Mercury contamination associated with small-scale gold mining in Tanzania and Zimbabwe

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Abstract

Mercury contamination associated with small-scale gold mining and processing represents a major environmental and human health concern in Eastern and Southern Africa. Approximately 200,000–300,000 persons are involved in small-scale gold mining activities in Tanzania and > 200,000 persons in Zimbabwe. Mercury (Hg) is used mainly for the processing of primary gold quartz veins and supergene gold mineralizations. Gravimetric material flow analyses show that 70–80% of the Hg is lost to the atmosphere during processing, 20–30% are lost to tailings, soils, stream sediments and water. For every 1 g Au produced, 1.2–1.5 g Hg are lost to the environment. Cumulatively, the anthropogenic Hg released annually into the atmosphere is approximately 3–4 t in the whole Lake Victoria Goldfields of Tanzania and > 3 t in Zimbabwe. Tailings are local 'hot spots' with high concentrations of As, Pb, Cu and Hg. Lateral and vertical dispersion of Hg lost to soils and stream sediments is very limited (laterally < 260 m, vertically < 20 cm). Dispersion of mercury from tailings is low because Hg is transported largely in the elemental, metallic form. In addition, Fe-oxide rich laterites and swamps appear to be natural barriers for the dispersion of metals in soils and streams. Ground and surface water quality data indicate very low dispersion rates during the dry season.

Keywords: Mercury; Gold mining; Tanzania; Zimbabwe; Soil; Tailings; Water; Stream sediments; Fe-oxides; Wetlands

1. Introduction

Mercury contamination associated with small scale gold mining and processing represents a major environmental and human health concern in many parts of the world. Many scientific data on the effects of using Hg to process gold have been published, mainly from South America (Malm et al., 1990, 1995; Pfeiffer et al., 1991, 1993; Bloom and Porcella, 1994; Cleary et al., 1994; Hacon et al., 1995; Veiga et al., 1994; Hacox, 1995; Veiga and Meech, 1995; Lacerta and Marins, 1997; Lacerta and Salomons, 1998; Roulet et al., 1998). In contrast, only very
few published data are available from Africa (Ikingura and Akagi, 1996; Kahatano and Mnali, 1997; Landner, 1997; Maponga, 1997; Ikingura et al., in press).

Small-scale gold mining activities are mainly manual, low tech, subsistence activities which employ large numbers of people. In Tanzania, this kind of mining employs approximately 200,000–300,000 largely unskilled persons contributing to poverty reduction in rural areas. The small-scale miners manually extract gold from rich and near-surface deposits, but in some places also from primary ores below the thick oxidized zone > 30 m. Mining of the primary gold quartz veins below this depth requires more sophisticated techniques and equipment.

Potentially, small-scale gold mining can contribute significantly to the economic development of a country. In Tanzania, gold production from the small-scale mining sector provided some 76% of Tanzania’s total mineral export in 1992. During 1992 small-scale miners sold 4525 kg gold worth US$40.4 million to the Bank of Tanzania.

Zimbabwe is a mining country with approximately 200 registered ‘formal’ medium-scale gold mines and thousands of small- to ‘micro-scale’ gold operations with approximately 200,000 ‘informal’ miners using similar techniques as in Tanzania. No official gold statistics exist for this sector in Zimbabwe, but Maponga (1997) estimated that up to 5 t Au are produced annually by small-scale miners. Official gold buying practices are largely ineffective, and most gold is smuggled out of the country.

The present study aimed at determining the extent of Hg contamination related to small-scale gold mining in selected areas of Tanzania and Zimbabwe, and to quantify the losses of anthropogenic Hg to air, soil, tailings and water in the study areas.

2. Materials and methods

2.1. Study and sampling areas

For the environmental studies, we selected sites in small-scale gold mining areas of the Lake Victoria Goldfields of northern Tanzania and in the Tafuna Hill area, near Shamva, Zimbabwe (Fig. 1). Geologically, both study areas are located in Precambrian granite/greenstone terrains: the Lake Victoria Goldfields in Tanzania (van Straaten, 1983; Barth, 1990; Borg et al., 1990; Gabert, 1990), and the Tafuna Hill deposits in the greenstone belt of the Bulawayan Group in Zimbabwe (Stidolph, 1977). In both areas Au is extracted from primary quartz veins and supergene enriched zones in the weathering environment. The small-scale miners use Hg to process the gold ore.

In the cause of general environmental impact assessments in Tanzania and Zimbabwe in 1997 and 1998 we measured losses of toxic metals by gravimetric mass balances studies, quantifying every material step during mining and processing (materials added, lost or recycled) with mechanical and electronic balances. This approach assisted in separating facts from ‘myths and preconceptions’ (Veiga et al., 1999) and provided the basis for practical interventions.

In soils, we studied the lateral and vertical dispersion of As, Cd, Cr, Cu, Pb, Zn and Hg at four small-scale gold mining camps in northern Tanzania, namely Mwakitolo, Katente, Kalole and Nyakafuru (Fig. 1). At Mwakitolo, the processing site is located in an area underlain by a thick laterite crust. The site lies close to the wide basin of the Isanga River which flows in a northerly direction into Smith Sound and Lake Victoria (Fig. 1). During the rainy season the processing site is located at the edge of the flooded Isanga river and there is a risk that the processing wastes are washed directly into Lake Victoria. At Mwakitolo, we collected soil samples along two transects, one along a 260-m-long N–S transect (three pits) in a catena starting from the Au–Hg processing site continuing down-gradient to the vertisols of the Isanga plains. The other transect (two pits) was perpendicular to the first transect. Other soil profiles were dug at the Kalole mining area, at Katente and Nyakafuru, located on gently undulating plains where soils are underlain by Fe-oxide-rich laterite within 1 m from the surface.
We analyzed 28 soil samples for pH, organic carbon, cation exchange capacity, P, as well as for contents of As, Cd, Cr, Cu, Pb, Zn and Hg.

Stream sediment samples were collected in two areas, one in Tanzania along the Bulyanhulu drainage system, and one in Zimbabwe in the Shamva area, with streams draining from various gold-mining centers, including Tafuna Hill. The Bulyanhulu river system represents a drainage system heavily affected by small-scale gold mining activities. Here, stream sediment and water samples were collected from 19 sampling sites along a 38-km-long stretch of the river system with sampling intervals of 1 km for the first 7 km downstream from the processing areas, and 5 km thereafter in the Igonzela swamp area until reaching Smith Sound, the southern extension of Lake Victoria (Fig. 1).

In Tanzania, we surveyed drinking water sources and domestic wells, processing water at mine sites, surface water and lake waters (n = 100). The water survey in Zimbabwe (n = 17) was restricted to ground water samples from Tafuna Hill and surface river waters along the Pote and Mazowe river systems for a distance of approximately 20 km downstream from Tafuna Hill. The surveys were undertaken during the dry season in 1997 with a few repeat samples collected during the start of the rainy season of 1998.

2.2. Sampling methods

Bulk samples from tailings and soil samples were collected manually, disintegrated with a clean glass rod, stored and sealed in clean polypropylene vials, and kept refrigerated until analysis at the Geoscience Laboratory, Sudbury, Canada. Stream sediment samples along contaminated rivers and streams were collected by advancing a 50-ml, polypropylene sample container into the silt-size river sediments. No preservatives were added to the samples. The samples were immediately placed into a cooler following sample collection, transferred to a refrigerator and shipped to Canada for analysis. Water samples were collected in sterile 50-ml polypropylene...
sample tubes. Two drops of ultrapure nitric acid (Grade AA) were added in the field for sample preservation. The samples were stored in coolers, immediately transferred to refrigerators and transported to the laboratory in Canada where they were analyzed within 1 week of arrival. Water samples were analyzed after filtration through a 0.45-μm filter by ICP-MS and replicate samples checked by cold vapor atomic fluorescence spectrometry using a PS analytical hydride/vapor generator coupled to a PSA Merlin detector (Chan and Bina, 1989). Soil, stream sediment and tailings samples were digested with aqua regia as received. Sub-samples were dried at 105°C to obtain moisture contents. Moist sub-samples (0.5 g) were weighed into dry 50-ml Nalgene tubes, pre-digested for 4 h with 3.0 ml HNO₃ at 22°C. On addition of 3 ml HCl the tubes were lightly sealed, transferred to a digestion block at 120°C for 2 h, cooled and brought to 50 ml with de-ionized water. All samples were analyzed by ICP-AES (Jarrel-Ash 61E) for major elements and by ICP-MS (Sciex-Perkin-Elmer ELAN-5000) for trace elements.

### 3. Results

The mass balance studies in Tanzania and Zimbabwe showed that most of the losses occur during Au–Hg amalgam roasting to remove excess Hg. The mass balance flow analysis (Table 1) illustrates that 70–80% of the Hg released to the environment is lost to the atmosphere through active amalgam roasting at the mine site and in goldsmiths' shops in towns. Between 20 and 30% are lost to tailings, soils and water. The amount of Hg released from the tailings by degassing is not known. The mass balance calculations showed that the ratio of Au extracted to Hg lost was in the range of 1:1.2–1.5 in both study areas.

The fate and pathways of Hg in the vapor phase could not be established in these studies. Some of the Hg emitted during amalgam roasting in houses or in goldsmith's shops is deposited on surfaces close to the site of emission, usually at the mudwall next to the fire place or on soot in the roof of the hut. A sample from the mudwall of a hut at Katente/Tanzania in which amalgam was roasted contained 324 mg Hg/kg, the soot of

<table>
<thead>
<tr>
<th>Process</th>
<th>Losses</th>
<th>Results</th>
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<tr>
<td>Extraction (mining)</td>
<td>100 kg ore</td>
<td></td>
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<tr>
<td>Crushing, grinding</td>
<td>−1.8 kg (dust/spillage)</td>
<td>98.2 kg ground ore</td>
</tr>
<tr>
<td>Gravity separation</td>
<td>−94.9 kg to tailings</td>
<td>3.3 kg concentrate</td>
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<tr>
<td>Amalgamation (in concentrate)</td>
<td>addition of 64.40 g Hg</td>
<td></td>
</tr>
<tr>
<td></td>
<td>recovery of 61.26 g Hg</td>
<td></td>
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<tr>
<td></td>
<td>total loss = 3.14 g Hg</td>
<td></td>
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<tr>
<td></td>
<td>−3.29 kg to tailings</td>
<td>4.47 g amalgam</td>
</tr>
<tr>
<td>Amalgam roasting</td>
<td>−2.08 g Hg lost as vapor (66%)</td>
<td>2.39 g raw gold</td>
</tr>
<tr>
<td>Refining, smelting of gold</td>
<td>−0.24 g Hg lost as vapor (7%)</td>
<td>2.15 g refined gold</td>
</tr>
</tbody>
</table>

In total: 
3.14 g Hg lost for 2.15 g Au won

1 Au: 1.46 Hg
the roof of a house at Tafuna Hill/Zimbabwe contained 104 mg Hg/kg, the soot in a goldsmith shop in Shinyanga/Tanzania analyzed 62 mg Hg/kg.

The small-scale gold miners in Tanzania and Zimbabwe produce two types of tailings: the ‘light’ fraction tailings discarded after gravity separation, and the concentrate tailings. The light fraction tailings commonly contain some metal concentrations from inefficient gravity separation. However, it usually contains no Hg as this has not been introduced in the process as yet. The concentrate tailings from which the miners removed the gold by amalgamation techniques is a heavy mineral concentrate and contains large amounts of metals including the introduced Hg. Unfortunately, the miners commonly combine the two tailings types into one pile where concentrations up to 1414 mg Hg/kg, 3350 mg As/kg, and up to 256 mg Pb/kg were measured. These tailings are not only ‘hot spots’ of contamination but often contain high concentrations of gold as well, either as free gold from poor processing techniques, or as refractory gold. The possible economic recovery of Au through reworking of the tailings should be combined with cleaning up the waste heaps from mercury and other toxic elements.

The results of the chemical analyses of soils from the Mwakitolyo profiles in Tanzania indicate high levels of As and Hg in the upper 10 cm at the gold processing site [88 mg As/kg, and 2.495 mg Hg/kg, in comparison to natural levels in soils, which are 11.3 mg/kg for As and 0.098 mg/kg for Hg (Ure and Berrow, 1982)]. The other heavy elements (Cd, Cr, Cu, Pb, Zn) measured in the profiles show background and not greatly varying concentrations. At a site, some 80 m down gradient from the processing area, where tailings material was probably deposited onto the soils from the site above by sheet wash both As and Hg values were also very high: 168.4 mg As/kg and 0.638 mg Hg/kg, respectively. Vertical distribution of As and Hg concentrations show a rapid decrease with depth. At 30 cm the As concentration was 6.5 mg/kg, Hg was 0.012 mg/kg. Other metals concentrations in soils are low: Cd, Cr, Cu, Pb and Zn concentrations are 0.1, 19–21, 15–23, 8–11, 24–35 mg/kg, respectively. In the third profile, located 260 m downgradient from the processing site, the metal concentrations are low, likely presenting background levels. The composite top soil sample from cultivated farmland < 50 m from the processing site contains very high concentration of As (269.2 mg/kg), and Hg (1.56 mg/kg).

The soil analyses of samples from two profiles at the mining village of Katente, are summarized in Fig. 2 and show that the top 12 and 20 cm, respectively, of top soils have slightly elevated Hg concentrations, the concentration of other elements are at background levels.

Results from the stream sediments and water surveys in the Tanzanian study area showed elevated concentrations of metals including Hg, Bi, As as well as Cu and Au in stream sediments only in the upper reaches of the river, specifically in the area around the processing sites of small-scale gold miners. Stream sediments in this area also contained considerable amounts of Au, a result of incomplete gold extraction and processing by small-scale gold miners. Stream sediments and water samples collected past the Igonzela swamp were generally low (Fig. 3 and Table 2). The dispersion of Hg is characterized by a marked reduction of Hg concentrations in sediments at the Igonzela swamp. Over a distance of 3 km the Hg concentrations in stream sediments dropped from 5.35 mg/kg to 0.17 mg/kg and further downstream to 0.02–0.01 mg/kg. The elevated levels of Cu (up to 1883 mg/kg), and Bi (> 17 mg/kg) in the stream sediments and water
Table 2
Stream sediment and water analyses from the Bulyanhulu River system

<table>
<thead>
<tr>
<th></th>
<th>Hg</th>
<th>Cu</th>
<th>Bi</th>
<th>As</th>
<th>Au</th>
</tr>
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<tbody>
<tr>
<td>Stream sediments up-gradient of the Igonzela swamp, near gold mining area ( (n = 5) ) (mg/kg)</td>
<td>0.17–5.35</td>
<td>80.1–1883.5</td>
<td>1.29–17.4</td>
<td>15–59</td>
<td>2.05–32.81</td>
</tr>
<tr>
<td>Stream sediments down-gradient of the Igonzela swamp ( (n = 5) ) (mg/kg)</td>
<td>0.01–0.02</td>
<td>6.7–11.4</td>
<td>0.11–0.31</td>
<td>0.07–0.8</td>
<td>n.d.</td>
</tr>
<tr>
<td>Water samples up-gradient of the Igonzela swamp, near mining area ( (n = 5) ) (μg/kg)</td>
<td>n.d.–0.4</td>
<td>n.d.–890.1</td>
<td>0.2–1.2</td>
<td>9.8–51.1</td>
<td>n.d.</td>
</tr>
<tr>
<td>Water samples down-gradient of the Igonzela swamp ( (n = 5) ) (μg/kg)</td>
<td>n.d.–0.1</td>
<td>5.5–41</td>
<td>n.d.</td>
<td>1.2–15.3</td>
<td>n.d.</td>
</tr>
</tbody>
</table>

Around Bulyanhulu are probably due to the inherited metal content in the extracted ore. For example, the high Cu concentrations in stream sediments close to the small scale mining area of Bulyanhulu are most likely related to Cu-sulphides found in the ore. Similar to the other metals, there is a steep drop in the Cu concentrations in stream sediments in the downstream area of the Igonzela swamp from 1514 mg/kg to 22 mg/kg over a distance of 5 km.

In the Zimbabwean study area the streams draining Tafuna Hill had elevated Hg concentrations (up to 0.7 mg Hg/kg) at the foot of the hill (<2 km from the source). At a distance of <4 km from the processing sites Hg concentrations are only 0.1–0.2 mg Hg/kg. Arsenic concentrations in stream sediments were elevated in the whole area around Shamva, with highest concentrations reported at the foot of Tafuna Hill (124–404 mg As/kg). Stream sediments from the Mazowe River, several kilometers downstream from various small- and large-scale gold mining operations, contained 26.2 mg As/kg.

Results of the water surveys in the Tanzanian and Zimbabwean study areas showed elevated metal concentrations only in areas of present and past small-scale gold mining activities, but drinking water samples were not contaminated.

In the Zimbabwean study area the Hg concentrations in rivers are in general 0.01–0.02 μg/l. Close to the gold processing sites at Tafuna Hill the Hg concentrations range from 0.02 to 0.65 μg/l. Only one sample (process water at amalgamation site at Tafuna Hill) had extremely high

Fig. 3. Mercury, arsenic and copper concentrations in stream sediments of the Bulyanhulu drainage system, northern Tanzania.
Hg concentrations (up to 2.33 μg/l). The mercury data in water obtained in this study are higher than background concentrations commonly described, possibly because of the presence of Hg adsorbed to very fine colloidal particles and dissolved organic material which passed the 0.45-μm membrane filter. However, the fact that selected results for the filtered samples were replicated by different methods does confirm the data accuracy for the samples as analyzed. In general, the dispersion of Hg in rivers draining from the mining sites is very low.

4. Discussion

The presented data show that Hg losses during amalgamation and Au–Hg roasting are high. The materials flow analysis illustrating emissions and losses to the environment especially to the atmosphere are similar to the ones recorded by Pfeiffer and Lacerda (1988). The recovered Au to Hg lost ratio is similar to that of other gold mining areas in the world, especially South America (Lacerda and Salomons, 1998). This ratio and the amount of gold produced using amalgamation techniques form the basis of the calculations of Hg losses. Detailed gravimetric measurements specify the compartments into which the Hg is lost, the atmosphere being the largest sink. The findings from the two surveys in Tanzania and Zimbabwe are significant for estimating global mercury losses from gold mining. The amount of Hg lost to the atmosphere in the study areas is considerably lower than the mean emissions of 78 t Hg/year from the more extensive gold mining activities in Brazil estimated by Lacerda and Marins (1997) reflecting lower total gold production from these areas.

In many soils, elevated concentrations of Hg and As were found only in the immediate surrounding of gold processing sites. The elevated Hg concentrations in the soils are probably a result of Hg physically lost by the small-scale miners during the amalgamation process. Elevated As concentrations in the soils are most probably related to As content of the inherent mineralogy of the ores which contain the mineral arsenopyrite (FeAsS₂). These results of high As and Hg concentration is soils close to amalgamation sites are consistent with findings in South America (Lacerda and Salomons, 1998).

In the study areas, the amount of Hg redeposited in soils and close by vegetation through short-range atmospheric transport is not known. Indications of Hg contamination from short-range volatile transport are the high Hg concentrations found in walls and roof portions of houses in which amalgam was roasted. In Brazil, a considerable amounts of Hg lost to the atmosphere are deposited in topsoil and vegetation very close to the sites of emission, specifically Au processing centers and goldsmith's shops in urban areas (Lacerda and Salomons, 1998). Mass balance studies by Roulet et al. (in press) and data presented by Lacerda and Salomons (1998) and Pfeiffer et al. (1993), show that gold mining and processing activities can result in severe direct Hg contamination of aquatic ecosystems and humans at the local scale.

The source–sink analyses of Hg in the soil studies in Tanzania suggest that the lateral and vertical dispersion of Hg is very limited and that the likely causes for the low apparent Hg mobility are related to the high adsorption capacities of Fe-oxy-hydroxides and soil organic matter. The strong association of Hg with Fe and Al oxy-hydroxides in tropical lateritic soils has been reported by many researchers (Semu et al., 1986; Roulet et al., 1998; Fostier et al., 1999; Ribeiro et al., 1999; Roulet and Lucotte, 1999). Roulet et al. (1998) and Roulet and Lucotte (1999) showed that Hg is preferentially accumulated in Fe–Al oxy-hydroxide rich oxisols in Brazil. Ribeiro et al. (1999) report a significant correlation between Fe complexes and Hg in sediments of the Alto Floresta region (Brazil). In a study on Hg contamination related to Hg mine tailings in California, Rytuba (1999) describes the effective adsorption of Hg on Fe-hydroxides over a pH range 2.2–7.8. Fostier et al. (1999) describe the ferruginous nodules in the oxidized zone of a soil profile in Brazil as ‘efficient traps for Hg’. Veiga and Meech (1995) propose to use lateritic soils as mitigation material for contaminated tailings.
The stream sediment survey in northern Tanzania, conducted in the dry season, showed that the dispersion of metals is characterized by a steep gradient from the metal contaminated source and that the Igonzela swamp along the Bulyanhulu river system forms — at present — a natural barrier, preventing heavy metals derived from mining activities to disperse into Lake Victoria. In Zimbabwe, the dispersion of metals along the Poti and Mazowe river system is also localized with very short dispersion patterns of metals along these drainage systems.

Most of the current gold mining and processing camps are located away from river or stream beds which flow mainly during the rainy season. Stream sediments from these intermittent streams showed only background concentrations of Hg, Cu, Pb, As indicating very low dispersion rates of Hg and other metals from these mining and processing sites.

There are several reasons for the apparent relative immobility of heavy metals in stream sediments and water. The reason for the sharp drop of Hg concentrations seems to be the fact that mercury from processing sites and tailings are likely to occur in the metallic form, as described by Lacerda et al. (1991). Another reason for the strong gradient of metal concentration in stream sediments seems to be the presence of large amounts of strongly metal sorbing iron and aluminum laterite crusts as well as the presence of large amounts of metal complexing organic matter.

All drinking water sources in the Shinyanga study area were within drinking water standards, even those water sources spatially very close to contaminated gold processing sites. At the point sources (processing sites, tailings) mercury is lost mainly as elementary metallic mercury with a very low dispersability and consequently only small amounts of Hg are mobilized into solution. These data are similar to the ones described by Lacerda et al. (1991). In addition almost all of the studied water wells were located in laterites, a medium which is known to have very high sorption capacities for heavy metals.

5. Conclusions

In the small-scale gold mining study areas of Tanzania and Zimbabwe the extent of Hg contamination in soils and stream sediments is localized, restricted to the immediate surrounding of gold processing sites only. Tailings are ‘hot spots’ with very high metal concentrations, but the Hg dispersion (horizontally and vertically) in soils is low, possibly due to the presence of Fe-oxyhydroxides and organic matter and due to the fact that mercury occurs mainly as metallic inorganic mercury. The Hg in filtered water samples of the study areas are within WHO guidelines. Mercury losses to the atmosphere are high, approximately 70–80% of Hg are released to the atmosphere, but fate and pathways of Hg in the atmosphere are so far unknown.

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