Short Communications

Aust. J. Soil Res., 1979, **17**, 355–60

Weathering Studies of Bougainville Mine Tailings in Lysimeters

Alan Hartley

Bougainville Copper Ltd, Panguna, North Solomons Province,
Papua New Guinea.

Abstract

Lysimeter studies indicated that under wet tropical conditions, there was significant sulfur depletion in the upper strata of low sulfide tailing deposits within three years of exposure.

The rate of acidification was influenced by drainage and aeration, and pH fell logarithmically with time.

Copper mobilized by acidification was redeposited within the profile where the pH remained in excess of 7.

Introduction

The weathering of tailings from the Bougainville Copper Ltd porphyry copper mine has been studied in 1 m diameter concrete lysimeters over a period of 4 years.

Run of mill, and hydrocycloned fines and coarse fractions (Table 1) were confined in separate exposed cylinders. Leachate was drawn fortnightly from three depths and solids were assayed after 37 and 45 months using cores removed *in situ.*

Panguna average rainfall is illustrated in Fig. 1. Monthly evaporation is between 80 and 100 mm. Temperatures range between 17°C and 29°C.

Results

Change in Leachate pH with Time

The pH fell logarithmically with time, the greatest changes taking place in the coarse sediments, and nearer the surface. The effect is illustrated in Fig. 2 for the coarse series, where levels D, C and B were respectively 109, 65 and 33 cm below the surface. Similar trends apply to the run of mill and fine series, though the degree of change was less. There was a strong correlation, as expected, between pH, base saturation and sum of exchangeable cations which was dominantly calcium.

Sulfate Concentration in Leachate

Sulfate was virtually the only anion appearing in leachate, with chloride assaying at less than 5% of the sulfate level. The sulfate content varied according to the rainfall in the interval preceding sampling, the dilution effect being greatest when heavy rain fell the night before. Over the time interval of 45 months there was a strong correlation between $[SO_4]^{2-}$ and $[Ca]^{2+}$, where base saturation exceeded 100%.

In the coarse sediments, for the C level leachate, the relationship was of the form:

$$\mathrm{SO_4\ (ppm)} = 2{\cdot}86\mathrm{Ca\ (ppm)} - 58{\cdot}86; \qquad r = 0{\cdot}99.$$

A regression series of Ca^{2+} against SO_4^{2-} indicated that Ca^{2+} was leached faster from coarse than from fine sediments, and from the surface faster than deeper strata.

Table 1. Particle size distribution in lysimeters
(mean of 8 cylinders %)

	Clay <0·02 mm	Silt 0·2–0·02 mm	Fine sand 2–0·2 mm	Coarse sand >2 mm
Coarse	1·0	8·4	36·5	54·1
Run of mill	2·7	22·7	45·5	29·1
Fines	10·0	45·5	44·1	0·4

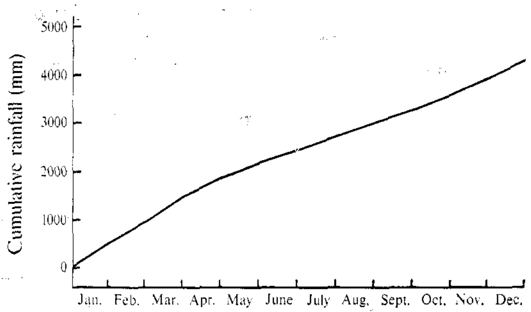

Fig. 1. Cumulative average annual rainfall—Panguna.

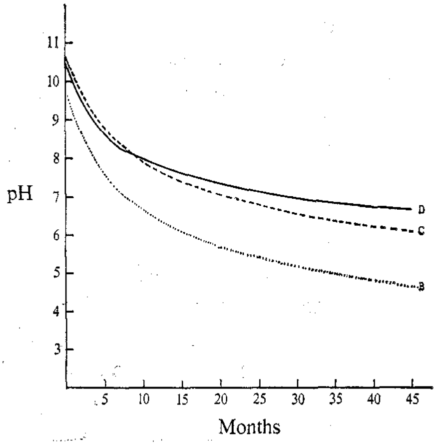

Fig. 2. Variation in pH with time at three depths:
B, 33 cm;
C, 65 cm;
D, 109 cm
in coarse sediment.
B: $\mathrm{pH} = 9{\cdot}76 - 1{\cdot}34 \ln(\mathrm{time})$; $r = -0{\cdot}87$.
C: $\mathrm{pH} = 10{\cdot}64 - 1{\cdot}19 \ln(\mathrm{time})$; $r = -0{\cdot}94$.
D: $\mathrm{pH} = 10{\cdot}34 - 0{\cdot}97 \ln(\mathrm{time})$; $r = -0{\cdot}93$.

When the pH fell below 7 and Cu^{2+} appeared in the leachate, the relationship recorded was:

$$\mathrm{SO_4\ (ppm)} = 6{\cdot}25\mathrm{Ca\ (ppm)} + 0{\cdot}94\mathrm{Cu\ (ppm)} - 10{\cdot}94; \qquad R^2 = 0{\cdot}99.$$

Copper and pH of Leachate

There was a strong negative exponential correlation between pH and Cu^{2+} in leachate at individual samplings. The same relationship did not hold over all samplings with time, since the actual copper concentrations were subject to dilution effects caused by rainfall. Copper was detected only in leachate when the pH fell below about 6·8. At the same sampling, more copper was present in leachate from the coarse series than from the run of the mill at similar pH levels. Typical copper concentrations are shown in Table 2.

The differences between the coarse and run of mill series were significant at the 1 % level.

An examination of total precipitation, wet and dry sequences and copper concentration revealed no consistent correlation.

Table 2. Copper concentrations (ppm) in leachate for coarse (C) and run of mill (R.O.M.) series

Months from start	pH: 4		5		6	
	C	R.O.M.	C	R.O.M.	C	R.O.M.
34·5	175·9	27·4	44·7	8·0	11·4	2·3
37·0	93·7	40·5	42·1	9·6	18·9	2·3
38·7	37·0	3·6	12·9	1·5	4·5	0·7
40·5	63·4	11·8	18·4	4·0	5·3	1·3
42·0	101·5	18·7	22·4	3·9	4·9	0·8
44·5	97·5	0·6	16·6	0·4	2·8	0·3

Potassium and Magnesium Levels in Leachate

The quantity of Mg^{2+} in leachate increased with time in the coarse series where there was strong acidification. The values ranged from about 3 ppm in the initial alkaline leachate to about 17 ppm in the B and C levels after 37 months.

Potassium concentrations have varied little with time or pH change, ranging generally between 20 and 40 ppm for all size fractions.

Silica in Leachate

At pH exceeding 7, silica levels were relatively constant in the range from 10 to 18 ppm. As acidity developed in the B and C levels of the coarse series, silica concentration in the leachate increased. The relationship between silica and pH below 7 was of the form:

$$SiO_2\ (\text{ppm}) = 152\cdot7 - 17\cdot84\text{pH}; \qquad r = -0\cdot96.$$

Aluminium, Iron and Zinc in Leachate

These three metals were not detected in leachate at pH levels above about 4·5.

Phosphate and Nitrogen in Leachate

Phosphate and nitrogen have been detected in leachate at very low levels. Phosphate is generally less than, or about, 0·1 ppm, while total nitrogen has been detected in the range 0·4–1·0 ppm. It is possible that algae growing in the collection tubes could be the source of detected nitrogen.

Sampling of Lysimeter Solids

In the coarse series a thin silty layer formed at the surface during initial filling, and it was consolidated by subsequent rainfall action. The rest was mainly sand sized, with a variation between grades of fine and coarse sand. Below about 50 cm (that is the C level) sand was saturated and thixotropic. In the run of mill series there was a stratification of fines (over 50% finer than 53 μm) around 20–30 cm (B level) beneath the surface.

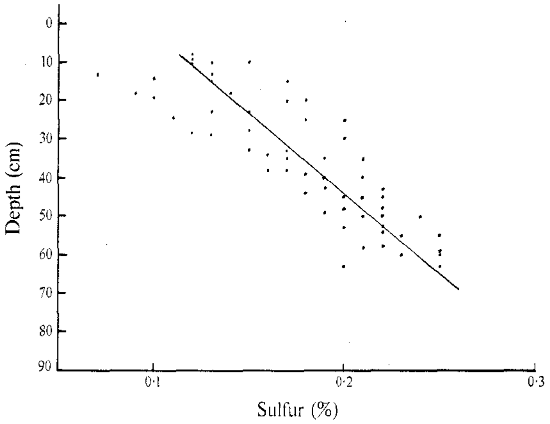

Fig. 3. Sulfur depletion with depth in coarse dediments (data from six lysimeters).

Sediment Colour

A colour gradient existed in all of the cylinders but most conspicuously in the coarse series. The original tailings colour was 10Y4/1 dark grey. This was preserved below the C level in the coarse series and below the B level in the run of mill series where the sediment was saturated at sampling. Nearer the surface the sediment was yellowish grey with a higher chroma in the upper layers:

0–20 cm 7·5Y5/2 yellowish grey
20–35 cm 7·5Y4/1 yellowish grey
35–55 cm 7·5Y5/1 yellowish grey
55 cm+ 10Y4/1 dark grey.

The colour variation is interpreted as an indicator of oxidizing conditions, there being little change apparent in the saturated lower strata.

Chemical Assays of Solids After 45 Months

Sediment pH fluctuated in the range 4·5–6·0 in the coarse series from the surface to about 55 cm depth. Below this there was a rapid increase to pH in excess of 8. This occurred in the zone where sediments became finer, where there was little indication of colour change, and where the sediment was water saturated.

Sulfur levels were strongly correlated with depth to the C level in the coarse series. The relationship is shown in Fig. 3 where the plotted curve is of the form:

$$S\% = 0{\cdot}1024 + 0{\cdot}0025 \text{ depth (cm)}; \qquad r = 0{\cdot}90.$$

Below the coarse C level, and throughout the whole column apart from the surface 10 cm of the run of mill series, sulfur levels varied between 0·20% and 0·29%, consistent with the initial assays.

Copper accumulated with depth in the interval between 5 cm and 60 cm with a relationship of the form:

$$\mathrm{Cu}\% = 0{\cdot}43+0{\cdot}0015 \text{ depth cm}; \qquad r = 0{\cdot}89.$$

Actual values ranged from 0·042–0·075% at 10 cm to 0·110–0·142% at 60 cm. The assays at 10 cm were within the range recorded for various particle sizes in the original tailings. There was evidence of copper contamination on the upper surface from wind-blown dust.

The total assays of calcium, magnesium, potassium, sodium and iron indicated no trends with depth, and lay within the range of values shown in Table 3.

Table 3. Assays of tailings

Values are percentages unless indicated otherwise

Te, Sb, In, Cd, As all <0·001

	Coarse	R.O.M.	Fine		Coarse	R.O.M.	Fine
Ba	0·038	0·047	0·047	Cr	0·016	0·018	0·021
Pb	0·009	0·009	0·008	TiO_2	0·52	0·42	0·40
Au	0·18 ppm	0·23 ppm	0·14 ppm	CaO	4·14	4·12	4·44
Ag	0·35 ppm	0·33 ppm	0·50 ppm	K_2O	1·85	1·70	1·85
Mo	0·002	0·002	0·002	Sr	0·045	0·05	0·055
Se	0·001	0·001	0·001	S	0·22	0·29	0·28
Zn	0·008	0·006	0·006	SiO_2	60·80	59·48	56·16
Cu	0·076	0·052	0·052	Al_2O_3	17·12	17·90	17·80
Ni	0·006	0·005	0·006	MgO	3·60	4·22	6·04
Co	0·005	0·004	0·005	Na_2O	4·70	4·48	4·60
Fe	4·57	4·71	5·29	V_2O_5	0·065	0·05	0·05
Mn	0·044	0·048	0·044				

Tailings as Soil

There was a low cation exchange capacity varying between 1 and 2 m.e./100 g. Values as high as 6 m.e./100 g in the surface 5 mm were measured where the moss *Brachymenuim exile* (Dox. et Mollc.) had colonized. The exchange complex was dominated by calcium, regardless of the degree of base saturation. The ratio of exchangeable calcium to exchangeable magnesium varied between 2 and 3 in unsaturated sediment, while the exchangeable potassium was less than 0·1 m.e./100 g. Exchangeable sodium was not generally detected. Where the base saturation exceeded 100%, exchangeable potassium lay in the range 0·1–0·27 m.e./100 g.

Organic enrichment under moss was restricted to the surface 5 mm. Carbon as high as 1·7% and total nitrogen 0·08% were recorded, but the subsurface levels were generally less than 0·04% carbon and less than 0·004% nitrogen. Phosphorus (bicarbonate extraction) was usually below the detection limit (<1 ppm), apart from some accumulation of up to 4 ppm at the surface.

Conductivity was low, ranging from 0·03–0·07 mS cm^{-2} in the coarse series, and 0·01–0·11 mS cm^{-2} in the run of mill sediments where there was a trend towards increasing conductivity with depth.

Discussion

The lysimeter studies indicated that the initially alkaline tailings (saturated with process lime) would undergo acid weathering given certain conditions. The system appeared dominated by the oxidation of sulfide and the removal of the sulfate anion and balancing cations from the weathering zone. This occurred in coarse tailings when there was natural wetting and drying with concomitant leaching and air-flushing. A significant sulfur gradient developed from the surface to the level of water-saturated and thixotropic sediment. Below this, and in run of the mill tailings where stratification with fines impeded water movement, there was no longer a sulfur gradient but only fluctuations within the original assay range (0·2–0·3% sulfur).

Within the profile, pH was influenced by physical factors affecting aeration. Impeded drainage caused by a high water table or by strata containing a high proportion of fines limited the removal of excess calcium in percolating water. The sulfate anion levels recorded from the base-saturated lower profiles probably originated mainly in the better aerated surface. Rapid oxidation has depleted the sulfide source and with it, the ultimate capacity for acid production. Slow oxidation has preserved the sulfide source, but the exchangeable cation store is also preserved especially where impeded water movement is the main cause.

Copper appeared in leachate when the pH fell below about 6·8. Regular assays of different size fractions of tailings have indicated that the residual copper content of the coarser fractions is higher than that of the finer. It is not clear from the available evidence whether this contributes to the higher release of copper at the same pH from the coarse as compared with the run of mill profiles. A bacterial examination of one coarse profile indicated that acidophilic sulfur oxidizing bacteria were present (counts up to 4×10^6 colonies/g from thiosulphate media at pH 4) in the upper 50 cm. Since oxygen supply is a factor limiting the effectiveness of bacterial acid leaching, the higher yield of copper from the coarse profile could also reflect more efficient bacterial action again resulting from better aeration. Vertical movement of copper in the profile has resulted in accumulation with depth in the sediment.

The results offer encouragement for the reclamation of tailings where acidification in the root zone could be limited by sulfide depletion. Alternatively, secondary enrichment and trapping of water-soluble copper in base saturated lower strata could provide an opportunity for subsequent metal recovery from suitable deposits.

Acknowledgment

The permission of Bougainville Copper Ltd to publish this communication is acknowledged.