

**IMPACT OF ACID MINE DRAINAGES FROM
ABANDONED MAMUT COPPER MINE ON
SELECTED METALS IN SEDIMENTS OF
MAMUT-LANGGANAN RIVER AND
BAMBANGAN RIVER,
RANAU, SABAH**



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MORIUS BIN BANTAS



UMS
UNIVERSITI MALAYSIA SABAH

**THESIS SUBMITTED IN
FULFILLMENT FOR THE DEGREE OF
DOCTOR OF PHILOSOPHY**

*PERPUSTAKAAN
UNIVERSITI MALAYSIA SABAH*

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2019**

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DECLARATION

I hereby declare that the materials in this thesis are my own except for quotations, excerpts, summaries and references, which have been duly acknowledged.

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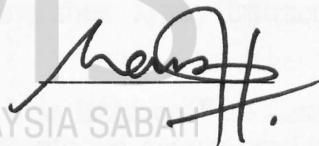
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ABSTRACT

The occurrences of acid mine drainages at abandoned Mamut Copper Mine, Ranau can adversely affect the surrounding rivers. The purpose of this study is to evaluate the impacts of AMD inputs on the status of copper, zinc, iron and aluminium, in the sediments of Mamut-Langganan River and Bambangan River, Ranau Sabah. The total concentration of heavy metals in six particle size fractions namely <0.063mm, 0.063-<0.125mm, 0.125-<0.25mm, 0.25-<0.50mm, 0.50-<1.00mm and 1.00-<2.00mm, metal's geochemical forms based on four-steps sequential extraction method, potential mobility, and solubility of metals at six different pH values namely 2, 3, 4, 5, 6 and 7 were determined. The concentrations of Cu, Zn, Fe and Al in AMD-impacted sediments of Mamut-Langganan River ranged 796-2610 µg/g, 95.7-256.1 µg/g, 4.04-13.6% and 3.85-5.76%, respectively and in Bambangan River ranged 73.5-1347 µg/g, 71.2-271.7 µg/g, 2.55-4.47% and 6.45-9.06%, respectively, and are relatively higher than the concentrations at non-AMD impacted sites (M1 and B1) with exception for Al in Mamut-Langganan River. The enrichment factor for Cu, Zn, Fe and Al ranged 20.7-68.0, 1.29-3.45, 2.01-6.76 and 0.47-0.71, respectively in Mamut-Langganan River, and 0.97-17.8, 1.02-3.89, 0.65-1.15 and 0.89-1.25, respectively in Bambangan River. Only in Mamut-Langganan River and for the metals Cu and Zn in sediments at immediate downstream of AMD input points, are dominantly present in the coarser particles (0.125-< 0.25 mm & 0.25-< 0.50 mm) instead of finest particles (< 0.063 mm). The proportion of metals, in particular Cu and Zn, associated with exchangeable and reducible forms in the sediment of both rivers are higher after inputs of AMD. The proportion of potentially mobile Cu and Zn in AMD impacted sediments ranged 80.4-91.7% and 75.0-89.8%, respectively in Mamut-Langganan River and 89.0-96.9%, 70.9-90.7%, respectively in Bambangan River, which is relatively higher than the proportion for non-AMD impacted sites. There is no apparent differences of the proportion of potentially mobile Fe and Al between non-AMD impacted and impacted sites in Mamut-Langganan River. However, the proportion of potentially mobile Fe and Al is relatively higher in AMD-impacted sediments of Bambangan River. Higher percentage of soluble Cu, Zn, Fe and Al in lower pH value. Regardless of pH value, in general, higher percentage of soluble Cu, Zn, Fe and Al in AMD impacted sediment compared with non-AMD impacted sediment in both rivers. Overall, inputs of AMD have a similar impact on the total concentration, distribution of metals according to particle size, distribution of metal according to geochemical form, and metal solubility according to pH for both Mamut-Langganan River and Bambangan River sediments. The impacts were more apparent for Cu and Zn, and at sites immediate downstream of the AMD input points and greater impact on Cu and Zn in sediments of Mamut-Langganan River compared to Bambangan River.

ABSTRAK

IMPAK SALIRAN ASID LOMBONG DARIPADA BEKAS LOMBONG KUPRUM MAMUT KE ATAS LOGAM BERAT TERPILIH DALAM SEDIMENT SUNGAI MAMUT-LANGGANAN DAN SUNGAI BAMBANGAN, RANAU, SABAH

Kewujudan saliran asid lombong di kawasan bekas lombong Tembaga Mamut, Ranau berupaya memberi impak negatif terhadap sungai-sungai di sekitarnya. Kajian ini bertujuan untuk menilai kesan input SAL terhadap status logam iaitu kuprum, zink, besi dan aluminium dalam sedimen Sungai Mamut-Langganan dan Sungai Bambangan. Kajian merangkumi jumlah kepekatan logam berat di dalam enam fraksi saiz sedimen ($<0.063\text{mm}$, $0.063-0.125\text{mm}$, $0.125-0.25\text{mm}$, $0.25-0.50\text{mm}$, $0.50-1.00\text{mm}$ and $1.00-2.00\text{mm}$), pemisahan bentuk geokimia dalam sedimen bersaiz $<0.063\text{mm}$ berdasarkan empat langkah pengekstrakan berperingkat, potensi pergerakan logam, dan kelarutan logam berat dalam enam nilai pH yang berbeza iaitu 2, 3, 4, 5, 6 dan 7. Kajian menunjukkan julat kepekatan Cu, Zn, Fe and Al dalam sedimen sungai terkesan SAL ialah masing-masing $796-2610\ \mu\text{g/g}$, $95.7-256.1\ \mu\text{g/g}$, $4.04-13.6\%$ dan $3.85-5.76\%$ bagi Sungai Mamut-Langganan dan $73.5-1347\ \mu\text{g/g}$, $71.2-271.7\ \mu\text{g/g}$, $2.55-4.47\%$ dan $6.45-9.06\%$ di Sungai Bambangan, yang secara relatif lebih tinggi daripada kepekatan Cu, Zn, Fe dan Al dalam sedimen yang tidak terkesan SAL kecuali Al di Sungai Mamut-Langganan. Nilai faktor pengayaan untuk Cu, Zn, Fe dan Al ialah masing-masing dalam julat $20.7-68.0$, $1.29-3.45$, $2.01-6.76$ dan $0.47-0.71$ bagi sungai Mamut-Langganan dan $0.97-17.8$, $1.02-3.89$, $0.65-1.15$ dan $0.89-1.25$ bagi sungai Bambangan. Dalam kes taburan logam berat mengikut saiz partikel, hanya di sungai Mamut-Langganan dan logam Cu dan Zn dalam sedimen di hilir paling hampir dengan input SAL, lebih dominan dalam partikel lebih kasar ($0.125-0.25\text{ mm}$ & $0.25-0.50\text{ mm}$) berbanding dalam partikel halus ($<0.063\text{ mm}$). Peratusan logam terutamanya Cu dan Zn, berisosiasi dengan fraksi berupaya bertukar dan boleh diturunkan dalam sedimen kedua-dua sungai, telah meningkat selepas kemasukan SAL. Julat peratus Cu dan Zn dalam sedimen terkesan SAL yang berpotensi bergerak bebas ialah masing-masing $80.4-91.7\%$ dan $75.0-89.8\%$, bagi sungai Mamut-Langganan dan masing-masing $89.0-96.9\%$, $70.9-90.7\%$, bagi sungai Bambangan yang mana secara relatif lebih tinggi berbanding peratus di lokasi yang tidak terkesan SAL. Tiada perbezaan ketara peratusan Fe dan Al yang berpotensi bergerak bebas antara sedimen terkesan SAL dan tidak di sungai Mamut-Langganan. Peratusan Fe dan Al yang berpotensi bergerak bebas secara relatifnya lebih tinggi dalam sedimen terkesan SAL di Sungai Bambangan. Lebih tinggi peratus logam berat yang larut dalam pH yang rendah bagi kedua-dua sungai. Walau bagaimanapun, lebih tinggi peratus Cu, Zn, Fe dan Al yang larut dari sedimen yang terkesan SAL dalam semua julat pH yang digunakan berbanding dari sedimen yang tidak terkesan SAL. Secara keseluruhan, input SAL mempunyai impak yang sama ke atas jumlah kepekatan logam, taburan logam mengikut saiz partikel, taburan logam mengikut fraksi geokimia, dan kelarutan logam mengikut pH untuk kedua-dua sedimen sungai Mamut-Langganan dan sungai Bambangan. Impak lebih ketara ke atas Cu dan Zn, dan di hilir paling dekat dengan punca input SAL dan ke atas sungai Mamut-Langganan berbanding sungai Bambangan

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LIST OF ABBREVIATIONS AND SYMBOLS

AMD	Acid mine drainage
MCM	Mamut Copper mine
Fe	Ferum/iron
Cu	Copper
Zn	Zinc
Al	Aluminum
Mn	Manganese
Ms	millisiemen
L	Litre
mL	millilitre
TDS	Total dissolve solid
SO_4^{2-}	Sulphate ion
CO_3^{2-}	Carbonate ion
OH^-	Hydroxide ion
CaCO_3	Calcium Carbonate
H_2SO_4	Sulphuric acid
HCl	Hydrochloric acid
NaOH	Sodium hydroxide
H_2O_2	Hydrogen peroxide
NH_4OAc	Ammonium acetate
HNO_3	Nitric acid
$\text{NH}_2\text{OH} \cdot \text{HCl}$	Hydroxylamine hydrochloride
HOAc	acetic acid
v/v	Volume/volume
XRD	X-Ray Diffractometer
AAS	Atomic Absorption spectrometer
ICP-OES	Inductively Couple Plasma Optical Emission Spectrometer
kg	Kilogram
mg	Miligram
g	Gram

ppm	Part per million
mm	Millimetre
mg/g	Milligram/gram
mg/kg	Milligram/kg
$\mu\text{g/g}$	Microgram/gram
mg/L	Milligram/litre
GPS	Global positioning system
SESB	Sabah Electricity Sendirian Berhad
EF	Enrichment factor
df	Dilution factor
+	Positive
-	Negative
=	Equal to
>	Greater than
<	Less than
\leq	Same or less than
\geq	Same or greater than
μ	Micro
\pm	Plus minus
\div	Division
\times	Times
$^{\circ}\text{C}$	Degree Celsius
Θ	Theta
Σ	Sum of
\approx	Approximate