with a phenolphthalein end point); water hardness (APHA 2340C); and chloride (TPS440 lab analyser with an ionode double junction reference electrode and an ionode chloride ion selective electrode) (Clesceri *et al*. 1989).

The concentrations of some heavy metals in a number of samples were found to be below the detection limit of the ICP-AES. It cannot be claimed that these heavy metals were not present, since they may have been present at concentrations below the limit. To ignore the samples altogether would lead to difficulties in statistical analyses. For the purposes of statistical analyses the concentrations of these heavy metals were set at the detection limit. The values were then carried through all the calculations performed on the heavy metal data.

Multiple analysis of variance (MANOVA) was used, where possible, to statistically analyse the data since the dependent variables were related (Coakes and Steed 1996). The assessment of significant differences by MANOVA was tested using Pillai's Trace because it is more robust than other available measures, such as Wilks'-lambda and Roy's Largest Root (Sharma 1996).

For some of the tests, covariance-variance matrices for all groups were not equal, which is one of the assumptions of the MANOVA test (Sharma 1996). For most of these tests, dependent variables were transformed to achieve univariate equality of variances. Where equality of variances was not obtained through transformations, a oneway ANOVA was used, or if equality of variances could still not be obtained, a non-parametric Kolmogorov-Smirnov test was used (Siegel 1956; Norusis 1995).

RESULTS

Open cut mining methods have shaped the eastern side of Big Cadia into a series of platforms (Ryan 1995). Figure 2 presents the results for each of the platforms in order, left to right, from the highest platform sampled to the creek bank level, labelled CBL. In soils from Big Cadia mean arsenic concentrations ranged from 9 to 46 mg/kg, exceeding the ANZECC and NHMRC (1992) guideline on some platforms and on the creek bank level by between 34 and 128%. Copper concentrations ranged from 108 to 258 mg/kg, exceeding the guideline on all platforms sampled and on the creek bank level by between 80 and 330%. Mercury concentrations ranged from 0.4 to 1.4 mg/kg, exceeding the guideline on Platform B and on the creek bank level by between 16 and 32%. Concentrations of the other metals analysed were below the ANZECC and NHMRC (1992) guidelines.

The mean pHs of the Big Cadia soil samples ranged from 3.4 to 4.1, well below the pH range given as background levels by the ANZECC and NHMRC (1992) guidelines. This means the sediments were more acidic than the background levels given in the guidelines.

The concentrations of heavy metals in the Big Cadia soil samples were compared with concentrations of heavy metals found by a survey carried out by Newcrest (Cadia Holdings Pty Ltd 1998), in surface soils from properties in the area. This comparison was used to help in determining background heavy metal levels, along with the Cadiangullong Creek and Panuara Rivulet creek bank soil survey. In general, higher levels of arsenic and copper were found in the present study of Big Cadia soils than were determined for local soils (Figures 3 and 4).

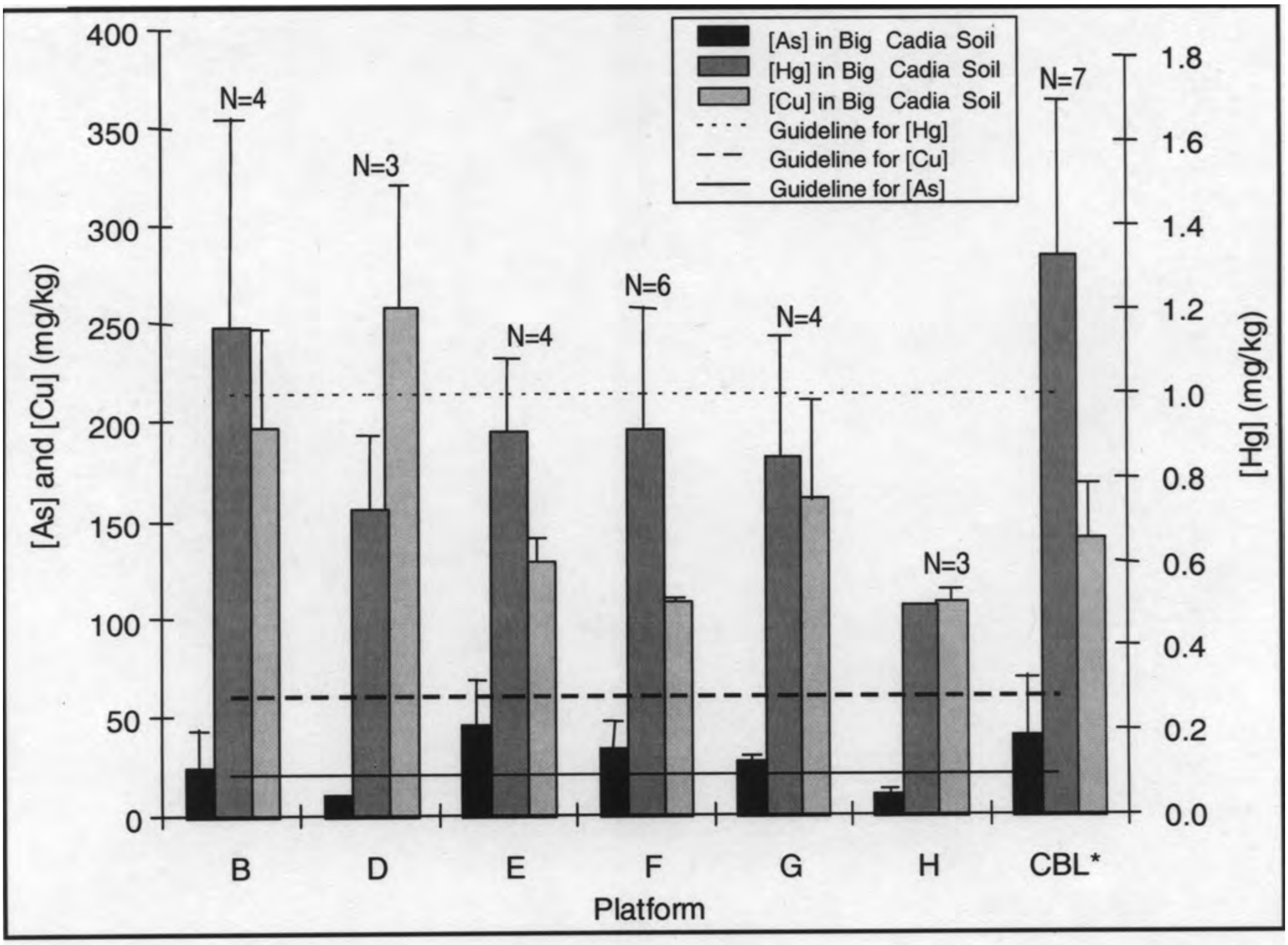
Mean cadmium, copper, manganese, nickel, lead and zinc concentrations in Big Cadia surface pools and runoff had levels on some or all of the Big Cadia platforms which exceeded the ANZECC & ARMCANZ (2000) guidelines' trigger level expected to protect 95% of species. Mean copper, manganese, nickel, lead and zinc had levels on some or all platforms which even exceeded the guidelines' trigger level expected to protect 80% of species. Copper exceeded the guidelines to the greatest extent, with concentrations in runoff ranging from 0.08 to 12.51 mg/L, exceeding the 95% trigger level by between 5,610 and 893,000%. In pools, copper concentrations ranged from 9.8 to 16.4 mg/L, exceeding the 95% trigger level by between 707,000 and 1,168,000%. The mean runoff pHs were acidic and ranged from 3 to 5.7.

Mean copper, manganese and nickel concentrations in creek bank soil samples from Cadiangullong Creek Site 3 (adjacent to Big Cadia) were statistically significantly higher than concentrations in creek bank soil samples from other sites along Cadiangullong Creek and Panuara Rivulet. Mean manganese concentrations at Cadiangullong Creek Site 1 (upstream of Big Cadia and below the dam wall) were also significantly higher than concentrations at the other sites except Site 2 ($F=2.801$, $df=30$, $p<0.001$) (Figure 5).

Mean copper concentrations in water ($F=10.268$, $df=24$, $p<0.001$), *Carex* sp. ($F=2.955$, $df=24$, $p<0.001$), biofilm ($F=18.092$, $df=12$, $p<0.001$) and sediments ($F=14.757$, $df=10$, $p<0.001$) at Sites 3 and 4 (adjacent to and immediately downstream of Big Cadia) were significantly higher than those at Sites 1 and 2 (above Big Cadia) (Figure 6).

Mean iron concentrations in water were significantly higher at Site 4 (immediately downstream of Big Cadia) than at Sites 1 and 2 ($F=10.268$, $df=24$, $p<0.001$). Mean iron concentrations in biofilm were significantly higher at Sites 3 and 4 than at Sites 1 and 2 ($F=18.092$, $df=12$, $p<0.001$). No significant differences were found between sites for iron in *Carex* sp. and in sediments, but general trends suggesting higher levels of iron at Site 3 than at Sites 1 and 2 can be seen (Figure 7).

Mean nickel concentrations in biofilm were significantly higher at Sites 3 and 4 than at Sites 1 and 2 ($F=18.092$, $df=12$, $p<0.001$). In sediments, significant differences were not found, but there was a general trend of higher concentrations of nickel at Site 4 than Sites 1 and 2 (Figure 8). In water, nickel concentrations rarely exceeded the detection limit of the ICP-AES, but they did exceed it at Sites 3 and 4. Nickel concentrations in *Carex* sp. never exceeded the detection limit.



* Creek bank level

Figure 2. *Concentrations of arsenic, copper and mercury in Big Cadia soils with the ANZECC and NHMRC (1992) guidelines.*

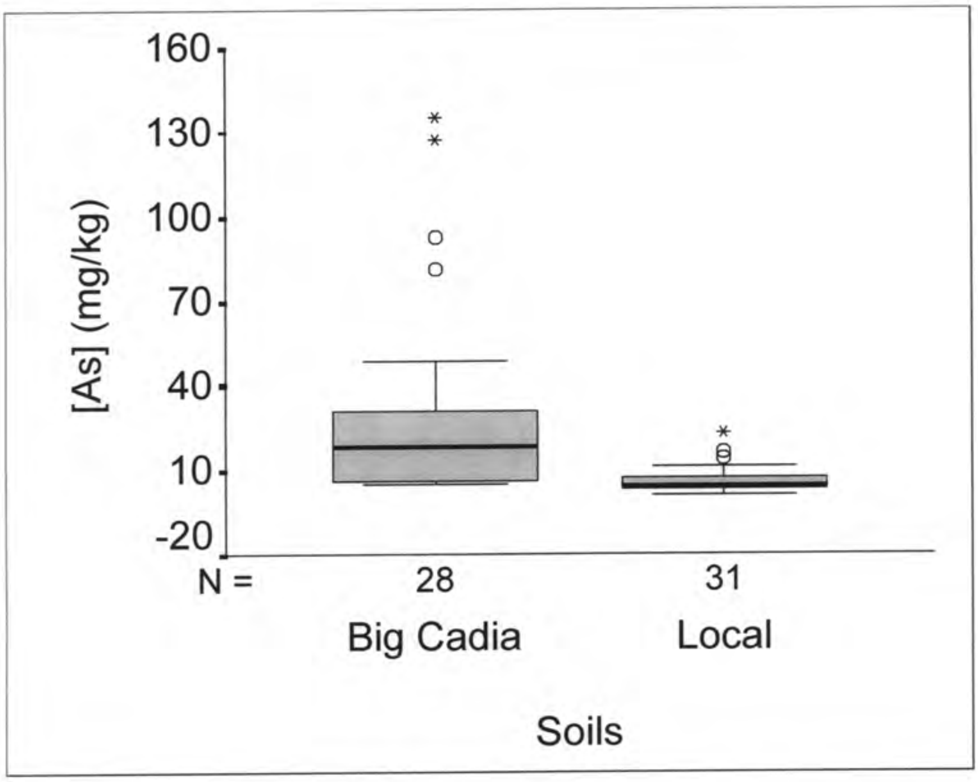


Figure 3. *Box and whisker plot comparing arsenic concentrations in Big Cadia soils (this study) with concentrations in local soils (Cadia Holdings Pty Ltd 1998).*

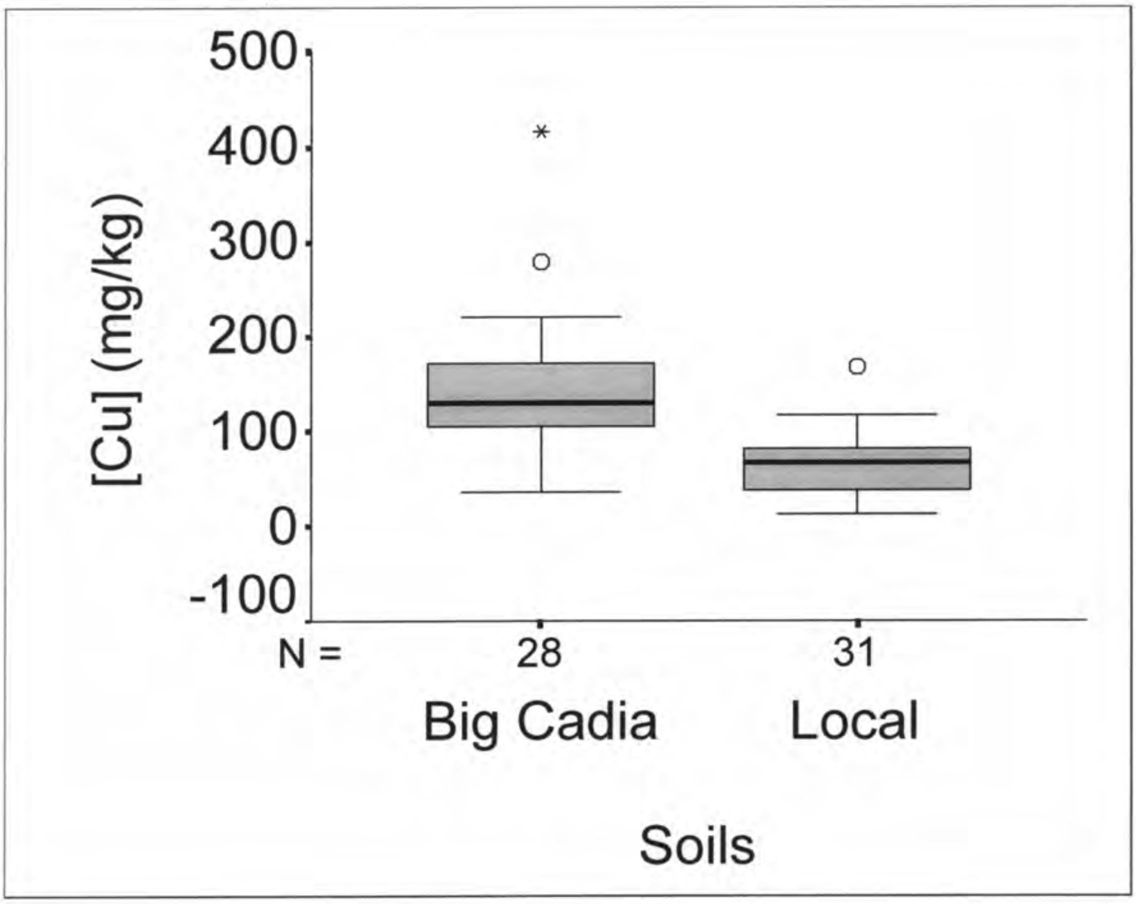


Figure 4. *Box and whisker plot comparing copper concentrations in Big Cadia soils (this study) with concentrations in local soils (Cadia Holdings Pty Ltd 1998)*

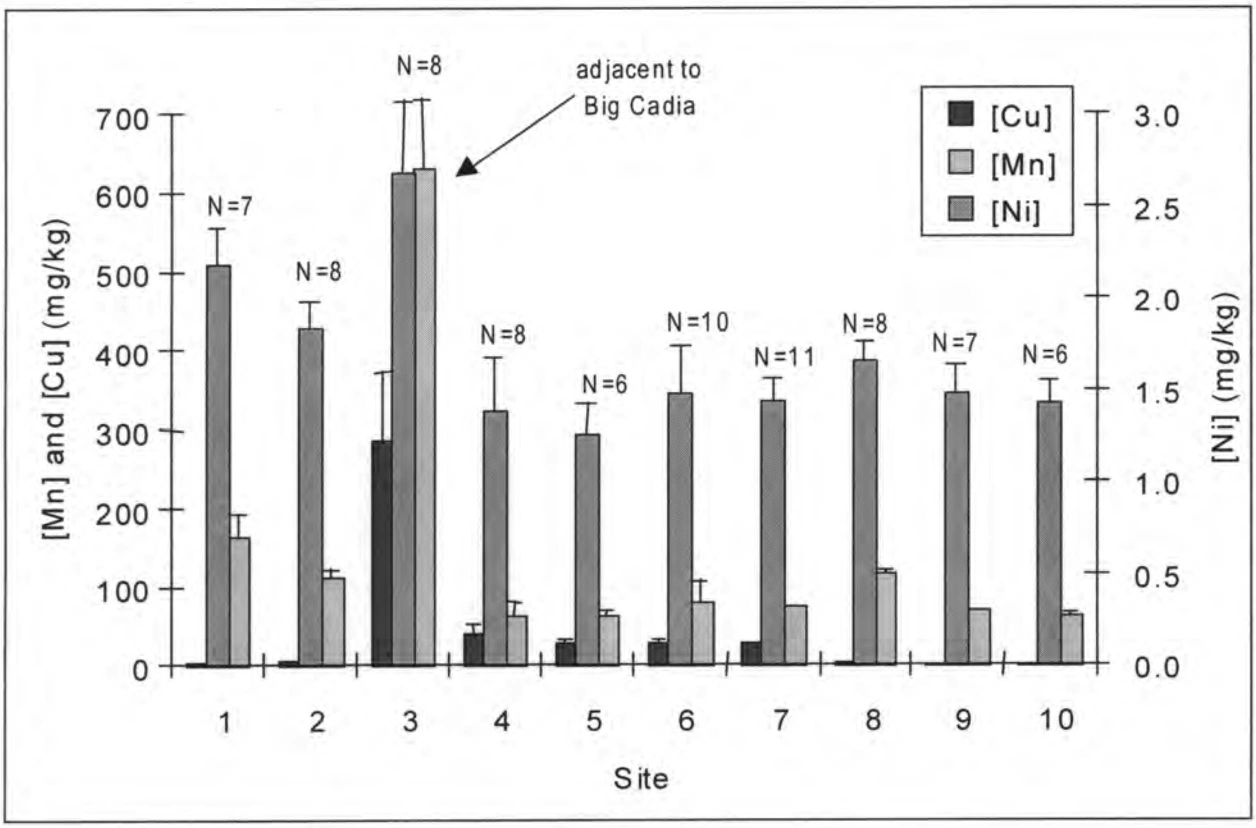


Figure 5. *Copper, manganese and nickel concentrations in soils from Cadiangullong Creek (Sites 1 to 7) and Panuara Rivulet banks (Sites 8 to 10).*

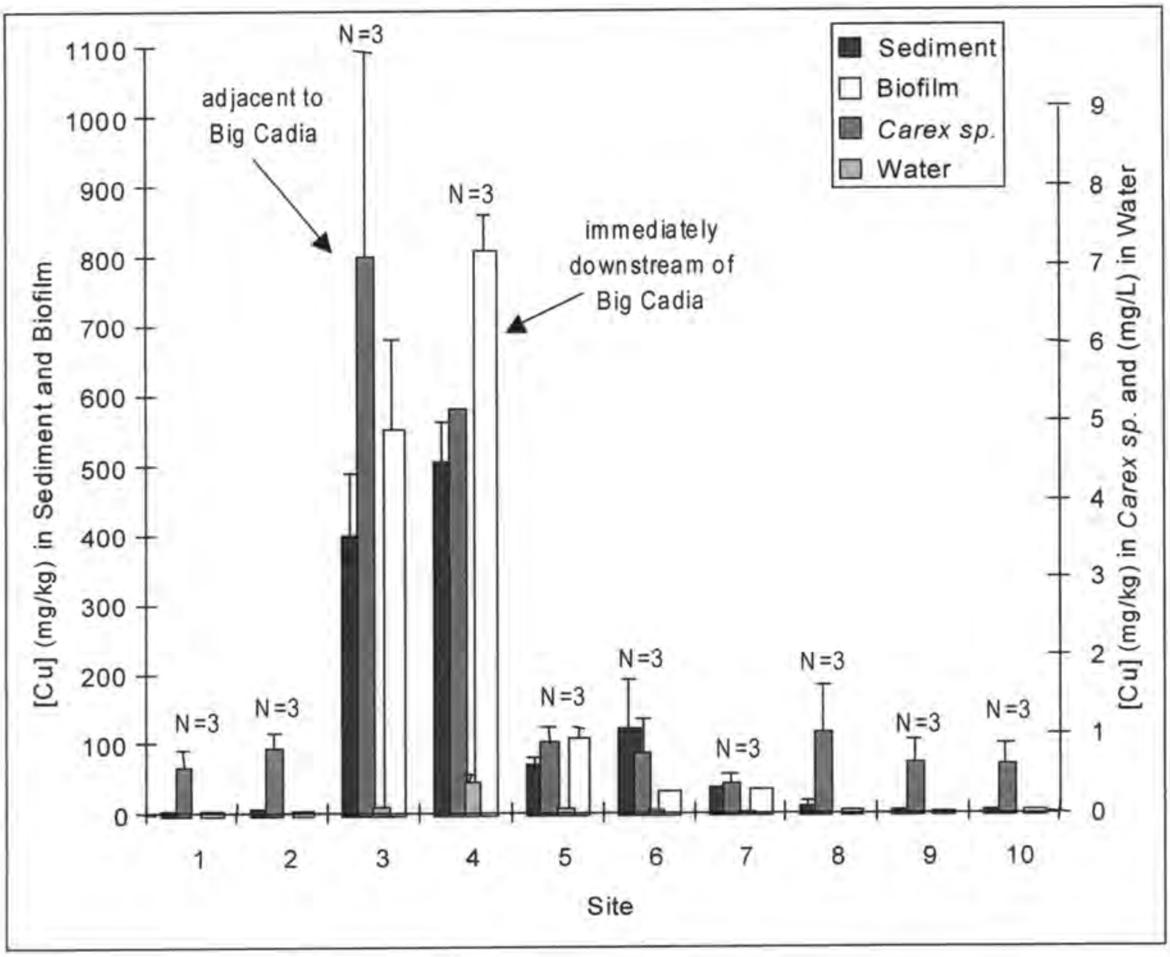


Figure 6. *Copper concentrations in water, Carex sp., biofilm and sediment from Cadiangullong Creek and Panuara Rivulet.*

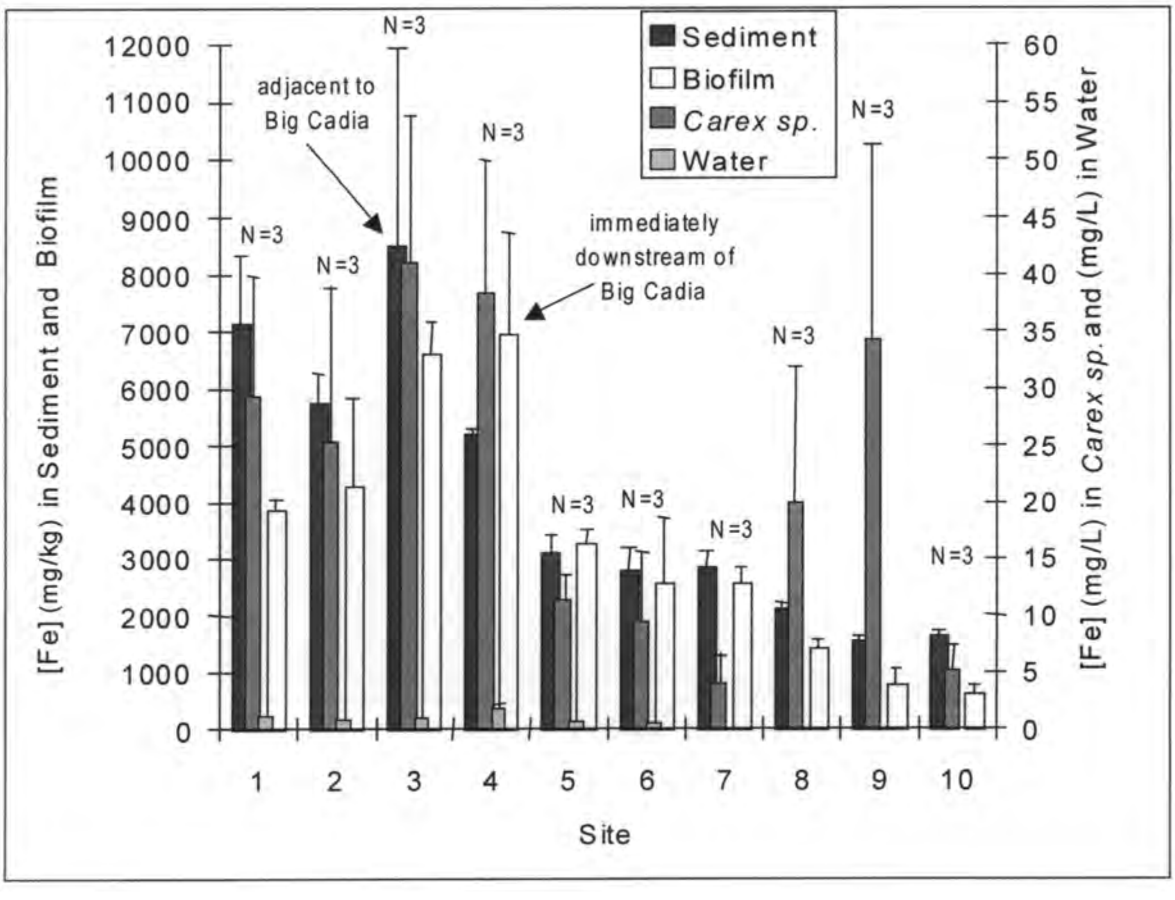


Figure 7. *Iron concentrations in water, Carex sp., biofilm and sediment from Cadiangullong Creek and Panuara Rivulet.*

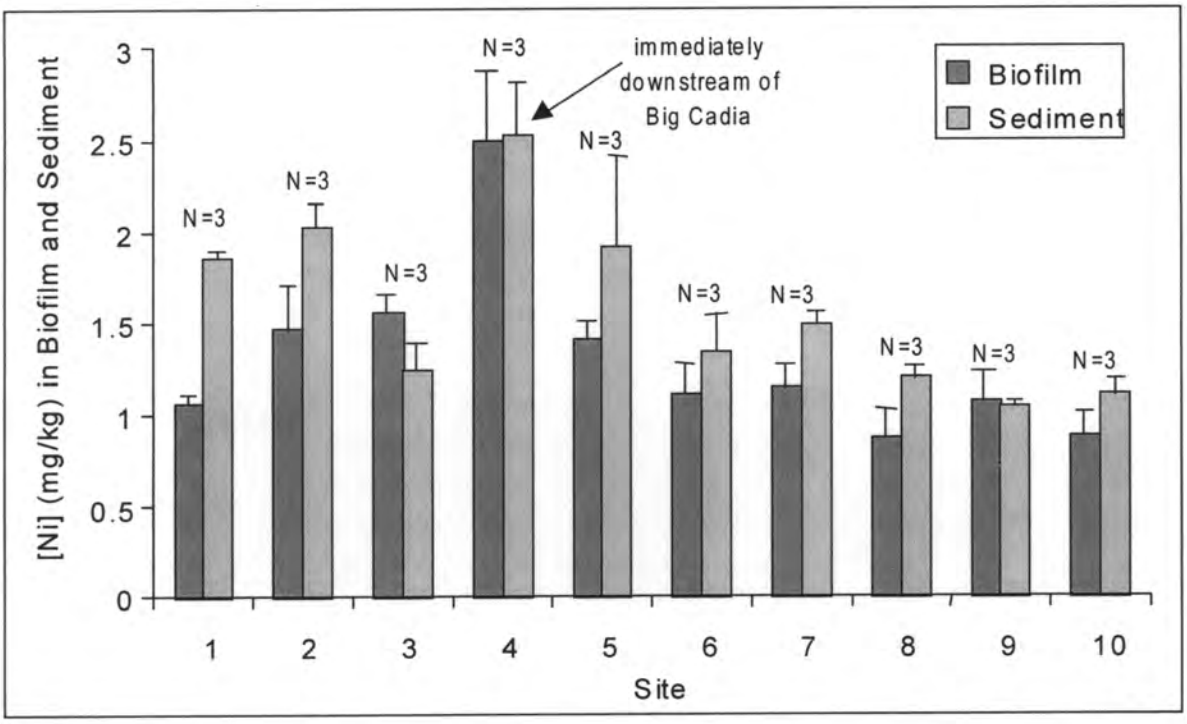


Figure 8. *Nickel concentrations in biofilm and sediment from Cadiangullong Creek and Panuara Rivulet.*

DISCUSSION

Big Cadia was found to have the potential to contribute heavy metals to Cadiangullong Creek. Open cut mining methods have shaped the hillside into a series of platforms stepping down towards the creek. Drainage lines and areas of severe erosion are clearly visible between these platforms. This topography allows runoff from each platform to flow into the creek.

Both soils and surface water within the Big Cadia site were found to be acidic. The solubility of most metals is increased by acid conditions (Bradshaw and Chadwick 1980; Salomons 1993). Therefore, heavy metals within Big Cadia have the potential to enter solution and be transported into Cadiangullong Creek.

Arsenic, copper and mercury concentrations in Big Cadia soils exceeded the levels recommended by the ANZECC and NHMRC (1992) guidelines. Big Cadia soils were also found to contain higher levels of arsenic and copper than the soils analysed by the Newcrest study from local properties (Cadia Holding Pty Ltd 1998). Mercury was not found during the Newcrest study, although the detection limits for mercury were the same for both studies. It can be concluded that mercury concentrations in Big Cadia soils are also greater than those found in local soils.

Cadmium, copper, lead, manganese, nickel, and zinc concentrations in runoff from Big Cadia exceeded the ANZECC & ARMCANZ (2000) guidelines' trigger level expected to protect 95% of species. All these metals are, therefore, of interest as potential contaminants in Cadiangullong Creek.

Copper, manganese and nickel concentrations in creek bank soils were enriched at Site 3, below the Big Cadia mine site, compared with other sites along Cadiangullong Creek.

Manganese levels were also high at Site 1, near the dam wall. Although no statistical significance was found, general trends showed iron and chromium levels at Site 1 to be high. These patterns suggest that sources of manganese, iron and chromium exist in the catchment above Big Cadia. No other general trends were identified from the creek bank soil survey. It can be concluded that sources of heavy metals within the catchment below Big Cadia are not contributing greatly to heavy metal concentrations within Cadiangullong Creek.

The instream survey found that copper, iron and nickel all showed patterns of increasing concentrations at Sites 3 and 4 (adjacent to and immediately downstream of Big Cadia). Other metals, such as manganese and zinc, appeared to increase at Sites 3 or 4, but such increases were not found to be significant. Therefore, concentrations of other metals cannot be directly attributed to Big Cadia. From the results of the present study, it can be concluded that the Big Cadia mine site is introducing significant amounts of copper, iron and nickel into Cadiangullong Creek.

This study was carried out concurrently with a fish survey by Laws (1999) in Cadiangullong Creek. Laws (1999) found only two fish species present in Cadiangullong Creek, the exotic Rainbow Trout (*Oncorhynchus mykiss*) and the native Mountain Galaxid (*Galaxias olidus*). There is a lack of data on heavy metal toxicity for *G. olidus*, but data are available for a related species, the Common Jollytail (*Galaxias maculatus*). The copper 48-h LC50 and 96-h LC50 for *O. mykiss* are 0.023 and 0.018 mg/L respectively and the copper 48h LC50 and 96-h LC50 for *G. maculatus* are 0.076 – 0.100 and 0.076 respectively (Skidmore and Firth 1983; Marr et al. 1998). These toxicity data show that the concentrations of copper in water from Cadiangullong Creek Sites 3, 4, 5 and 6 (Figure 6) are potentially toxic to *O. mykiss*. The concentrations at Sites 3 and 4 would be potentially toxic to *G. maculatus*. It is not known whether *G. olidus* has a similar or higher tolerance to copper than *G. maculatus*, but it is certainly possible that the water would also be toxic to *G. olidus*.

CONCLUSIONS

The following conclusions were drawn from this study.

(i) The topography of the Big Cadia mine site and the acidity of sediments and runoff showed that Big Cadia was capable of contributing heavy metals to Cadiangullong Creek.

(ii) Arsenic, copper and mercury in sediments from the Big Cadia mine site were enriched above background levels. Big Cadia runoff was found to contain high levels of cadmium, chromium, copper, iron, lead, nickel and zinc. Copper, manganese and nickel concentrations in creek bank soils were enriched immediately below the Big Cadia mine site.

(iii) Copper, iron and nickel concentrations in Cadiangullong Creek were greater at Sites 3 and 3a than at other sites along the creek.

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