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Environmental impact of former gold mining on the Orangi river, Serengeti N.P., Tanzania

R. J. BOWELL[1], A. WARREN[2], H. A. MINJERA[3] and N. KIMARO[3]
[1] *7 Sheldrak Gardens, Hordle, Lymington, Hants, UK;* [2] *Department of Zoology, The Natural History Museum, London, UK;* [3] *Maji, Arusha P.O. Box 3020, Tanzania*

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Abstract. The Orangi river is an important all-year source of water for wildlife in the northern part of the Serengeti National Park. At two points along the river in the Banagi area, tributaries draining the adit and tailings of the Kilimafeza mine impact the Orangi. The former Au-Cu mine is subject to occasional wet season flooding leading to the release of iron ochres from the adit and physical as well as chemical mobilization of tailings material. The unpolluted river chemistry is essentially Na-Ca-HCO_3^- and well-buffered. Drainage waters from the tailings are characterized by low pH (2.3) and high concentration of sulphate (up to 3280 mg/l), aluminium (275 mg/l), arsenic (324 mg/l), copper (125 mg/l), iron (622 mg/l), lead (21 mg/l), manganese (65 mg/l), and zinc (126 mg/l). Adit-drainage waters are typically of a lower pH (4.6) and have a lower concentration of sulphate (up to 1840 mg/l) and metals (up to 25 mg/l Al, 92 mg/l As, 42 mg/l Cu, 258 mg/l Fe, 9.6 mg/l Pb, 53 mg/l Mn, and 102 mg/l Zn). Mixing of these acidic waters with the alkaline river leads to rapid metal precipitation as Fe-ochre coatings on clastic sediment. This effect is more noticeable in the dry season. Consequently, although the tributaries draining the two sources are heavily contaminated, the effective buffering of the mine drainage waters restricts any potential pollution to within 1 km of the mine workings. Faecal coliforms show an antipathetic relationship to low pH and high metal conditions. The only mobile metals in the water outside this area are Mn and Zn and their contamination can be biologically monitored using a protozoan-based bioassay.

Introduction

The Serengeti National Park in northern Tanzania is one of the most populated wildlife reserves in Africa with a large mammal population in excess of 2 million. It is also an important source of revenue for tourism. The area is part of the Lake Victoria goldfields (Foster & Piper 1993) and active gold mining occurred from 1933 to 1966 producing 8810 kg Au (Barth 1990). The legacy of this mining can be observed in the Banagi area where the remains of the Kilimafeza mine (the largest mine within the parks boundary) are still present. These consist of a small pile of sulphide tailings (the exact dimensions unknown as much of this is partly revegetated) and an open adit. The area is an important focus for the Serengeti's elephant population due to the combination of acacia woods and open grasslands. The impact of potentially toxic elements on wildlife is poorly understood, although toxic metal pollution has been cited as a cause of 'paralysed trunk disease' in elephants around Lake Kariba, Zimbabwe (Jordan 1994). Aqueous discharge from mines can seriously affect local groundwater quality. Oxidation of sulphide ores, particularly those rich in iron-bearing sulphides, introduces high concentrations of metals, hydrogen ions, and sulphate ions into waters fed by mine discharge (Lowson 1982; Nordstrom 1982; Chapman et al. 1983; Rampe & Runnells 1989; Blowes & Jambor 1990; Davis et al. 1991; Blowes et al. 1992; Fuge et al. 1993). These acidic waters affect aquatic life, and in some instances, the quality of drinking water supplies.

Faecal pollution is another major factor affecting the quality of both surface and groundwater in Tanzania (Warren et al. 1993). Several important diseases of man and animals are caused by waterborne pathogens, e.g. dysentery, cholera, polio etc., all of which are usually transmitted by the consumption of contaminated water. testing water for the presence of all such pathogens individually would be costly and impractical. Therefore, indicator organisms are used to signify the presence of faecal contamination and, therefore, the possible presence of enteric pathogens. The most commonly used indicator organisms are faecal coliform bacteria. Faecal coliforms are enteric bacteria that normally reside harmlessly in the large intestine. They are excreted in large numbers when animals defaecate. The presence of any faecal coliforms in waters used for drinking or for irrigation would be of concern since the WHO guideline value is zero/100 ml in drinking water. Comparatively little attention has been given to the effect of acid mine drainage on the survival of faecal coliforms in the aquatic environment. One of the aims of this research is to investigate the distribution of faecal coliforms in the Orangi river-Kilimafeza mine system in order to assess the impact of acid mine drainage on these important indicator organisms.

Effective management and remediation of acid mine drainage is possible only if the processes that influence metal release and transportation are fully understood. From a long-term modelling study of the geochemistry of sulphide-rich tailings at the Heath Steele mine, New Brunswick, the processes affecting the mobilization of metals and production of acid mine drainage were found to be continuously active (Blowes et al. 1992). This suggests that, providing dissolved oxygen and water can circulate throughout the tailings pile, these processes can continue on geological time scales (Blowes et al. 1992). The influence of seasonal changes has also been shown to effect geochemical processes in uncontaminated stream sediments (Ridgeway & Dunkley 1988; Bowell & Ansah 1993). Consequently sampling was conducted in wet (April 1992) and dry (October 1992) seasons in the Banagi area. This study aims to assess the contribution of metals from the mine site to the Orangi river, metal speciation, and fluctuations in the impact due to seasonal variations.

Ecology

The woodlands in the area studied are dominated by acacia (*Acacia xanthophloea, A. clarigera, A. tortilis* and *A. mellifera*) and average rainfall is 5500 mm p.a. (recorded at Seronera Lodge, J. Saar per. comm.). Flooding of the Orangi river occurs between March and May with the maximum flow recorded as 0.53 m^3/s. The average air temperature in the area is 24 °C with the range being 11 to 30 °C. The Serengeti is host to a mammal biomass population in excess of two million which is described in great detail elsewhere (Hanby 1988).

Geology

The Archaean rocks of the Serengeti, which host the gold mineralization, have been described by numerous workers (recent comprehensive reviews given by Cahen & Snelling 1984; Barth 1990; Foster & Piper 1993, amongst others). The sequence comprises amphibolite, feldspathic quartzite, migmatitic quartz-feldspar gneiss, and kyanite schists which are representive of an ancient granitoid-greenstone terrain (Fig. 1). Younger granitoid intrusives, dated at ca. 2850–2250 Ma, puncture this late Archaean terrain (Cahen & Snelling 1984; Foster & Piper 1993). Two major successions have been recognized in the greenstone belts, which host the gold: the lower Nyanzian system, dated at 2850 to 3118 Ma, comprises a mafic volcanic group, an andesitic to felsic volcanic group, and a clastic group. The overlying Kavirondian has been dated at 2560 to 2850 Ma and is dominately clastic (Geosurvey 1979, 1981; Cahen & Snelling 1984). The volcanics in the Nyanzian, which host the gold mineralization, have been metamorphosed to lower greenschist facies. Gold mineralization consists of quartz veins and disseminated sulphides in quartz-carbonate-chlorite shear zones (Van Straaten 1984; Barth 1990). The major sulphide present is pyrite with minor arsenopyrite, galena, and sphalerite. Gold occurs predominately as native gold. Where the weathered remnants of mineralization are preserved, azurite, böhmite, cyanotrichite, gibbsite, goethite, hematite, jarosite, malachite, and scorodite have been reported (Geosurvey 1979, 1981; MADINI 1984–5; Van Straaten 1984; Barth 1990). In this study the only secondary minerals containing heavy metals identified in gossan and tailings material were azurite, böhmite, gibbsite, goethite, hematite, jurbanite, jarosite, and scorodite.

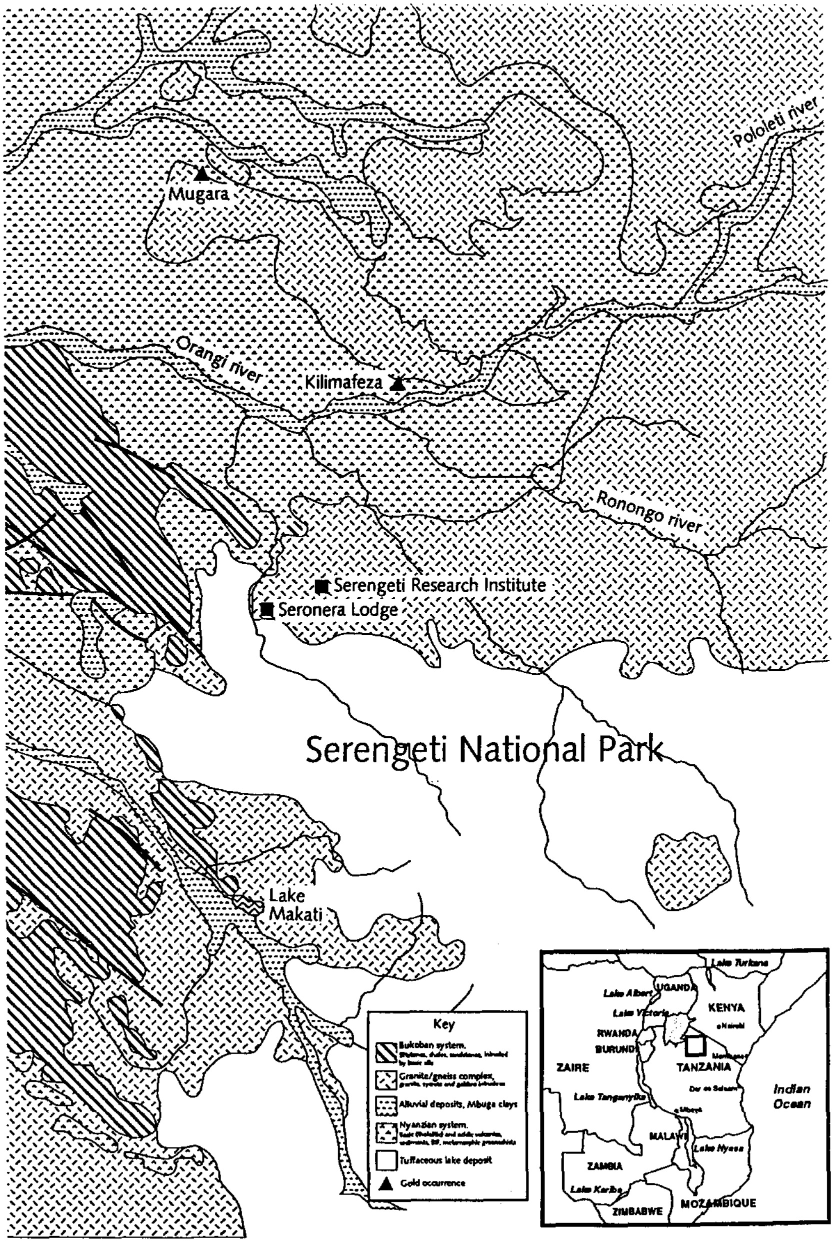

Fig. 1. Geology of the Serengeti goldfields (after Barth 1990).

Sampling methodology

A total of 6 sites were established in the vicinity of the Kilimafeza mine and a further 8 along the Orangi river along with a control site 2 km upstream

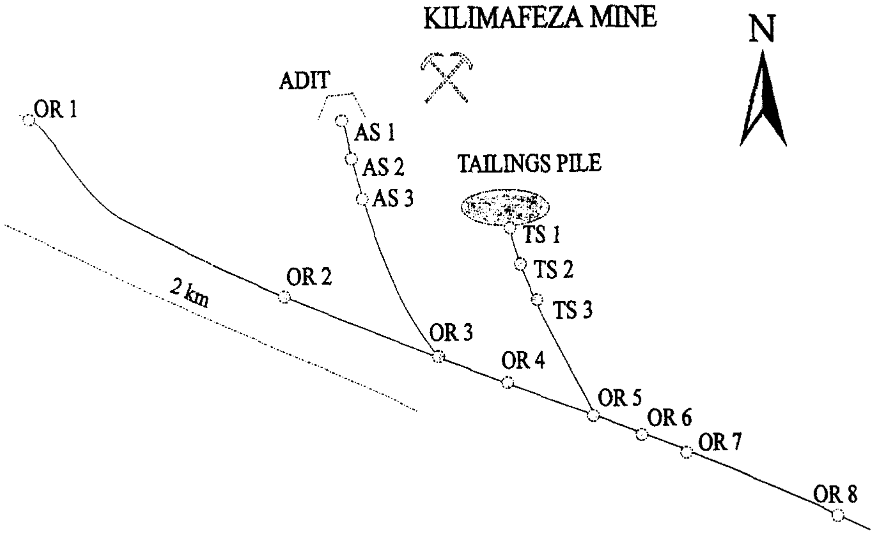

Fig. 2. Sample location map of Kilimafeza mine and Orangi river.

(Fig. 2). At each site 0.5 kg of sediment was collected by a hand-held auger and 3 × 1 l water samples were collected; one unfiltered; unfiltered and fixed with 10% HNO_3 acid; filtered through a 0.45 mm Durapore membrane filter and acidified with 10% HNO_3 acid. Measurements of Eh, pH, electrical conductivity (E.C.), temperature and dissolved oxygen were made at each sample site using an Oakton field kit.

Faecal coliforms were enumerated in the field using a DelAgua water testing kit (DelAgua Ltd., University of Surrey, Guildford, UK). Immediately upon collection, water samples (100 ml, 50 ml or 10 ml) were passed through a 0.45 μm sterile filter membrane. Each membrane was placed onto a pad containing Membrane Lauryl Sulphate broth, and incubated for 14 to 18 hours at 44 oC. Colony forming units of faecal coliform bacteria were then counted.

Additionally sulphate and copper concentration measurements were made in the field to monitor sample degradation. Sulphate analysis was by titration using a conditioning reagent of sodium chloride and hydrochloric acid and barium chloride. The level of turbidity produced was then compared to a sulphate comparator with 25 standards (PSAT kit 7778, LaMotte Chemical Company). Copper was analyzed by a field colourimeter using sodium diethyldithio-carbamate as a reagent. For all samples in which both were present, a good agreement was found between field and laboratory results (Table 1) inferring that little sample degradation had occurred.

Table 1. Comparison of field and laboratory results. All concentrations in mg/l.

Site	Field result	Laboratory result
OR 1d Cu	0.2	0.19
SO_4	11.0	11.5
pH	7.3	7.29
OR 5d Cu	10.2	8.9
SO_4	>300	385
pH	6.0	6.05
OR 7d Cu	0.4	0.36
SO_4	65	78.2
pH	6.8	6.83
TS 1d Cu	>100	125
SO_4	>300	3280*
pH	2.3	2.26
TS 3d Cu	27	26.3
SO_4	>300	2160*
pH	5.8	5.85
AS 1d Cu	40	42
SO_4	>300	1840*
pH	4.6	4.56
AS 3d Cu	10	9.2
SO_4	>300	730*
pH	5.9	5.87

* measurement after dilution
Cu and sulphate concentrations in mg/l

Mineral identification was carried out by optical and electron microscopy and confirmed by X-ray diffraction (XRD using PC IDENTIFY) and Fourier transform infra-red spectroscopy. Chemical analysis of the ochres was carried out by inductively coupled plasma atomic emission spectrometry (ICPAES, by Analabs (UK) Ltd) and the detection limits are given along with results in Table 2. Trace elements were extracted from 5 g of ochre by digestion with 15 ml of nitric acid (70%) and 15 ml of perchloric acid (70%).

Natural waters were analyzed by ion-chromatography (IC, Dionex-300) using for anion analysis an AS4A–AMMS column with Na_2CO_3 (1.8 m/M) eluent at a flow rate of 2.5 ml/min, and for group I/II cation analyses CS 12 column with methane sulphonic acid eluent (20 mM) at a flow rate of 2 ml/min

Table 2. Partial geochemistry of ochres and sediment from Orangi river (OR) and Kilimafeza adit (AS) and tailings (TS) drainage.

Site	Fe g/kg	Al g/kg	Mn g/kg	Pb mg/kg	Cu mg/kg	Zn mg/kg	As mg/kg
TS 1d	205	113	1.9	711	146	248	422
TS 1 w	189	112	1.2	740	139	233	413
TS 2 d	469	98.1	4.2	395	159	255	659
TS 2 w	423	87.6	3.6	400	151	253	642
TS 3 d	310	236	8.5	210	364	265	365
TS 3 w	333	269	5.9	234	263	259	322
OR 5 d	153	116	5.3	86	174	165	58
OR 5 w	108	112	4.9	93	211	172	63
AS 1 d	255	125	3.6	580	321	208	198
AS 1 w	260	127	3.8	650	386	265	229
AS 2 d	283	139	5.1	310	336	198	552
AS 2 w	323	162	5.3	310	339	220	600
AS 3 d	203	160	4.2	630	481	143	390
AS 3 w	209	190	4.9	660	560	165	456
OR 3 d	92.5	55.5	2.8	138	115	142	39
OR 3 w	96.2	67.5	3.8	142	126	167	57

d = dry season
w = wet season

and detection by a pulsed electrochemical detector in conductivity mode. Transition metal analysis was accomplished with a Dionex CS 5 column with pyridine-di-carboxylic acid eluent and 4(2-pyridylazo) resorcinol post column derivitisation and measurement by a variable wavelength detector in the range 520–530 nm. The natural waters were also analyzed by ICPAES. Detection limits are shown along with the results in Table 3.

In order to assess the level of metal impact on forage in the Banagi area samples of acacia (*Acacia xanthophloea*), toothbrush sage (*Salvadora persica*) and sisal (*Sansevieria robusta*) were collected and analyzed for metal content. The bush and tree samples were split into bark, leaves, and roots prior to ashing at 150 °C for 4 hours. Metals were concentrated by nitric acid-perchloric acid digest of the ash and analyzed by IC. The results are given in Table 4.

Toxicity due to heavy metals in the water samples was assessed using the protozoan-based bioassay developed by Forge et al. (1993). In this technique, toxicity is measured as a function of growth inhibition of the ciliated protozoan

Table 3. Water geochemistry of the Orangi river and Kilimafeza mine drainage waters.

Orangi river Sample		pH	alkal	F	Cl	SO_4	Na	K	Ca	Mg	FC
Or 1	dry	7.3	270	7.9	14.9	11.6	121	3.9	28.6	23.6	>250
	wet	7.8	260	6.4	36.2	9.8	138	6.2	30.3	26.2	
Or 2	dry	7.5	266	6.1	19.0	12.2	124	4.9	27.6	24.6	>250
	wet	7.9	253	4.3	30.8	11.3	142	5.9	30.3	27.2	
Or 3	dry	6.3	160	3.2	20.6	271	108	3.1	26.3	21.9	180
	wet	6.9	160	2.9	29.8	240	108	3	28.5	23.2	
Or 4	dry	8.3	180	3.1	26.6	228	106	3.9	26.1	22.8	172
	wet	6.9	158	2.4	31.8	210	110	6.2	27.9	24.1	
Or 5	dry	6	110	2.5	28.4	385	88	3.2	21.3	16.3	68
	wet	6.6	105	2.4	32.6	257	76	6.4	22.9	18.6	
Or 6	dry	5.5	112	2.0	16.3	186	92	3.3	20.0	23.4	150
	wet	7.6	201	2.8	30.0	149	98	5.4	28.9	20.9	
Or 7	dry	6.8	149	3.6	16.3	76.8	112	4.7	26.4	26.2	>250
	wet	7.4	223	6.1	33.6	63.6	117	0.2	30.3	20.1	
Or 8	dry	7	271	6.0	17.6	30.3	118	8	38.6	27.6	>250
	wet	7.6	208	5.8	36.8	20.6	190	6	30.0	30.8	
Ts 1	dry	2.3	13	1	12.9	3280	38	2.1	6.5	4.3	0
	wet	4.1	19	1.1	35.0	2780	42	2.0	6.7	6.6	
Ts 2	dry	6.4	19	1.3	16.6	2150	45	3.3	2.0	4.9	22
	wet	5.9	20	1.4	33.7	1850	49	3.9	10.8	6.1	
Ts 3	dry	5.8	22	1.2	16.3	890	53	3.6	11.8	6.7	33
	wet	8.3	24	1.6	34.9	626	58	4.1	12.5	6.4	
As 1	dry	4.6	27	1.2	8.5	1840	61	3.9	7.9	6.3	28
	wet	6.1	29	1.2	34.2	1780	69	4.2	8.0	7.9	
As 2	dry	5.5	30	1.4	12.6	1320	73	4	11.2	10.8	41
	wet	6.1	20	1.6	36.1	1267	77	4.3	13.9	12.5	
As 3	dry	6.9	46	1.5	10.9	730	89	4.5	13.5	11.3	49
	wet	6.3	43	1.8	33.8	560	96	4.9	14.8	14.9	

Table 3. Continued.

			Al (tot)	Al (fil)	Fe (tot)	Fe (fil)	Mn (tot)	Mn (fil)	As (tot)	As (fil)	Cu (tot)	Cu (fil)	Zn (tot)	Zn (fil)	Pb (tot)
Or 1	dry		0.18	<0.1	0.21	<0.1	0.013	0.006	<0.6	<0.6	0.00	0.03	0.88	0.38	<0.5
	wet		0.11	<0.1	0.16	<0.1	0.009	0.006	<0.6	<0.6	0.00	0.02	0.76	0.33	<0.5
Or 2	dry		0.16	<0.1	0.18	<0.1	0.01	0.006	<0.5	<0.6	0.49	0.19	0.83	0.37	<0.5
	wet		0.11	<0.1	0.14	<0.1	0.005	<0.005	<0.5	<0.5	0.20	0.09	0.72	0.3	<0.5
Or 3	dry		0.21	<0.1	28.5	1.0	4.96	2.38	9.23	4.25	6.2	2.44	16.3	8.73	1.82
	wet		3.12	<0.1	26.3	1.3	3.96	1.89	3.98	2.52	2.9	1.32	14.8	7.89	1.25
Or 4	dry		11.5	<0.1	14.8	0.96	2.11	1.07	0.80	<0.5	4.9	2.41	16.2	8.23	1.19
	wet		10.2	<0.1	11.2	0.58	1.0	0.70	0.03	<0.5	3	1.48	13.8	6.89	1.06
Or 5	dry		23.4	1.0	58.3	6.21	6.86	3.88	13.78	6.47	18.9	9.77	21.9	11.28	5.78
	wet		16.3	0.5	52.0	4.21	4.83	2.79	12.81	4.90	9.8	4.79	16.4	8.54	4.92
Or 6	dry		10.3	<0.1	26.7	3.2	4.29	2.47	0.79	<0.5	8.3	2.61	16.2	7.41	1.59
	wet		7.2	<0.1	21.3	2.9	3.90	1.78	6.56	<0.5	2.1	0.98	12.0	6.25	0.98
Or 7	dry		5.3	<0.1	8.21	<0.1	1.10	0.58	0.03	<0.5	0.36	0.16	6.3	2.72	0.78
	wet		3.8	<0.1	0.95	<0.1	0.87	0.31	<0.5	<0.5	0.14	0.03	4.8	2.33	0.51
Or 8	dry		0.96	<0.1	0.89	<0.1	6.43	0.21	<0.5	<0.5	0.11	0.03	2.29	0.58	0.62
	wet		0.72	<0.1	0.39	<0.1	3.42	0.13	<0.5	<0.5	0.06	0.02	2.05	0.61	0.62
Ts 1	dry		176	47.1	622	176.3	6.58	5.73	324	286	125	110	126	98.8	21
	wet		143	36.8	699	185.6	6.26	6.49	262	221	100	96.2	117	89.7	10
Ts 2	dry		119	16.8	198	45.8	8.08	4.97	50	42	116.7	84.8	118	90.3	14.6
	wet		90	13.3	197	34.8	6.86	4.32	62	30.0	100.7	73.4	112	83.7	8.6
Ts 3	dry		36	26	112	20.2	6.1	4.68	43	23.8	41.7	21.3	99.5	73.6	10.2
	wet		27	1.5	93	13.0	5.25	3.00	36	12.3	20.3	13.1	96.7	84.1	6.9
As 1	dry		26.4	1.9	266	20.3	5.3	4.30	22.3	64.0	42	30.2	102	80.8	9.0
	wet		21.0	1.10	2.43	23.6	6.26	4.36	86.3	63.2	27	23.8	80.3	74.1	9.2
As 2	dry		22.3	1.42	102	9.5	4.75	3.03	71.2	44.6	19.0	14.0	96	77.9	7.6
	wet		18.7	0.83	80	7.8	4.58	3.65	64.6	38.6	8.3	4.93	79.8	63.8	6.8
As 3	dry	12.2	0.65	68	6.4	4.73	4.06	20.9	11.8	9.2	4.58	83	80.7	6.3	
	wet		8.6	0.17	62	2.8	4.63	3.75	15.2	8.4	3.7	1.73	76.9	65.2	4.5

All concentrations in mg/l alkal = alkalinity; F.C. = faecal coliform in countries per 100 ml;
fil = filtrate concentration; tot = total concentration

Table 4. Three element geochemistry of plant samples from Orangi river. All concentrations in mg/kg, collected in the dry season.

Species	Site	Na	Ca	Fe	Al	Mn	Cu	Zn	As
Acacia	OR 1	212000	18590	5900	155	233	9.1	42.3	< 0.5
	OR 5	188000	16320	6230	255	265	12.8	65.2	0.91
	OR 8	233000	17800	5810	158	181	10.2	45	<0.5
Sage	OR 1	18500	15960	6050	146	218	6.3	40.8	< 0.5
	OR 5	143200	13200	6230	221	248	11.9	47.1	0.75
	OR 7	179000	16980	6180	186	167	8.1	42.3	<0.5
	TS 2	142900	9600	7320	2550	258	85	165	15.5
	TS3	153000	11200	7220	1950	271	63	158	14.2
Sisal	OR 1	179000	14300	6970	236	259	11.7	68.7	<0.5
	OR 5	81000	8250	6580	275	286	14.8	78.9	1.26
	OR 7	145000	13600	6380	218	219	12.5	65.2	<0.5
	OR 8	189500	15600	6210	187	196	11.3	68.2	<0.5
	TS 2	66500	4020	8920	5500	365	112	196	26.3
	TS 3	71100	5650	8630	5320	271	98	183	18.9

Colpoda steinii. To each well of a 24-well polystyrene tissue culture plate was added 0.025 ml of a suspension of *C. steinii* containing ca. 100 cells in mineral water (Volvic), 0.025 ml of a dense suspension of the bacterium *Pseudomonas fluorescens* as food for the *C. steinii*, and 0.45 ml of water sample. Four replicates were prepared for each sample. The plates were incubated at 25 °C for 24 hours and the cells were fixed by adding 0.05 ml Lugol's iodine. The number of cells in each well was estimated by counting the number of cells in a 0.1 ml subsample in a Sedgwick-Rafter counting chamber. For each sample the mean number of *C. steinii* cells per well was expressed as a fraction of the mean number of cells per well for controls grown in mineral water. Fisher's least significant differences of means with $P < 0.05$ were calculated.

Results and discussion

Geochemistry of unpolluted Orangi river

The unpolluted surface waters are an alkaline Na-Ca-bicarbonate brine with high F (between 3–8 mg/l) and are typical of Serengeti waters (MAJI 1972, 1988). The high alkalinity and pH of the waters will influence the rapid precipitation of metals and partition into sediment minerals. Subsequently this would limit the impact of metal-rich waters on the river system. Total dissolved oxygen content is high in the uncontaminated waters ranging from 20–23 vol %. Fish are abundant and aquatic faunas such as crocodiles and hippopotamuses are present in the river leading to eutrophic conditions with high nitrate and phosphate. The mineralogy of the sediments is very similar at both control sites and is dominated by quartz and other clastic minerals with minor amounts of clay minerals. Trace minerals such as ilmenite and rutile are most likely derived from the greenstone while garnet, kyanite, and apatite are probably transported from the breakdown of the Archaean basement rocks and intrusive granites. The impact of mine released waters is noticeable in the drop in pH and rise in metals at sites OR 3 and OR 7 (Fig. 3).

Geochemistry of adit-drainage stream

Drainage from the mouth of a partially blocked adit at Kilimafeza has led to the release of acidic waters with elevated levels of heavy metals. The rate of discharge from the adit remains constant throughout the year around 0.008 m^3/s and at the adit mouth is non-turbid. It flows approximately 100 m before impacting the Orangi river. Amorphous orange precipitates and abundant dendritic macroscopic colonies of Fe-oxidizing bacteria occur along the tributary. At the catchment point the build-up of precipitates, dominated by iron oxyhydroxides, jarosite, kaolinite, quartz, and smectite, greatly increases turbidity. The sediments are enriched in arsenic (700 mg/kg), copper (560 mg/kg), lead (310 mg/kg), and zinc (220 mg/kg) compared to other sites (Table 2) suggesting that sediment minerals are effective at scavenging metals from adit drainage waters. As pH increases along the tributary feeding into the Orangi the aqueous concentrations of all heavy metals identified (Cu, Pb, Zn, As, Mn, Fe, and Al) drop rapidly. In the very acidic waters, metals are present as dissolved species except Fe, Pb, and Al which show a strong partition into the particulate fraction (total concentration/filtered concentration) which increases as pH increases (Table 3).

Geochemistry of tailings drainage channel

The downstream source of polluted water is at the base of a tailings pile. Geochemical analysis of mine waste (TS 1) revealed very high levels of As, Pb, and Zn (Table 2). The surface flow of water varied between wet and dry seasons from 0.017 m^3/s in the dry season up to 0.55 m^3/s during a storm in the wet season. The influence of sub-surface drainage from tailings could not be assessed, although there is good evidence that it exists (see below). Therefore these results probably represent an underestimate of the heavy metal impact on

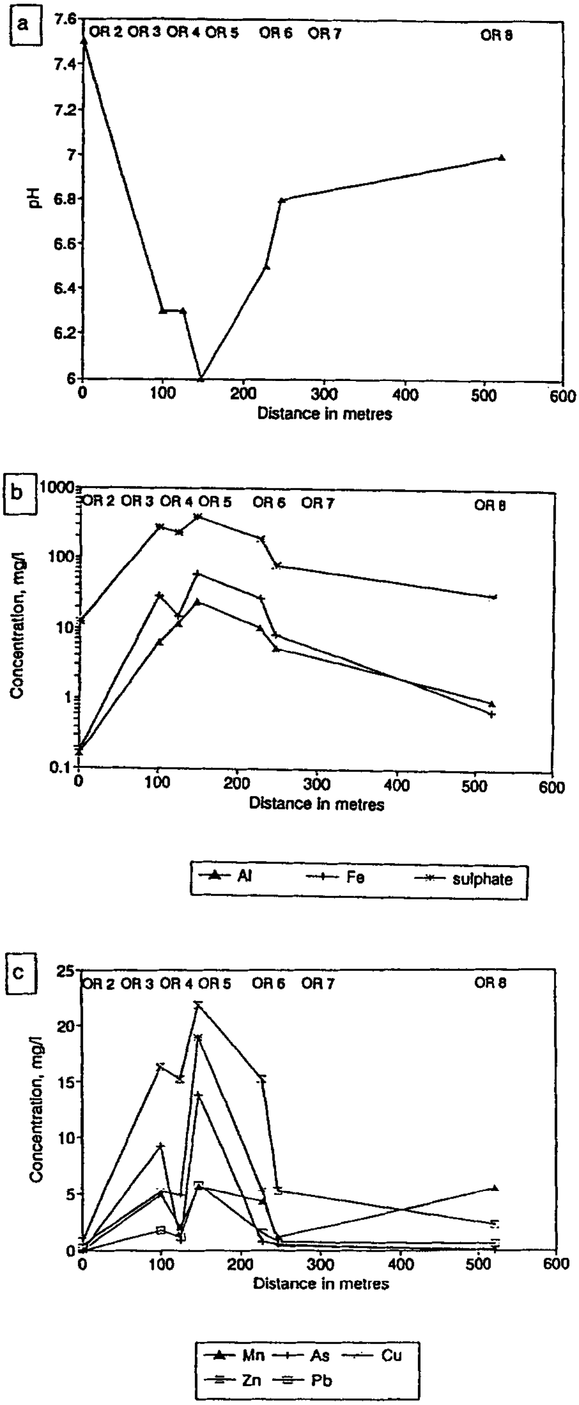

Fig. 3. Orangi river dry season hydrogeochemistry (total element concentration) a: pH b: Al, Fe, SO_4 c: Mn, As, Cu, Zn, Pb (sample sites are shown and refer to locations in Fig. 2).

the aquatic environment. A greater seasonal response is shown for the tailings drainage than for adit drainage waters with much higher metal concentrations (particulate and filtered) in the dry season samples. Heavy metal levels are in a similar range to those reported for other areas of acid mine drainage (Chapman et al. 1983; Davis et al. 1987; Blowes & Jambor 1990; Blowes et al. 1992; Fuge et al. 1993). The water pH is extremely acidic, reaching a low value of 2.3 in the dry season with water chemistry essentially Na-Mg-Fe-sulphate. Apart from Fe, the other major heavy metals are Al, As, Cu, Mn, and Zn. Despite high levels of Pb in the sediment, aqueous concentrations were much lower being below the detection limit. For Fe and Al the concentration in the particulate fraction was much greater in all samples than for the dissolved fraction (Table 3). Field measurements showed that drainage waters were high in dissolved oxygen, suggesting that oxidation and low pH are responsible for the release of tailings contaminants. In contrast to the adit drainage, tailings drainage waters are oxic, more acidic and consequently mobilize a significantly higher concentration of metals. This is reflected by the greater impact on the Orangi river of these drainage waters at their point of confluence with the river than that of the adit drainage (Fig. 3). The precipitation of metals probably involves oxidation of ferrous Fe as well as neutralization and could be microbially assisted (see below). Metal precipitation from solution could also be assisted by the presence of humic acids in the sediments and dispersed in the water column. Equally these acids could also assist in mobilization of the metals as dissolved species or particulate matter (Schnitzer 1978; Tipping & Cooke 1982; Ephraim & Marinsky 1986).

Seasonal variations in heavy metal dispersion

By comparing data from the two field seasons a pattern of metal behaviour is observed with a sequence of chemical reactions occurring downstream (Figs. 4–7). This is most vividly observed during the dry season when the tributaries feeding the Orangi river from both pollutant sources consists of a series of pools connected by minor surface flow. This phenomenon has been observed in similar studies in arid environments (Chapman et al. 1983; Rampe & Runnells 1989). A light brown transparent drainage water feeds from the adit mouth into large pool. The water flowing into this pool is clear and very acidic (ASd 1, Table 3). Downstream geochemical reactions cause successive pools to discolour. In the large pool the clastic sediment has an orange-coloured Fe-ochre coating which is enriched in As (ASd 2, Table 2). Downstream the pools of water become blue-green, less turbid and have a flocculent grey precipitate. Analysis by XRD suggested that the precipitates were largely amorphous with traces of smectite but are chemically rich in Al and Cu with lower levels of As and Fe (Table 2). Where the tributary impacts the Orangi river (100 m from the adit pile) the precipitated ochres are mainly goethite with traces of smectite, jarosite, quartz, and hematite. The waters at this point of confluence are acidic, although not as acidic as at the source of pollution, and contain elevated levels of Fe, Al, As, Zn, and Mn in the sediments (ORd 3, Table 2) and in the water (ORd 3, Table 3). These changes in the mineralogy and chemistry of ochre precipitates and water chemistry downstream are the result of acidic, reducing, metal-rich waters mixing with alkaline, oxidizing waters leading to neutralization. This is a natural equivalent of liming (Fuge et al. 1994; Sperring 1994) as waters in the area are essentially Na-Ca-bicarbonate composition (Table 3). As pH increases from the adit (pH 4.6) along the channel, sediments are enriched in Fe and As (Fig. 5). Sediment concentrations of Al and Cu also increase as pH increases towards neutrality followed by Mn and Zn (Fig. 5). The impact was observed in the field by the rise in stream turbidity, from <5 JTU to >1200 JTU, and by the drop in dissolved oxygen gas levels, from 23 vol % to less than 18 vol % between sites AS 1 and AS 2 (Fig. 8). These observations would be consistent with a mixing model of ferrous Fe oxidation and neutralization of acidic waters by the alkaline stream waters (Nordstrom 1982a; Chapman et al. 1983; Rampe & Runnells 1989; Davies et al. 1991; Fuge et al. 1994).

As with the adit drainage waters, drainage from the tailings pile to the Orangi also precipitate a series of ochres in the shallow drainage streams with a drop in metal concentration from source to the point of confluence with the Orangi river (site OR 5, Fig. 6). These ochres are largely ferric oxyhydroxides with minor Al-minerals and deposit a higher metal concentration in the sediments (Table 2). A similar trend of metal precipitation, to that observed from the adit drainage can be observed in the tailings stream sediments (Fig. 7).

The pH increases in the pools from 2.3 at the base of the tailings to 5.4 in pool TS 2 where ferrihydrite is precipitated, to 5.8 in pool TS 3 where Al and Cu are deposited in the smectite-amorphous ochre precipitate. The rise in pH (from 2.3 to 6) is reflected by a drop in water Fe concentration (Table 3) such that aqueous Fe concentration drops from over 600 mg/l total Fe at the source to less than 60 mg/l at the impact point (Fig. 6). The increase in precipitated Fe in the sediments shows a close correlation to precipitation of As and the drop in Fe and As concentrations in water over the same interval (Figs. 6–7). Deposition of Fe also correlates in part to a drop in sulphate concentration (Fig. 6) which would be expected with the precipitation of jarosite and also possibly Fe oxyhydroxide-sulphates. This latter amorphous phase has been postulated as occurring in iron ochres as the products of Fe-oxidizing bacteria (Bigham et al. 1990). Arsenic is largely present in oxic natural waters as arsenate (Haswell et al. 1985; Bowell et al. 1994). The

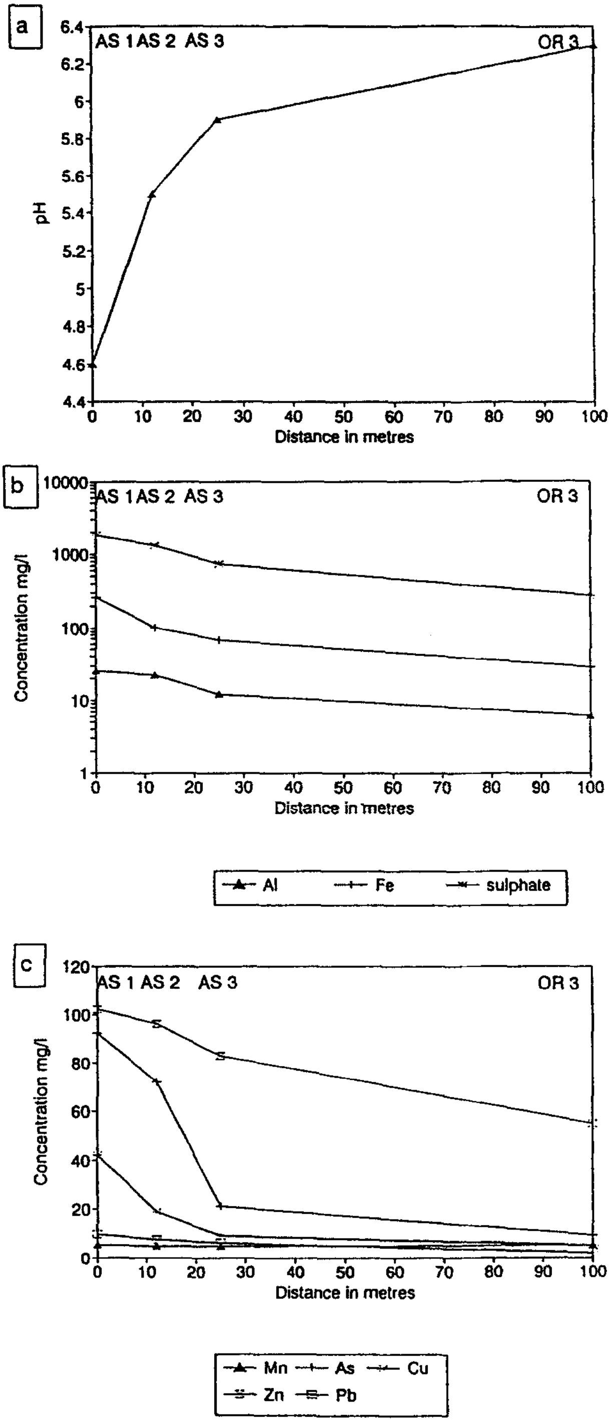

Fig. 4. Adit stream dry season hydrogeochemistry (total element concentration) a: pH b: Al, Fe, SO_4 c: Mn, As, Cu, Zn, Pb (sample sites are shown and refer to locations in Fig. 2).

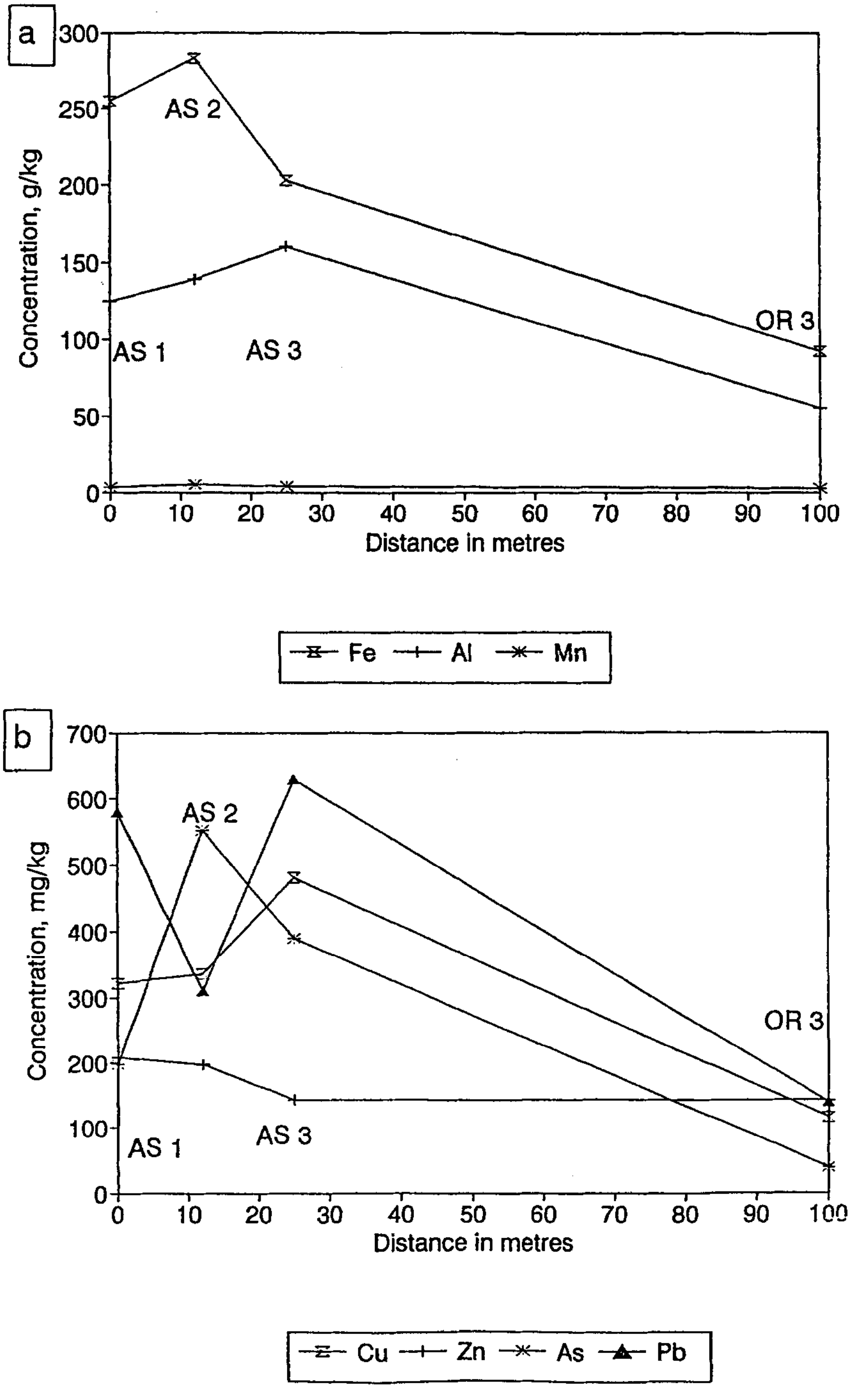

Fig. 5. Adit stream sediment geochemistry (total element concentration) a: Al, Fe, Mn b: As, Cu, Zn, Pb (sample sites are shown and refer to locations in Fig. 2).

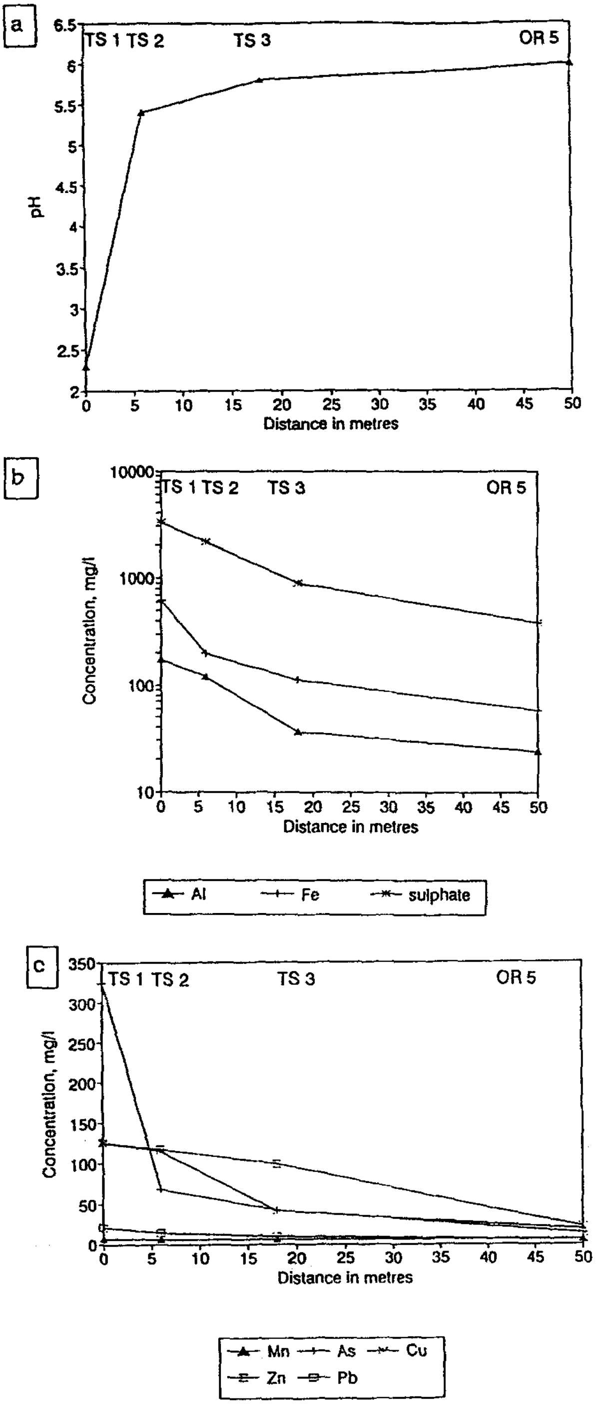

Fig. 6. Tailings stream dry season hydrogeochemistry (total element concentration) a: pH b: Al, Fe, SO_4 c: Mn, As, Cu, Zn, Pb (sample sites are shown and refer to locations in Fig. 2).

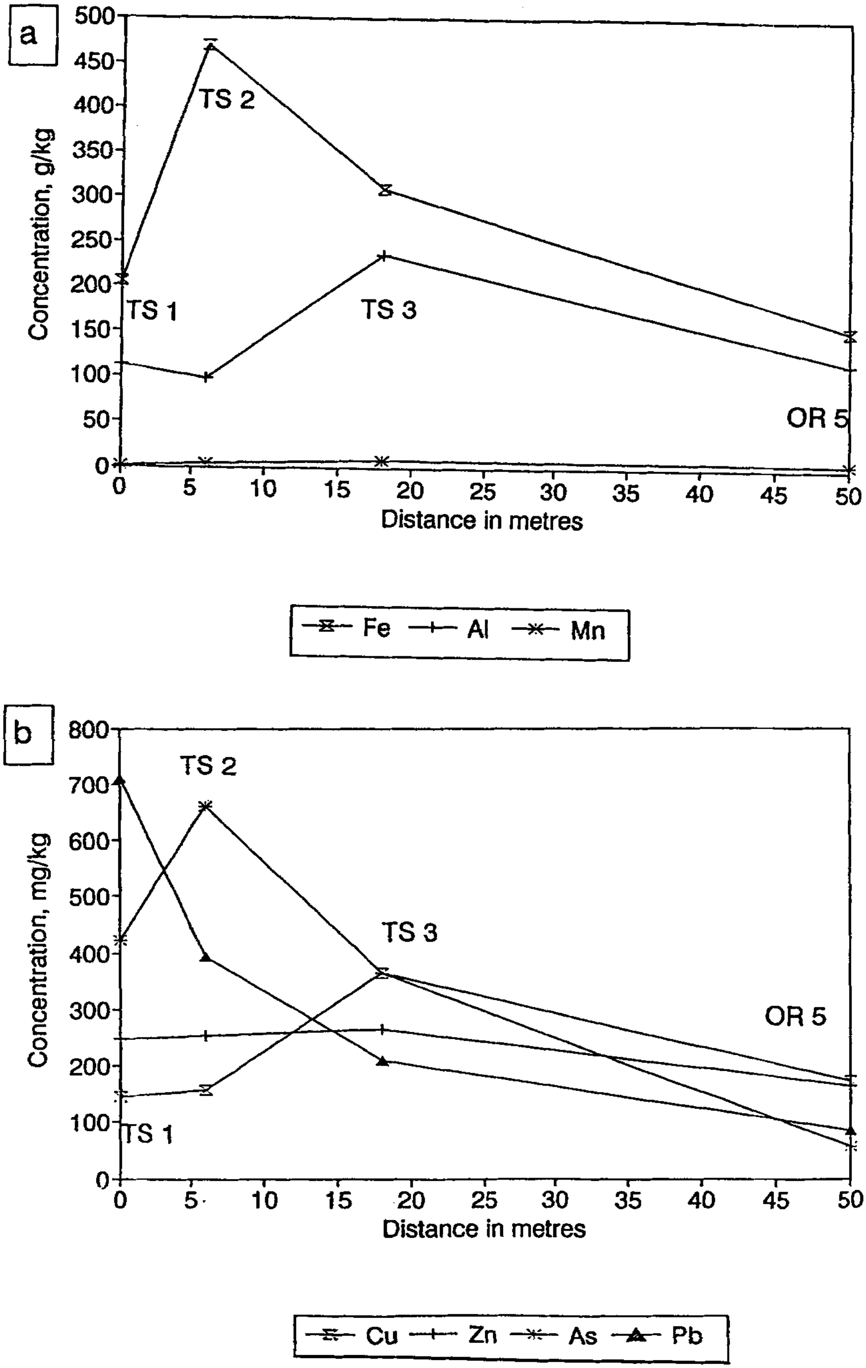

Fig. 7. Tailings stream sediment geochemistry (total element concentration) a: Al, Fe, Mn b: As, Cu, Zn, Pb (sample sites are shown and refer to locations in Fig. 2).

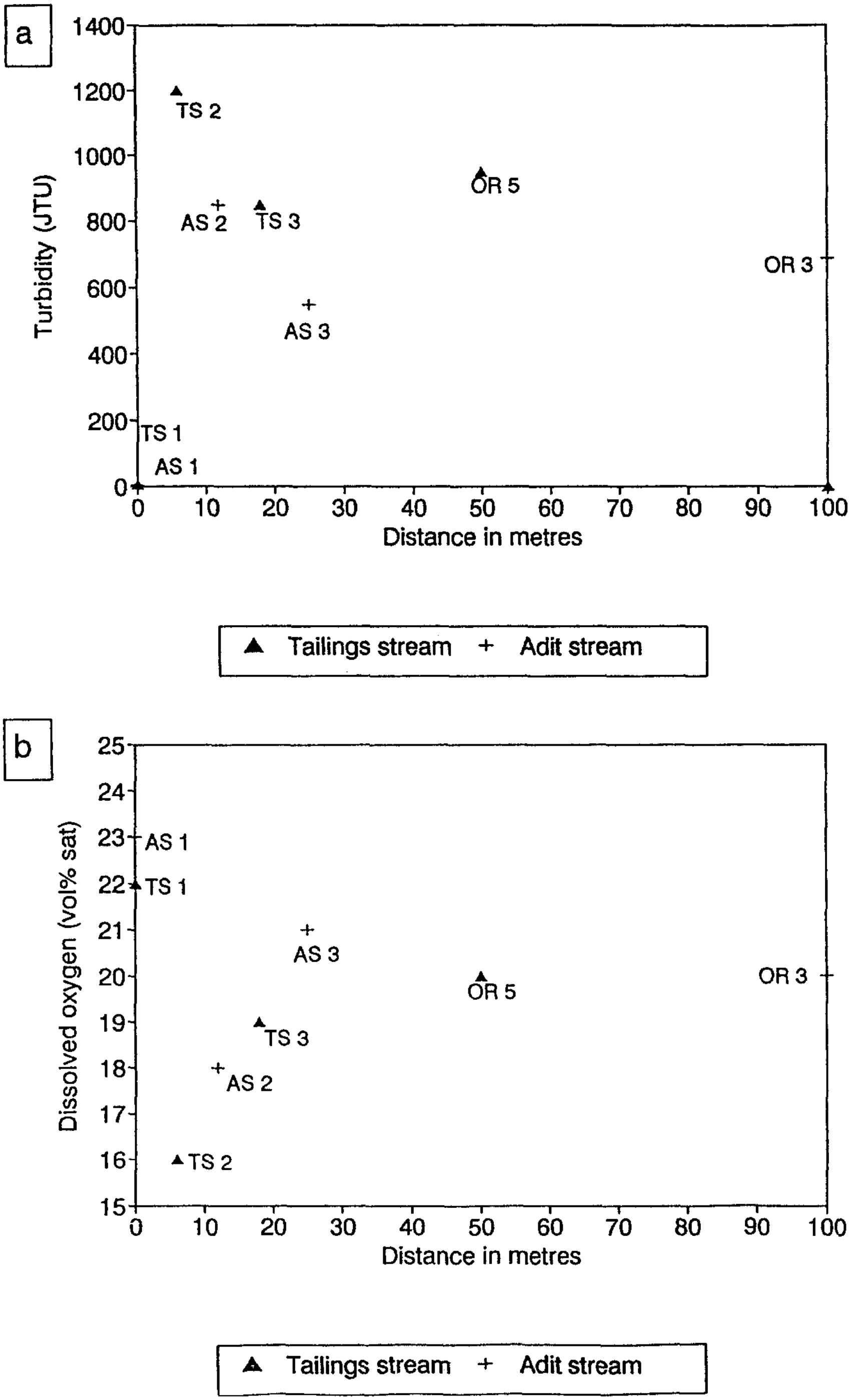

Fig. 8. Turbidity (a) and dissolved oxygen (b) in mine drainage streams (dry season) (sample sites are shown and refer to locations in Fig. 2).

presence of high As (up to 124 mg/l, TS 1) in the waters is of obvious concern as to its impact on the Banagi ecosystem. Since the only identified arsenate mineral, scorodite, appears restricted to the tailings pile it is likely that elsewhere the precipitated As is sorbed to the precipitated Fe-ochres which have a high affinity for arsenate ions at low pH (Sigg & Stumm 1980; Xu et al. 1991; Waychunas et al. 1993; Bowell 1994). This affinity increases closer to the isoelectric point, which for Fe oxides and hydroxides is between pH 5 to 8 (Parfitt 1980).

Aluminium, copper, and to a lesser degree zinc, show a similar trend downstream with their removal from drainage at a higher pH than Fe and As (Figs. 4, 6). Significant concentrations of all three metals, however, are still discharged into the Orangi river (23.4 mg/l Al, 18.9 mg/l Cu, and 21.9 mg/l Zn at OR 5). Prior to the point of confluence sediment concentrations (at site OR 5, Fig. 7) of all three elements are high (236000 mg/kg Al, 174 mg/kg Cu, and 165 mg/kg Zn). From bulk mineralogy of the sediments at this site a number of Al minerals are present in the ochres, boehmite, gibbsite, jurbanite, as well as smectite. It is worth noting that sulphate in the sediments at sites OR 3 and 5 are enriched in sulphate as well and that the precipitation of Fe and Al sulphate phases could be a major control on limiting Fe and Al solubility in the waters. Theoretical modelling on Al has highlighted the importance of jurbanite in controlling Al solubility in acidic waters (Nordstrom 1982b). No secondary Cu or Zn minerals were recorded and it is likely that Cu and Zn are absorbed by smectite (Slavek & Pickering 1981; Johnson 1986).

The highest level of Pb in the tributary is at the tailings seep (TS 1, 740 mg/kg) but decreases rapidly away from this site. Although high Pb values were recorded along most of the tailings channel, no Pb was recorded in the water samples. Consequently it is assumed that most of the transported Pb has been mechanically dispersed, possibly during flooding or other such high-energy events. Certainly Pb values in all the sediments in the wet season are higher than in the dry season in contrast to the other metals (Table 2). Manganese, and to a lesser extent Zn, are more mobile than other elements with elevated levels up to 100 m downstream in the Orangi (OR 7), where levels of all other trace metals are below 1 mg/l (Fig. 3).

The impact of the two point sources of contamination is much more noticeable in the dry season. During the wet season constant flushing of the area by rainwater and elevated river levels dilutes the mine drainage leading to a relatively constant high pH for all waters (4–6.5 for mine drainage waters and 7–8 for the Orangi river). Consequently sulphate and metal content in the water is lower and pH higher (Table 3). A major influence on this will be the contribution to the bulk volume of water in the tributaries. During the dry season with low precipitation only drainage from the mine site will feed the tributaries, whilst a dilution effect will be experienced during the wet season. This highly seasonal pattern is also reflected in the sediment geochemistry with higher metal concentration during the dry season, possibly in response to metal saturation in the concentrated drainage and precipitation of salts as pH rises along the drainage streams. The lower concentrations in the wet season could be due to mechanical erosion of stream sediments with the 'stripping' of dry season ochres from the sediments. Another factor will be 'dilution' of the sediments by the deposition of new clastic material (Fuge et al. 1993). As noted above the exception to this is Pb which has a higher concentration in the sediments during the wet season (Table 2).

Heavy metal speciation

Water samples were analyzed for total (i.e. non-filtered) and dissolved (filtered at 0.45 mm) metals with metals in the particulate fraction estimated as:

total metal concentration – metal concentration in filtrate

At the source sites tailing and adit drainage waters are highly acidic (pH < 3). Metals at such a low pH are predominately in the dissolved state (Table 4). The exception to this are Fe, Al, and Pb which show an important particulate fraction at all pH values (Table 5). In the wet season data (Table 5), pH is higher for all samples and where pH is above 5 for both wet season and dry season data sets, the metal concentration in the particulate fraction increases.

Biogeochemistry of Orangi river

In order to assess the level of metal impact on forage in the Banagi area samples of acacia (*Acacia xanthophloea*), toothbrush sage (*Salvadora persica*) and sisal (*Sansevieria robusta*) were collected and analyzed for metal content. Control samples were collected from close to Serona valley to determine metal accumulation away from the mine site. Higher levels of heavy metals are present in the vegetation in the dry season than in the wet season with sisal being the most effective accumulator of heavy metals (Table 4). In the wet season only the sisal and sage contained elevated metal content and then only in close proximity of the mine (Table 4). In general acacia showed elevated values of Fe, Al, Mn, and Zn compared to the control site vegetation with elevated As and Cu only at the Kilimafeza mine. If preferential uptake occurs of these metals, then essential nutrients like Ca, K, and Mg may be inhibited (Phipps 1981; Thurman 1981). From the analysis of the vegetation a possible correlation between high Fe, Mn, and Zn and low Ca and Na can be observed. However, this could merely be a pH effect with the alkalis being less abundant in the strongly acidic mine waters than in the alkaline river water.

Table 5. Proportion of total metal concentration in <0.45 μm filtrate. All values expressed as a % of total metal concentration (for Pb < 1% of total concentration was present in the filtrate for all sites).

Sample	pH	Fe	Al	Mn	As	Cu	Zn
OR 1 d	7.3	na	na	62.0	na	33.0	44.2
OR 1 w	7.8	na	na	na	na	33	43.4
OR 2 d	7.5	na	na	60	na	37.5	44.5
OR 2 w	7.9	na	na	na	na	34.6	41.7
OR 3 d	6.3	6.7	na	48.0	46.1	46.9	53.6
OR 3 w	6.9	4.9	na	48.0	29.2	45.6	53.5
OR 4 d	6.3	6.5	na	50.5	na	49.3	54.1
OR 4 w	6.9	5.2	na	47.6	na	51.7	49.9
OR 5 d	6.0	10.6	6.8	68.7	39.7	48.9	53.7
OR 5 w	6.3	8.0	3.5	57.8	38.5	47.2	52.1
OR 6 d	6.5	11.9	na	57.8	na	46.7	48.8
OR 6 w	7.1	13.2	na	45.8	na	44.4	48.4
OR 7 d	6.8	na	na	50.0	na	21.4	51.3
OR 7 w	7.4	na	na	46.4	na	27.3	48.5
OR 8 d	7.0	na	na	38.7	na	27.3	29.8
OR 8 w	7.5	na	na	37.7	na	na	29.8
TS 1 d	2.3	28.3	15.3	87.0	81.8	88.2	78.4
TS 1 w	4.1	27.2	15.0	87.0	78.4	88.2	76.7
TS 2 d	5.4	23.6	7.8	81.7	60.9	73.3	76.5
TS 2 w	5.8	18.6	7.2	73.7	58.8	72.9	74.7
TS 3 d	5.8	18.3	7.2	76.7	55.3	51.2	73.9
TS 3 w	6.3	14.6	5.6	76.0	53.6	49.8	73.9
AS 1 d	4.6	11.4	7.1	82.8	75.4	86.2	83.7
AS 1 w	5.1	9.7	5.4	82.8	73.2	85.9	83.0
AS 2 d	5.5	9.3	6.4	82.4	62.0	78.4	81.0
AS 2 w	6.1	8.4	4.4	82.5	59.8	59.3	79.9
AS 3 d	5.9	7.9	5.3	79.5	56.5	50.7	73.1
AS 3 w	6.3	4.5	2.0	77.9	55.2	46.8	73.1

na = not applicable as filtrate concentration was below detection limit

Faecal coliforms were isolated from all eight sites along the Orangi river (Table 3). The highest numbers were recorded at sites unaffected by drainage from the mine (OR1, OR2) and at sites well downstream (OR7, OR8). Concentrations at these sites were >250/100 ml, or too numerous to count (TNTC) with the enumeration technique employed. This is indicative of gross pollution and would present a high risk if used as a source of untreated drinking water. Somewhat lower concentrations (58–172/100 ml) were recorded at sites closer to the mine site, i.e. OR3, OR4, OR5 and OR6 (Fig. 9).

Faecal coliforms were also isolated along the adit-drainage stream and the tailings drainage channel, although concentrations at these sites were all <50/100 ml (Table 3). Two possible reasons for this reduction in concentration of faecal coliforms are (i) that the stream and channel are less polluted than the river or (ii) that the physicochemical conditions in the stream and channel, such as the high concentrations of heavy metals and low pH, adversely affect the survival of faecal coliforms. Previous studies have shown that there is a rapid decline in the survival of the faecal coliform bacterium *Escherichia coli* in natural waters below pH 5.5 (McFeters & Stuart 1972). Similar trends have also been reported in soils and sediments (Reddy et al. 1981; Gold et al. 1992). Certainly, the complete absence of faecal coliforms at site TS1 (pH 2.3) can be explained by acidity alone since complete die-off of these organisms occurs below pH 3.0 (McFeters & Stuart 1972). The high concentrations of heavy metals in the adit-stream and tailings drainage channel are also likely to be sufficiently high to affect the survival of faecal coliforms although these effects are less well understood. There appeared to be a strong relationship between concentrations of faecal coliforms, heavy metals and pH within the system (Fig. 10). It should be noted, however, that different microorganisms may have vastly different tolerances to extremes of pH and heavy metal concentrations. Some are adapted to conditions such as those found in the Orangi river-Kilimafeza mine system and may even play an important role in heavy metal mobilization in the water (Warren et al. 1993). Furthermore, some waterborne pathogens may be less sensitive than faecal coliforms to low pH-high heavy metal conditions. Therefore, the absence of faecal coliforms in such waters should not be interpreted as being indicative of microbiological safety.

The results of the protozoan-based bioassay revealed that growth of *C. steinii* was inhibited by samples taken from all six locations tested (OR1, OR7, TS1, TS3, AS1 and AS3). In each case the concentration of heavy metals in the water was thought to be the cause. In order to be biologically available for uptake by microorganisms, it is likely that metals have to be in solution (Alloway & Jackson 1991). Therefore, the results of the bioassay are interpreted in the light of concentrations of dissolved heavy metals, i.e. in filtered water samples, rather than total metal concentrations.

The most dramatic effects were seen with samples TS1, TS3, AS1 and AS3, all of which killed cells of *C. steinii* almost immediately on contact. In all four cases this effect was almost certainly due to the high concentrations of heavy metals such as Cu (up to 110 mg/l) and Zn (up to 98.8 mg/l) present in the filtered water samples (Table 3). These concentrations are around two

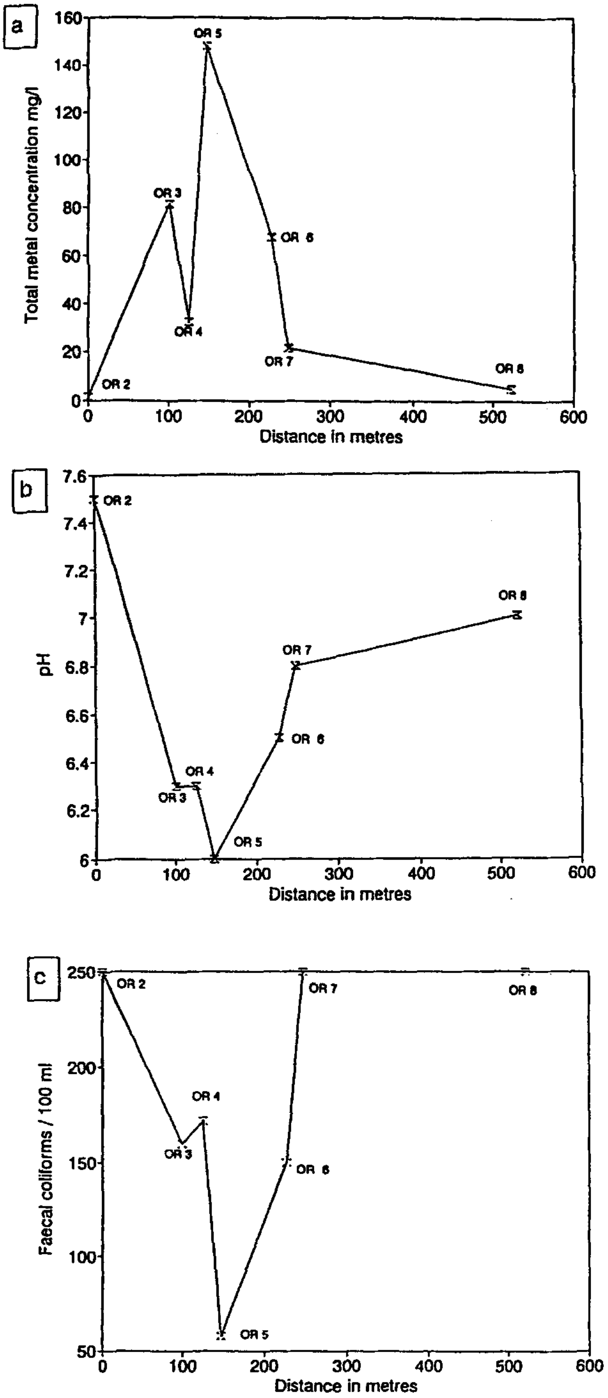

Fig. 9. Distribution of Total heavy metals (a), pH (b), and Faecal coliform counts (c), along the Orangi river (dry season) (sample sites are shown and refer to locations in Fig. 2).

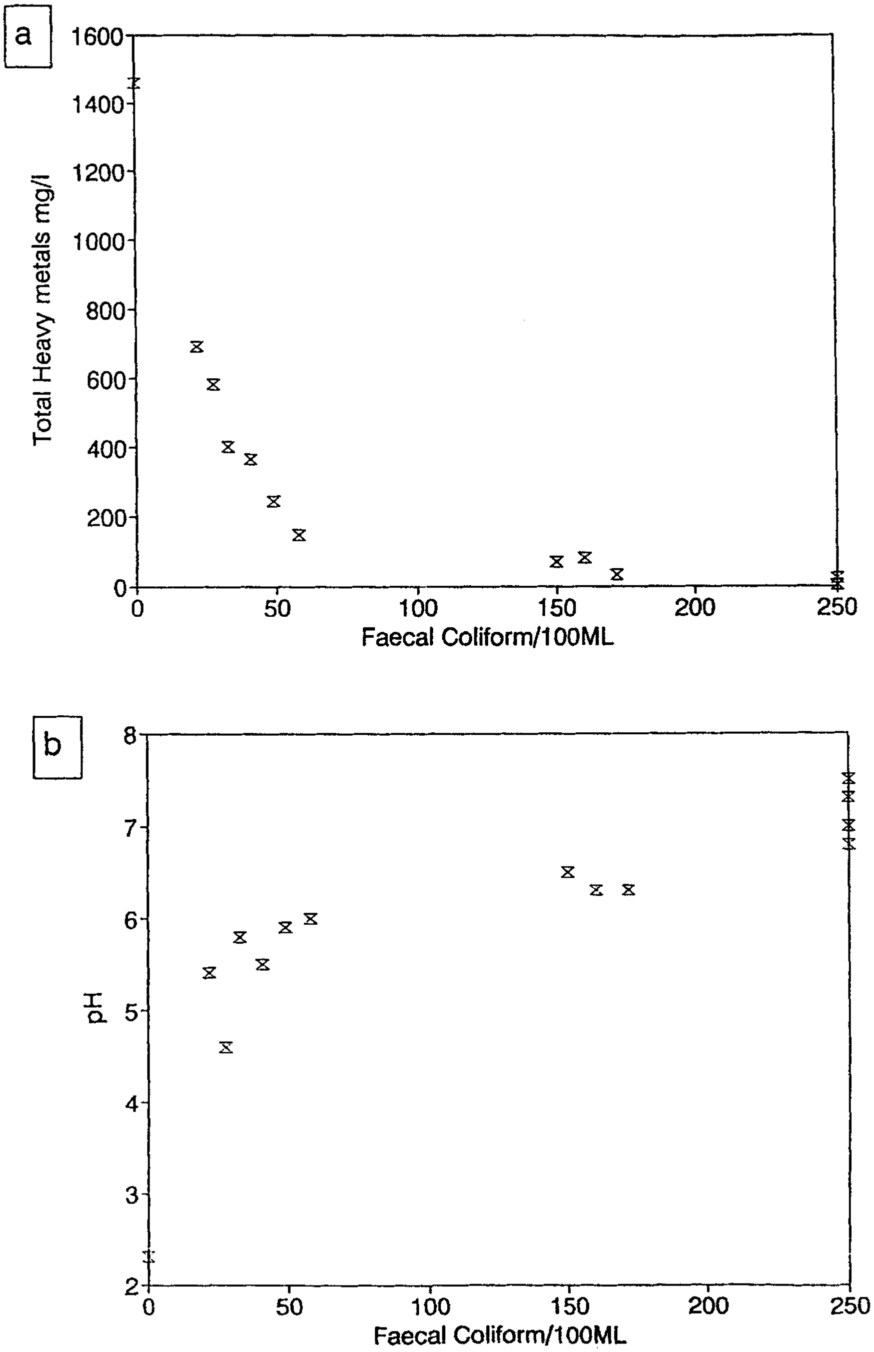

Fig. 10. Relationship between Faecal coliform counts and; a: total heavy metals; b: pH For all dry season water samples.

orders of magnitude higher than those reported by Forge et al. (1993) to reduce the growth of *C. steinii* by 50% (RG_{50}), i.e. Cu 0.25 mg/l, Zn 0.85 mg/l. In the case of TS1, however, death of *C. steinii* might also have been due to the low pH (2.3) since the lower pH tolerance of *C. steinii* is reported to be 4.0 (Bick 1972).

By contrast, *C. steinii* was able to grow over a 24-hour period in samples from both OR1 and OR7 although with reduced efficiency compared with the control. The greater effect was seen in OR7 where growth of *C. steinii* was only 44% of that in the control. This could be explained by the concentration of Zn alone (2.72 mg/l) which is greater than its RG_{50} for *C. steinii*. In sample OR1 the growth of *C. steinii* was 61% of that in the control reflecting the lower concentrations of heavy metals at this site (Table 3).

Conclusions

From the analysis of water chemistry along the Orangi river two point sources of metal contamination are evident from the Kilimafeza mine. The first impact is from adit-drainage waters which are characterized by low pH and high metal and sulphate content. The second point source occurs as tailings drainage waters from a small mound of decomposing sulphide-rich mine tailings which produce a more acidic, metal-rich effluent. The impact of the two sources varies seasonally with a more pronounced effect in the dry season.

During the wet season constant flushing of the area by rainwater and elevated river levels dilutes the mine drainage leading to a relatively constant high pH for all waters (6–6.5 for mine drainage waters and 7–8 for the Orangi river). Consequently sulphate and metal content in the water is low. However, as shown above, in the dry season water drainage from the tailings pile and from the adit dominate the water chemistry which impacts the Orangi river in the study area. Consequently, as flow is lower, residence time for water in the tailings pile is greater so pH is lower and metal and sulphate content higher. This highly seasonal pattern is also reflected in the sediment geochemistry with higher metal concentration during the dry season, possibly in response to metal saturation in the concentrated drainage and precipitation of salts as pH rises. The lower concentrations in the wet season could be due to mechanical erosion of stream sediments with the ‘stripping’ of dry season ochres from the sediments. Another factor is likely to be ‘dilution’ of the sediments by the deposition of new clastic material. As noted above the exception to this is Pb which has a higher concentration in the sediments during the wet season.

At the source sites tailing and adit drainage waters are highly acidic (pH < 3). Metals at such a low pH are predominately in the dissolved state. The exceptions to this are Fe, Al, and Pb which show an important particulate fraction at all pH values. In the wet season pH is higher for all samples and where pH is above 5 for both wet season and dry season data sets, the metal concentration in the particulate fraction increases (as shown in Table 5).

Faecal coliform bacteria appeared to be adversely affected by the low pH-high heavy metal conditions present in the acid mine drainage waters. These findings could have important implications for the monitoring of mine-impacted waters since other waterborne pathogens may be less sensitive than faecal coliforms to such conditions. The results of the protozoan based bioassay suggest that this may be used as a rapid, low-cost method for monitoring the biological availability of heavy metals in waters, especially when combined with chemical analyses.

Although it is difficult to quantify the impact of the mine drainage on the Banagi area, the spatially limited elevated metal content in sediments, water, and vegetation would suggest that any impact is restricted to a small area (less than 1 km^2). Since most animals which graze or browse in this area, do so for a short time, then the impact is negligible. The metals Pb and As, although present at toxic levels in the tailings, are strongly sorbed by sediment minerals and ochres so have a very limited availability. The only animals at risk would be those which reside in the immediate vicinity of the mine site and are subjected to long term exposure of elevated heavy metals in water and vegetation, particularly in the dry season. Among those animals most at risk are very mature bull elephants which forage over a small part of the Banagi area and have a high bulk requirement of vegetation and water. A long term monitoring study, involving analysis of animal blood as well as water and vegetation, is required in order to assess the lasting impact of the mine tailings on the Banagi area.

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