

Technical Article

Lake George as a Sink for Contaminants Derived from the Kilembe Copper Mining Area, Western Uganda

Tina Hartwig¹, Michael Owor², Andrew Muwanga², Dieter Zachmann¹, and Walter Pohl¹

¹Institute of Env Geology, Technical Univ, PO Box 3329, 38023 Braunschweig, Germany; ²Dept of Geology, Makerere Univ, Kampala, Uganda; corresponding author's e-mail: walter.pohl@tu-bs.de

Abstract. The copper mine at Kilembe in the Ruwenzori Mountains in western Uganda ceased to operate in 1978 but a steady flow of contaminants, including Cu, Co, Ni, Zn, Cd, and sulphate, continues to enter the Nyamwamba-Rukoki River, which passes through Queen Elizabeth National Park, and finally flows into Lake George. Lake George is quite shallow, alkaline, and highly eutrophic. Measuring mass-flow of contaminants, water, and suspended solids in the Rukoki River near Kasese allowed us to estimate their input into Lake George. Grid sampling of lake sediments indicated that the contaminants settle near the two mouths of the river; low concentrations in a drill core in the centre of Lake George indicate that further dispersion within the lake is small. Sequential extraction experiments on lake sediments and lake water analyses suggest low bioavailability of the heavy metals. We conclude that Lake George is a highly resilient system that efficiently immobilises contaminants. Though there is no health risk for the population under present environmental conditions, a reduction of the contaminant load is desirable.

Key words: Contaminant immobilisation; eutrophic; heavy metals; Kilembe copper mine; Lake George; lake sapropels; sulphur contamination; Uganda

Introduction

Kilembe lies on the eastern slopes of the Ruwenzori Mountains, in western Uganda. The stratabound Cu-Co sulphide deposits of Kilembe are hosted by metamorphic sediments with intercalated minor basic metavolcanics (Warden 1985). About 16 Mt of ore were produced at Kilembe from 1956 to 1978, yielding approximately 270,000 t of copper metal. Because of political unrest, production ceased but the mine has not yet been closed. There was always hope that one day the mine might be reactivated. Because of this situation, underground mine openings, several smaller open pits, ore heaps near the concentrator plant, waste rock heaps, and tailings dams were left exposed to weathering and mobilisation of contained metals.

From 1956 to 1978, copper ore was mined at Kilembe and processed at Kasese (Figure 1). Both sites are heavy metal contaminant sources, due to acid mine drainage, weathering and erosion of waste rock and floatation tailings at Kilembe (Muwanga 1997), and erosion and weathering of a large stockpile of cobaltiferous pyrite at Kasese. The latter is presently being reprocessed by bioleaching to recover the cobalt in an environmentally responsible manner. Before this, pyrite eroded from the stockpile into the bed of the Kamulikwezi, a small seasonal stream that flows into the Kahendero Bay of Lake George. This contamination has not yet been rehabilitated.

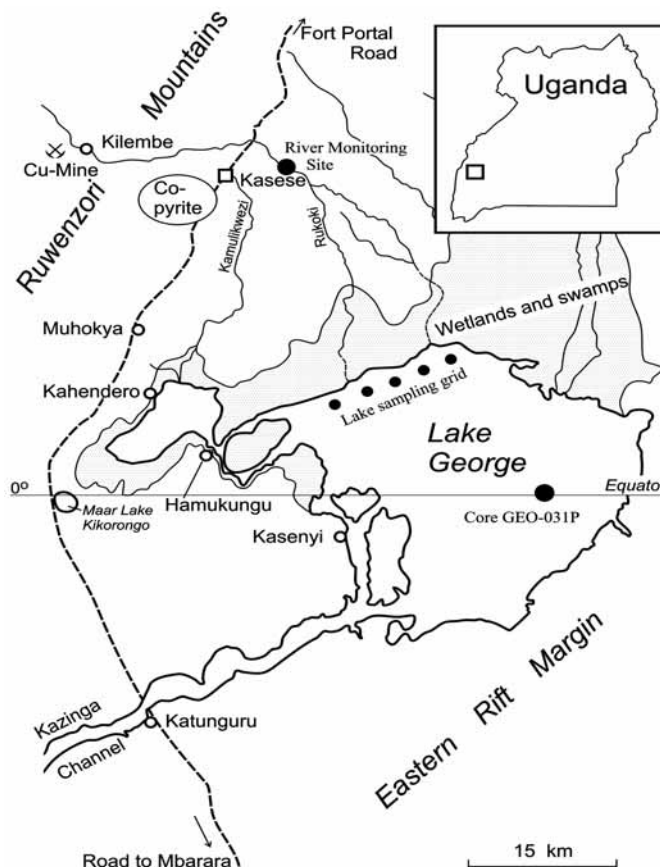


Figure 1. Sketch map of Lake George and the surrounding area, showing Kilembe mines, the site of the Kasese Cobalt Company Ltd cobalt leaching plant (Co-pyrite), and the sampling sites referred to in this paper

Table 1. Ore and trace metal contents of Kilembe ore and of floatation tailings (Muwanga 1997)

	Ore (n = 2)	Tailings mean (n = 49)	Tailings standard deviation
Fe	20.8%	10%	2
Mn	0.17%	0.17%	0.038
S (calc)	17.5%	4.6%	n.d.
Cu	2.37%	1725 ppm	1302
Co	1.0%	983 ppm	839
Ni	487 ppm	405 ppm	151
Zn	906 ppm	142 ppm	49
Cd	17.5ppm	3 ppm	0.7

General Geological and Physical Setting of Lake George

Lake George is part of the chain of the Great Lakes of the western branch of the East African Rift System. It formed in the southern part of a failed rift section (the George Rift), east of the Ruwenzori Mountains, which ends about 30 km north of Lake George (Laerdal and Talbot 2002). The main (Albert) rift is continuous from Lake Edward to Lake Albert along the western flank of the Ruwenzoris.

Lake George, 914 m above sea level (asl) has an average depth of about 2.4 m, with a very flat lake floor, which is tilted towards the east, reaching a depth of ≈ 11 m depth near the eastern escarpment. Extensive papyrus and reed swamps lie north of the lake, especially along the eastern rift escarpment (Figure 1). The lake is alkaline (pH 9-10), eutrophic, and biologically very productive, dominated by blue-green algae (cyanophytes and chlorophytes, especially *microcystis*). This is the base for a rich fishery that reached its productivity peak in the early 1960s. The lake is essentially well mixed, but oxygen deficiency near the bottom occurs during the day due to solar radiation and biological activity (Greenwood 1976). Its ecosystem has been very stable with only small fluctuations (Beadle 1981; Laerdal et al. 2002; Russell et al. 2003) for at least 2,000 years, although human land use changed its surroundings from forests to savannah, and finally to grassland. The environs of the lake are now part of the Queen Elizabeth National Park.

Sediments are brought into the lake by several rivers from the north. Near the northern shore, sand and silt are deposited as well as organic-rich pelitic mud. Most of the lake bottom is covered by organic-rich diatomaceous sapropel muds.

Lake George lies on the equator. Therefore, two rainy and two dry seasons mark its climate, the rainy season in April-May being stronger than the second in October-November (Viner and Smith 1973). Due to the rift valley position, yearly rainfall over the lake (≈ 1000 mm/a) is quite low for an equatorial climate.

With the predominating easterly trade winds, warm and humid air flows towards the Ruwenzori Mts. (Beadle 1981), and as it rises, cools and produces high precipitation. This is the source of nearly all inflow into Lake George. In contrast, little drainage takes place from the East into the George basin, because of low rainfall, a steep escarpment and small catchment areas.

The hydrological balance of Lake George is not well constrained (ILEC 2000; Lehmann 2002; Viner and Smith 1973). The lake's surface is 250 km². Water inflow is dominated by rivers and amounts to ~ 440 Mm³/a. Half of this total is contributed by the Rukoki River (average flow of ~ 7 m³/s, based on systematic measurements taken from 1954 to 1983). Rainfall over the lake (about 1000 mm/a) adds about 250 Mm³/a. Evaporation from the lake (~ 1800 mm/a) is estimated at 450 Mm³/a. As the lake level hardly changes, the lake's outflow through Kazinga Channel into Lake Edward is about 240 Mm³/a. The length of the Kazinga Channel between Lakes George and Edward is about 40 km, and the difference in elevation between the two lakes is only ~ 2 m. Combined with the large width and considerable depth of the channel, this leads to an extremely sluggish flow that is only noticeable during wet seasons (Viner and Smith 1973).

The Ruwenzori Mts. rise to elevations of $>5,000$ m asl, with a steep western side and a gentle slope towards the east. Therefore, the Nyamwamba-Rukoki River has a large catchment area and has built up an extensive alluvial fan in the rift north of Lake George. Although well vegetated, middle reaches of this fan are moderately active, as witnessed by frequent changes of river courses during the last 40 years. Annual flooding leaves widespread silt deposits that are observable in LANDSAT images. Near Kasese, as the Nyamwamba River enters the rift (and is then called Rukoki), stream sediments change over about 1 km from coarse gravels with boulders to sand and silt. The Nyamwamba is obviously drowning in coarse sediment in its lower reaches above Kasese, while the Rukoki is incised into older alluvium, laterite and volcanic tuffs near that town. This is interpreted to

reflect both very limited neotectonic deformation on the Kasese Fault bordering the Ruwenzoris and relatively low annual run-off in the Nyamwamba River compared with earlier Holocene times.

Materials and Methods

In March 2003, water and sediment samples were collected for this study from several stations along the Nyamwamba-Rukoki Rivers and from Lake George. Sampling equipment comprised a HTH-gravity corer, a UGT-sediment trap, and a Nordmeyer water sampler. Sample locations were recorded with a handheld Garmin-GPS device. The analytical work performed at Technical University Braunschweig was undertaken by the Geochemical Laboratory of IUG (Environmental Geology Institute). The analytical methods are verified by regular participation of the laboratory in official quality controls.

Physico-chemical parameters of waters (pH, Eh, T, EC, dissolved oxygen, turbidity) were measured on site with WTW (Wissenschaftlich-Technische Werkstätten, Weilheim) portable equipment. Water samples were collected in polyethylene bottles cleaned by acid steam stripping. The samples were stored cool and dark for transport to the laboratory. In the laboratory, water samples were filtered ($<0.2 \mu\text{m}$) and acidified with suprapure nitric acid. Main and trace element analyses were performed using ICP-OES and ICP-MS.

Sediments were collected in polyethylene bags and refrigerated. In the laboratory, sediments were wet-sieved, dried at 60°C , and then prepared for geochemical and/or mineralogical (XRD) analysis. Most analyses were carried out on the sieve fraction $\leq 63 \mu\text{m}$, with a number of controls based on five fractions and on unsieved material $< 2 \text{ mm}$. Digestion of 0.2 g aliquots was carried out at $80\text{--}100^\circ\text{C}$ with 20 mL aqua regia. Main, minor and trace element determinations were performed using ICP-OES (Bausch and Lomb ARL Type 3520, and Fisons Maxim) and ICP-MS (Micromass Platform). TC (total carbon) was measured using a LECO CS 144 instrument. In a number of control experiments, the sample was first treated with HCl in order to dissolve any inorganic (carbonate) carbon. The results with and without leaching being identical showed that Lake George sediments do not contain carbonate. The pH of the sediments was determined with a pH-meter (WTW pH325), according to the procedure described in ISO 10390 (International Organization of Standardization 2005). Minerals in the sediments were identified by X-ray diffraction (Phillips PW 1730/10). Sequential extraction experiments were carried out on Lake George north shore sediment samples following the method of Jakob et al. (1990),

which is a modified version based on Tessier et al. (1979). All data from the present study are presented on a dry weight basis unless stated otherwise.

Results

Transport of Contaminants from the Mine to the Lake

River water, sediment in the active flow channels, and soil in inundation areas are contaminated downstream from the mine. Immediately below the mining area, average metal contents in river sediments are 1150 ppm Cu, 167 ppm Co, 81 ppm Ni, 1.7 ppm Cd and 108 ppm Zn, most frequently associated with Fe-Mn-oxides or with phyllosilicates. Sequential elution in six steps showed the highest mobilisation potential for Cu and Co (Muwanga 1997). Pre-mining Cu contents in stream sediments at Kilembe were about 30-50 ppm (Hawkes and Webb 1962).

About 25 km downstream from Kilembe, at the railway bridge over Rukoki River near Kasese (Figure 1), a sampling site was established by the Kasese Cobalt Company Ltd (KCCL) for monitoring water chemistry and particulate transport. Unfortunately, there are no concurrent flow data at the site. Mass flow of pollutants at this site (see below) can therefore only be estimated using earlier hydrographic data combined with water analyses from Table 2. This approach probably results in a good estimate, as mass transport and annual mean flow are generally highly correlated in rivers (Sarmiento et al. 2004). Higher mass flow may dilute dissolved pollutants but causes more particulate transport.

At the KCCL monitoring site, the river is an incised narrow channel with a flow cross section about 5 m wide and 1 m deep. Active sediments along this part of the river are fine grained sand and silt with very little clay, correlating with a flow velocity of $\sim 1.15 \text{ m/s}$ (at a mean water level).

Suspended solids were sampled at this site in March 2003 with a sediment trap (Kozerski 2002), sieved and analysed for major and trace elements. Compared to coarser grain size fractions, C_{org} , Fe, Mn and sulphur are clearly enriched in the $\leq 63 \mu\text{m}$ fraction. This fraction is also the main carrier of the heavy metals, with 425 ppm Cu, 167 ppm Co, 64 ppm Zn, and 49 ppm Ni. There was little difference in metal content of suspended solids between normal and flood conditions, although the amount of sediment trapped was smaller at high flow velocity. Combining these data with the long term measurements of suspended solids (Table 2) allows an upper bound estimate of mass transport of contaminants in solid

form (see below). Colloidal transport could not be investigated. Table 2 shows that River Rukoki water is essentially of drinking water quality, although it carries a considerable load of dissolved contaminants from Kilembe mine to Lake George (Table 3).

The suspended solid contaminant load carried by the Rukoki River can be estimated based on mean flow data, the contaminant concentration of sediment trap samples, and on the mass of suspended solids in river water (Table 2). Although these data cannot represent the complexity of particulate transport in the river system, they are thought to approach the order of magnitude. Except for iron, contaminants are mostly transported in their dissolved form (Tables 3 and 4).

A considerable flow of contaminants passes through from Kilembe at the Kasese monitoring site. Dissolved plus particulate mass amounts to about 7,500 t/a of SO₄, 650 t/a of Fe, 27 t/a of Mn, 30 t/a of Cu, 16 t/a of Co, 5 t/a of Ni, 10 t/a of Zn, and at least 13 kg/a of Cd. Thus, there is nearly three times the mass of sulphur than what is needed to bind the metals as sulphides.

Access to the river is very difficult downstream from this site, because of dense vegetation and restrictions

posed by Queen Elizabeth National Park. Sampling at a mid-point between the 35 km distance to Lake George (Figure 1) confirmed that little change takes place in the main characteristics of the river. At about this location, however, the river enters the impenetrable wetlands belt that fringes the northern shore of Lake George.

The wetland vegetation is characterised by tall reeds and *Cyperus papyrus*, which screens the river channel from view in aerial photographs and LANDSAT images. Generally, wetlands are considered to be extremely efficient traps of metals (Taylor et al. 1998). In principle, cations and sulphur may be permanently immobilised by microbial reduction or adsorption to organic matter, and subsequent burial. The efficiency of these processes is controlled by physical, microbial and chemical parameters (Ho and McKay 2004).

From our data on sediment and water samples from Lake George near its northern shore, however, it is clear that the flux of contaminants from Kilembe enters the lake with little diminution (see below). We assume that the river maintains its rapid flow through the wetlands even at low water levels, and produces break-through channels during flood water surges.

Table 2. Range of variation and averages of parameters measured in River Rukoki water at Kasese railway bridge (KCCL unpublished data 2000-2002; n = 89)

	Mean	Minimum	Maximum	Standard deviation (sd)	WHO (2004) guideline values
pH	7.3	6.6	8.0	0.31	not of health concern
EC, $\mu\text{S}/\text{cm}$	120.9	48.4	217	39.7	not of health concern
Fe, mg/L	0.6	0.1	2.0	0.26	not of health concern
Mn, mg/L	0.1	0.0	0.3	0.04	not of health concern
SO ₄ ²⁻ , mg/L	33.6	7.0	61.0	9.5	no adverse effects
Cu, mg/L	0.113	0.001	0.300	0.053	2
Co, mg/L	0.07	0.0	0.16	0.04	no comment provided
Ni, mg/L	0.02	0.0	0.12	0.02	0.02
Zn, mg/L	0.04	0.0	0.1	0.02	3
Cd, $\mu\text{g}/\text{L}^*$	0.06	0.03	0.16	-	3
Susp. Solids, mg/L	39.7	3.8	490	76.41	not of health concern

*Cd analyzed at Geochemical Laboratory IUG-TUBS (n = 5)

Table 3. Calculated dissolved contaminant load (t/a), and standard deviation (sd) in the Rukoki River (based on long term mean flow of $\sim 7 \text{ m}^3/\text{s}$ at Kasese)

	Fe	Mn	SO ₄	Cu	Co	Ni	Zn	Cd*
Conc, mg/L	0.6	0.1	33.6	0.113	0.07	0.02	0.04	0.00006
sd	0.26	0.04	9.5	0.053	0.04	0.02	0.02	n.d.
t/a mean	132	22	7,390	25	15.4	4.4	8.8	0.0132
sd	57.2	8.8	2,090	12	8.8	4.4	4.4	n.d.

*Note: n = 89 except for Cd (n = 5)

Table 4. Estimated particulate contaminant load (t/a, rounded), and standard deviation (sd) in the Rukoki River at Kasese (based on mean mass of suspended solids 8,734 t/a with a standard deviation of 16,810 t/a)

	Fe	Mn	SO ₄	Cu	Co	Ni	Zn	Cd
Concentration, ppm	59,654	644	4,578	425	167	49	64	b.d.l. (0.000015)
Load, t/a, mean	520	5.6	40	3.7	1.5	0.4	0.6	-
Load, t/a, sd	1,000	10.8	77	7.1	2.8	0.8	1.0	-

Heavy Metals and Sulphur in Lake George

An irregular sampling grid of 46 stations (controlled by GPS) was established in northern Lake George near both mouths of the Rukoki River (Figure 1). Lake bottom muds, and near the river mouths, silt and sand, were recovered with a gravity corer. The youngest sediments were 5-10 cm of liquid muds, with more cohesive samples taken from 5-15 cm depth and estimated to be between 5 to 15 years old, due to the high sedimentation rate near the river mouths. The available equipment did not allow slicing, so all data refer to this admittedly imprecise age bracket, which, however, is undoubtedly younger than active mining at Kilembe. Water was sampled at mid-depths to the bottom, the latter being generally at 1.5-2 m below surface. Due to the high level of biological activity, Lake George waters are very turbid (34-63 nephelometric turbidity units--NTU). The analytical results will be presented in detail by Owor et al. (in preparation).

In March 2003, the lake water had a pH of 9 – 10 at temperatures of 28 – 32°C. The redox potential varied from +44 to -20 mV, probably due to different levels of organic activity. Electrical conductivity was between 260 and 222 $\mu\text{S}/\text{cm}$, which indicates a slight evaporative concentration. The water contains 15-20 mg/L both of Ca and Na, and 5-10 mg/L of Mg and K. Zn (up to 45 $\mu\text{g}/\text{L}$) and Cu (16 $\mu\text{g}/\text{L}$) reached the highest concentration of the Kilembe-derived contaminants (sulphur was not measured). Cadmium was below the detection limit at 0.015 $\mu\text{g}/\text{L}$.

Sediments are mainly clayey silts with a high content of organic matter (sapropels). The uppermost layers are probably resuspended and redistributed frequently due to storms producing ground waves. The sediments are homogeneous without any layering or banding. Fine sand and silt banks occur only near the river mouths. Minerals identified by XRD-analyses in nearshore lake sediments are quartz, anorthite, albite, biotite, kaolinite, and muscovite/illite. Neither crystalline Fe-S phases nor iron or manganese oxides were found. Sediment pH varies between 4.5 and 7, with a mean of 5.45. Therefore, carbonate phases of Ca, Mg, Mn, or Fe are rarely stable. In most samples, total carbon content equals C_{org} .

Kilembe-derived contaminants in recent Lake George sediments (Table 5) clearly reflect the input from the Rukoki River. Ratios between mean concentrations of pollutants in the lake compared to river sediments show little change, e.g. $\text{Cu}/\text{Co}_{\text{Lake}} : \text{Cu}/\text{Co}_{\text{Rukoki}} = 2.5:2.5$, $\text{Cu}/\text{Zn}_{\text{Lake}} : \text{Cu}/\text{Zn}_{\text{Rukoki}} = 5.6:6.6$, and $\text{Fe}/\text{Mn}_{\text{Lake}} : \text{Fe}/\text{Mn}_{\text{Rukoki}} = 82:92$. The small deviations may either be spurious or may indicate that more Fe and Cu are

trapped in the wetlands compared to Mn and Zn. Absolute concentrations in lake sediments are generally lower, however, than in Rukoki sediments or in suspended solids (compare Tables 4 and 5). Maximum concentrations cluster near the river mouths, which is attributed to rapid precipitation of pollutants on entering the lake. Exceptions are S and Zn. Zn seems to suffer no diminution while passing through the wetlands. Sulphur increases dramatically, which is due to wastewater from the KCCL cobalt plant, which is pumped into River Rukoki below our Kasese observation site. However, bio-oxidation and leaching of the cobaltiferous pyrite is planned to end in about 2010. Correlation analysis shows that C_{org} and S have a strong positive covariance, implying that apart from sulphide phases, a second major sink for S is sulphurized organic matter.

Contaminant Precipitation in Lake George Sediments

Six Lake George bottom sediment samples were sequentially extracted to assess the bioavailability and binding strength of the contaminants. Although sequential extraction is subject to readsorption, redistribution, and a lack of selectivity, it still provides a practical tool for elucidating the complex dynamics existing in sediments (Landner and Reuther 2004). Solubility was determined in six steps (Jakob et al. 1990): I = exchangeable ions (leaching with ammonium acetate, pH 7), II = carbonate phase (sodium acetate, pH 5), III = easily reducible phase (hydroxyl ammonium chloride, pH 2), IV = moderately reducible phase (ammonium oxalate acid, pH 3), V = sulphide or organically bound phase (hydrogen peroxide/ammonium acetate, pH 7), and VI = residual, mineral phase (aqua regia). The analytical results will be presented in detail by Owor et al. (in preparation).

Up to 20% of the iron in recent lake sediments occurs as oxyhydroxides and oxides (elution steps III and IV); most of the rest is bound in sulphide/metal-organic phases (V). Simpson et al. (2000) have shown that rapid sulfidation produces surface coatings of FeS on amorphous Fe(III) oxyhydroxide particles. This may explain the occurrence of oxidized Fe and Mn in these anaerobic sediments. Easily exchangeable manganese (step I), in contrast, reaches 40%. The remainder is bound in oxyhydroxides and oxides (III and IV), little sulphide, and in the residual phase (IV). Up to 20% of sulphur occurs together with Ca in fraction (I), probably as a very soluble Ca-sulphate. As expected in these black muds, the remaining sulphur is bound in sulphide or organic form (V).

The distribution of copper in the various phases is quite erratic in different samples, but generally, levels were low in steps I, II, and III, adding up to only 20%;

Table 5. Organic carbon (in %), and Kilembe mines-derived contaminants (ppm) in Lake George sediments near the north shore (n = 47)

	Fe	Mn	S	Cu	Co	Ni	Zn	Cd	C _{org} %
mean	32,107	392	11,580	346	137	63	62	0.3	7.2
max	51,584	649	22,506	787	529	238	127	1.0	17.4
min	21,507	244	673	37	18	25	41	0	0.9
sd	4,834	76	4,313	253	95	33	12	0.1	2.6

most of the Cu is hosted in less reducible phases (IV) and in sulphides/metal-organic phases (V). The distribution of nickel and zinc is very similar to copper. Cobalt reaches 30% in step I, about 10% each in steps III and IV, with the remainder mainly in the sulphide/metal-organic phases (V). Cadmium was found in phase VI, but this is probably an analytical artefact; most probably, it is exclusively present as a sulphide (O'Day et al. 2000).

Overall, these results reveal that even in recent Lake George sediments, only a small part of the Kilembe-derived contaminants is bound in exchangeable form. Most of the total mass is fixed in sulphides and organic matter, or in Fe-Mn oxyhydroxides and oxides. Therefore, their bioavailability is low, though it is possible that diagenetic processes or environmental change could induce conditions that would release part of the heavy metals (see discussion below).

Heavy Metals in Core GEO-031P

An important aspect of this work was to investigate lake sediment cores in order to compare recent anthropogenic contamination with natural pre-mining (before 1957) concentrations derived from the Kilembe ore bodies. The University of Minnesota Limnological Research Center kindly provided samples of core GEO-031P, drilled in May 2003 in the centre of Lake George. We analysed these samples for major and trace elements (especially Kilembe-derived heavy metals) by ICP after hydrofluoric acid digestion (aliquot 0.2 g, 5 mL conc. nitric acid, 2 mL conc. perchloric acid, and 7 mL conc. hydrogen fluoride at 140°C), as well as C_{org}, S, and mineral phases by XRD as described above. The analytical results will be presented in detail by Owor et al. (in preparation).

Core GEO-031P was drilled about 20 km southeast of the two mouths of River Rukoki (Figure 1). This is beyond the distance reached by flood plumes from the river. The core was drilled to a depth of 630 cm from the lake bottom, at a water depth of 5.5 m (Figure 2). At 4.8 m below the lake floor, a peaty clay layer was encountered, representing low-standing swamp sediment. Underlying mottled soils are products of sub-aerial weathering. The overlying clayey sapropels are unstratified and indicate continuous

sedimentation. Radiocarbon dating of this core suggests sedimentation rates at this site vary between about 0.2 and 0.5 cm/yr (Jim Russell and Doug Schnurrenberger, written comm., 2004). This puts the 1950 (pre-mining) level in core GEO-031P at a depth between 11 and 27 cm. We received seven sample slices irregularly spaced from 2-111 cm below the lake floor. The deepest sample should have an age of about 1780 AD, assuming the higher sedimentation rate, or 1450 AD with the lower rate. Obviously, these samples reach well back into pre-mining time.

From the desiccation surface at 4.8 m below the lake floor upwards, the water content of the sapropel increases from 60 to over 97%. Within the section covered by our samples (Figure 2), water content increases from 95 to 97%. Minerals identified by XRD include quartz, muscovite/illite, kaolinite, biotite, and pyrite. In the uppermost two samples, zirconium content increases by about 30%, from a mean of 53 ppm to 67 ppm. This is interpreted to reflect higher availability of comminuted rock material due to mining at Kilembe. At the same time, most major and minor elements derived from Ruwenzori rocks (including Fe, Mn, Ca, K, Na, and Ti) decrease; dilution by quartz is assumed to be responsible for this. It is interesting to note that plagioclase is absent in these mid-lake sediments.

Among the Kilembe-derived contaminants, Cu, Co, Ni, and Cd increase moderately in the youngest sediments (Table 6). Concurrently, As, Pb, and especially Sb are enriched, reflecting a general, not mining-derived anthropogenic contamination (e.g. Kasese town). In contrast, levels of Fe, S, Mn, and Zn are lower in the uppermost two samples. This cannot be due to reduced mobilisation from the source. The explanation may be either retention along the transport-path (for Fe and Mn), or remobilisation by oxygenated lake water during storm waves (S and Zn). In addition, de-watering of the sediment may transport Fe²⁺ upwards, while H₂S diffuses downwards (Jorgensen et al. 2004). In Core GEO-031P, the sulphur-iron ratio is slightly above 1, indicating that most iron is sulfide-bound. The reverse has been found in the north-shore lake sediments, where iron is in surplus.

Among the Kilembe-derived contaminants, Cu, Co, and Ni contents are lower in these sediments than the

crustal average, Zn is about equal, while S and Cd are above the crustal average (Table 6).

Discussion and Conclusions

Yearly dissolved plus particulate mass flow from Kilembe via the Rukoki River amounts to about 7,500 t of sulphate (not considering the temporary input from KCCL), 650 t of Fe (see below, however), 27 t of Mn, 30 t of Cu, 16 t of Co, 5 t of Ni, 10 t of Zn, and at least 13 kg of Cd. That includes nearly three times the mass of sulphur than what is needed to bind the metals in sulphide form.

Geochemical data from north shore sediment and water samples indicate that the Rukoki River discharges sediment and water directly into Lake George. At flood conditions during rainy seasons, plumes of river water and suspended sediment may extend several hundred metres out into the lake. Further distribution of contaminants occurs due to the slow movement of water from the north shore towards the Kazinga outlet in the south. Living organisms, including fish, will also be a vector of dispersion. Storms with ground waves may be an agent of resuspending and redistributing the fluffy sediment. However, the rapid decrease of Kilembe-derived elements from the north shore to the centre of the lake (core GEO-031P) and to the south (Lwanga et al. 2003) clearly demonstrates that contaminants are efficiently immobilised in Lake George sediments.

Due to an extremely high organic production in the lake, the sediments are brown or black, muddy sapropels with about 30% of organic carbon. As Fe, Mn, Zn, Cu, and Co are micronutrients, import of these elements from Kilembe may enhance organic productivity. Various inorganic and biologically-mediated processes take place in these sediments, including anaerobic microbial methanogenesis at depth, bacterial sulphate reduction near the surface,

and consequent formation of amorphous or meta-stable FeS-phases, which stabilise Kilembe mine-derived metals. The sulphides of Cd, Cu, Co, Ni, and Zn are highly insoluble so these metals are immobilised and not bio-available (Ankley et al. 1996). Methane-producing archaea and bacteria are probably a source of free protons, as the availability of Ni in the sapropels may favour formation of Fe-Ni-hydrogenase (Armstrong 2004). Resuspension of sediment in oxygenated lake water can provoke renewed oxidation of sulphides and consequent liberation of contaminants, although generally, sulphides are resistant to temporary oxidation (Simpson et al. 2000). In the lake water, aerobic biogenic methane oxidation produces CO₂. This is converted by photosynthesis to organic matter and so adds to the alkalinity of Lake George that is provided by evaporation.

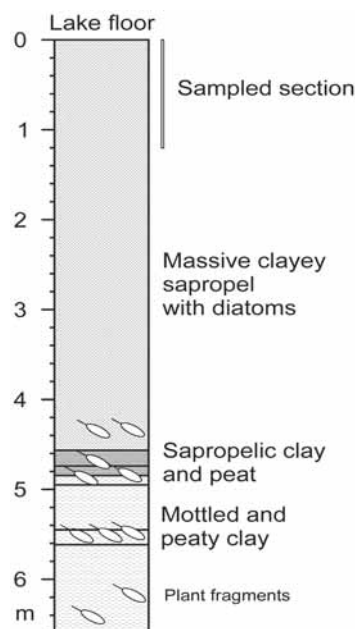


Figure 2. Generalised lithology of core GEO-031P (after Russell and Schnurrenberger, written comm. 2004) with the section sampled for this work

Table 6. Kilembe-derived contaminants and organic C in core GEO-031P

sample depth, cm	Fe, ppm	Mn, ppm	S, ppm	Cu, ppm	Co, ppm	Ni, ppm	Zn, ppm	Cd, ppm	C _{org} , %
2-4	37,246	455	23,031	50	17	27	61	1.2	27.8
8-10	36,356	444	33,278	46	11	20	58	0.8	27.9
16-19	43,273	513	37,401	43	13	17	75	1.1	27.2
25-27	46,642	526	38,192	44	13	18	76	0.8	25.4
41-44	43,130	474	38,284	32	13	26	74	1.0	29.8
68-71	44,552	613	40,995	34	11	28	71	0.8	30.5
108-111	41,443	688	39,447	30	13	28	78	0.9	31.8
Crustal abundance*	50,000	900	500	60	25	80	70	0,18	n.a.

* consensus figures from Smith & Huyck 1999

Methanogenesis may take place through the whole sapropel column (e.g., during drilling, large amounts of CH₄ are released and bubble up to the surface). In contrast, H₂S production probably peaks at the interface of sulphate-reduction and methanogenesis zones (Jorgensen et al. 2004). In the latter, anaerobic reduction of any remaining sulphate takes place.

The reaction between H₂S and iron leads to formation of amorphous iron monosulphide (and possibly metastable greigite and mackinawite) in the sediments; these can gradually convert to pyrite. Reduced sulphur may also form organic and organo-metallic compounds. At low sulphur availability, reduced pore waters may dissolve Fe, Mn, and other metals. Dewatering of the sediments can in that case cause vertical transport of metals, but this is not recognisable in the analytical data of core GEO-031P (Table 6). As illustrated by sequential extraction experiments, Cu, Co, Ni, and Zn form organo-metallic compounds, or are included in FeS-phases. Similar to Ba and P, Cd is enriched in living organisms and is characteristic for organic matter from high productivity waters.

The intermittent vertical spacing of samples from core GEO-031P made it impossible to identify the exact onset of Kilembe-derived pollution. As shown above, concentrations of Kilembe ore elements seem to

indicate that only the uppermost two samples are anomalous, implying the lower sedimentation rate. Several element ratios, however, produce distinct clusters that include the uppermost four samples, suggesting that the onset of mining-induced import is located between 27 and 41 cm depth (Figure 3). This supports the higher sedimentation rate of about 0.5 cm/a.

It is not possible to quantitatively describe the dispersion and fate of Kilembe-derived contaminants in Lake George samples with the data presented. Assuming, however, that the siting of core GEO-031P in the centre of the lake predisposes it as representative of the mean concentration, a simple working model may be devised as follows: assuming an area of 250 km², a sedimentation rate of 0.5 cm/a (with 97% water and a density of 1.1 g/cm³), the yearly dry mass deposited is about 40,000 t. Based on the data from 2-4 cm depth, a theoretical total contaminant deposition posterior to active mining at Kilembe may be calculated (Table 7). A comparison with the mass input by the Rukoki River reveals remarkable differences. Obviously, Cu, Co, and Ni and sulphur are efficiently immobilised as they enter the lake, as most heavy metals precipitate at the transition to an alkaline and reducing environment. Only a small amount of these contaminants reaches the site of core GEO-031P. The more mobile elements, Mn and Zn, show a wider dispersion. Iron, in contrast, seems to be enriched, but

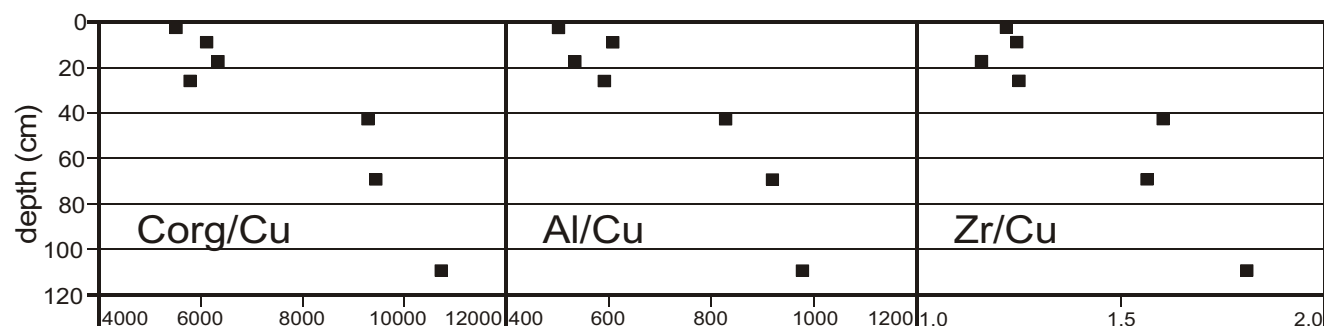


Figure 3. Ratios of Al, Zr, and C_{org} to Cu against depth in core GEO-031P, illustrating clusters of anomalous values in the uppermost four samples, presumably due to onset of copper mining at Kilembe in 1957

Table 7. Theoretical total yearly contaminant deposition in Lake George calculated from core GEO-031P data, compared to the Rukoki River input

Core GEO-031P	A: theoretical mass/year deposited subrecently in Lake George (calc)	B: Rukoki input/year (measured/estimated mean \pm standard deviation)	Ratio A/B (mean)
S (2.3%)	950 t	2,500 \pm 700 t	0.38
Fe (3.7%)	1,540 t	650 \pm 1,100 t	2.4
Mn (455 ppm)	19 t	27 \pm 20 t	0.7
Cu (50 ppm)	2 t	29 \pm 19 t	0.07
Co (17 ppm)	0.7 t	17 \pm 12 t	0.04
Ni (27 ppm)	1 t	5 \pm 5.5 t	0.2
Cd (1.2 ppm)	48 kg	>13 kg*	3.7
Zn (61 ppm)	2.5 t	9 \pm 5.5 t	0.28

* Note that the cadmium input by the Rukoki River is not well constrained.

we conclude that our data underestimates the import of Fe by the Rukoki River, probably occurring in colloidal phases $>0.2 \mu\text{m}$ (Ranville and Schmiermund 1999). Overall, core Geo-031P is clearly not representative of the average contamination of Lake George, and instead highlights the very limited dispersion of contaminants within the lake.

Heavy metal levels in Rukoki River and Lake George water are not an immediate danger to animal and human life, though copper levels are potentially high enough (above $15 \mu\text{g/L}$) to cause stress to aquatic life. Fish sampled in Lake George do show signs of stress (Lwanga et al. 2003), but this is probably due to feeding on suspended particles and biota rather than uptake of dissolved ions. The reason for this is that dissolved organic matter decreases metal toxicity by lowering the free ion activity (Vink 2002).

Metal contents in lake sediments near the northern shore clearly reflect mine-derived contamination. At present environmental conditions, however, a heavy metal transfer from sediment to water is very restricted because of the high alkalinity of the lake water. The highest toxic risk in Lake George are at present not mine-derived heavy metals but cyanobacterial blooms that can produce toxins (*cyanotoxins*), which in high concentrations are fatal to humans, livestock, and wildlife (Huisman et al. 2005).

In conclusion, the Lake George system is evidently a highly resilient system that efficiently immobilises contaminants from Kilembe. Yet, diminution of the contaminant load is desirable. This may be achieved by retreatment of the tailings and recovery of the contained metals, concurrently allowing environmental clean-up of Kilembe mines. Until then, most contaminants are trapped in reduced sediments near the lake's north shore. Only drying out of the lake would lead to a dramatic release of heavy metals and of sulphur from the sediments. Paleoclimatic studies in many parts of Africa have shown that warming at higher latitudes that may result from global change will induce a stronger monsoon in Africa and concomitantly more humidity, while cooling would cause lake level regressions (Gasse 2001; Stager et al. 2005). Higher lake levels under a more humid climate are therefore expected to increase sedimentation rates and concurrently, burial and immobilisation of the pollutants. Accordingly, a catastrophic drying-out scenario appears highly improbable in the foreseeable future.

Acknowledgements

This research was supported by Volkswagenstiftung (Hannover). Jim Russell and Doug Schnurrenberger, Limnological Research Center, University of Minnesota, kindly provided samples of core GEO-031P. KCCL (Kasese Cobalt Company Ltd) assisted with equipment and time series data on River Rukoki waters measured at the Kasese railway bridge.

References

- Ankley GT, Di Toro DM, Hansen DJ (1996) Technical basis and proposal for deriving sediment quality criteria for metals. *Environ Toxicol Chem* 15: 2056-2066
- Armstrong F (2004) Hydrogenases: active site puzzles and progress - current opinion. *Chemical Biology* 8: 133-140
- Beadle LC (1981) The inland waters of tropical Africa. Longman, New York, 433 pp
- Gasse F (2001) Hydrological changes in Africa. *Science* 292: 2259-2260
- Greenwood PH (1976) Lake George, Uganda. *Phil. Trans R Soc London B*. 274: 375-391
- Hawkes HE, Webb JS (1962) Geochemistry in mineral exploration. Harper & Row, New York, 415 pp
- Ho YS, McKay G (2004) Sorption of copper(II) in aqueous solution by peat. *Water, Air and Soil Pollution* 158: 77-97
- Huisman J, Matthijs HCP, Visser PM (eds) (2005) Harmful cyanobacteria. Springer, New York, 243 pp
- ILEC (2000) <http://www.ilec.or.jp> – World Lakes Database (July 2005)
- Jakob G, Dunemann L, Zachmann D, Brassler T (1990) Untersuchungen zur Bindungsform von Schwermetallen in ausgewählten Abfällen. *Abfallwirtschaft J* 2 (7/8) : 451-457
- Jorgensen BB, Böttcher ME, Lüschen H (2004) Anaerobic methane oxidation and a deep H_2S sink generate isotopically heavy sulphides in Black Sea sediments. *Geochim Cosmochim Acta* 68: 2095-2118
- Kozerski HP (2002) Determination of areal sedimentation rates in rivers by using plate sediment trap measurements and flow velocity-settling flux relationship. *Water Research* 36: 2983-2990
- Laerdal T, Talbot MR (2002) Basin neotectonics of Lakes Edward and George, East African Rift. *Paleogeography, Paleoclimatology, Paleoecology* 187: 213-232

- Laerdal T, Talbot MR, Russell JM (2002) Late Quaternary sedimentation and climate in the Lakes Edward and George area, Uganda-Congo. In: Odada EO, Olago DO (eds), *The East African Great Lakes: Limnology, Paleolimnology and Biodiversity*, Kluwer, Dordrecht, p 429-470
- Landner L, Reuther R (2004) *Metals in society and in the environment*. Kluwer, Dordrecht, 406 pp
- Lehmann JT (2002) Application of satellite AVHRR to water balance, mixing dynamics and chemistry of Lake Edward, East Africa. In: Odada EO, Olago DO (eds), *The East African Great Lakes: Limnology, Paleolimnology and Biodiversity*. Kluwer, Dordrecht, p 235-260
- Lwanga MS, Kansiime F, Denny P, Scullion J (2003) Heavy metals in Lake George, Uganda, with relation to metal concentrations in tissues of common fish species. *Hydrobiologia* 499: 83-93
- Muwanga A (1997) Environmental impacts of copper mining at Kilembe, Uganda: a geochemical investigation of heavy metal pollution of drainage water, stream sediments and soils in the Kilembe Valley in relation to mine waste disposal. *Bswg geowiss Arb* 21, Institut für Geowissenschaften, TU Braunschweig, 140 pp
- O'Day PA, Carroll SA, Randall S, Martinelli RE, Anderson SL, Jelinski J, Knezovich, JP (2000) Metal speciation and bioavailability in contaminated estuary sediments, Alameda Naval Air Station, California. *Environ Sci Technol* 34: 3665-3673
- Ranville JF, Schmiermund RL (1999) General aspects of aquatic colloids in environmental geochemistry. In Plumlee GS, Logsdon MJ (eds), *The environmental geochemistry of mineral deposits*. *Rev in Economic Geol* 6: 183-199
- Russell JM, Johnson TC, Kelts KR, Laerdal T and Talbot MR (2003) An 11,000-year lithostratigraphic and paleohydrologic record from equatorial Africa: Lake Edward, Uganda-Congo. *Palaeogeography, Palaeoclimatology, Palaeoecology* 193: 25-49
- Sarmiento AM, Nieto JM, Olias M (2004) The contaminant load transported by the river Odiel to the Gulf of Cadiz (SW Spain). *Applied Earth Sci.* 113: 117-122
- Simpson SL, Apte SC, Batley GE (2000) Effect of short-term resuspension events on the oxidation of cadmium, lead, and zinc sulphide phases in anoxic estuarine sediments. *Environ Sci Technol* 34: 4533-4537
- Smith KS, Huyck HLO (1999), Abundance, relative mobility, bioavailability, and human toxicity of metals. In Plumlee GS, Logsdon MJ (eds), *The environmental geochemistry of mineral deposits*. *Rev in Economic Geol.* 6, p 29-70
- Stager JC, Westwood J, Grzesik D, Cumming BF (2005) A 5500-year environmental history of Lake Nabugabo, Uganda. *Paleogeography, Paleoclimatology, Paleocology* 218: 347-354
- Taylor GH, Teichmüller M, Davis A, Diessel CFK, Littke R, Robert P (1998) *Organic Petrology*. Borntraeger, Berlin, 704 pp
- Tessier A, Campbell PGC, Bisson M (1979) Sequential extraction procedure for the speciation of particulate trace metals. *Anal Chem* 51: 844-851
- Viner AB, Smith IR (1973) Geographical, historical and physical aspects of Lake George. *Proc Royal Soc London* 184: 235-270
- Vink JPM (2002) Measurement of heavy metal speciation over redox gradients in natural water-sediment interfaces and implications for uptake by benthic organisms. *Environ Sci Technol* 36: 5130-5138
- Warden AJ (1985) Reappraisal of geological setting and potential of Kilembe copper mine, Uganda. *Trans. Instn Min Metall* B94: 94-105
- WHO (World Health Organization)(2004) *Guidelines for drinking-water quality, 3rd edit, Annex 4, Chemical summary tables*. Geneva

Submitted May 11, 2005; accepted July 17, 2005