Investigation of trace metal concentrations in soil, sediments and waters in the vicinity of “Geita Gold Mine” and “North Mara Gold Mine” in North West Tanzania

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Investigation of trace metal concentrations in soil, sediments and waters in the vicinity of gold mines in North West Tanzania

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Abstract

The “Geita Gold Mine” (GGM) and “North Mara Gold Mines” (NMGM) have developed large quarries in the middle of settlements in fertile agricultural lands. Possible hazardous impact on the natural recourse have impelled a pilot study on the trace element contents in soils, sediments and natural waters carried out in June 2009. The need to follow up these studies is obvious as several sites have accumulated potentially hazardous contents of trace elements. The previous accidental spill that took place from the NMGM in May 2009 has seriously contaminated sediments and waters nearby. However, other sites more remote from this site are affected, as particularly the contents of Arsenic, As, was found to be unacceptably high in both sediments and waters. Although the contamination situation was less severe in the area near GGM, also findings from this place show that As contents in sediments is worrying. Detailed biogeochemical studies at both places are highly recommended. Along with such studies, there is a pressing need for an extensive study of the population, their diet and agricultural management practices, to map particularly the As sources, its transport in the food chain and finally its content in human tissue samples of different groups of the populations.

Introduction

In Tanzania large scale mining started in the 1998. From that time on Tanzania has witnessed an ever expanding sector with its concomitant increase in its contribution to the economy. At the moment Tanzania is third behind South Africa and Ghana. There are three Goldfields in Tanzania the most prominent being Lake Victoria that is spreads in Tabora, Shinyanga, Mwanza and Kagera regions. This goldfield produce s about 90% of all gold that is produced in Tanzania. Geita Gold mine GGM and North Mara Gold Mine (NMGM) fall in this Goldfield. Although gold mine seems to be one of the major contributors to foreign earnings, this is not without problems. There have been complains that both Geita Gold Mine and North Mara Gold Mine at some points discharged wastes into nearby water sources in their areas of operation A study carried out in Nyakabale, a village adjacent to one of the waste rock damps in Geita reported levels of heavy metals that were higher than those accepted by WHO (Bitala 2008). In 2009 a spill from a tailing dam and leachate from was reported in the media to have occurred around North Mara Gold mine. A small study that followed this event assessed concentration of trace metals and Cyanide in water and soils and sediments. Results obtained indicated that in some places levels of trace metals were higher that what is permissible by WHO, US EPA and Tanzania government standards (Bitala et al., 2009). The concern about trace elements is that they pose both environmental and health consequences problems. It is on this basis that WHO, and governments have set up critical limits loads of trace metals that should
not be exceeded. Parallel to those investigations, frequent reports with uneven quality of
documentations has brought forward strong indications of severe human sufferings of different
characters and seriousness. These findings are of great concern, and flowingly this particular study
was initiated as a pilot project to investigate the contents of trace elements (metals and semimetals)
and cyanide in terrestrial and aquatic environments around the two named mining projects. This
study is planned to be followed up by a broad study of human and animal health.

The main objective for the project was to determine the total concentrations of trace metals in
sediments, soils and drinking waters. Secondly, total cyanide was to be determined in selected water
and sediment samples. Environmental samples were planned withdrawn from sites near the residual
waste tailings and from sites at increasing distance from the installations, to investigate possible
regional impact from large scale mining. Samples were also taken from land and water sites of
different management practises.

Materials and Methods

Soils, sediment and water sampling and metal analysis:
An extensive storage of discharged cyanide in large dams, and the storage of residual waste material
in open-air rock piles, is placed near villages and other settlements. Hence, sampling was carried out
at increasing distances from these installations. Samples were withdrawn from rivers, ponds and
from cultivated lands. Sampling was also carried out at remote areas relative to the mining sites as
control. The sites are shown in the Google-earth images shown in figures 1-4 and the site are
recognized by the numbers. Coordinates are displayed in Table 1. The coordinate format is in
UTM/UPS, WGS 84, and the units are in metric system. Results are reported in tables 2-14. Soils were
sampled in pairs at 2-5 cm and near 30 cm depths, whereas sediments were collected one by one
from top sediment layers. Water samples were taken from different open-air water bodies. Soils and
sediments were stored in 250 ml High Density Poly Ethylene (HDPE) plastic containers closed with a
lid. Water samples were collected by a High Density Poly Propylene (HDPP) syringes and the water
was immediately transferred through 0.45 μm polyethersulfone membrane filters to poly propylene
(PP) test tubes. The filters and the test-tubes were provided by VWR International (vwr.com). These
tubes are lidded when leaving the producer and not opened before filtration. Blanks of MilliQ H₂O
(electric resistance higher than 18.2 MΩ cm⁻¹) was stored in equal tubes for 48hr. and filtered
through the same membrane filters before analysis.
Figure 1. This image from Google Earth indicate the sampling sites near Geita city and the control site 17.

Figure 2. This is an image from Google Earth indicting the sampling sites in Nyakabale. The installations are currently larger than shown in the image. Site 16 and 17 are located outside this image. See figure 1.
**Figure 3.** This image from Google Earth indicates the sampling sites near Tarime city and the control site 36.

**Figure 4.** This is an image from Google Earth indicating the sampling sites in east of Tarime. The installations are currently covering a much larger area than shown in the image. Site 36 is outside this image. See figure 3.
Table 1. The table shows sampling site coordinates and altitude together with the date and time of day positioned by a handheld GPS.

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Metal analysis

All soil and sediment samples were air dried and subsamples were prepared for total metal extractions. These subsamples were grinded in an agate mortar and forwarded for soil carbon and total soil metal determination. The non-grinded soils and sediment subsamples were used for soil pH determination. Water samples were stored cold (4-5°C) pending analysis. The metal extraction from soils and sediments were carried out using Microwave digestion by stepwise heating dry material with concentrated ultra pure HNO₃ up to 250dgr. C, using a Milestone Ultraclave, for 1 hr. and 15 min. The metal concentrations in water samples and soil/sediment extracts were determined using two different analyzers, namely a Perkin Elmer Sciex Elan Inductively Coupled Plasma Mass Spectrometry (ICP-MS) and a Perkin Elmer Optima 5300 DV Inductively Coupled Plasma Optic Emission Spectrometry (ICP-OES). The use of two different instruments managed by two different analysts assures high quality control. Moreover, the ICP-MS is most suitable for very dilute solutions and the ICP-OES the most suitable for more concentrated solutions. Blanks and reference material was used. Standard reference material (SRM) for soils used were the SRM 2709 (National Institute of Standards & Technology, 2003), whereas the SRM used for water analysis where the SRM 1643e (National Institute of Standards & Technology, 2004).

The pH in water samples were estimated in field using pH strips, however, only used as indicator of pH. Since field-pH meter was not applied, metal speciation was carried out based on the crude estimations. We investigated, however, the pH effect on the chemical speciation by changing the input pH up and down one unit relative to the estimations while carrying out the speciation modeling. Chemical speciation was carried out using WHAM/Model VI version 6.0 for waters (Tipping, 1998). Analysis of pH in river waters is problematic due to possible oversaturation of dissolved C in pore water. When such water drains continuously into running water, dissolved carbon escapes due to reduction of CO₂ saturation in open atmosphere compared to soil atmosphere, and hence pH may increase significantly after some time of storing, at least if pH is not very low. In still waters, the pore water has equilibrated with atmosphere prior to analysis, and pH is less unstable. Hence, water pH will not be discussed in depth here but the effect on chemical speciation of trace metals will be explored.

Other analysis of soils, sediments and waters:

The inorganic carbon was determined as the difference between total carbon in soil and total organic carbon. Parallel soil and sediment samples were repeatedly washed with 2M HCl to remove inorganic carbon, before total organic carbon was determined using a LECO CHN -1000 analyzer. Soil pH was
measured in deionised water with solid:solution ratio at 1:2.5. The concentrations of \( \text{Cl}^- \), \( \text{NO}_3^- \) and \( \text{SO}_4^{2-} \) in water samples was determined by a Lachat IC 5000 ion chromatograph, and the amount of dissolved carbon (organic and inorganic) by the use of a Shimadzu TOC-5000 analyser.

**Sampling of waters and sediments and analysis of total cyanide:**

Water samples were sampled withdrawn for analysis of total cyanide immediately fixed with NaOH. Cyanide in water and in the soil was determined using Pyridine-pyrazolene method as described by Allen (1989) delivered from Greenberg (1985). Thus cyanide was distilled from the samples after addition of acid cuprous and then collected in a flask containing 5ml of 1M Sodium hydroxide. To the distillate was added to bring it to a pH of 7.0 after a 0.2 ml chloramine T was added. The samples were mixed by inverting them 3 times and left for 2 minutes. To each of the samples 5ml of pyridine-pyrazolene reagent was added. The mixtures were left for 20 minutes after which the absorbance of each mixture was taken at 620nm using Jenway 6305 spectrophotometer. Cyanide standards were made from potassium cyanide. Calibration curve prepared from standard reading was used to calculate the concentration of cyanide in each of the samples.

**Results and discussions**

*Generally*

The trace metal/semimetal contents in soils, sediments and waters are individually reported in tables arranged separately between the two investigated areas. The two areas are for simplicity from now on referred to as Nyakabale and Tarime. Soils were sampled in two depths, whereas sediments are only withdrawn from the top layer. Waters samples are not taken from soil solution, apart from one place, but from sites potentially being important for drinking water, general house hold use and watering of domestic animals. Generally we recovered negligible contents of CN in water and sediment samples, and hence the reporting of data will focus on distribution of metals and semimetals.

**Results from sampling in Nyakabale**

*Soil.*

Out of the metals determined, the variation in As, Cr and Cu is greatest between sites (Table 2). In addition, soil from one site (site 6) was high in Pb. The content of Zn (range: 6.3-47.8 mg/kg in top soil and 7.1-121.6 mg/kg in sub soil) is also higher in soil from site 6, but not very high. The highest content of Cr (range: 6.4-120.8 mg/kg in top soil and 7.4-89.9 mg/kg in sub soil) and Cu (range: 2.1-245.5 mg/kg in top soil and 2.3-333.8 mg/kg in sub soil) was recovered in soil samples from site 8 and
9 towards river Nyakabale. Site 8 is located downhill a residual waste tailing, where rock material unfit for processing is piled up. Site 9 is located further down from site 8, whereas site 10, 11 and 12 follows at increasing distance from site 8 and 9. In addition to Cu and Cr, some As (range: nd-19.6 mg/kg in top soil and from nd-17.3 mg/kg in sub soil. “nd” means under detection limit) was determined in soils. In table 3 the soil pH is shown and the lowest pH values are found in soils from site 7 and 8. Site 7 is also near to the residual waste tailing. The soil pH was also slightly lower than other sites at site 1. The total content of soil sulfur is not shown in tables, but at site 7 and 8, the amount of total S was in the range of 2.6 to 6.8 g/kg, but one sample contained as much as 62.0 g/kg. There are also significant amount of S in soils from site 6: 3.1 and 25.0 g/kg. The content of soil S in control sites was in the range of 0.05-0.50 g/kg.

Sediments.
The trace element contents in sediments are shown in table 4. Also sediments from site 8 contains the highest contents of Cr (range: 11.8-75.0 mg/kg), Cu (range: 6.2-208.4 mg/kg), and Ni (range: 6.4-40.5 mg/kg), whereas the highest content of As (range: 5.0-49.3 mg/kg) was found in sediments withdrawn from site 6. Some As was determined in sediments at site 8, 12 and 13. Sediment pH was lowest in samples from site 8, 12 and 14 (Table 5). The sulfur contents in sediments from natural sites ranged from 01.-2-2 g/kg (site 6). At site 8 the sediment content was as high as 7.4 g/kg.

Water.
The water samples taken from site 8 contained the highest concentrations of most metals analyzed, even Al (table 7). This was also the only site where the concentration of Cd is significant. This is not a natural watering pond, since the site is in a restricted area, but no fence is set up, and domestic animals and game might accidentally use the water as they cannot distinguish safe water from contaminated water. The concentrations of some of the elements in water at this site are above the WHO drinking water recommendations (WHO, 2008). The WHO standards are indicated in the last rows of the table. Water taken from site 10 is not free water but soil solution (filtered through 0.45μm membrane filter), and hence not comparable with drinking water standards. Apart from water at site 8, and the high Hg concentration in water at site 12, the trace element concentrations in other water samples taken in Nyakabale were lower than WHO recommendations. Other water chemical parameters used for speciation calculations are displayed in table 6.
Table 2. The table shows the total contents of trace elements in soils taken from areas near Nyakabale village, opposite the Geita gold mine (GGM) project. Soils are withdrawn from two depths.

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<td>21.0</td>
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<td>11.9</td>
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<td>26.5</td>
<td>9.6</td>
</tr>
<tr>
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<td>0.033</td>
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<td>nd</td>
<td>3.7</td>
<td>26.1</td>
<td>14.2</td>
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<td>nd</td>
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<td>-</td>
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<td>-</td>
<td>nd</td>
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<td>27.3</td>
<td>6.5</td>
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<td>19.7</td>
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<td>25-30</td>
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<td>-</td>
<td>nd</td>
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<td>7.4</td>
<td>2.8</td>
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<td>5.17</td>
<td>13.8</td>
<td>7.1</td>
<td>nd</td>
</tr>
</tbody>
</table>

*nd: under detection limits.*

* -: not analyzed

*a: all metal contents in soils are given in mg/kg apart from Al shown in g/kg.

*b: CNTOT was also determined in sub-soil from site 9 but the content was under detection limit*
Table 3. The table shows soil pH, total soil-C, total soil organic-C and total soil-N in soils withdrawn from Nyakabale village opposite the Geita gold mine (GGM) project. Soils are withdrawn from two depths.

<table>
<thead>
<tr>
<th>Site</th>
<th>Depth</th>
<th>Soil pH</th>
<th>Total Soil-C</th>
<th>Total Soil org. C</th>
<th>Total Soil-N</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>%</td>
<td></td>
<td></td>
</tr>
<tr>
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<td>25-30</td>
<td>4.48</td>
<td>0.82</td>
<td>0.69</td>
<td>0.03</td>
</tr>
<tr>
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<td>6.39</td>
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<td>2.84</td>
<td>0.24</td>
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<td>0.50</td>
<td>0.01</td>
</tr>
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<td>0.13</td>
</tr>
<tr>
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<td>6.24</td>
<td>1.42</td>
<td>1.27</td>
<td>0.05</td>
</tr>
<tr>
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<td>5.20</td>
<td>1.29</td>
<td>1.06</td>
<td>0.04</td>
</tr>
<tr>
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<td>2.19</td>
<td>1.86</td>
<td>0.14</td>
</tr>
<tr>
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<td>0.55</td>
<td>0.47</td>
<td>0.01</td>
</tr>
<tr>
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<td>6.27</td>
<td>0.87</td>
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<td>0.00</td>
</tr>
<tr>
<td>7</td>
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<td>3.36</td>
<td>0.15</td>
<td>0.11</td>
<td>0.00</td>
</tr>
<tr>
<td></td>
<td>25-30</td>
<td>3.35</td>
<td>0.13</td>
<td>0.23</td>
<td>0.00</td>
</tr>
<tr>
<td>8</td>
<td>0-5</td>
<td>4.16</td>
<td>1.14</td>
<td>0.92</td>
<td>0.04</td>
</tr>
<tr>
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<td>4.20</td>
<td>1.96</td>
<td>1.64</td>
<td>0.07</td>
</tr>
<tr>
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<td>0.42</td>
<td>1.27</td>
<td>0.01</td>
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<tr>
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<td>0.42</td>
<td>0.40</td>
<td>0.01</td>
</tr>
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<td>1.28</td>
<td>0.07</td>
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</tr>
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<td>8.22</td>
<td>7.38</td>
<td>0.52</td>
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<tr>
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<td>6.22</td>
<td>1.71</td>
<td>1.58</td>
<td>0.12</td>
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<td>2.46</td>
<td>2.13</td>
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</tr>
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<td>5.73</td>
<td>0.38</td>
<td>0.36</td>
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</tr>
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<td>25-30</td>
<td>5.86</td>
<td>0.36</td>
<td>0.30</td>
<td>0.01</td>
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</table>

Table 4. The table shows the total contents of trace elements in sediments taken from Nyakabale village opposite the Geita gold mine (GGM) project. Sediments are withdrawn from top layer.

<table>
<thead>
<tr>
<th>Site</th>
<th>Depth</th>
<th>Al (g/kg)</th>
<th>Hg (mg/kg)</th>
<th>As (mg/kg)</th>
<th>Cd (mg/kg)</th>
<th>Co (mg/kg)</th>
<th>Cr (mg/kg)</th>
<th>Cu (mg/kg)</th>
<th>Ni (mg/kg)</th>
<th>Pb (mg/kg)</th>
<th>V (mg/kg)</th>
<th>Zn (mg/kg)</th>
</tr>
</thead>
<tbody>
<tr>
<td>2</td>
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<td>26.5</td>
<td>-</td>
<td>5.0</td>
<td>nd</td>
<td>4.6</td>
<td>31.4</td>
<td>9.0</td>
<td>13.4</td>
<td>10.7</td>
<td>44.2</td>
<td>13.2</td>
</tr>
<tr>
<td>4</td>
<td>0-5</td>
<td>33.7</td>
<td>-</td>
<td>17.2</td>
<td>nd</td>
<td>6.7</td>
<td>34.1</td>
<td>14.6</td>
<td>19.3</td>
<td>11.5</td>
<td>35.9</td>
<td>27.0</td>
</tr>
<tr>
<td>5</td>
<td>0-5</td>
<td>34.3</td>
<td>-</td>
<td>5.0</td>
<td>nd</td>
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<td>31.9</td>
<td>23.0</td>
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<td>13.9</td>
<td>38.9</td>
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</tr>
<tr>
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<td>13.0</td>
<td>-</td>
<td>49.3</td>
<td>nd</td>
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<td>50.6</td>
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<td>69.7</td>
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<td>nd</td>
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<td>63.1</td>
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<td>42.5</td>
<td>0.019</td>
<td>5.0</td>
<td>nd</td>
<td>12.2</td>
<td>42.8</td>
<td>40.5</td>
<td>31.4</td>
<td>14.5</td>
<td>41.7</td>
<td>33.0</td>
</tr>
<tr>
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<td>0-5</td>
<td>44.6</td>
<td>0.015</td>
<td>21.5</td>
<td>nd</td>
<td>6.1</td>
<td>39.1</td>
<td>24.4</td>
<td>22.7</td>
<td>17.7</td>
<td>60.1</td>
<td>35.7</td>
</tr>
<tr>
<td>13</td>
<td>0-5</td>
<td>12.5</td>
<td>-</td>
<td>10.4</td>
<td>nd</td>
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<td>53.0</td>
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<td>-</td>
<td>5.0</td>
<td>nd</td>
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<td>11.8</td>
<td>6.2</td>
<td>6.4</td>
<td>9.4</td>
<td>14.7</td>
<td>12.1</td>
</tr>
</tbody>
</table>

nd: under detection limits. -: not analyzed. a: all metal contents in sediments are given in mg/kg apart from Al shown in g/kg. b: CNTOT was also determined in sediment from site 14 but the content was under detection limits.
Table 5. The table shows sediment pH, total sediment-C, total sediment organic-C and total sediment-N in samples withdrawn from Nyakabale village opposite the Geita gold mine (GGM) project. Sediments are taken from the top layer.

<table>
<thead>
<tr>
<th>Site</th>
<th>Sed pH</th>
<th>Total sed-C</th>
<th>Total sed org. C</th>
<th>Total sed. N</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>%</td>
<td></td>
<td></td>
</tr>
<tr>
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<td>6.84</td>
<td>0.60</td>
<td>0.05</td>
<td>0.01</td>
</tr>
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</tr>
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<td>7.37</td>
<td>2.19</td>
<td>1.86</td>
<td>0.14</td>
</tr>
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<td>7.81</td>
<td>1.02</td>
<td>0.70</td>
<td>0.02</td>
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<tr>
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<td>1.56</td>
<td>1.27</td>
<td>0.06</td>
</tr>
<tr>
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<td>2.24</td>
<td>0.21</td>
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<td>4.58</td>
<td>7.78</td>
<td>6.04</td>
<td>0.54</td>
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<td>1.15</td>
<td>0.04</td>
</tr>
<tr>
<td>14</td>
<td>4.11</td>
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<td>1.25</td>
<td>0.07</td>
</tr>
</tbody>
</table>

*: not enough material

Table 6. The table shows contents of dissolve organic C (DOC), chloride (Cl), nitrate-N (NO₃-N) and sulfate (SO₄²⁻) in water samples from Nyakabale village opposite the Geita gold mine (GGM) project.

<table>
<thead>
<tr>
<th>Site</th>
<th>DOC</th>
<th>Cl</th>
<th>NO₃-N</th>
<th>SO₄²⁻</th>
</tr>
</thead>
<tbody>
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<td></td>
<td>mg/l</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
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<td>nd</td>
<td>158.0</td>
</tr>
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<td>2.56</td>
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<td>nd</td>
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<td>0.022</td>
<td>1.1</td>
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</tbody>
</table>

*nd: under detection limits.*
Table 7. The table shows the total concentrations of trace elements in selected water samples from rivers and ponds in areas near Nyakabale village opposite the Geita gold mine (GGM) project. The WHO guidelines are given for comparison with the analytical results.

<table>
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<th>Site</th>
<th>$CN_{TOT}$</th>
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<th>Ag</th>
<th>As</th>
<th>Au</th>
<th>Cd</th>
<th>Co</th>
<th>Cr</th>
<th>Cu</th>
<th>Hg</th>
<th>Ni</th>
<th>Pb</th>
<th>Se</th>
<th>Th</th>
<th>U</th>
<th>V</th>
<th>Zn</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>mg/l</td>
<td>mg/l$^a$</td>
<td>µg/l</td>
<td></td>
<td></td>
<td></td>
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<td>0.0</td>
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<td>0.07</td>
<td>1.0</td>
<td>0.01</td>
<td>0.03</td>
<td>0.4</td>
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WHO:  

| µg/l | 0.07$^d$ | - | 10 | 3 | 50 | 2000 | 6 | 70 | 10 | 10 | - | 15 | 3000$^3$ |

nd: under detection limits.  
-: not analyzed  
a: all metal concentrations are given in µg/l apart from Al shown in mg/l.  
b: The concentrations presented as 0.0 are not exactly 0, but values are too low to be shown in µg/l.  
c: This “water sample” is not taken from running water but from soil solution, and hence these concentrations should NOT be directly compared with concentrations of metals in free waters like rivers and lakes.  
d: There are no accepted standards in WHO for this element at the moment.  
e: Derivation of guideline values is not required at this time, however, drinking-water containing Zn at levels above 3 mg/l (equal to 3000 µg/l) may not be acceptable to consumers (WHO, 2008).
Comments to the results from Nyakabale investigation

The findings do not support an extensive atmospheric deposition of trace elements to soils in this area, as significant difference in trace element contents between top-soil and sub-soil could only be found at some sites. At those sites, there was a top-soil increase of Cr and Cu in soils taken from sites 9, 10 and 11. This finding fits with the slightly higher contents of Cr in sediments from site 11, 12 and 13, compared to the most remote site 14, indicating surface transport of suspended or dissolved Cr over land were sites 9-12 are located. When water is flooded over dry soil, very little will penetrate the soil profile. Some of the eroded material might be trapped in vegetation and sediment on top soil, but most will be carried away with the water, down to an aquatic recipient where it will be trapped for shorter or longer time. Although the metal contents in soils and sediments are not alarmingly high, the visual (deposition of eroded material downside site 8) and chemical findings indicate clearly that erosion from the residual waste tailing has occurred.

It is important to note that water samples, in particular those withdrawn from running water, only reports the situation the very moment they are collected! Residence time of still water in these areas was not determined, but residence time in shallow waters is normally weeks, maximum months. Hence, the findings in sediments should be assessed seriously as the quality of sediments will indicate the chemical impact on sediments from occasional flooding in the catchment area. The finding of As in sediments collected from different sites should be of concern although the concentration in the water bodies, at sampling time, was not very high.

Results from sampling in Tarime

The sampling in around North Mara Gold Mine (NMGM) Project in Tarime district was concentrated in three areas namely; Nyabigena, Nyabilama and Ikwinyunyi. The lowest site numbers 18-22 refers to the Ikwinyunyi area where an accidental spill from a tailing dam apparently occurred on the 8th May, 2009, whereas the other site numbers refers to sampling near Nyabigena where red leachate flowing rock piles was evident and Nyabilama with a channel towards river Mara. The later two sites are opposed the waste rock piles. Site 36 refers to a control area in river Mara (Kirumi bridge) not far from Lake Victoria. The tables report findings after total analysis of soils, sediment and water samples.

Soil.

In general, the contents of trace elements are slightly higher in soils sampled from this area than those taken from Nyakabale. The major difference is that As (range: nd- 126.1 mg/kg in top soil and nd- 45.0 mg/kg in sub soil) was found in almost all samples and, and that the content is generally
higher (Table 8). The content of Zn (range: 15.6-252.0 mg/kg in top soil and 15.8-207.9 mg/kg in sub soil) and Ni (range: 4.5-65.7 mg/kg in top soil and 4.4-34.3 mg/kg in sub soil) in also found to be higher, whereas the Co, Cr, Pb and V contents are lower (see table 8). The contents of As, Co, Cr Cu, Ni, V and Zn were found to be generally higher around NMGM area than at the control site 36. The results show that the highest contents were found in soils taken from site 18, and to some degree in soils from sites 29-32. Arsenic contents were highest in soil from site 22, as expected, but also high in soils from sites 29 and 32. The topsoil at site 24, had been over flooded and suspended mineral material was deposited on top. The content of Ni was notably high at this site, higher than any other places investigated.

Table 8. The table shows the total contents of trace elements in soils taken from areas surrounding North Mara Gold Mine (NMGM) project in Tarime district. Soils are withdrawn from two depths.

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<th>Cd</th>
<th>Co (mg/kg)</th>
<th>Cr (mg/kg)</th>
<th>Cu (mg/kg)</th>
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nd: under detection limits.
-: not analyzed.
a: all metal contents in soils are given in mg/kg apart from Al shown in g/kg.
The soil pH varied, and the lowest pH values were found in soils collected from site 18, 21 and 22, where the spill had taken place (Table 9). It is noteworthy that pH in soil 19 is higher, and this site is located upstream from where the spill had entered the Tighite river. The total contents of S in soils varied from 0.1 g/kg at site 36 to 21.0 g/kg at site 21.

**Table 9.** The table shows soil pH, total soil-C, total soil organic-C and total soil-N in soils withdrawn from areas surrounding North Mara Gold Mine (NMGM) project in Tarime district. Soils are withdrawn from two depths.

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<td>1.78</td>
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<td>0.43</td>
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<td>2.76</td>
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<td>0.69</td>
<td>0.55</td>
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<td>2.91</td>
<td>2.69</td>
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<td>2.32</td>
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<td>25-30</td>
<td>7.10</td>
<td>0.94</td>
<td>0.99</td>
<td>0.06</td>
</tr>
</tbody>
</table>

* not enough material

**Sediment.**

When investigating the results from sediments, it is clear that the contents of As (range: nd- 522.5 mg/kg) is very high in the sediments of Tighite river, site 18, table 10. These sediments were collected from the river sediments close to the soils referred to by the same number. The same increase is not clear for the other elements presented. Note, however, that As occur also further
down in the valley. There is no detectable As in sediments from site 36 (Kirumi bridge). Although the differences between sites were not as great for the other elements as for As, all trace element contents in sediment are significantly higher than in the sediment sample collected from site 36.

Table 10. The table shows total contents of trace elements in sediments taken from areas surrounding North Mara Gold Mine (NMGM) project in Tarime district. Sediments are withdrawn from the top layer.

<table>
<thead>
<tr>
<th>Site</th>
<th>Depth</th>
<th>Al</th>
<th>Hg</th>
<th>As</th>
<th>Cd</th>
<th>Co</th>
<th>Cr</th>
<th>Cu</th>
<th>Ni</th>
<th>Pb</th>
<th>V</th>
<th>Zn</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>g/kg</td>
<td></td>
<td>mg/kg</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>18</td>
<td>0-5</td>
<td>32.5</td>
<td>0.015</td>
<td>522.5</td>
<td>nd</td>
<td>33.5</td>
<td>69.0</td>
<td>25.2</td>
<td>34.2</td>
<td>16.4</td>
<td>55.2</td>
<td>97.6</td>
</tr>
<tr>
<td>19</td>
<td>0-5</td>
<td>28.4</td>
<td>-</td>
<td>57.9</td>
<td>nd</td>
<td>16.1</td>
<td>72.2</td>
<td>12.9</td>
<td>31.5</td>
<td>18.9</td>
<td>53.6</td>
<td>59.5</td>
</tr>
<tr>
<td>21</td>
<td>0-5</td>
<td>15.8</td>
<td>0.771</td>
<td>98.7</td>
<td>nd</td>
<td>10.8</td>
<td>27.3</td>
<td>18.2</td>
<td>27.2</td>
<td>14.0</td>
<td>15.5</td>
<td>107.2</td>
</tr>
<tr>
<td>22</td>
<td>0-5</td>
<td>23.7</td>
<td>-</td>
<td>425.9</td>
<td>nd</td>
<td>4.8</td>
<td>20.0</td>
<td>17.3</td>
<td>14.5</td>
<td>7.6</td>
<td>21.4</td>
<td>73.7</td>
</tr>
<tr>
<td>25</td>
<td>0-2</td>
<td>25.7</td>
<td>0.086</td>
<td>10.0</td>
<td>nd</td>
<td>17.8</td>
<td>43.9</td>
<td>10.4</td>
<td>24.3</td>
<td>17.3</td>
<td>51.8</td>
<td>57.7</td>
</tr>
<tr>
<td>26</td>
<td>0-2</td>
<td>11.5</td>
<td>0.008</td>
<td>14.1</td>
<td>nd</td>
<td>7.5</td>
<td>38.9</td>
<td>5.0</td>
<td>13.7</td>
<td>7.8</td>
<td>30.0</td>
<td>27.3</td>
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<tr>
<td>27b</td>
<td>0-5</td>
<td>11.6</td>
<td>-</td>
<td>17.9</td>
<td>nd</td>
<td>4.84</td>
<td>13.4</td>
<td>3.1</td>
<td>6.4</td>
<td>8.8</td>
<td>12.7</td>
<td>14.6</td>
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<td>29b</td>
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<td>53.5</td>
<td>0.025</td>
<td>33.1</td>
<td>nd</td>
<td>9.4</td>
<td>35.8</td>
<td>10.9</td>
<td>18.9</td>
<td>12.3</td>
<td>21.1</td>
<td>89.0</td>
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<tr>
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<td>0-5</td>
<td>43.1</td>
<td>-</td>
<td>12.2</td>
<td>nd</td>
<td>7.5</td>
<td>32.1</td>
<td>12.1</td>
<td>19.1</td>
<td>16.5</td>
<td>23.1</td>
<td>123.8</td>
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<tr>
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<td>0-5</td>
<td>31.2</td>
<td>-</td>
<td>35.5</td>
<td>nd</td>
<td>7.1</td>
<td>43.4</td>
<td>7.7</td>
<td>20.0</td>
<td>9.0</td>
<td>16.7</td>
<td>49.0</td>
</tr>
<tr>
<td>33</td>
<td>0-5</td>
<td>22.8</td>
<td>-</td>
<td>12.6</td>
<td>nd</td>
<td>3.7</td>
<td>22.1</td>
<td>5.1</td>
<td>10.2</td>
<td>6.9</td>
<td>12.1</td>
<td>25.7</td>
</tr>
<tr>
<td>34</td>
<td>0-5</td>
<td>42.8</td>
<td>-</td>
<td>nd</td>
<td>nd</td>
<td>7.6</td>
<td>28.2</td>
<td>11.4</td>
<td>18.3</td>
<td>18.2</td>
<td>20.4</td>
<td>136.7</td>
</tr>
<tr>
<td>35</td>
<td>0-5</td>
<td>49.6</td>
<td>-</td>
<td>nd</td>
<td>nd</td>
<td>8.1</td>
<td>35.2</td>
<td>16.5</td>
<td>19.4</td>
<td>17.5</td>
<td>25.5</td>
<td>142.3</td>
</tr>
<tr>
<td>36</td>
<td>0-5</td>
<td>11.1</td>
<td>-</td>
<td>nd</td>
<td>nd</td>
<td>2.1</td>
<td>5.7</td>
<td>3.1</td>
<td>2.7</td>
<td>7.4</td>
<td>13.6</td>
<td>21.8</td>
</tr>
</tbody>
</table>

nd: under detection limits. - : not analyzed. a: all metal contents in sediments are given in mg/kg apart from Al shown in g/kg. b: CTOT was determined in sediments from sites 27, 29 and 32 and the contents were 0.005 mg/l, 0.004 mg/l and under detection limits, respectively.

Sediment pH in site 18 was 3.3, and closer to the mine tailing, sites 21 and 22, the sediment pH was as low as 2.9 (Table 11). The sediment pH at other sites ranged from 6.2-8.3. Also the content of sediment S was highest in sediments associated to the accidental spill, where site 21 contains sediments with 68.0 g S/kg. The S content at site 18 was not higher than 1.8g/kg, whereas the total S ranged from 0.1 at site 36 to 1.5 at site 29.
Table 11. The table shows sediment pH, total sediment-C, total sediment organic-C and total sediment-N in samples withdrawn from areas surrounding North Mara Gold Mine (NMGM) project in Tarime district. Samples are withdrawn from the top layer.

<table>
<thead>
<tr>
<th>Site</th>
<th>Sed pH</th>
<th>Total sed-C</th>
<th>Total sed org. C</th>
<th>Total sed. N</th>
</tr>
</thead>
<tbody>
<tr>
<td>18</td>
<td>3.26</td>
<td>0.80</td>
<td>0.65</td>
<td>0.04</td>
</tr>
<tr>
<td>19</td>
<td>7.34</td>
<td>1.35</td>
<td>1.09</td>
<td>0.08</td>
</tr>
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<td>21</td>
<td>2.93</td>
<td>1.29</td>
<td>1.12</td>
<td>0.12</td>
</tr>
<tr>
<td>22</td>
<td>2.91</td>
<td>1.58</td>
<td>1.34</td>
<td>0.10</td>
</tr>
<tr>
<td>25</td>
<td>7.44</td>
<td>0.68</td>
<td>0.54</td>
<td>0.03</td>
</tr>
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<td>26</td>
<td>7.57</td>
<td>0.08</td>
<td>0.04</td>
<td>0.00</td>
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<td>27</td>
<td>8.03</td>
<td>0.21</td>
<td>0.16</td>
<td>0.01</td>
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<td>7.66</td>
<td>2.88</td>
<td>2.66</td>
<td>0.21</td>
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<tr>
<td>31</td>
<td>6.38</td>
<td>1.70</td>
<td>1.58</td>
<td>0.12</td>
</tr>
<tr>
<td>32</td>
<td>7.84</td>
<td>0.62</td>
<td>0.58</td>
<td>0.03</td>
</tr>
<tr>
<td>33</td>
<td>8.32</td>
<td>1.50</td>
<td>1.23</td>
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</tr>
<tr>
<td>34</td>
<td>7.54</td>
<td>3.62</td>
<td>3.22</td>
<td>0.30</td>
</tr>
<tr>
<td>35</td>
<td>6.84</td>
<td>1.42</td>
<td>1.23</td>
<td>0.09</td>
</tr>
<tr>
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<td>6.19</td>
<td>2.82</td>
<td>2.41</td>
<td>0.16</td>
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</table>

Water.

The concentrations of trace elements in the different water samples are shown in table 12. Some CN was detected in two sites, although the levels are not very high. Apart from Al, all element concentrations are presented in μg/l. For comparison, the WHO recommendations are presented in the last rows of the table. It is very clear that the concentrations of As, Cd, Cr, Ni, Se, U and Zn vastly crosses the WHO recommendations in some of the sites. As expected, water taken from sites associated to the spill in May 2009, contain the highest concentrations. It is, however, noteworthy that also water sampled from sites 27, 29, 32, and 38 and to some degree 31 (for As), contained disturbing high concentrations of As and Co. WHO does not provide any drinking water recommendations for Co, however, the proposed Canadian guidelines for chronic exposure aquatic life is in the range of 4μg/l (Nagpal, 2004). Now, it is important to point out that this value is not connected to drinking water qualities for humans, due to lack of literature available, but to aquatic life. It is recommended that to prevent fresh water environment from acute toxicity, Co concentration should not exceed 110 μg/l, and that over longer period of time not to exceed 4μg/l to prevent chronic effects. Nevertheless, the concentrations of As and Co reaches concentrations beyond internationally issued environmental and human health recommendations, not only near the site where the accidental spill had occurred, but also at sites not directly connected to that particular site at all. The concentration of sulfate, nitrate and chlorine concentrations shown in table 13 follows the same pattern, indicating acid character of spill has occurred. Although solution pH was only measured using pH paper, those tests indicated pH values at 2.5-3.0 for water at sites 18, 21 and 22 (at site 19 the river water pH was 5.5).
### Table 12. The table shows the total concentrations of trace elements in selected rivers and ponds in areas surrounding North Mara Gold Mine (NMGM) project in Tarime district. The WHO guidelines are given for comparison with the analytical results.

<table>
<thead>
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<th>Site</th>
<th>CN</th>
<th>TOT</th>
<th>Al</th>
<th>Ag</th>
<th>As</th>
<th>Au</th>
<th>Cd</th>
<th>Co</th>
<th>Cr</th>
<th>Cu</th>
<th>Hg</th>
<th>Ni</th>
<th>Pb</th>
<th>Se</th>
<th>Th</th>
<th>U</th>
<th>V</th>
<th>Zn</th>
<th>WHO:</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>mg/l</td>
<td>mg/l</td>
<td>µg/l</td>
<td></td>
<td></td>
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<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>18</td>
<td>-</td>
<td>363.00</td>
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<td>307.0</td>
<td>0.4</td>
<td>108.00</td>
<td>4583.0</td>
<td>254.2</td>
<td>1670.1</td>
<td>0.5</td>
<td>9962.3</td>
<td>7.80</td>
<td>111.4</td>
<td>33.8</td>
<td>464.5</td>
<td>1.44</td>
<td>43473.3</td>
<td></td>
<td></td>
</tr>
<tr>
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<td>-</td>
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<td>0.1</td>
<td>1.1</td>
<td>nd</td>
<td>0.01</td>
<td>0.1</td>
<td>nd</td>
<td>1.1</td>
<td>0.4</td>
<td>0.9</td>
<td>0.07</td>
<td>1.0</td>
<td>0.2</td>
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<td>0.98</td>
<td>1.9</td>
<td></td>
<td></td>
</tr>
<tr>
<td>21</td>
<td>-</td>
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<td>149.00</td>
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<td>0.0</td>
<td>224.00</td>
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<td>169.2</td>
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</tr>
<tr>
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<td>-</td>
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<td>0.0</td>
<td>0.1</td>
<td>nd</td>
<td>0.02</td>
<td>0.1</td>
<td>0.3</td>
<td>0.3</td>
<td>0.1</td>
<td>1.3</td>
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<td>0.0</td>
<td>0.12</td>
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<td>0.6</td>
<td>2.6</td>
<td>0.1</td>
<td>20.9</td>
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<td>0.0</td>
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</tr>
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<td>nd</td>
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<td>22.4</td>
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<td>0.4</td>
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<tr>
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<td>319.0</td>
<td>nd</td>
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<td>12.0</td>
<td>0.00</td>
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<td>0.1</td>
<td>9.1</td>
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<td>0.0</td>
<td>1.4</td>
<td>0.2</td>
<td>0.01</td>
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</tbody>
</table>

**WHO:**

- mg/l: 0.07
- µg/l: 10 - 3 - 50 2000 6 70 10 10 - 15 - 3000

*a*: all metal concentrations are given in µg/l apart from Al shown in mg/l.

*b*: The concentrations presented as 0.0 are not exactly 0, but values are too low to be shown in µg/l.

*c*: There are no accepted standards in WHO for this element at the moment.

*d*: Derivation of guideline values is not required at this time, however, drinking-water containing Zn at levels above 3 mg/l (equal to 3000 µg/l) may not be acceptable to consumers (WHO, 2008).
Table 13. The table shows contents of dissolve organic C (DOC), chloride (Cl), nitrate-N (NO₃-N) and sulfate (SO₄²⁻) in water samples from areas surrounding North Mara Gold Mine (NMGM) project in Tarime district. Sediments are taken from the top layer.

<table>
<thead>
<tr>
<th>Site</th>
<th>DOC</th>
<th>Cl</th>
<th>NO₃-N</th>
<th>SO₄²⁻</th>
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<tr>
<td>18</td>
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<td>24.0</td>
<td>54.3</td>
<td>5770.0</td>
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<td>7.7</td>
<td>-</td>
<td>1.6</td>
<td>5.9</td>
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<td>21</td>
<td>6.5</td>
<td>26.0</td>
<td>30.5</td>
<td>8290.0</td>
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<td>-</td>
<td>-</td>
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<td>0.01</td>
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<td>nd</td>
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<tr>
<td>36</td>
<td>15.8</td>
<td>14.8</td>
<td>nd</td>
<td>1.6</td>
</tr>
</tbody>
</table>

nd: under detection limits.
-: not enough material

Comments to the results from Tarime investigation

As in Nyakabale, there is no reason to believe that there is a significant impact of atmospheric deposition of trace elements to the soils in the district. At some sites, one can find a pattern of elevated concentrations of metals in top-soil compared to sub-soil, and those sites are 21, 22 to some degree site 24 (very clear for Ni there), but also at sites 32 and 35. Again, this is taken as indication of surface transport of suspended material. This was visible when investigating the fields that had been over flooded with spills from the mine tailings near sites 18-22. Hence there is no doubt that the spill has caused severe toxicity of water, and to some extent degradation of soils and sediments nearby, particularly due the high As content. The investigation of other sites, indicates that trace elements have been transport by flooded rivers depositing sediments on cultivated land and riverbanks. Generally, the soil concentrations of trace metals are not extreme, but the high contents of As in soils and sediments is of great concern!

Although the soil contents of trace metals are not very high, the water analyses reveal disappointingly extreme concentration of toxic metals and As. The mining activity is therefore a chemical threat of extreme character to the nearby land due to its storage of CN, trace metals and semimetals, and possibly other elements in toxic concentrations.
Chemical speciation of soil solution taken from both sites

All living cells take up trace elements through a semi permeable membrane, and the transport is generally controlled by the membrane, flux of solutes (solute transport to membrane and internalization in the cell), and the solute chemistry. The latter is often referred to as chemical speciation. Hence, the chemical form of the element of interest is of great importance as that again is controlled by the chemical environment in the solution outside the living cells. In soil solution and natural waters the solution pH and concentration of other solutes has a great impact. It boils down to that we need to estimate the free ion activity (FIA) to know the potential bioavailability of the potentially toxic or essential elements. In this particular study the lack of precise information about solution pH is limiting. However, the use of well developed speciation models, like the speciation model WHAM/Model VI, helps to predict important estimates of the speciation anyhow (Almås et al., 2007). The in situ solution pH was estimated only in water samples from areas surrounding NMGM using pH strips. The lack of solution pH information in Nyakabale is compensated for by estimating a default pH based on soil pH, and then we have tested the effect of changing pH up and down one unit. The results from speciation modeling can be seen in table 14. Without going too far in discussing the outcome, there is a general tendency for rather high fraction of metals being in the free ion activity form, which again is the most biologically relevant specie for uptake. This is most clear in the low-pH samples. Another important factor controlling metal speciation is the contents of dissolved organic carbon (DOC). DOC is a constituent of dissolved organic acids that can bind and prevent uptake. The low content of DOC in these waters samples may have had an impact on the rather high fraction of FIA of metals in water. Further investigation of these waters with regards to metals speciation and bioavailability would be of great interest.
Table 14. The table shows an estimate of percent free ion activity (FIA) out of total in water, and the effect from changing the pH one unit down (pH-1) and one unit up (pH+1) relative to the estimated pH in field (pH0). The estimated pH is shown in the first column.

<table>
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<tr>
<th>Site</th>
<th>pH</th>
<th>pH0</th>
<th>pH1</th>
<th>pH0</th>
<th>pH1</th>
<th>pH0</th>
<th>pH1</th>
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<th>pH1</th>
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<td>41.3</td>
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<td>16.1</td>
<td>60.5</td>
<td>60.8</td>
<td>59.7</td>
<td>58.6</td>
<td>59.0</td>
<td>57.8</td>
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<td>59.2</td>
<td>56.7</td>
<td>29.9</td>
<td>46.4</td>
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</table>

a pH was not measured in field water in Nyakabale, hence the pH is set to 5.0 as a qualified estimate based on soil pH.
Conclusion from both studies

Generally, there is no reason to dispute the impact of large scale mining on the local environments, but the study shows a great variety in type of elements and intensity spilled. We have not been speculating nor discussed the direct health effect connected to these findings as that is outside our expertise. The only link to human health has been the comparison of element concentrations in water with WHO drinking water recommendations (WHO, 2008). Our evaluation is that fertile lands of generally good quality accumulate spills containing varying concentrations of potentially hazardous elements. The soil analysis shows, apart from the accident site in Tarime, not very high contents of trace elements, whereas the waters and sediment samples contained higher contents of the investigated elements. The fact that we only at some locations detected differences between top and sub layers contents of trace elements in soils, indicates no or only little atmospheric deposition of trace elements. We see to varying degree that there is a significant surface transport of trace elements, very likely eroded from a contamination source. Along those erosion and transport pathways we find some accumulation of metals in topsoil. Suspended matter seems to be carried over land in episodes of heavy rainfall or accidental leaching of waste. Additionally, suspended material loaded with trace elements seems to have been carried by flooded rivers later being deposited on cultivated land. It is worth while noticing that even in Nyakabale, where no recent episodes of catastrophic spills have been reported, the contents of several trace elements are substantially higher in sediments than in soils. Hence, the metals and semi-metal contents in sediments are not eroded from the natural soils in the catchment areas.

It is important to stress that bioavailability of elements in soils and sediments have not been evaluated, and that other chemical parameters than total contents (usually extracted by strong acid at high temperature) controls the transfer of element from solid phase to solution. The concentration in solution is neither the best indicator for plant and microbial uptake, as it is generally accepted that the FIA of trace elements in water is the dominating specie taken up by cells. However, speciation is naturally less important for drinking waters, as the whole body of water containing all solutes is used for drinking when not pretreated by purification. Bioavailability of trace metals in environmental samples collected from this area should be investigated further.

Finally, the local population base most of their diet on locally produced food and animals that are feed and watered using local sources not regularly controlled or treated by purification. It is important to stress that the quality of land and water is critical for the local population! Even if drinking water is purified, the use of contaminated water for watering of crops is not recommended.
as contaminants will be transferred to the cultivars (Rahman et al., 2003; Huq and Naidu, 2005). This investigation, particularly the one part carried out around NMGM in Tarime, explicitly shows the hazardous potential mining activities pose on the local population food and drinking sources. Episodes of undesired spills may have dramatic consequences as the installation are not 100% water tight and since no effective alarm service system seems to be effectuated preventing undesired exposure to unsuspecting consumers.

The high concentrations of As widespread should be investigated further. Several places the concentrations of As was above 10 µg/l, currently set as recommended drinking water limit by the WHO. Canada is, however, contemplating about reducing this limit down to 5 µg/l, as there have been published several studies indicating that 10 µg/l is not protective. Typical exposure to As leads to an accumulation of As in tissues as skin, hair and nails, resulting in various clinical symptoms such as hyperpigmentation, and keratosis. There has also been reported increase of skin and internal organ and lung cancers from As exposure. Moreover, fatal losses and premature delivery, decrease in birth weight of infants can occur even at low (<10 µg/l) exposure levels (Kapaj et al., 2006).

Acknowledgments

We want to give thanks to the Tanzanian Episcopal Conference (TEC), the Muslim Council of Tanzania (BAKWATA) and the Christian Council of Tanzania (CCT) for initiating and inviting the authors to carry out the study. Moreover, we want to bring forward a special thanks to the Norwegian Church Aid (NCA), Gunstein Instefjord and Kjetil Abildsnes for establishing scientific contact between the University of Dar es Salaam and Norwegian University of Life Sciences and for funding the whole study. We also want to extend our thanks to rev. Godfrey Walalaze at NCA for assistance in providing research clearance for Almås and arranging the logistic needed for a smooth field work and for connecting the team with key persons at the respective sites. Last but not least we also owe the local district bishops and reverends a great hand, the local villagers our gratitude for invaluable assistance during sampling and finally our thanks goes to our cheerful driver Athuman Seif for his sound driving and patience during the whole period of field work.
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<td>Arsenic</td>
</tr>
<tr>
<td>C</td>
<td>Carbon</td>
</tr>
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<td>Cd</td>
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<td>Cl</td>
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<tr>
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