Historical pollution variability from abandoned mine sites, Greater Blue Mountains World Heritage Area, New South Wales, Australia

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Abstract Core and surface sediments from the Tonalli River, a tributary of the artificial lake, Lake Burragorang, in the Blue Mountains National Park, New South Wales, Australia, were studied to evaluate the spatio-temporal distribution of pollutants from the Yerranderie silver–lead–zinc mine site, abandoned in the late 1920s. A sediment core was collected in the mouth of the Tonalli River, at its junction with Lake Burragorang, and surface sediment samples were collected in the Tonalli River and its tributaries. The concentrations of Pb, As, Zn, Cu, Cd, Hg and Ag in the sediments were determined by ICP-MS and ICP-AES techniques. Temporal variability of metal concentrations was established through ^{210}Pb dating of the core sediments and compared with published historical records, rainfall records and bushfire data. Metal concentrations in core sediments showed an overall increase around the year 1950 as well as increases coincident with heavy rainfall. Spatially, metal concentrations were up to 400 times the guideline limit around mine sites but decreased rapidly with distance downstream of the mines.

Keywords ^{210}Pb dating · Pollution · Sediments · Trace metals · Australia · New South Wales · Tonalli River · Yerranderie · Galena mining

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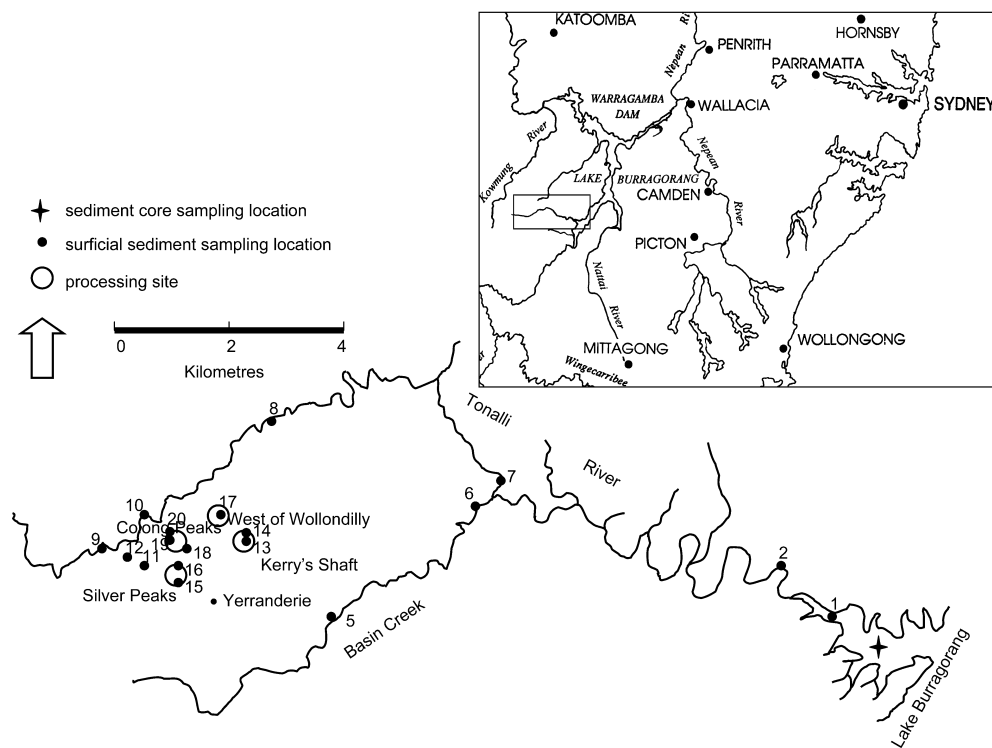
Introduction

Extraction of metals from sulfide ores generally results in 90% of the minerals being discarded as tailings (Moore and Luoma 1990). Tailings and associated overburden materials may contain elevated concentrations of copper, zinc, cadmium and lead, and are a potential source of localised ground and surface water contamination. The concentrations of heavy metals in waters are controlled by factors such as: precipitation – dissolution, coprecipitation reactions, adsorption – resorption and solid solution substitutions (Blowes and Jambor 1990). Once loaded into the waters, metal contaminants are sequestered by settling particulate matter and removed from the water column (Kolak and others 1998). Thus, surface sediments and sediment cores can be studied to determine present day and historical contaminant loading. This is especially useful when dealing with point-sources, such as mine and processing sites.

Because of the complex interplay of many physico-chemical and geomorphological parameters that influence the distribution of heavy-metal contamination in specific areas, analytical work on soils, surface and core sediments from streams, pore-water and water samples has been undertaken in many industrialised and urban areas, attempting to identify and predict the patterns of contamination. The importance of such type of studies is being increasingly recognised in Australia (Thoms and Thiel 1995; Birch and Taylor 2000).

Approximately 80% of drinking water in the Sydney region is supplied by Lake Burragorang (McCotter 1996), impounded by Warragamba Dam. Among the tributaries of Lake Burragorang is the Tonalli River, whose catchment area contains many abandoned galena (PbS) mines and four ore processing sites (Fig. 1). Since cessation of mining, little has been done to prevent run-off and sediment transport from the mine and processing sites. During periods of heavy rainfall, metal contaminants are transported to the Tonalli River and, from there, to Lake Burragorang. Studies of stream sediments in the Tonalli River from and around the processing areas have shown that the levels of lead, zinc, cadmium and arsenic are well above the Australian and New Zealand Environment and Conservation Council and Agriculture and Resource Management Council of Australia and New Zealand (ANZECC and ARMCANZ 2000) guidelines for stream sediments (O'Hagan, unpublished data, 1999). The Sydney Catchment Authority (SCA) conducted a study

which revealed measured levels of nitrogen and metal concentrations well in excess of ANZECC and ARMCANZ guidelines in samples of river water and soil collected from two mining processing areas (Capararo, unpublished data, 1999).

A 100-year temporal record of pollution fluctuation in the Tonalli River downstream of the mines and processing sites was determined. Over this time period, major mining operations in the Tonalli River catchment commenced and ceased. Historical records and climate variability records such as rainfall and bushfire were available throughout this time frame and were compared with the results of this study. Surface sediments from the Tonalli River, Basin Creek and minor tributaries were used to determine the spatial distribution of pollution in the catchment.

Heavy metal concentrations, including lead, arsenic, zinc, copper, cadmium, mercury and silver in both core and surface sediments, were determined. The age of core sediments was obtained using the ^{210}Pb method (Goldberg 1963; Oldfield and Appleby 1984), and thus the variation of metal concentration in the core sediments with age could be established.

Materials and methods

The Tonalli River is approximately 100 km southwest of Sydney, New South Wales, Australia, and is a sub-catchment of Lake Burragorang (Fig. 1). Construction of the Warragamba Dam commenced in 1948, leading to the formation of the artificial Lake Burragorang. The dam was completed and the reservoir full by 1960.

Before dam construction, most of the Burragorang Valley was rough pastoral land. The sparse population was steadily growing as access routes improved (Barrett 1995). In 1872 the mineral galena (PbS) was discovered in the Tonalli River catchment (Barrett 1995). Around this time, many mines were established along with the town of Yerranderie and there were four main processing sites in operation: Colong Peaks, Silver Peaks, West of Wollondilly and Kerry's Shaft. The town of Yerranderie, the mines and the processing sites have been abandoned since cessation of mining in 1928, brought about by the depression and dam construction.

Rainfall and bushfire records were investigated as indicators of climate variability which was suspected to influence the distribution of metal pollution. Monthly rainfall records from 1908 to 2000 were available, while bushfires have been formally documented since 1957. Since dam construction, the SCA use preventative measures against bushfires in the catchment region so it was concluded in this study that bushfire records were not a reliable indicator of climate variability.

The rainfall data in this study are reflective of the Southern Oscillation Index (SOI). The SOI is formulated by taking the difference between each of the monthly mean sea level pressure (MSLP) anomalies at Tahiti and Darwin and normalising each of the differences by dividing them by the standard deviation of the monthly difference in MSLP anomalies at both locations and multiplying by 10 as described by Allan and others (1996). Periods of strong protracted negative values relate to El Niño episodes while periods of strong protracted positive values relate to La Niña episodes. Most El Niño events are associated with drought over eastern Australia while La Niña events are

associated with above-average rainfall and flooding (Power and others 2000). The three largest La Niña events in the last century occurring between 1945 and 1955, 1973 and 1975 and 1998 and 2000 caused widespread flooding throughout Australia.

The sediment core was collected in 15-m water depth using a modified hammer-driven piston corer (Neale and Walker 1996). The coring device was lowered from the boat to about 50 cm above the sediment/water interface. The core barrel was driven into the sediment using the hammer which was activated on the boat by a cable. The piston, kept in a fixed position 50 cm above the sediment/water interface, passed through the core barrel as the sample was collected, allowing the sediment/water interface to be sampled with little disturbance. The sediment core was sampled at 0.5-cm intervals in a vertical position on the boat. Controlled pressure from a water pump applied at the base of the core slowly extruded the sediment from the core barrel, allowing small sampling intervals. Surface sediments were collected using a small plastic spade. All sediment samples were dried in a <30 °C oven and gently disaggregated using a mortar and pestle. All samples underwent trace metal, grain size and organic matter analysis. The trace metal concentrations were determined by ICP-MS and ICP-AES analyses of bulk sediment digested by a mixture of HNO_3, HCl and HF. Measurements of trace metal concentrations in the reference materials MESS-2 (National Research Council Canada) and RTS-3 (Canadian Certified Reference Materials Project) were routinely carried out during the study. The data obtained from the reference materials analyses are reported in Table 1. Grain size was determined using a Malvern Mastersizer with a 300RF lens, 0.05–900 μm detection range and a beam length of 2.40 mm. The sediments were dispersed in water and ultrasonicated in the sample dispersion unit before being pumped into the detection cell. The organic content of the sediment samples was the difference in weight before and after being heated in a muffle furnace at 450 °C for 7 h. The ages of core sediment slices were obtained by the ^{210}Pb method, first described by Goldberg (1963) and first applied to the dating of lake sediments by Krishnaswamy and others (1971). The total ^{210}Pb activity was determined by measuring its granddaughter [210]Po, which was assumed to be in secular equilibrium with [210]Pb. Supported ^{210}Pb was approximated by measuring [226]Ra activity.

Approximately 2 g of each dried sediment sample was spiked with [209]Po and [133]Ba yield tracers. Each sediment sample was subsequently leached with hot concentrated acids to release polonium and radium. Polonium was autoplated onto silver disks after reduction of Fe^{3+} with ascorbic acid and analysed by high resolution alpha-spectrometry. Radium was co-precipitated with $BaSO_4$ on a filter-source. The chemical yield was determined by measuring the artificial tracer [133]Ba on an HPGe-gamma detector.

The unsupported ^{210}Pb activity was calculated by subtracting the [226]Ra activity from the total ^{210}Pb activity. The sedimentation rates were calculated using the constant initial concentration (CIC) method (Robbins and Edgington 1975; Goldberg and others 1977) with the formula:

$$t_I = 1/y * Ln (A_0/A_t) \tag{1}$$

where A_0 is unsupported ^{210}Pb activity at the sediment surface in dpm g^{-1} of dry sediment; A_t is unsupported ^{210}Pb activity at time t in dpm g^{-1} of dry sediment; y is the decay constant of [210]Pb (0.03114 year^{-1}); and t_I is the difference in age of sediment surface and sediment at depth in years.

The sedimentation rate was determined from the slope of the least-square fit for ^{210}Pb excess values plotted versus depth. The calculations assumed biological and physical mixing to be restricted to the mixed surface layer (MSL). The CIC-model dictates that the ^{210}Pb activity must decline monotonically with increasing depth. However, in some cases the sediment core displays linear-segments within a non-linear profile; in these cases the individual slopes were used to construct a sedimentation history with varying initial concentrations as discussed by Brugam (1978).

Results and discussion

Sediment core

The grain size distribution and the organic content of the sediment core are shown in Fig. 2a. The finer material (<63 μm) dominates the top few centimetres followed by a transition region between 10 and 20 cm down-section where sandy material (>63 μm) becomes the dominant phase. A steady decrease in organic content is observed in the top 15 cm and the trend is similar to that of the <63 μm material (r^2=0.930).

Concentrations of Pb, Zn, As and Cu within the sediment core both increase and fluctuate above a core depth of

Table 1
Comparison of reference material values with obtained results. All values are in parts per million. *lld* Lower than detection limit=0.2

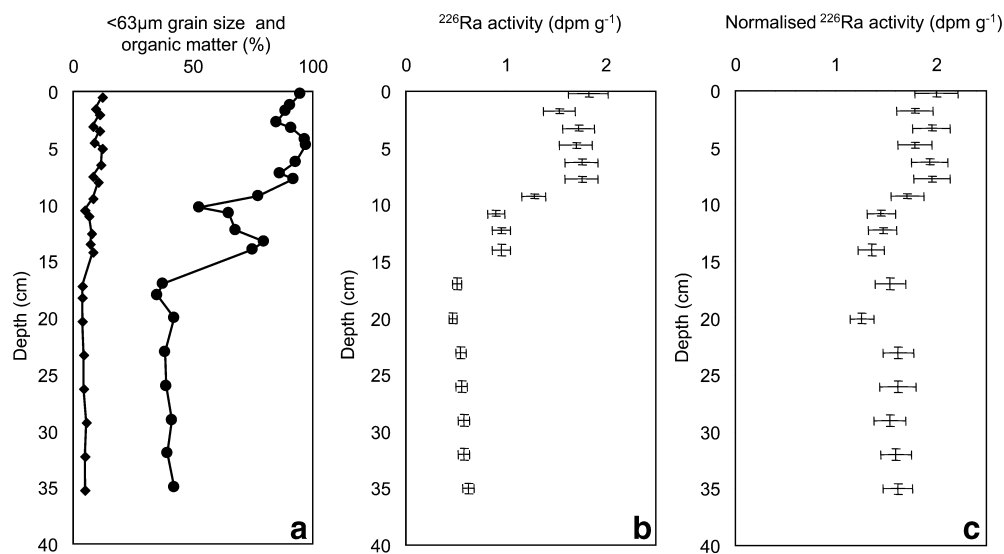| Element	Pb	As	Zn	Cu	Cd	Hg	Ag
RTS-3							
Certified values	146.0±20.0	9.1±2.6	1,850.0±80.0	2,820.0±90.0	9.1±1.6	No data	<8
Obtained results	158.0±5.2	10.2±0.4	1,890.0±66.4	2,812.0±107.5	8.5±0.0	7.2±0.8	6.7±0.0
MESS-2							
Certified values	21.9±1.2	20.7±0.8	172.0±16.0	39.3±2.0	0.24±0.01	0.09±0.01	0.18±0.02
Obtained results	21.0±2.0	20.7±1.1	163.5±13.7	37.8±3.5	0.3±0.1	lld	lld

Fig. 2
a Percentage of <63 μm sediment fraction versus depth; **b** ^{226}Ra activity versus depth; and **c** ^{226}Ra activity normalised using <63 μm fraction versus depth
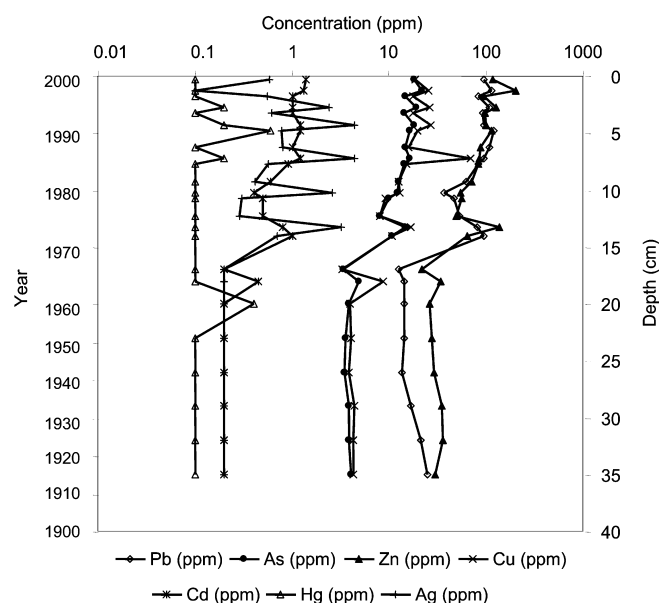

Fig. 3
Changes in metal concentration versus age and core depth

20 cm (Fig. 3). Downsection from this point the metal concentrations are lower and do not appear to fluctuate. This change coincides with changes in the <63 μm grain size fraction and organic matter content. Trace metal concentrations were normalised (Arakel 1995; Birch and Taylor 2000; Birch et al. 2001) to determine whether the grain size and organic matter content influenced the observed changes. The overall increase in metal concentrations was found to be influenced by the increase in finer sediments and the increase in organic matter, while the concentration fluctuations were not. It was concluded that although the large increase in fine sediment above a core depth of 20 cm had increased the overall concentration of the metals, fluctuations present in the core sediments were a result of other physical or chemical processes.

Interpretation of the ^{210}Pb activity profile was a stepwise process as the uptake of ^{210}Pb in sediments has been shown in previous studies to be influenced by the grain size of the sediments (Eisma and others 1987; He and Walling 1996). Supported ^{210}Pb activity, approximated in this study by ^{226}Ra activity, is derived from erosional input and should theoretically be constant (Oldfield and Appleby 1984). The deviation from constant activity shown in Fig. 2b provides a good indication that grain size may have an influence. Using the <63 μm grain size fraction data for the core, the ^{226}Ra activity profile was normalised. The data spread was significantly reduced (σ =0.25 c.f. σ=0.54) as shown in Fig. 2c, suggesting that ^{210}Pb, which adsorbs to the surface of sediment particles, would also be influenced by the percentage of the <63 μm grain size fraction and hence change in sediment surface area in each sediment slice.

Unsupported ^{210}Pb, derived from atmospheric input, was therefore assumed to be affected by the varying percentages of <63 μm material in the individual sediment slices. Thus, the unsupported ^{210}Pb activities, estimated by subtraction of un-normalised supported ^{210}Pb from total ^{210}Pb, were normalised using the percentage of <63 μm grain size fraction in the corresponding sediment samples. Once normalised, the unsupported ^{210}Pb activity profile (Fig. 4a) was used to determine the sediment accumulation rate and the age of the sediment slices from the cores. The unsupported ^{210}Pb profile (Fig. 4a) is non-linear (plotted on a logarithmic-linear scale), indicating that the sediment accumulation rates have changed over time (Brugam and Speziale 1983; Eisma and others 1987). Using the modified CIC method described by Brugam (1978), zones of constant sedimentation and hence constant ^{210}Pb flux into the system were distinguished. Sedimentation zones were identified by the linear trends among the data points. Two trends were identified, each having different slopes, and intersecting at approximately 20 cm depth within the core (Fig. 4a). The data points (n=12) in the 0 to 20 cm depth zone have a correlation coefficient r^2=0.929. The data points (n=5) in the >20 cm

depth zone have a correlation coefficient r^2=0.841. The different slopes of the two individual correlation lines indicate that two periods of sedimentation characterised by different rates of deposition occurred in the time interval recorded by the sediments in the core.

Crucial to this modified CIC method for determination of sedimentation rates with unsupported ^{210}Pb is the assumption that the flux of unsupported ^{210}Pb (A_0) to the sediment has been constant through time over broad sections of the core. Once A_0 values were established for each zone, they were substituted into Eq. (1) to calculate t-values for each corresponding A_t value. The depth of each sediment slice and the corresponding calculated t-values were plotted as shown in Fig. 4b and the slopes of the lines are the sedimentation rates in centimetres per year. The calculated sedimentation rates were 0.26±0.04 cm year^{-1} for the top part of the core and 0.07±0.03 cm year^{-1} for the bottom of the core. The change in sedimentation occurred at an age of 50 years ago or, in calendar years, 1950.
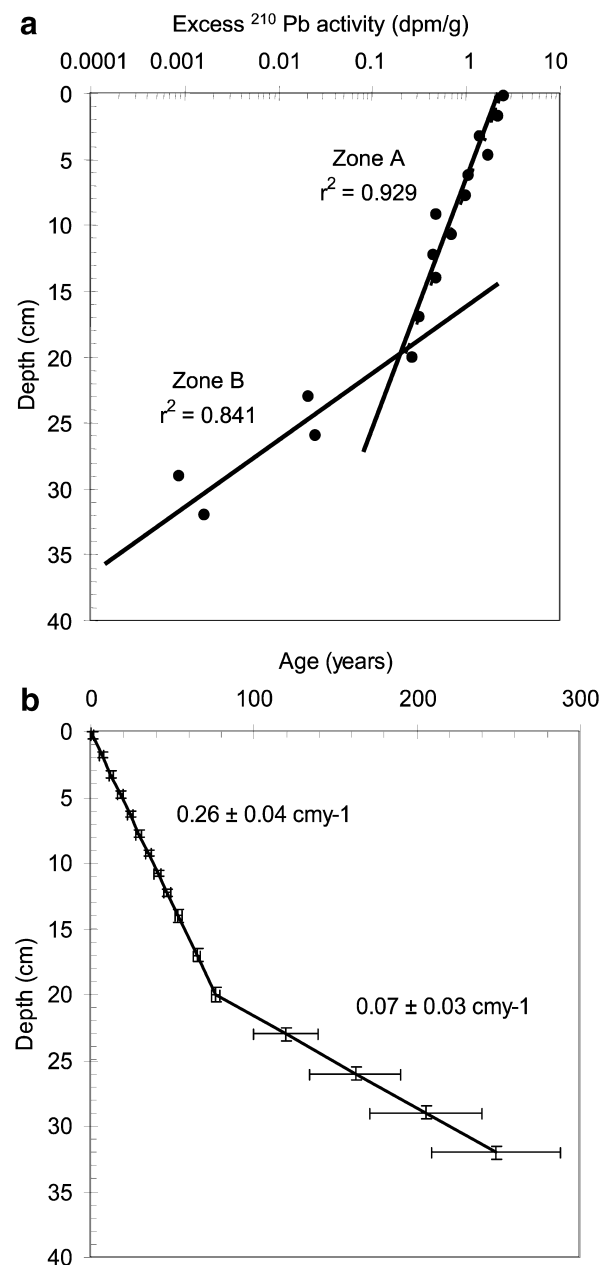
The ages thus determined for the core sediments were used to construct a geochronology of changes in metal concentrations (Fig. 3). Rainfall was investigated as a possible influence on the change in metal concentration. It was found that the major concentration fluctuations in the Pb, Zn, As and Cu profiles were coincident with heavy rainfall preceding drought periods. The two largest changes in metal concentration occurred between 1935 and 1953 and 1993 and 2000 and coincide with La Niña events. Two smaller fluctuations are evident at 1987 and 1982, the latter being coincident with a La Niña event. An interesting relationship between Ag and rainfall was observed and is shown in Fig. 5. There is a sudden increase in the concentration of Ag in the sediments when the annual rainfall exceeds a value of 1,800 mm.

A chronology of core sediment parameters such as grain size and organic matter content are indicative of changes in the catchment and depositional environment over the last 100 years. The major change in grain size distribution and organic content of the sediments is coincident with dam construction between the years 1940 and 1960. Prior to dam construction, the mouth of the Tonalli River was a higher energy fluvial environment dominated by a stream bed and some fluvial terraces. Now a lacustrine environment, the lower energy promotes the deposition of finer grained material. Along with the change in sediment type, the rate at which the sediment deposited also changed. Prior to dam construction, coarse sediments deposited at a slower rate of 0.07 cm year^{-1}, typical of fluvial flats, while the more recent fine sediments are depositing at the higher rate of 0.26 cm year^{-1}. The metal concentration record in

Fig. 4
a Unsupported ^{210}Pb activity profile showing the two zones of sedimentation and corresponding correlation coefficients and **b** calculated t-values for the sediment core versus depth and average sedimentation rates
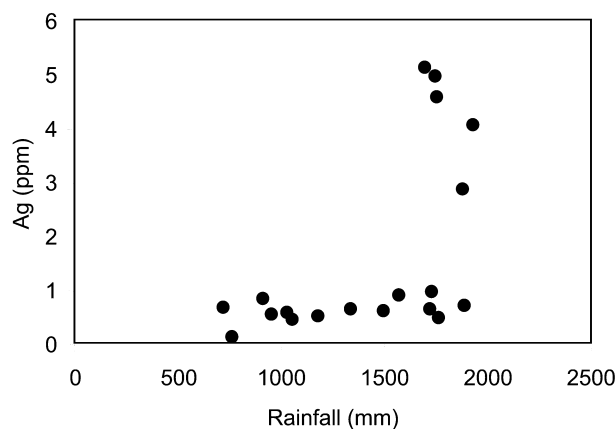

Fig. 5
Relationship between Ag concentration in sediments and annual rainfall

the sediment core does not indicate that the level of contamination has substantially decreased in time, even though mining terminated in 1928. There appears to be some correlation with grain size as there has been an overall increase in metal concentrations in response to the deposition of finer sediments after dam construction. The fluctuation of some metals such as Pb and Zn showed a simple correlation with climate variability; increased metal concentration in the sediments coincide with heavy rainfall following long periods of drought. Therefore the impact is much more pronounced because the sediments depositing are effectively concentrated by lower water levels. In the case of Ag, the relationship was not a simple one, there appeared to be a threshold rainfall value above which Ag becomes incorporated in higher concentrations into the sediments. This evidence suggests that although rainfall is one of the driving forces behind pollution distribution in the catchment, there are at least two different modes of pollution transport to the river: direct particulate transport from the processing sites and the leaching of metals from the tailing material caused by the acidic nature of the water.

Surface sediment

The results for grain size distribution of the surface sediment samples varied considerably. The percentages of sand (>63 μm) and mud (<63 μm) fractions were determined and these results are summarised in Table 2. Surface sediment samples did not appear to show a downstream trend in finer grain size; rather, fine sediments were restricted to the tailing dumps at the processing sites and areas receiving direct runoff from them. The results for the percentage of organic matter in the surface sediment samples were varied and a trend was not apparent (Table 2). All surface sediment samples exhibit-

ing a relatively high organic matter content (>10%) were collected in areas of the Tonalli River with either a large proportion of aquatic plants (site #1) or below trees and shrubs that overhung the river (sites #7, #9 and #10). Concentrations of metals in surface sediments were greater in sediment samples collected in minor tributaries that carry direct runoff from the processing sites during rainfall events (sites #14, #16 and #20) and also in the Tonalli River at site #10. Samples collected upstream of the mining influence (sites #9 and #11) have low metal concentration values. Other surface sediment samples collected in both the Tonalli River and Basin Creek exhibit low metal concentrations. Sediments consistently contained higher levels of Pb, As and Zn than any other metal investigated and the spatial distribution of these metals is shown in Fig. 6. Low pH levels in water were observed around tailing dumps, and surface river sediment in close proximity to these dumps contained levels of Pb, Cu, As, Zn, Hg, Ag and Cd well above ANZECC sediment quality guidelines (400×, 14×, 200×, 22×, 3×, 11× and 11×, respectively). Figure 7 compares the concentrations found in the surficial sediments near each of the processing sites with ANZECC sediment quality guidelines.

Spatially, the majority of the investigated pollutants, originating from the tailings, stay nearby, creating a potential
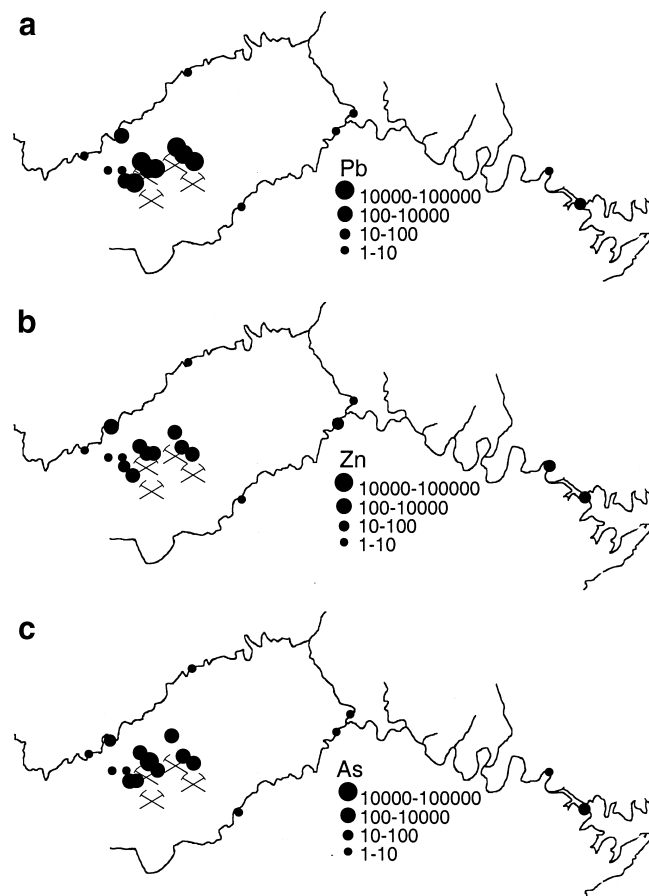
Table 2
Grain size and organic matter content results for surface sediment. Errors estimated by coefficient of variation (CV) determined by multiple analysis of #10

Site number	Percent <63 μm	Percent >63 μm	Percent organic matter
1	47.5±0.5	53±1	12.7±0.4
2	19.1±0.2	81±2	1.20±0.04
3	33.1±0.3	67±2	7.9±0.2
4	5.4±0.1	95±2	1.31±0.04
5	13.7±0.1	86±2	3.7±0.1
6	3.99±0.04	96±2	1.43±0.04
7	63.0±0.7	37±1	15.6±0.5
8	4.63±0.05	95±2	0.97±0.03
9	52.0±0.5	48±1	15.5±0.5
10	73.5±0.8	27±1	21.1±0.6
11	13.9±0.1	86±2	5.8±0.2
12	13.1±0.1	87±2	6.6±0.2
13	3.61±0.04	96±2	1.9±0.1
14	29.3±0.3	71±2	5.2±0.2
15	35.3±0.4	65±2	6.2±0.2
16	56.1±0.6	44±1	8.8±0.3
17	14.9±0.2	85±2	2.3±0.1
18	7.0±0.1	93±2	1.34±0.04
19	95.4±1.0	4.6±0.1	5.0±0.2
20	2.12±0.02	98±2	1.62±0.05

Fig. 6
Distribution of **a** Pb, **b** Zn and **c** As in the Tonalli River surface sediments. All concentrations are in parts per million
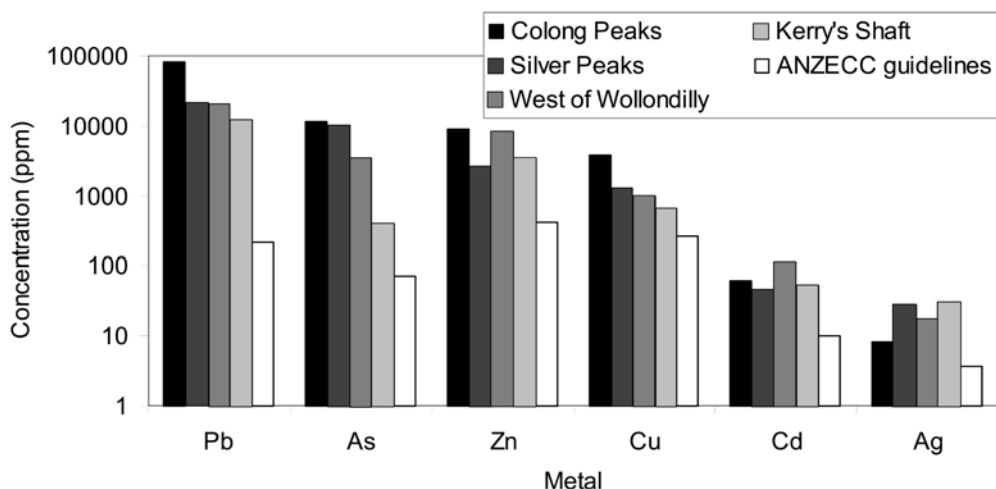
Fig. 7
Metal concentrations in surficial sediment collected in minor tributaries carrying runoff from Kerry's Shaft, Silver Peaks, West of Wollondilly and Colong Peaks

hazard of localised levels of toxic material, resulting from alteration of natural parameters such as the water acidity, causing serious environmental damage to smaller-scale ecosystems. The tailings continue to be a source of heavy metal contamination to the river, but distance from the mine-works is an important factor in the mitigation of pollution. The surface stream sediments collected downstream from the tailing dumps at the processing areas contained progressively lower amounts of heavy metals. This trend does not correlate with the grain size distribution of the sediments, suggesting that the levels of pollution still measured to this day are not influenced by the energy of the environment in a simple fashion.

The finer sediments of the Tonalli River located at the junction with Lake Burragorang still contained high pollutant concentrations, highlighting the necessity to not rely on water alone as an indicator of aquatic pollution. To obtain a better understanding of the potential hazard, it would be necessary to determine the bioavailable fraction of metals within the sediments of the Tonalli River.

To minimise further pollution of the Tonalli River catchment, remediation of the mine sites is essential. The results obtained in this project demonstrate that the best way to solve the local pollution problem is by directly addressing the mechanical and chemical erosion of the tailings at the processing areas. While it may be relatively simple and inexpensive to provide mechanical barriers to the input of finer particles into the river, this can be only a temporary solution. A detailed study of the chemistry and mineralogy of the tailings may provide a longer-term solution to the problem.

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