

Geochemical modelling of metal toxicity in the Tui mine catchment, Te Aroha, NZ.

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Abstract

The toxicity and bioavailability of metals in acid mine drainage (AMD) and neutral mine drainage (NMD) from old mine sites can be predicted using geochemical modelling. Such modelling of aluminium toxicity in West Coast streams was found to be very consistent with ecological assessments in the same streams, clearly identifying the threshold free aluminium ion concentrations at which impacts on aquatic ecology were observed. The focus of this research has now shifted to modelling the toxicity of trace metals in streams draining the recently rehabilitated Tui mine site in Te Aroha, in the Coromandel region of New Zealand.

Prior to its rehabilitation in 2013, the Tui mine was NZ's most contaminated site. While metal run-off from the mine tailing dam to Tui Stream has been significantly reduced by the rehabilitation works (liming and capping of the tailings dam), NMD discharging to the adjacent Tunakohia Stream still results in high concentrations of manganese, cadmium and zinc. Geochemical modelling using PHREEQC indicated that high proportions (70% - 96%) of these metals are present as toxic free metal ions. Ecological surveys in Tunakohia stream show a greatly reduced taxonomic richness, and the absence of mayflies. A fish survey in the Tunakohia showed an absence of fish in the upper and middle reaches. By contrast, in Tui Stream, the concentrations of these metals (again predominantly present as free ions) have been reduced to much less than their concentrations in Tunakohia Stream. Ecological surveys show that benthic invertebrate communities have recovered in Tui Stream to a greater degree than in Tunakohia Stream.

Keywords: Acid mine drainage, neutral mine drainage, geochemical modelling, PHREEQC, Tui Mine.

Introduction

Acid mine drainage (AMD) is an environmental issue associated with mining activities (Banks et al., 1997; Blowes et al., 2003; Nordstrom and Alpers, 1999). AMD occurs when the ore being mined contains iron sulphides which are oxidised when they come into contact with water and oxygen (Evangelou, 1995). This results in acidic mine drainage waters which contain high levels of sulfate ions and trace metals. If there is sufficient neutralising capacity in the rocks surrounding the mined ore deposit, the acid can be neutralised but the mine drainage waters still contain significant levels of sulfate ions and some trace metals (Banks et al., 1997). When this occurs the mine drainage water is said to be neutral mine drainage (NMD). Neutral mine drainage is often present in catchments draining lead and zinc mines which have a lower iron sulfide content and therefore a lower acid generating capacity (Blowes et al., 2003; Warrender et al., 2011). In both AMD and NMD waters, trace metal concentrations can be high enough to be toxic to aquatic organisms. Most water quality guidelines specify total or dissolved metal concentrations. However, as acknowledged in the free ion activity model (FIAM) and biotic ligand model (BLM), it is mainly the un-

complexed, dissolved free metal ion that is toxic (to aquatic species) as it can pass through the cell membrane (Campbell, 1995; Hassler et al., 2004). The concentration of the free metal ion is difficult to determine analytically, however, geochemical speciation modelling programs, such as PHREEQC (Parkhurst et al., 1980), can predict the proportion of complexed and free metal ions.

Previous work by the authors in AMD catchments on the West Coast of New Zealand found that a reliable threshold for aluminium toxicity to aquatic life could be found by plotting macroinvertebrate community values against free ion (Al^{3+}) concentrations (Waters and Webster-Brown, 2013). This paper presents the results of similar research to see whether speciation is as valuable as an indicator of trace element toxicity downstream of a heavy metal ore deposit, which is leaching zinc, manganese and cadmium into the environment.

Study area

The former Tui mine is located on the western slopes of Mt Te Aroha, near the township of Te Aroha, Waikato, New Zealand. Two streams, Tui Stream and Tunakohia Stream drain the mine area. Tui Stream drains the catchment on the west side of mine and receives discharge from the tailings dam. Tunakohia Stream drains the east side of the mine and receives discharge from several mine adits. These two streams flow through the township of Te Aroha before discharging into the Waihou River, which flows into the Firth of Thames. Tui mine was operational between 1966 and 1974 as a lead and zinc mine but was abandoned when the company operating the mine went into liquidation (Morrell, 1997). However, the mine adits and tailings dam continued to leach metals into the nearby streams.

In 1979-80, metal concentrations in the stream below the tailings dam, discharging into Tui Stream, were found to be up to 20mg/L Mn and 50mg/L Zn, while pH was as low as 3.2 (Tay, 1980). Metal concentrations in the upper reaches of Tunakohia Stream were not as high, at up to 1.67mg/L Mn, 23.8mg/L Zn and 0.14mg/L Cd, for a near neutral pH of between 6.89 and 7.84 (Tay, 1980). Almost 20 years later, metal concentrations in both streams remained elevated with up to 11.4 mg/L Mn and 44.1 mg/L Zn and an acidic pH (2.8) in Tui Stream (Webster, 1995), and up to 8.17mg/L Zn and 0.063mg/L Cd in Tunakohia Stream (Hickey and Clements, 1998). By 2002, water quality around the mine site had deteriorated even further, with surface drainages near the tailings dam containing up to 25.4 mg/L Mn, 470mg/L Zn and 5.4mg/L Cd, with pH as low as 2.45, and neutral pH adit discharges containing up to 1.17mg/L Mn, 20.2mg/L Zn and 0.16mg/L Cd (Harvey, 2003). Partial remediation of the tailings dam to stabilise the tailings pile in 2006 reduced drainage seeping through the dam, but did not significantly improve water quality with heavy metal concentrations in the Tui Stream tributary still acidic and with up to 18mg/L Mn, 46mg/L Zn and 0.2mg/L Cd (Sharplin, 2008).

Consequently the Tui Mine became recognised as New Zealand's most contaminated heavy metal site (Sabti et al., 2000). This ranking, along with perceived ongoing instability of the tailings dam in a major seismic event, prompted the local regional and district councils to rehabilitate the site (WRC, 2013). Rehabilitation works were carried out at the site between 2010 and 2013, when the adits were plugged and flooded with a limestone slurry and the tailings dam was stabilised, capped and planted with grasses (WRC, 2013). Ecological and water quality monitoring is still being undertaken at the site (by consultants to Waikato Regional Council), to evaluate progress being made towards recovery of the water quality of the Tui and Tunakohia streams.

Methods

In this study water samples were collected at ten sites in this catchment (Fig. 1). Four sampling sites were located along Tui Stream including a reference site (TuiC) upstream of the input from tailings dam. Four sampling sites were located along Tunakohia Stream as well as a reference site (TunaC) on the south branch of the Tunakohia Stream, which is unaffected by mining works at the Tui Mine. One other reference site was located on the Wairongomai Stream which is in a separate catchment with similar geology. This site was included in order to compare with historic sampling at this site (Hickey and Clements, 1998). Samples were collected in November 2015 in order to avoid the effects of high rainfall during the winter months and the lower stream flow of the summer months.

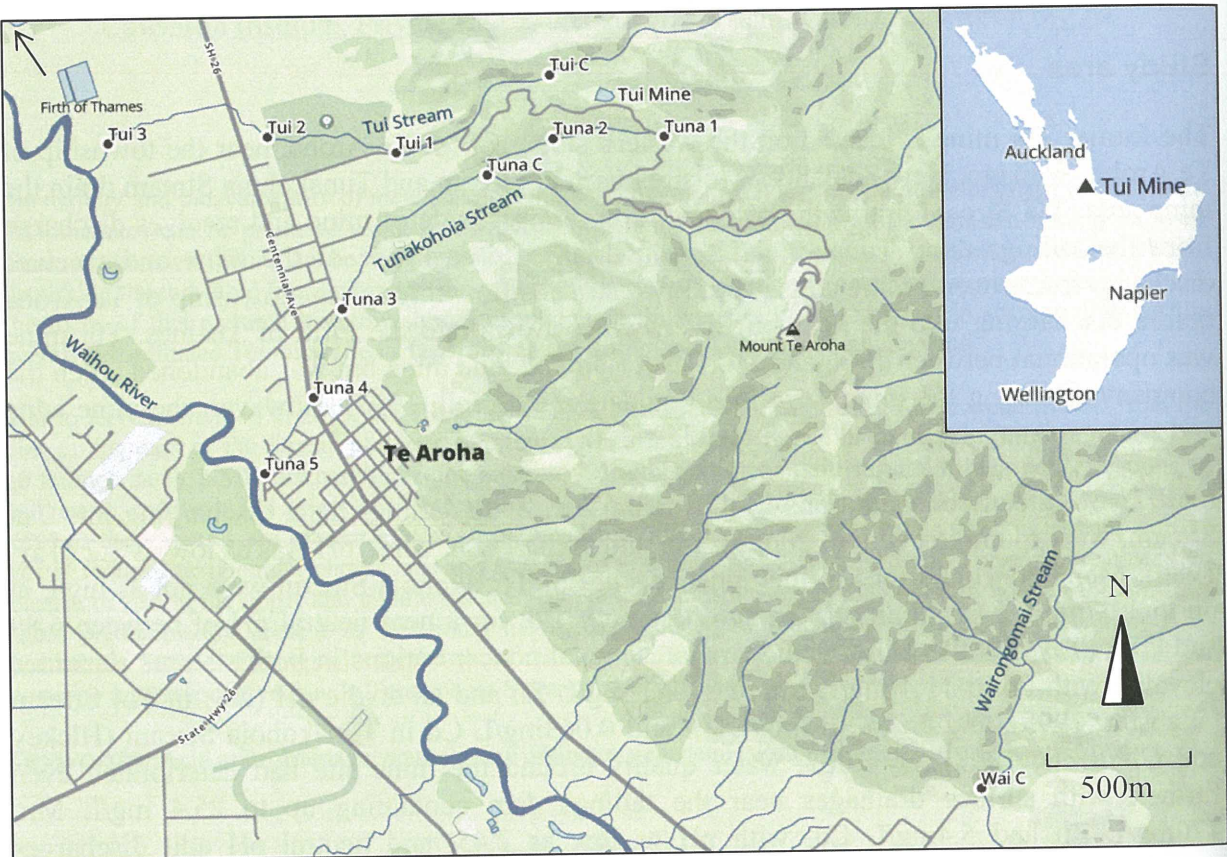


Figure 1. Map of study area near Te Aroha, Coromandel, showing water sampling sites used for this research.

Measurements of temperature, pH, conductivity and dissolved oxygen were made *in situ* using a HACH HQ40d portable multi parameter meter. Three 50mL water samples were collected in sterile HDPE centrifuge tubes; one water sample for major ions and carbonate determination, and two water samples (one unfiltered and one filtered through 0.45 µm) acidified with Aristar concentrated nitric acid for trace element analysis. Samples were analysed for major cations and trace elements by ICP-MS (University of Canterbury) and for major anions by HPLC (Lincoln University). Carbonate concentrations were determined by IRGA at the WCFM laboratories at Lincoln University.

Speciation modelling for the dissolved metals was carried out using the geochemical modelling program PHREEQC (Parkhurst et al., 1980), using the WATEQ4F thermodynamic database.

Macroinvertebrate samples were collected in December 2014 according to the protocol of Stark et al. (2001). Samples were preserved in the field with 70% ethanol. Samples were then sieved through 500µm mesh in the laboratory. Invertebrates were counted and identified to the lowest taxonomic level using identification guides of Winterbourn et al. (2006).

Results and discussion

Water chemistry

Table 1 shows the water chemistry at the sites during the November 2015 sampling survey.

Table 1. Chemistry of the Tui and Tunakohia Streams in November 2015. For trace metals, the dissolved fraction (<0.45µm) is shown, and concentrations denoted in **bold** are above ANZECC (2000) trigger value aquatic ecosystem guidelines for the protection of 95% aquatic species, as shown at the base of the table.

Site	pH	Temp (°C)	DO (mg/L)	Cond (µS/cm)	Cl ⁻ (mg/L)	NO ₃ ⁻ (mg/L)	SO ₄ ²⁻ (mg/L)	HCO ₃ ⁻ (mg/L)	Na ⁺ (mg/L)	Mg ²⁺ (mg/L)	K ⁺ (mg/L)	Ca ²⁺ (mg/L)
Tuna1	7.42	13.7	9.69	666	10.2	0.38	285	69.5	13.1	18.1	1.07	109
Tuna2	7.12	12.2	10.1	409	11.8	0.66	154	32.9	13.1	13.7	1.24	50.3
Tuna3	6.84	13.8	9.88	308	12.2	1.03	93.6	34.1	12.7	10.2	1.12	34.6
Tuna4	6.92	15.5	9.39	249	12.7	1.16	65.4	41.7	11.7	7.84	1.04	25.5
Tuna5	7.52	19.6	10.1	252	12.9	0.92	65.6	39.0	11.7	7.52	1.10	24.5
TunaC	7.07	12.1	9.85	102	11.0	0.19	14.6	26.8	7.60	2.81	0.52	8.03
Tui1	7.40	11.9	10.3	150	10.7	0.19	29.2	32.2	9.44	4.38	0.84	12.9
Tui2	7.23	12.9	9.94	150	10.8	0.22	29.6	29.2	9.47	4.37	0.91	12.4
Tui3	7.31	15.9	8.98	150	11.7	0.78	28.6	32.1	10.0	4.65	1.09	11.6
TuiC	7.21	11.4	9.97	125	10.6	0.31	20.1	31.8	8.90	3.34	0.68	10.8
WaiC	7.28	14.3	9.75	141	9.27	0.04	36.7	18.1	7.20	4.30	0.69	12.5

Site	Fe (µg/L)	Mn (µg/L)	Cu (µg/L)	Pb (µg/L)	Zn (µg/L)	Cd (µg/L)	Ni (µg/L)
Tuna1	5.06	477	2.77	2.30	2470	15.9	8.24
Tuna2	3.14	75.3	3.87	2.47	1840	13.3	3.75
Tuna3	9.30	1.75	0.92	0.13	647	6.49	1.35
Tuna4	13.0	4.03	0.49	0.10	342	3.89	0.89
Tuna5	26.1	20.5	1.14	0.10	181	2.40	0.66
TunaC	8.29	0.48	0.33	<0.03	0.81	0.02	0.17
Tui1	2.88	0.40	0.84	0.05	45.7	0.36	0.59
Tui2	20.2	10.5	0.90	<0.03	53.9	0.39	0.56
Tui3	66.7	47.8	0.94	0.09	37.6	0.23	0.60
TuiC	2.77	0.20	0.17	<0.03	9.63	0.06	<0.04
WaiC	11.7	9.75	0.25	0.04	2.57	0.03	2.21
ANZECC	-	1900	1.4	3.4	8.0	0.2	11

Dissolved iron and manganese concentrations both show an initial decrease in the upper catchments, followed by an increase in the lower reaches of the Tui and Tunakohia Streams. Copious precipitation of brown iron oxide is evident in the upper reaches of both catchments, which would reduce the dissolved iron concentration (any iron oxide solids suspended in the water column of greater than 0.45µm will be excluded from this measurement). It appears likely that precipitates carried into the lower reaches may re-dissolve to a degree, leading to higher iron and manganese concentrations downstream. For manganese, there may also be additional municipal and agricultural sources of contamination to the lower Tunakohia stream. There was little significant difference between iron and manganese concentrations for the different catchments.

Dissolved zinc and cadmium concentrations decrease along the length of the Tunakohia Stream, from the sampling site at the top of the stream near adit 4 to where the stream enters the Waihou River. However, all sites (except for the reference site on the south branch of the stream) remain well above the ANZECC (2000) guidelines for the protection of 95% of the aquatic species. Similarly, for Tui Stream, dissolved zinc and cadmium concentrations vary downstream but remain above ANZECC (2000) guidelines. Even the reference site (TuiC) is just above the ANZECC (2000) guideline for zinc, and this is upstream of any tailings drainage input so may reflect naturally elevated levels from host rock weathering. The difference between the zinc concentrations in the two streams is close to being statistically significant (at $P=0.079$).

Dissolved lead and nickel concentrations are highest in the upper Tunakohia Stream catchment, but remain below the ANZECC (2000) guideline trigger values. Lead and nickel concentrations decrease to levels similar to that of the reference sites in the lower reaches of Tunakohia Stream. ANZECC (2000) guidelines for copper are exceeded only at sites Tuna1 and Tuna2, and while concentrations do remain elevated relative to the reference site in the lower reaches of the stream, they are below the guideline trigger value. In Tui Stream, while dissolved lead, copper and nickel concentrations at impacted, upper catchment sites are elevated above the background levels observed at the reference site, they are consistently below ANZECC (2000) guideline trigger values for moderately impacted streams.

Metal speciation

Figure 2 shows PHREEQC calculated dissolved metal speciation for those metals exceeding ANZECC (2000) guidelines; zinc, cadmium and copper, and for manganese which is also very elevated in the upper Tunakohia catchment. The speciation of dissolved zinc and cadmium (Figs 2a and 2d) shows that, despite the concentration changes along the length of the Tunakohia Stream, free metal ions (Zn^{2+} and Cd^{2+}) dominate the speciation at all sites and remain orders of magnitude higher than the ANZECC (2000) guidelines for the protection of aquatic species at all sites, except for the reference site. Less than 30% of dissolved zinc and cadmium is complexed, mainly by sulfate ligands. Similarly in the Tui Stream catchment, free Cd^{2+} and Zn^{2+} ions dominate the speciation of dissolved cadmium and zinc respectively, with less than 15% complexed by sulfate and other ligands. Concentrations are lower than in Tunakohia Stream but still exceed ANZECC (2000) guidelines.

The speciation of dissolved copper (Fig. 2b) indicates a lesser role for the free metal ion, which constitutes only up to 40% of the dissolved copper concentrations in Tunakohia Stream and up to 15% in the Tui Stream. Complexation of copper, mainly by the hydroxyl ion, has reduced the concentration of free Cu^{2+} ion to below ANZECC (2000) guidelines at all sites.

Dissolved manganese concentrations are below ANZECC (2000) guidelines for ecosystem health, but do exceed ANZECC (2000) guidelines for contact recreation (Fig 2c). Free Mn^{2+} ion makes up the majority (>80%) of dissolved manganese at all sites. At Tuna1 manganese complexed by sulfate does make a reasonable contribution (20%) to the overall dissolved manganese concentration, but Mn^{2+} is still highest at this site and remains above the contact recreation guidelines.

In assessing the benefits of remediation, it appears that the addition of lime slurry to the adits draining to the Tunakohia has done little to reduce the toxicity of dissolved cadmium and zinc. Neither the concentration of the dissolved metal nor the concentration of the free metal

ion have been significantly reduced by this action. This may reflect the fact that pH of adit drainage has always been high (near neutral) relative to the more acidic drainage from the tailings dam (Tay, 1980; Webster, 1995; Sharplin, 2008).

Dissolved and free metal ion concentrations in Tui Stream have been reduced to a greater extent as a result of remediation of the Tailings dam in this catchment. However, while dissolved and free copper ion concentrations are below ANZECC guidelines in Tui Stream, free cadmium and zinc concentrations do still exceed these guidelines.

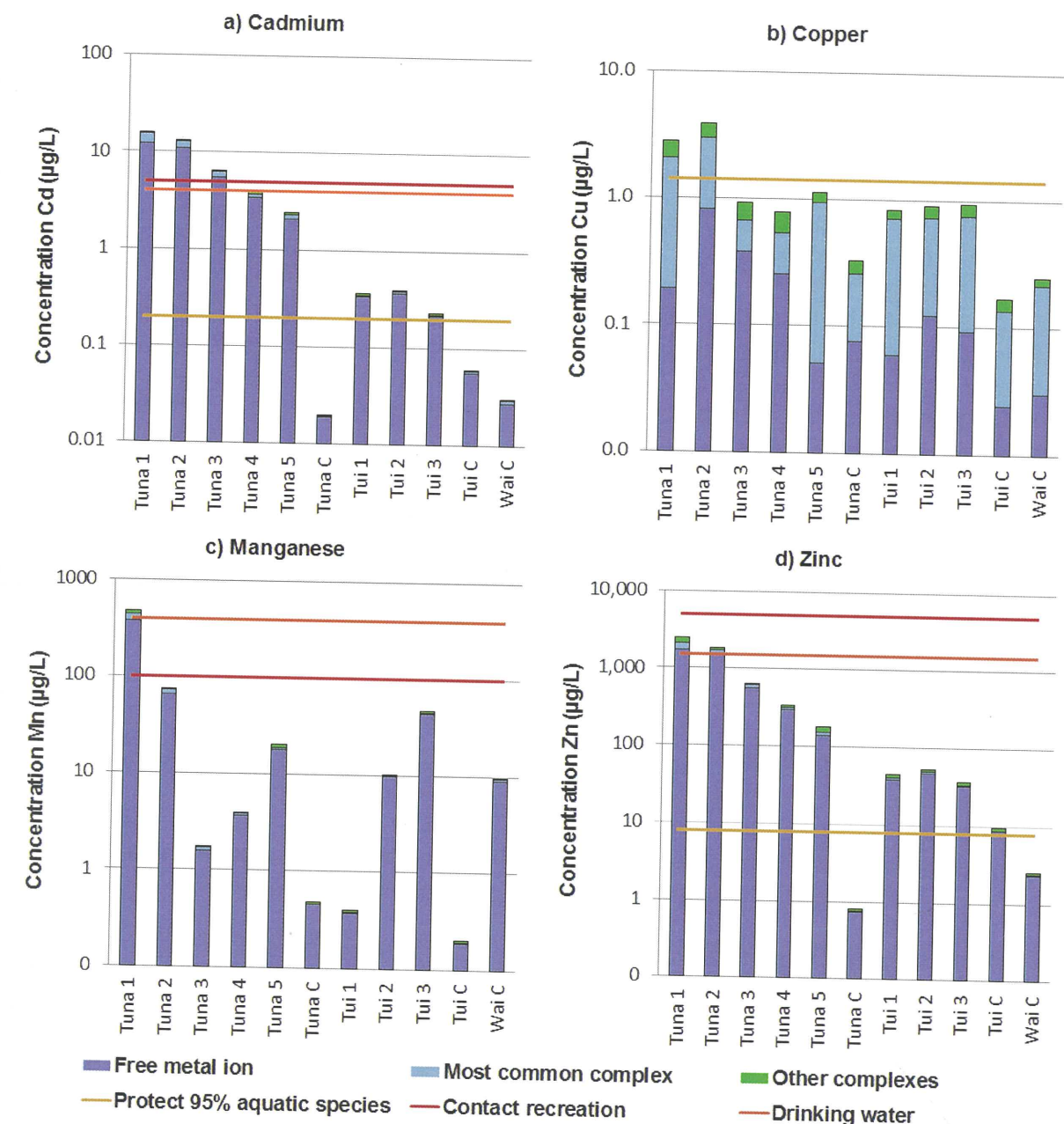


Figure 2. PHREEQC predicted speciation for dissolved metals which exceed ANZECC (2000) guideline for aquatic ecosystem protection or for contact recreation (ANZECC, 2000). The concentrations specified as MAVs in the NZ DWS (MoH, 2008) are also shown. Note the log scale on the y axis.

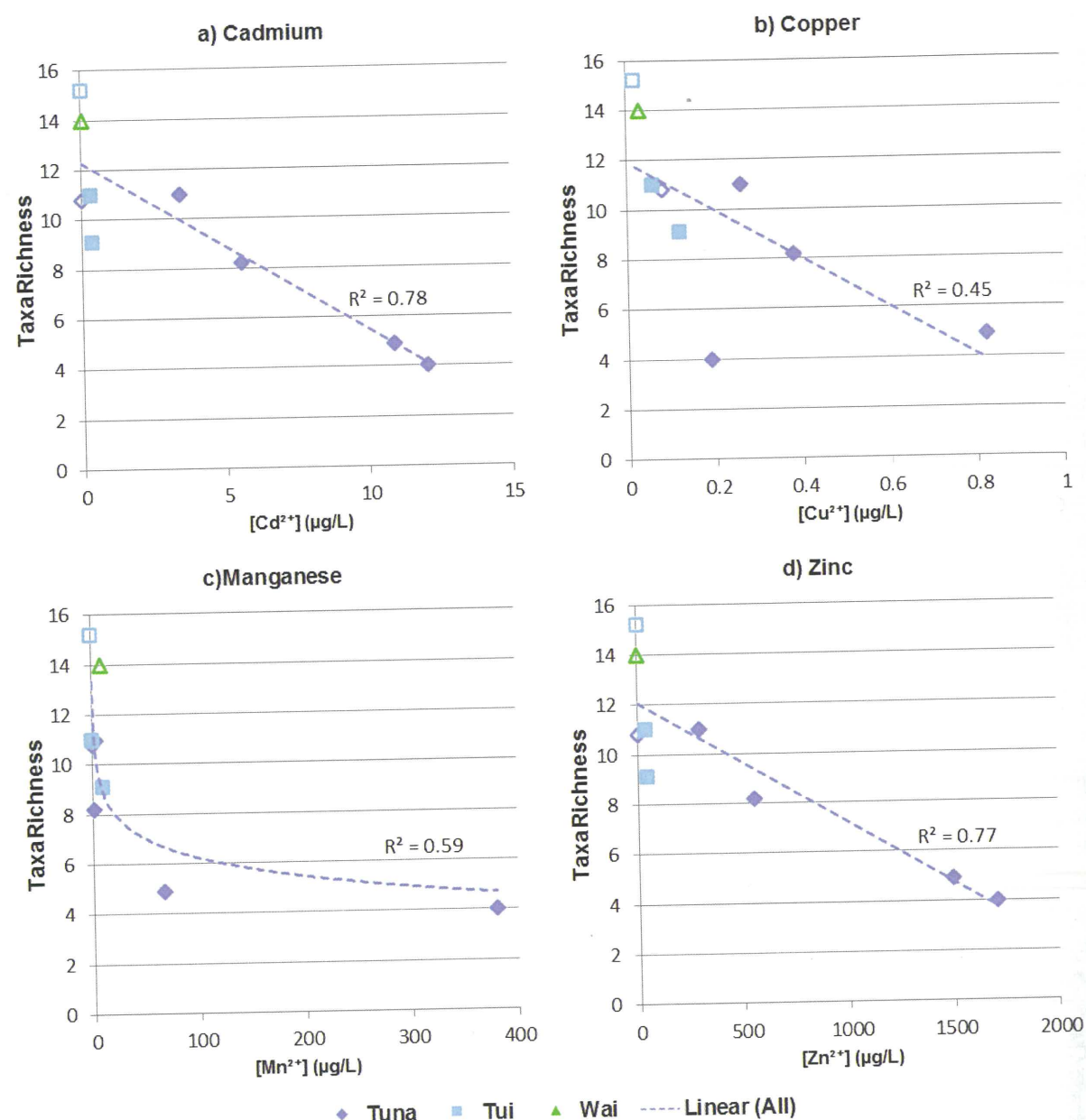


Figure 3. Free metal ion concentrations plotted against macroinvertebrate taxa richness. Open data points denote reference sites.

The relationship between macroinvertebrate taxa richness and the dissolved free metal ion concentrations is shown in Fig. 3. Free Cd^{2+} and Zn^{2+} ion concentrations show a moderate linear relationship with taxa richness (Figs 3a and 3d) with $R^2 = 0.78$ Cd, $R^2 = 0.77$ respectively. Because free Cd^{2+} and Zn^{2+} ions make up a significant proportion of the total metal concentrations, taxa richness will also show a similar relationship with total or dissolved cadmium and zinc concentrations.

A linear relationship between taxa richness and free Cu^{2+} ion concentration (Fig. 3b) is not evident ($R^2=0.47$). Given that the majority of dissolved copper is predicted to be complexed, and all Cu^{2+} values are below ANZECC (2000) guidelines for ecosystem protection, this confirms copper concentrations are not having a significant impact on the aquatic ecosystems of these catchments. The relationship between free Mn^{2+} ion concentration and taxa richness (Fig. 3c) appears stronger (though not linear), but this is heavily influenced by the Tuna1 site

which has a very high free Mn^{2+} concentrations and dictates the logarithmic trend ($R^2 = 0.59$). As Mn^{2+} is only high at Tuna1, while low macroinvertebrate numbers were observed at both sites Tuna1 and Tuna2, this is likely due to factors other than manganese toxicity.

The data collected for aluminium in West Coast streams by Waters and Webster-Brown (2013) showed a threshold below which taxa richness increased rapidly. The approach taken by Waters and Webster-Brown (2013) was to fit a trend line through two data sets, one for high free metal ion concentrations which showed a steep gradient, and a second for lower free metal ion concentrations with a shallower gradient. Where these trends met a “threshold” for Al^{3+} toxicity was identified. This was found to be 0.46mg/L , which corresponds to approximately 0.6mg/L for dissolved aluminium as Al^{3+} makes up a significant proportion of dissolved Al in these streams. This is well above the ANZECC (2000) guideline value for protection of aquatic life of 0.055mg/L and is not intended to be an overall guideline value, but instead acts as a reasonable remediation target.

In this study, however, a reduction in taxa richness with increasing free zinc and cadmium ion concentration was shown but no clear threshold value was observed. Instead a consistent linear relationship for all metal concentrations was observed. As a first approach, we have identified arbitrary thresholds as those which maintain taxa richness at $>50\%$ of the reference site taxa richness. The free ion concentrations at which this threshold is passed in Tunakohia Stream are approximately $10\mu\text{g/L}$ for cadmium, and $1300\mu\text{g/L}$ for Zn. However it must be noted that cumulative effects of high Zn and Cd concentrations (as well as possibly sub-toxic levels of Mn and Cu) have not yet been considered. In Tui Stream taxa richness remains above this arbitrary threshold in both of the sites downstream of the tailing dam, consistent with the lower levels of metals and free metal ions at these sites. Remediation of the tailing dam, and the subsequent reduction in acidity and heavy metal concentrations in Tui Stream, has clearly been more effective in terms of ecosystem recovery, than remediation works in the Tunakohia Stream catchment.

Conclusion

The preliminary findings from this study indicate that remediation efforts at Tui Mine have improved the water quality in Tui Stream such that dissolved and free metal ion concentrations are now below ANZECC (2000) guidelines for aquatic ecosystem protection for all metals except cadmium and zinc. Water quality improvements have enabled macroinvertebrate species to recolonise the upper reaches of the Tui Stream.

However, dissolved copper, cadmium, manganese and zinc concentrations in the Tunakohia Stream still exceed ANZECC (2000) guidelines for aquatic ecosystem protection, and for all except copper, free metal ion concentrations also exceed these guidelines. Copper has been complexed sufficiently by hydroxide ligands to fall below the ANZECC (2000) guideline value. Tunakohia Stream still has low macroinvertebrate taxa richness where free zinc and cadmium ion concentrations are high, exceeding the threshold values proposed of $10\mu\text{g/L}$ for Cd, and $1300\mu\text{g/L}$ for Zn. Further work is required to separate the cumulative effects of multiple elevated metals.

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