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Exploratory geochemical assessment of stream water and sediment contamination in Gatumba tin and tantalum mining district, Rwanda.

by Judith Flügge, Andrew Muwanga, Kerstin Trümper, Dieter Zachmann, Walter Pohl, Braunschweig and Kampala (Uganda)

with 4 Figures and 2 Tables in the Text

Abstract: Stream waters are not contaminated with dangerous inorganic elements. According to WHO guidelines, these waters are acceptable for lifelong consumption without risk to health. Note, however, that microbial waterborne disease vectors were not investigated. Stream sediments generally have relatively low contents of most elements that may be derived from pegmatites. Of mine-related contaminants, only uranium and arsenic exceed their respective average crustal abundance. Both appear to be higher near former mine sites but this is probably not caused by mining. Causes are rather primary hydrothermal and secondary supergene dispersion, and recent mobilisation because of intensive agricultural use. Yet at present, the welfare of the local population is not threatened. The most serious hazard is human interference with groundwater by abstracting drinking water either from wells in valley fill or from deeper horizons of the regolith. Overall, our work suggests that past mining at Gatumba caused little dispersion of deleterious elements.

1. Introduction

Apart from physical changes of a landscape, mining may leave a heritage of less visible hazards to humans and environment. Within an interdisciplinary cooperative pilot project ("Coltan Environmental Management") the consequences of former and ongoing tin and tantalum mining in Central Africa are currently investigated. As a sample area for a pilot project, the mining district of Gatumba was chosen.

Central Africa hosts one of the few tantalum ore provinces of the world. In this region, tantalum deposits are of the pegmatitic type (POHL 1994, 2005) and are exploited by small scale or by artisanal mining methods. Commonly, the ore mineral is tantalum-rich columbite $[(Fe,Mn)(Ta>Nb)_2O_6]$ which contains about 52 to 86 weight% Ta_2O_5. Tantalum pegmatites are part of the Li-Cs-Ta-rare-metal-group of pegmatites and granites. Cassiterite is often the dominating ore, while lithium, beryllium, caesium, and feldspar or kaolinite are occasional co-products. Minor and trace minerals associated with ore include pyrite, arsenopyrite, sulfides, and sulfosalts (with Pb, Zn, Cd, etc.). Therefore, deleterious elements which may be present in primary or in placer deposits of tantalum include arsenic, fluorine, sulfur, and base metals. In addition, some tantalites have elevated traces of uranium (and its radioactive daughter nuclides, most notably 222radon, 226radium, 214bismuth, and stable lead).

In this paper, results of exploratory screening for the mentioned potential pollutants (excepting the unstable isotopes of the uranium decay series) in drainage water and sediments of part of the Gatumba area are reported.

2. Mining, Geological and Physical Setting of Gatumba district

Coltan (and tin ore) mining at Gatumba was active in a semi-industrial manner from 1929 to 1985. During this period, some 18,000 tonnes of cassiterite and columbotantalite concentrate were produced. As average grades of ore in situ are assumed at 0.5 kg/m³, the total mass of rock moved can be estimated at 36 Mm³ or about 50 million tonnes. Because mining took place mainly by hydraulic methods, most of this mass was moved downslope from hillsides into nearby valleys. Quartz boulders and smaller fragments down to sand size form the bulk of the waste rock left behind. Minor amounts of white mica are ubiquitous, as are iron oxide particles. Bottoms of stream valleys adjacent to mines were buried by this mass of loose sediment. Stream gradients were locally changed.

A sizeable part of the ore was extracted from alluvial placers in the valleys proximal to primary deposits. In that case, the stream sediments were also shifted downvalley, but only for tens of metres. Alluvial mining disturbed the natural bedding of the valley fill and commonly left an irregular hummocky landscape.

Typical former mine sites at Gatumba are elongate hillside quarries with a width of about 50 m and a length of 200 m. These quarries mirror the occurrence of long dikes of mineralised pegmatites that generally strike South-North and have a nearly vertical dip. The thickness of the pegmatites varies up to 40 m, but is most often only 10 m. Therefore, the opencut mines are narrow ravines that are partly refilled with rock fall from barren pegmatite and country rock schists. Mining started by exploitation of residual placers that formed near the base of an ancient (Cretaceous to Tertiary?) soil cover above the mineralised pegmatites. Subsequently, weathered (kaolinised) parts of in-situ pegmatites were extracted but not hard rock ore because grades were too low. Since 1985, mining in the district was limited to very small scale artisanal activities, targeting local high-grade pockets. Recently, exploration work was started in order to define ore bodies for industrial mining.

Gatumba is one of many pegmatite mining districts in the Kibara orogen (POHL 1994). The Kibaran is an intracontinental fold belt that formed when the supercontinent Rodinia (ca. 1100-800 Ma: TORSVIK 2003) was consolidated. The rocks involved are mainly clastic marine sediments that have been deposited and folded in the mid-Proterozoic (before ca. 1100 Ma). Folded shales, sandstones and quartzites are intruded by large syntectonic batholitic granites. The common grade of metamorphism is very low, but approaching the batholiths rises to high-T/low-P facies. Between 1000 to 900 Ma, a multitude of rather small granites intruded throughout the Kibaran in anticlinorial and anticlinal positions. Many of these bodies parented pegmatites and hydrothermal quartz vein fields mineralised with cassiterite or wolframite. The genesis of these granites may be related to the peculiar heat regime in mantle and crust within supercontinents.

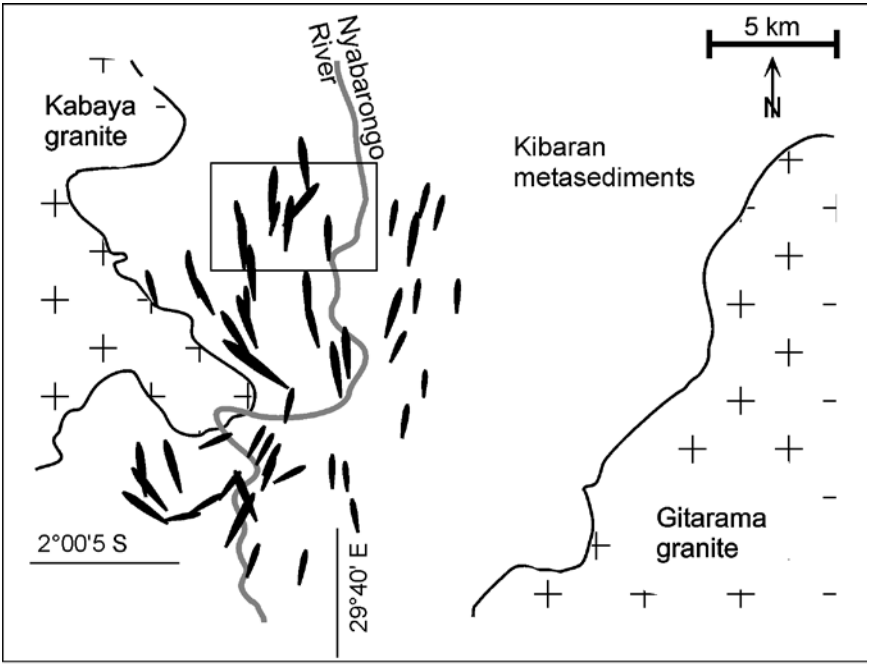

Figure 1 – Geological situation of Gatumba tin and tantalum mining district (modified from ZISERMAN 1982). Black lenses: pegmatites. Frame indicates position of study area (Figs. 2 and 3).

The Gatumba district lies on the western, east-dipping flank of a narrow synclinorium (GÉRARDS 1965) between two large granitic batholiths (Fig. 1). Sericite-quartz schists (with microscopic garnet, biotite and chlorite) are the prevailing rocks. Intercalated bands of quartzites and lenticular doleritic (metabasaltic) sills seem to form less than five percent of the rock mass. Bedding planes and secondary anticlinal folds strike South-North as do the pegmatite dikes. A compressive tectonic phase may have controlled the formation of fractures intruded by pegmatites (Lomamian orogeny, POHL 1994). In the Southwest of the district, the roof of a granite is exposed, with pervasive quartz-fluorite veining. This may be related to the granite at depth that supposedly is the source of the Sn-Ta pegmatites.

To the West of Gatumba, mountains rise to the Congo-Nile water divide at around 3,000 metres above sea level. This is due to uplift during formation of the Central African rift. Water courses in the Gatumba mining district are small to medium-sized creeks that all drain into Nyabarongo river (Fig. 2). In the pilot project study area around Nganzo, drainage is from West to East, falling from about 1650 m to 1420 m above sea level, with an average gradient of ~60 metres/km. In reality, streams exhibit long stretches of low gradient interrupted by short rapids. Alluvial fill of the valleys is only one or two metres thick in the western, upper reaches, but attains an exposed thickness of >6 metres in the middle sections. The lower parts of the valleys are geologically of quite recent age, alluvial sediments are patchy and the creeks often cut through fresh rock.

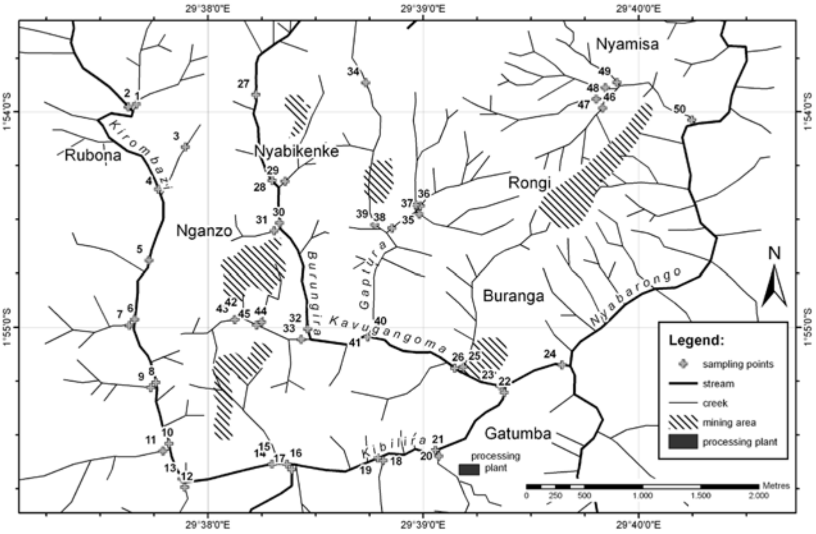

Figure 2 – Stream water and sediment sample sites in northern Gatumba mining district, Rwanda.

Underneath recent gravels, and overlying either bedrock or older alluvium, a bed of organic-rich, plastic black clay occurs along the Kirombasi and Kibilira creeks. This is thought to be a sediment of Pleistocene Nyabarongo lake that developed when the Virunga volcanos dammed the former bed of paleo-Nyabarongo river that earlier flowed into the Kivu-Edward rift basin.

3. Materials and Methods

In April 2007, water and sediment samples were collected for this study from the active bed of streams in the northern Gatumba mining district (Fig. 2). 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 (Institute of Environmental Geology). The analytical methods are verified by regular participation of the laboratory in official quality controls.

Physico-chemical parameters of waters (pH, Eh, T, EC) 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 μm, cellulose-acetate) and acidified with suprapure nitric acid. Main and trace element analyses were performed using ICP-OES and ICP-MS. Fluorine was measured with an Ion Sensitive Electrode (ACME Laboratories, Canada).

Stream sediments were collected in polyethylene bags and refrigerated. In the laboratory, sediments were sieved, dried at 60^{o}C, and then prepared for geochemical and/or mineralogical (XRD) analysis. Most analyses were carried out on the sieve fraction ≤63 µm. Digestion of 0.2 g aliquots was carried out at 120-160°C with hydrofluoric acid. The residue was dissolved in 20 mL 1N nitric acid. 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). Total equal to organic carbon (Corg) was measured using a LECO CS 144 instrument. Tests with hydrochloric acid and X-ray diffractometry did not reveal carbonates in the sediments. 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). All data from the present study are presented on a dry weight basis unless stated otherwise.

4. Results

Water of mountain streams at Gatumba is near-neutral, oxygen-saturated, but very turbid due to soil erosion. Suspended matter was not investigated. A summary of selected chemical properties is presented in Table 1. Our investigations did not cover microbial waterborne disease vectors.

Table 1. Range of variation and averages of parameters measured in stream water samples of northern Gatumba mining district (n = 50), compared to WHO guideline values.

Parameter [µg/L]	Mean	Minimum	Maximum	Standard deviation (sd)	WHO (2006) guideline values
pH [-]	7.2	6.7	7.8	0.23	no guideline
EC [µS/cm]	46.7	28.6	84.9	13.4	no guideline
Fe	525	29.6	3,676	706	no guideline
Mn	103	2.9	552	110	400
Na	3,722	645	7,215	1,667	no guideline
As	0.42	a	3.39	0.62	10
S	1,888	16.4	48,116	6,957	no guideline
Cd	0.03	a	0.15	0.04	3
Cu	2.58	0.46	9.78	1.92	2,000
Pb	1.9	0.02	48.1	7	10
Zn	3.9	0.2	19.2	3.7	no guideline
F	219	140	310	43	1,500
U	0.16	a	1.2	0.2	15

Note: a = concentration below detection limit (As 0.034; Cd 0.015; U 0.003 µg/L)

Active (most recent) sediments of the mountain streams at Gatumba are mainly sand and pebbles; boulders are evidence of occasional flash floods. Silt and clay size sediments are recessive. This mud fraction of stream sediments at Gatumba consists mainly of quartz and muscovite, with variable but smaller contents of kaolinite, illite and plagioclase, as revealed by XRD in quick scan mode. Nearly all sediments collected are brown, obviously due to their origin by erosion of oxidised soil, with very little direct contribution from bedrock. Rounded limonite nodules are common in all grain size fractions. Most are probably derived from lateritic ferricrete or from pisolitic laterite, but some may represent oxidised sulfide pockets in pegmatite. A summary of selected chemical properties of stream sediments is presented in Table 2.

Table 2. Range of variation and averages of parameters measured in stream sediment samples of northern Gatumba mining district (n = 50), compared to crustal abundance and the “Holland List” (MHSPE 2000).

Parameter [ppm or mg/kg]	Mean	Minimum	Maximum	Standard deviation (sd)	Crustal abundance*	Holland List intervention values
pH	6.5	5.6	7.1	0.4	-	ng
C_{org} %	0.6	0.1	2.1	0.4	-	ng
Fe %	3.38	1.7	5.4	1.0	5	ng
Mn	333	a	937	255	900	ng
K %	1.9	0.75	3.55	0.6	2.6	ng
Ca	3,809	666	13,485	2,507	30,000	ng
Mg	4,354	a	17,551	3,511	21,000	ng
As	37.2	14.1	101	21.3	2	55
S	139	33	323	58	500	ng
Cu	9.4	a	88	18.2	60	190
Pb	14.4	5.5	133	17.8	16	530
Zn	38.6	9.16	99.3	18.2	70	720
$U^{\#}$	6.2	3.7	15.6	2.3	3	ng

Note: ng signifies “no guideline”; a signifies “concentration below detection limit”; *consensus figures from SMITH & HUYCK 1999; $U^{\#}$ calculations exclude sample no. 20.

5. Discussion

Stream waters at Gatumba are not contaminated with any dangerous inorganic elements. Measured concentrations are below WHO and European drinking water guidelines (WHO 2006, EU 1998). Note that WHO guidelines describe a quality of water that is acceptable for lifelong consumption without risk to health. It is also of interest that WHO (2006) has removed guideline figures for several substances from earlier versions (eg iron, sulfate). At Gatumba, only manganese contents slightly exceed recommended levels (Table 1). This does not pose a health hazard, however, and is probably an artefact of sample transfer from the field to the laboratory. In spite of precautions, the stored water may have suffered lowering of Eh by consumption of dissolved oxygen.

Stream sediments generally have relatively low contents of most elements that may be derived from pegmatites. Because much of the stream sediment mass must be derived from soil and ultimately from the sericite schists or metabasalts we propose to discuss first the maximum values (Table 2). This may help to identify possibly mine-derived elements:
Both iron and manganese reach contents just above the crustal average, most probably inherited from metabasalt. Maximum potassium contents are clearly higher in stream sediments than its crustal average. The abundance of pegmatite-derived muscovite in the stream sediments easily explains this observation. Calcium and magnesium contents remain below the crustal average. Neither element can be derived from the pegmatites that are enriched in the alkalis potassium (in microcline and muscovite) and sodium (in albite). Calcium and magnesium are most certainly provided by erosion of soil that formed above metabasalt. The same source – the metabasalts – appears to be responsible for copper, lead and zinc maxima exceeding the respective crustal abundance. It is unfortunate that the extension of metabasalts beneath the soil cover at Gatumba is little known so that anomalies cannot be confidently tied to these mafic rocks.

Uranium is interesting because its mean content in stream sediments is twice, and its maximum (apart from sample no. 20) is about five times the crustal average; note, however, that the maximum value of 31.5 ppm is singular in sample no. 20 that was collected from the little creek passing by the processing plant (Table 2, Fig. 2). Recommended Upper Crustal values (Rudnik & Gao 2003) of 2.7±0.6 ppm confirm that even minimum uranium contents in stream sediments at Gatumba are above its crustal average. Obviously, there is a generally elevated background, and the location of higher contents is controlled by the proximity to mining areas (not illustrated). However, the precise source of uranium in Gatumba stream sediments remains vague. The anomalous sample taken near the ore processing plant implies that one or more minerals in the tailings (and by implication in the pegmatites) are uraniferous. Columbite, for example, is known to contain up to 1000 ppm uranium. However, columbites are remarkably resistant hosts to uranium and its daughter products, and Ta_2O_6-grades in pegmatites generally are only about 400 g/t. Low uranium contents in Gatumba stream waters (Table 1) seem to be inconsistent with high contents in stream sediments. We suggest that uranium (VI) sorbed to amorphous iron hydroxides and other soil-derived colloidal suspended particles may explain this disparity. Natural soils with 0.8 to 11 ppm uranium are generally considered non-hazardous for agricultural use. Legal guideline limits for uranium in soil are not available.

Arsenic is the second element that at Gatumba clearly exceeds its average crustal abundance (Table 2) estimated at about 2 ppm (range 1.7 to 5) similar to contents in average granite (1.5) and basalt (2 ppm) (Smith & Huyck 1999). In the Upper Crust, 4.8±0.5 ppm is the recommended value of average arsenic content (Rudnik & Gao 2003). Sediments contain more arsenic than magmatic rocks, with highest contents in pelites (~13 ppm). In that case, organic matter contents are the main control on arsenic fixation. As arsenic was not measured in earlier national geochemical surveys, the distribution of arsenic in the Earth's surficial materials is poorly known (Plant et al. 2005).

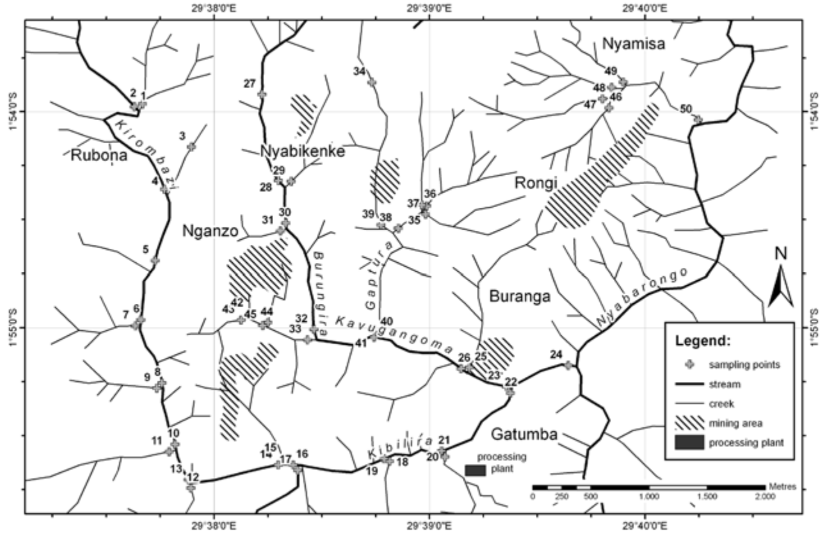

Figure 3 – Arsenic contents in active stream sediments of northern Gatumba mining district, Rwanda.

Arsenic at Gatumba is generally enriched above normal background values. Its mean content in stream sediments is nearly twenty times, and its maximum is about fifty times the crustal average of SMITH & HUYCK (1999). Similar to uranium, higher contents are located near former mining sites (Fig. 3), but contrary to uranium, the maximum was not found adjacent to the processing plant but in drainage within the northeast corner of the study area. In a cumulative frequency plot, arsenic contents form a nearly straight line, indicating that there is only one population of data (Fig. 4). This is interpreted as the result of a general and widespread hydrothermal arsenic halo surrounding the pegmatite swarm at Gatumba, probably modified by secondary dispersion due to weathering. The origin of the data set starts at about 12 ppm As that might be the background value of the quartz sericite country rocks. The near-absence of organic matter in these rocks, however, suggests that the schists should have a much lower As-content.

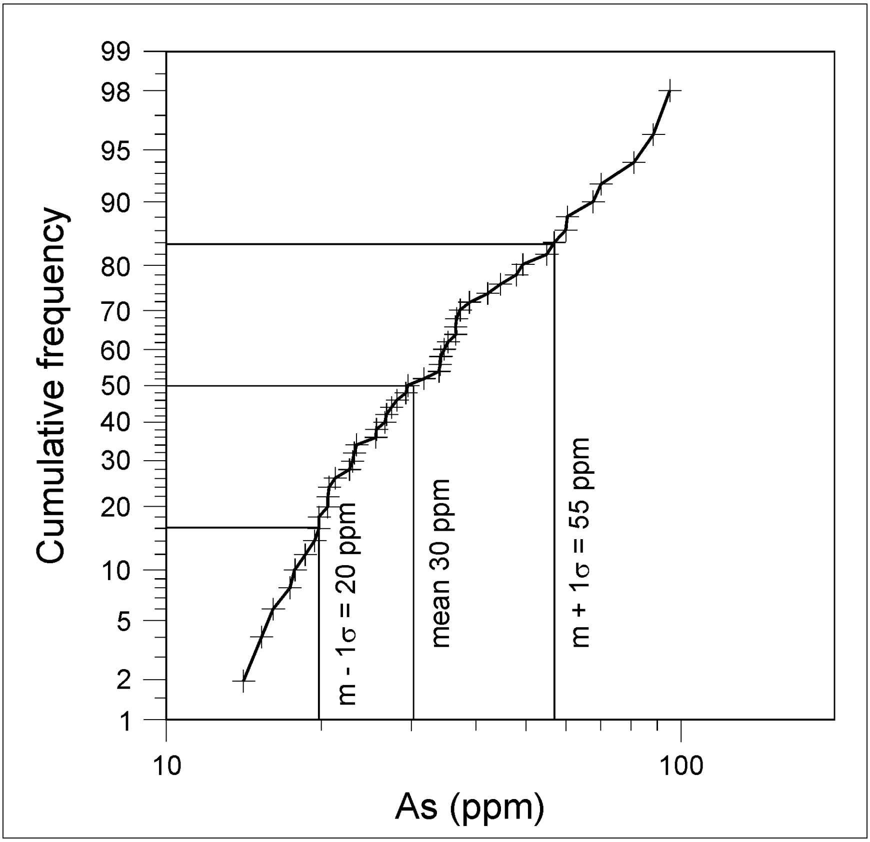

Figure 4 – Cumulative frequency plot of arsenic in stream sediments of northern Gatumba mining district, Rwanda.

Arsenic is geochemically peculiar as it may form both anions or cations. It occurs at oxidation levels -3, 0, +3 and +5. Its most common mineral is arsenopyrite (FeAsS). Weathering of arsenian iron sulfides produces intermediate products realgar and orpiment, and arsenates including the greenish scorodite ($FeAsO_4.2H_2O$). In the end, arsenic is moderately mobile in oxidic water as As^{+5}, but also in strongly reduced groundwater as As^{+3}. Mine-derived waters are often contaminated even when the ore has only traces of arsenic (ZHU ET AL. 2003). Mobile arsenic tends to be immobilised and geochemically enriched in iron oxides, in siderite concretions hosted by claystone, in phosphorites and black shales. Actually, arsenic is hardly mobile to immobile under oxidising conditions in the presence of abundant iron-rich particulates and at pH>5 to circum-neutral. In that case, arsenic-mobility is not in solution (compare As-contents in Gatumba water, Table 1) but in eroded particulates (SMITH & HUYCK 1999).

Trivalent arsenic is more toxic than other forms. Doses of more than 100 mg of arsenic will cause acute poisoning, while chronic exposure impairs health and increases the risk of cancer. The main path of arsenic exposure of humans is drinking water. Plants cultivated on contaminated soils are less frequently a source of critical intake. Therefore, acceptable maximum levels in drinking water are generally set at 10 µg per litre (eg WHO 2006). The intervention level for soil in residential areas is 55 mg/kg dry mass ("Holland List", MHSPE 2000) and the inspection value for agricultural land 200 mg/kg (BMJ 1999). In small concentrations, however, arsenic is an essential element (OREMLAND & STOLZ 2003). Healthy humans contain about 18 mg As, that must be continually replaced because of the short biological half life (10-30 h) of the element in the body (LINDH 2005).

Unrelated to mining, natural arsenic concentrations in aquifers and/or groundwater of Holocene lowland valleys are a hidden hazard. In the Ganges delta this caused Humanity's biggest mass poisoning (NORDSTROM 2002, MEHARG 2005, SMEDLEY & KINNIBURGH 2005). Earlier, millions of Bangladeshi people had relied on surface water, but from 1980 to 1990 thousands of drinking water wells were drilled to provide "safe", that is microbially clean water. Of these, nearly 50% had As>10 micrograms/litre. Sadly, this was only recognized after numerous people had developed skin and internal disorders, including cancer. With 2-20 ppm, As-contents in the Ganges river sediments are quite ordinary. The reason for spots of elevated dissolved arsenic (III) seems to be locally strong reduction of the groundwater (SMEDLEY & KINNIBURGH 2005).

The precise source of arsenic in Gatumba stream sediments remains unknown. Considering the relatively high mobility of arsenic in hydrothermal systems at small sulfur activities, the higher concentrations in the vicinity of mining areas (Fig. 2) may well be due to primary dispersion from the pegmatites into country rock schists. Weathering-induced secondary dispersion in soil is affirmed by Reetsch (in preparation). Most probably, arsenic in the streams is derived from a variable combination from i) arsenopyrite exposed in mines, ii) the natural primary geochemical halo of mineralisation, and iii) natural secondary weathering dispersion in soil. Conceivable vectors are erosion of soil and of weathered rock, and interflow water in permeable soil horizons. Groundwater flow in hard rock beneath the regolith is very limited because of low permeability. Present mobility of arsenic is probably more enhanced by deforestation, intensive agriculture and consequent disturbance and erosion of soil, than by mining that ended 30 years ago.

6. Conclusions and Recommendations

Stream waters at Gatumba are not contaminated with dangerous inorganic elements. According to WHO guidelines these waters are acceptable for lifelong consumption without risk to health. However, suspended matter should be removed before intake. Fluorine contents are too low, as healthy drinking water should contain between 0.7 and 1.5 mg/l F (EDMUNDS & SMEDLEY 2005). Microbial waterborne disease vectors were not investigated.

Stream sediments at Gatumba have relatively low contents of most deleterious elements that might be derived from the tin-tantalum pegmatites, including lead, zinc, cadmium, and sulfur. An exception are uranium and arsenic, with quite elevated and anomalous contents. For both elements, the pegmatites and a primary hydrothermal geochemical halo are the primary source, but later dispersion by supergene alteration and recently, by agriculture is probably the main factor of present mobilisation. The extent of the contribution by former mining of tin and tantalum cannot be quantified. Data indicate that the pegmatite swarm is surrounded by a wide halo of elevated arsenic contents.

However, the precise nature of arsenic and uranium sources at Gatumba have yet to be determined. The pathways between sources and streams should be investigated. The adsorption partners, and speciation of arsenic and uranium in the river sediments wait to be ascertained. Samples of older (deeper) parts of alluvial sediments and of deep groundwater in the valleys should be analysed for arsenic, pH, Eh, and H_2S, in order to avoid a repetition of the tragic errors made in Bagladesh. A program of water testing should be carried out for all water drawing points dispersed in the hills, as it is possible that some drain water from arseniferous regolith or soil material. Possible pathways of arsenic from river sediment to food plants, animals and humans should be studied. Considering the elevated uranium contents, an exploratory survey of radioactivity (APPLETON 2005) at Gatumba is advisable, especially around the processing plant.

Overall, our work suggests that past mining at Gatumba caused no widespread dispersion of deleterious elements. The cause for elevated arsenic and uranium contents in soil and stream sediments are probably natural processes. Gatumba district is essentially a large geogenic geochemical anomaly as are many mineralised areas (POHL 2005). The welfare of the local population is not threatened. Yet, human interference with soil (by agriculture), water (by abstracting drinking water from valley fill) and bedrock (by irresponsible mining) may be hazardous. For a full understanding of the present environmental state of the mining district and possible hazards, more detailed investigations are recommended.

7. Acknowledgements

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