



## Contribution to the evaluation of the level of mercury pollution of the soils of gold mining sites in the territory of Fizi, Eastern of Democratic Republic of Congo

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### Abstract

Soils collected from the sites studied were studied during a 16-month cycle (August and December 2016 to August and December 2017) in order to evaluate their degree of mercury pollution in dry seasons and rainfall. The total mercury was determined by atomic absorption spectrophotometry (AAS) while the indices of mercury pollution were successively calculated using the appropriate formulas. The results thus obtained revealed that, with the exception of the soils of the Lulimba reference site (non-mining site), the soils of all the gold panning sites in the Fizi territory are largely polluted by mercury with regard to their contents. Mercurial pollutants, in this case mercury enrichment factor (EF), mercury contamination factor (CF), mercury geoaccumulation index (Igeo), Ecological Potential Risk Factor of Mercury (EPRF) and the Ecological Risk Index for Mercury (ERI) that deviate from the standards recommended by the Canadian Council of Ministers of the Environment. This reported pollution results from anthropogenic gold mining activities that generate effluents and elemental mercury that pollute terrestrial and other ecosystems.

## 1. Introduction

Gold mining generally uses three types of gold ore processing processes, the choice of which is influenced by the location of extraction (rivers or mines), the degree of oxidation of the ore, the size of the grain, and the nature of ores containing gold. This is the amalgamation method, the gravitational method and the cyanidation method [1]. Of these three methods of gold extraction, only amalgamation extraction uses mercury. It involves contacting the gold of the ore with mercury to form a gold-mercury amalgam (Au-Hg). This amalgam thus obtained is then heated to separate the gold residue from the mercury, which can be reused. This method is widely used and is generally used for artisanal mining and semi-industrial or "small-scale" mines which would represent approximately 20 to 30 % of the world's gold production, or about 550 to 800 tons per year [2].

During the last decades, a quantity of mercury estimated between 800 and 1000 tons is consumed annually by the artisanal exploitations in the world for the recovery of gold by amalgamation [3].

Then, during the handling of mercury to form the gold-mercury amalgam (Au-Hg), a quantity of mercury can accidentally fall on the ground and pollute it; likewise another quantity of mercury can evaporate at the temperature and immediately pollutes the air and subsequently the soil by wet or dry atmospheric deposition. In addition, open-air incineration of gold-mercury amalgam can generate mercury vapor that precipitates in the form

of acid rain or can be spread by the action of wind and contaminate soils, plants, aquatic plants, stream, and the food chain [4].

Mercury is poorly mobile in soils and is rapidly immobilized by reactions with metal oxides (iron, aluminum and manganese) and especially with organic matter. It can undergo microbial or chemical methylation reactions, with formation mainly of monomethylmercury, which will promote its bioaccumulation. These processes are mainly controlled by sulphide ion concentration, pH, oxidation-reduction potential, cation exchange capacity (CEC), chloride concentration, aeration, soil mineral composition, soil texture and the amount of organic matter [5]. In recent times, a number of studies have focused on assessing the impacts of mercury generated during gold panning on soils in several countries [6]. These studies have revealed that mercury-induced environmental pollution from gold panning can become a major source of mercury pollution of soil in Korea [7] and [8], in the United States of America [9], from Brazil [10], Indonesia [11], Mexico [12], Senegal [13], Ghana [14], etc.

Curiously, at present, there are no gold washing sites in the province of South Kivu in general, and those in the territory of Fizi in particular, with information on the level of mercury pollution.

However, the soil quality of gold panning sites in the Fizi territory is deteriorating, especially since in this region, gold-mercury amalgam (Au-Hg) is incinerated in the open air on gold panning sites.

The effluents resulting from the artisanal mining of gold are dumped on the soils of gold panning sites. Thus, this study wants to fill these gaps mentioned above. Its main purpose is to assess the degree of mercurial pollution of the soils of gold panning sites in Fizi territory using total mercury content and indices of mercury pollution, including the mercury enrichment factor (EF), mercury contamination factor (CF), the mercury geoaccumulation index (Igeo), the ecological potential risk factor for mercury (EPRF) and the ecological risk index for mercury (ERI).

## 2. Material and Methods

### 2.1. Study area

This study was carried out on soils sampled from 8 gold panning sites in Fizi territory (Misisi, Lubichako I, Lubichako II, Tulonge, Ngalula, Kuwa, Makungu and Nyangé) and a reference site or non-mining site called Lulimba.

This territory is part of eight territories that make up the province of South Kivu located east of the Democratic Republic of Congo (DR Congo).

Geographically, the territory of Fizi is situated between 3 ° 30 'and 4 ° 51' 32 " South latitude, 27 ° 45 'and 29 ° 14' 10 " longitude East.

It is limited to the North by the territory of Uvira, to the West by the territory of Mwenga and Shabunda, to the South by the territory of Kalemie (Tanganyika), and finally to the East by Lake Tanganyika [15]. The information related to the location of different points of soil sampling in the sites studied is highlighted in Figure 1.

### 2.2. Methods of sampling and analysis of sediment samples

The samples of the studied soils were collected during 4 Campaigns (August 2016, December 2016, August 2017 and December 2017). The choice of soil sample collection sites was dictated by the presence or absence of pollution sources capable of modifying the soil quality of the artisanal gold mining areas of Fizi territory or those which are far from gold washing sites.

For each sampling point chosen, soil samples were collected every first two weeks of the month of August (in the dry season) and December (in the rainy season) of 2016 and 2017.

A clump was taken at a depth of 20 cm by pushing the soil probe in the direction of progress of a screw from left to right while turning it in the opposite direction.

This sampling operation was repeated twice and the samples made were mixed to form a composite sample that reduces the variation of the results and the uncertainty surrounding the average value obtained during the laboratory analysis of the desired parameters.



**Figure 1:** Map of sampling points for soil samples at non-mining and mining sites in Fizi territory

The resulting composite soil core was packaged in a clean 500 mL plastic box that had been labeled in advance. All boxes containing the composite soil core were stored in situ at 4 ° C in coolers before being sent to the laboratory for analysis.

The measurements that made it possible to evaluate the degree of mercurial pollution of the soils collected in the studied sites concerned six variables including the total mercury content as well as the indices of pollution or mercurial contamination such as the mercury enrichment factor (EF), mercury contamination factor (CF), mercury geoaccumulation index (Igeo), ecological potential risk factor for mercury (EPRF) and mercury ecological risk index (ERI).

Total mercury content was determined using Atomic Absorption Spectrophotometry (AAS). In addition, the Enrichment Factor (EF) was calculated by the formula below:

$$EF = \frac{[Me]_{mes} / [Al]_{mes}}{[Me]_{Th} / [Al]_{Th}}$$

With,  $[Me]_{mes}$  = the concentration of the element dosed (Hg for this case) in the soil sample taken from sites where the anthropic activities were carried out.

$[Al]_{mes}$  = the concentration of aluminum in the soil sample taken from sites where human activities were carried out.

$[Me]_{Th}$  = the concentration of the element dosed (Hg for this case) in the soil sample of the reference site, natural site or site without human activities. This is, in a way, the reference concentration of the dosed element (Hg for this case). This content is usually called the content of the natural background.

$[Al]_{th}$  = the concentration of aluminum in the soil sample of the reference site, natural site or site without human activities.

In the case of this study, the chosen reference site is indeed the village of Lulimba where no gold panning activity is carried out.

Hence, the total mercury and aluminum levels of arable soil in this field were considered reference concentrations to determine the mercury enrichment factor in all river sediment samples from artisanal mining sites in the Fizi territory.

In addition, it should be noted that the aluminum element has been used for geochemical normalization as usual for the following reasons: aluminum, generally measured, is sparingly soluble and, until now, unaffected by pollution. In addition, this element is generally considered conservative.

Thus, the pollution intensity scale based on enrichment factor (EF) interval values has five classes:  $EF \leq 2$  (no or low anthropogenic enrichment);  $2 < EF < 5$  (Moderate enrichment);  $5 < EF < 20$  (Significant enrichment);  $20 < EF < 40$  (very strong enrichment) and  $EF \geq 40$  (extreme enrichment) [16].

The geoaccumulation index ( $I_{geo}$ ) was calculated using the formula proposed by Müller in 1979:

$$I_{geo} = \log_2 \left( \frac{[Me]_{mes}}{[Me]_{Th}} \times 1,5 \right)$$

The coefficient 1.5 is a correction factor that takes into account natural fluctuations in the content of a given metal that can be attributed to mineralogical changes in the soil.

This geoaccumulation index is associated with a pollution scale of seven classes below:  $I_{geo} \leq 0$  (Unpolluted Soils or Background);  $0 < I_{geo} < 1$  (unpolluted to moderately polluted soil);  $1 < I_{geo} < 2$  (moderately polluted soils);  $2 < I_{geo} < 3$  (moderately to heavily polluted soils);  $3 < I_{geo} < 4$  (highly polluted soils);  $4 < I_{geo} < 5$  (heavily to highly polluted soils);  $I_{geo} > 5$  (extremely polluted soils) [17].

The fact factor for mercury contamination was determined by the following relationship:

$$CF = \frac{C_{Hg}}{B_{Hg}}$$

With, CF = Mercury contamination factor;  $C_{Hg}$  = mercury concentration in soil samples from anthropized sites;  $B_{Hg}$  = Concentration of mercury in the soil sample of the reference site, natural site or site without possible human activities (soil of Lulimba village).

This is actually the reference concentration of this metal element. The mercury contamination factor is related to a four-class pollution scale:  $CF < 1$  (contamination / pollution absent or absent);  $1 \leq CF < 3$  (contamination / moderate pollution);  $3 \leq CF < 6$  (concomitant contamination / pollution);  $6 \leq CF$  (very high contamination / pollution) [18].

The ecological potential risk factor (EPRF) was determined by the product of the values of the mercury toxic response factor with those of the mercury contamination factor. Hence the notation below:  $EPRF = Tf_{Hg} \times CF$  [18].

With  $Tf_{Hg}$  = mercury toxic response factor = 40; CF = Mercury contamination factor. It should be noted that the ecological potential risk factor for mercury is related to a pollution scale of five classes:  $EPRF < 40$  (contamination / pollution with low ecological risk);  $40 \leq EPRF < 80$  (contamination / pollution with moderate potential

ecological risk);  $80 \leq \text{EPRF} < 160$  (contamination / pollution with considerable potential ecological risk);  $160 \leq \text{EPRF} < 320$  (contamination / pollution with high potential ecological risk);  $\text{EPRF} > 320$  (contamination / pollution with a very high ecological risk) [19].

Finally, the ecological risk index of mercury (ERI) was determined via the sum of the values of the ecological potential risk factors of the metallic element [20]. Hence the following relation:

$$\text{ERI} = \sum_{i=1}^n \text{EPRF}_i$$

The ecological risk index for mercury is collected on a pollution scale of four classes:  $\text{ERI} < 150$  (contamination / pollution with low ecological risk);  $150 \leq \text{ERI} < 300$  (contamination / pollution with moderate ecological risk);  $300 \leq \text{ERI} < 600$  (contamination / pollution with considerable ecological risk);  $\text{ERI} \geq 600$  (contamination / pollution with a very high ecological risk) [19].

### 2.3. Statistical treatment of data

The data was encoded and processed using Microsoft Excel 2010. R version 2.15.1 software was used for descriptive data analysis while XLSTAT 2016 was used to separate the means of different factors at the level of significance of 5 %.

## 3. Results and discussion

### 3.1. Total mercury content (HgT) in the soils studied

The results in relation to the total mercury content in the soils of the sites studied are presented in Table 1. The results presented in Table 1, reveal that with the exception of the soils of the Lulimba reference site (non-mining site), the soils of all the artisan gold mining sites in the Fizi territory have been polluted by mercury from as much as their total mercury levels have exceeded the required standards by a high of  $0.5 \text{ mg.Kg}^{-1}$ .

In addition, the total mercury concentrations of the soils of the sites studied varied significantly from one site to another (P-value = 0.000).

Thus, with the exception of the Lulimba reference site, whose soils had the lowest mean total mercury levels ( $0.042 \pm 0.032 \text{ mg.Kg}^{-1}$ ), the soils of the Misisi gold panning site showed the average mercury concentration highest in relation to those of other gold washing sites ( $22.401 \pm 0.652 \text{ mg.Kg}^{-1}$ ) alternately followed by soils of the gold panning sites of Nyangé ( $19.618 \pm 0.556 \text{ mg.Kg}^{-1}$ ) and Makungu ( $18.427 \pm 0.554 \text{ mg.Kg}^{-1}$ ) with mean total mercury concentrations statistically identical; Lubichako I ( $16.056 \pm 0.458 \text{ mg.Kg}^{-1}$ ); Lubichako II ( $13.771 \pm 0.318 \text{ mg.Kg}^{-1}$ ); Kuwa ( $11.548 \pm 0.194 \text{ mg.Kg}^{-1}$ ); Ngalula ( $8.775 \pm 0.162 \text{ mg.Kg}^{-1}$ ); Tulonge ( $6.953 \pm 0.026 \text{ mg.Kg}^{-1}$ ).

In the same way, the total mercury concentrations of the soils of the Fizi sites studied also varied in a highly significant way from one investigation Campaign to another (P-value = 0.000) while increasing considerably from the Campaign 1 (August 2016) in Campaign 4 (December 2017). The Campaign 4 (December 2017) showed the highest average total mercury concentrations for all the sites studied above ( $19.335 \pm 0.353 \text{ mg.Kg}^{-1}$ ) followed respectively by the Campaign 3 (August 2017) for which the average levels of total mercury are of the order of  $14.805 \pm 0.335 \text{ mg.Kg}^{-1}$ , the Campaign 2 (December 2016), for which the average total mercury concentrations were  $9.870 \pm 0.324 \text{ mg.Kg}^{-1}$  and lastly, the Campaign 1 (August 2016) for which the average total mercury concentrations were of the order of  $8.253 \pm 0.300 \text{ mg.Kg}^{-1}$ .

These results are clearly in agreement with those of other researchers, namely [21], [12] and [22] stipulating respectively that the levels of total mercury varied very significantly according to the sites and Campaigns of investigations while being very derisory in the soils of non-mining sites (reference sites) and very high in the soils of gold panning sites in the Kédougou region (Eastern Senegal), artisanal gold mining sites in the North-West Central Mexico as well as gold mining sites in the central region of Mongolia.

**Table 1:** Total mercury content (HgT) in the soils studied

The results in relation to the total mercury content in the soils studied are presented in Table 1.

Sites / campaigns and Parameter	Kuwa	Lubichako I	Lubichako II	Lulimba*	Makungu	Misisi	Ngalula	Nyangé	Tulonge	Mean
Hg T										
(mg.Kg <sup>-1</sup> )										
				P-value for the Rivers : 0.000		p-value forthe Campaigns : 0.000				
Campaign 1	7.298±0.124	10.136±0.425	8.743±0.300	0.025±0.010	11.618±0.532	14.166±0.636	5.580±0.149	12.382±0.521	4.332±0.007	8.253±0.300d
Campaign 2	8.683±0.200	12.155±0.463	10.421±0.319	0.046±0.032	13.952±0.547	16.938±0.647	6.639±0.158	14.852±0.541	5.144±0.010	9.870±0.324c
Campaign 3	13.103±0.214	18.223±0.470	15.612±0.321	0.047±0.042	20.925±0.562	25.424±0.658	9.941±0.165	22.280±0.567	7.689±0.015	14.805±0.335b
Campaign 4	17.108±0.238	23.709±0.475	20.308±0.331	0.050±0.046	27.211±0.574	33.077±0.667	12.941±0.178	28.960±0.596	10.647±0.071	19.335±0.353a
Mean	11.548±0.194e	16.056±0.458c	13.771±0.318d	0.042±0.032h	18.427±0.554b	22.401±0.652a	8.775±0.162f	19.618±0.556b	6.953±0.026g	13.066±0.328

\* : Non-mining site (reference site); averages with the same letters on the line or column are not statistically different at the significance level alpha = 0.05; Campaign 1: August 2016; campaign 2: December 2016; Campaign 3: August 2017; Campaign 4: December 2017; HgT: total mercury content

### 3.2. Mercury Enrichment Factor (EF) Values

The results for mercury enrichment factors (EF) are shown in Table 2.

**Table 2:** Mercury Enrichment Factor (EF) Values

Sites / campaigns and Parameter	Kuwa	Lubichako I	Lubichako II	Lulimba*	Makungu	Misisi	Ngalula	Nyangé	Tulonge	Mean
EF	P-value for the Rivers : 0.000				p-value for the Campaigns : 0.000					
Campaign 1	8.767±0.711	12.687±0.507	11.600±0.397	0.343±0.038	13.630±0.514	10.556±0.814	6.784±0.760	18.350±0.400	5.427±0.756	9.794±0.544c
Campaign 2	10.359±0.736	13.614±0.512	12.687±0.412	0.354±0.114	14.524±0.519	77.085±0.836	8.217±0.774	15.671±0.421	6.623±0.748	17.682±0.564bc
Campaign 3	22.874±0.742	26.879±0.532	25.474±0.417	0.364±0.122	28.376±0.522	23.969±0.841	21.880±0.780	31.672±0.439	21.662±0.732	22.572±0.570b
Campaign 4	46.003±0.745	53.878±0.541	50.887±0.428	0.375±0.123	55.669±0.532	48.588±0.856	43.562±0.784	58.707±0.426	44.761±0.724	44.714±0.573a
Mean	22.001±0.734b	26.765±0.523ab	25.162±0.414ab	0.359±0.099c	28.050±0.522ab	40.050±0.837a	20.111±0.774b	31.100±0.422ab	19.618±0.740b	23.691±0.563

\* : Non-mining site (reference site); averages with the same letters on the line or column are not statistically different at the significance level alpha = 0.05; Campaign 1: August 2016; campaign 2: December 2016; Campaign 3: August 2017; Campaign 4: December 2017; EF: Enrichment Factor

The results in Table 2 show that the average values of mercury enrichment factor in soils of all the sites studied were of the order of  $23.691 \pm 0.563$ . Nevertheless, it was found that the mercury enrichment factors of the sites studied (Lulimba, Misisi, Lubichako I, Lubichako II, Tulonge, Ngalula, Kuwa, Makungu and Nyangé) varied in a highly significant way in one site to the other ( $P$ -value = 0.000). As a result, the soils of the Lulimba reference site (non-mining site) presented the lowest value of mercury enrichment factor ( $0.359 \pm 0.099$ ) relative to the soils of other sites studied. In addition, the soils at the Misisi gold panning site had the highest average value of the mercury enrichment factor compared with the average values of the mercury enrichment factors in the soils of other gold mining sites ( $40.050 \pm 0.837$ ) followed successively by the soils of the gold mining sites of Nyangé ( $31.100 \pm 0.422$ ), Makungu ( $28.050 \pm 0.522$ ), Lubichako I ( $26.765 \pm 0.523$ ) and Lubichako II ( $25.162 \pm 0.414$ ), whose enrichment factor values in mercury are statistically identical; and finally Kuwa ( $22.001 \pm 0.734$ ), Ngalula ( $20.111 \pm 0.774$ ) and Tulonge ( $19.618 \pm 0.740$ ) whose mercury enrichment factor values are statistically similar. In terms of the intensity of the mercurial pollution of these sites, it was noted that the anthropogenic mercury enrichment was non-existent or low in the soils of the Lulimba reference site (non-mining site), especially since their average value of enrichment factor was less than 2. In addition, anthropogenic mercury enrichment was respectively extreme in the soils of the Misisi gold panning site (as  $EF \geq 40$ ); very strong in the soils of gold mining sites of Nyangé, Makungu, Lubichako I, Lubichako II, Kuwa and Ngalula (because  $20 < EF < 40$ ); significant in the soils of the gold panning site of Tulonge.

Similarly, mean values of soil mercury enrichment factors at the study sites varied significantly from one Campaign to another ( $P$ -value = 0.000) while increasing significantly from the Campaign 1 (August 2016) to the Campaign 4 (December 2017). The Campaign 4 (December 2017) presented average values for the highest mercury enrichment factors for all sites studied ( $44.714 \pm 0.573$ ), followed respectively by the Campaign 3 (August 2017) for which the mean values of mercury enrichment factors are of the order of  $22.572 \pm 0.570$ ; the Campaign 2 (December 2016) for which the average values of mercury enrichment factor values were  $7.682 \pm 0.564$  and lastly, the Campaign 1 (August 2016) for which average values of mercury enrichment factors were  $9.794 \pm 0.544$ . These results are similar to those of other researchers such as [6] as well as [21] who respectively found that anthropogenic mercury enrichment was very low in the non-mining sites near gold panning sites in Mongolia and Kédougou while it was either extreme, very strong or significant in the soils of gold panning sites of Kédougou and Mongolia. They also found that the factors of anthropogenic mercury enrichment varied very significantly according to the gold washing sites and the study Campaigns.

In view of the results in Table 3, it appears that the average values of mercury geoaccumulation indices in the soils of the sites studied (Lulimba, Misisi, Lubichako I, Lubichako II, Tulonge, Ngalula, Kuwa, Makungu and Nyangé) were of the order of  $3.938 \pm 0.328$ . It has been noted that the values of the mercury geoaccumulation indices ( $I_{geo}$ ) in the soils of the studied sites varied in a highly significant manner from one site to another ( $P$ -value = 0.000). Thus, the soils of the Lulimba reference site (non-mining site) presented the average value of the lowest geoaccumulation index ( $-0.667 \pm 0.060$ ) relative to the soils of the other sites studied. In addition to this, the soils of the Misisi gold panning site had the highest average value of the highest mercury geoaccumulation index ( $I_{geo}$ ) compared with the average values of mercury geoaccumulation indices from soils at other sites gold panning ( $4.909 \pm 0.714$ ) alternately followed by soils of the gold panning sites of Nyangé ( $4.748 \pm 0.398$ ) and Makungu ( $4.708 \pm 0.319$ ) whose mercury geoaccumulation index values are statistically similar; then Lubichako I ( $4.555 \pm 0.316$ ) and Lubichako II ( $4.475 \pm 0.480$ ) whose values for mercury geoaccumulation indices are statistically the same; Kuwa ( $4.327 \pm 0.091$ ); Ngalula ( $4.221 \pm 0.332$ ) and Tulonge ( $4.163 \pm 0.238$ ), whose mercury geoaccumulation index values are statistically identical.

Concerning the intensity of the mercurial pollution of these sites, it was noted that the soils of the Lulimba reference site (non-mining site) were not polluted by mercury because the average value of their geoaccumulation indices mercury was far less than zero. In addition, the soils of all gold mining sites in Misisi, Nyangé, Makungu, Lubichako I, Lubichako II, Kuwa, Ngalula and Tulonge were highly polluted ( $4 < I_{geo} < 5$ ). In addition, the average values of soil mercury geoaccumulation indices in the study sites also varied significantly from one investigation Campaign to another ( $P$ -value = 0.000) while undergoing a considerable increase in Campaign 1 (August 2016) to Campaign 4 (December 2017).

### 3.3. Values of the mercury geoaccumulation index (Igeo)

The results in relation to the soil mercury geoaccumulation index values are presented in Table 3.

**Table 3:** Values of the mercury geoaccumulation index (Igeo)

Sites / campaigns and Parameter	Kuwa	Lubichako I	Lubichako II	Lulimba*	Makungu	Misisi	Ngalula	Nyangé	Tulonge	Mean
Igeo	P-value for the Rivers : 0.000				p-value for the Campaigns : 0.000					
Campaign 1	3.143±0.017	3.393±0.278	3.351±0.432	-0.621±0.003	3.407±0.275	3.737±0.700	3.172±0.218	3.387±0.352	3.126±0.200	2.899±0.275c
Campaign 2	3.337±0.031	3.583±0.310	3.481±0.475	-0.657±0.014	3.627±0.315	3.664±0.712	3.212±0.248	3.601±0.400	3.095±0.214	2.994±0.302c
Campaign 3	4.197±0.157	4.275±0.320	4.236±0.500	-0.677±0.117	4.447±0.325	4.548±0.719	4.148±0.568	4.800±0.418	4.097±0.265	3.786±0.377b
Campaign 4	6.632±0.160	6.970±0.354	6.833±0.511	-0.713±0.126	7.350±0.362	7.685±0.726	6.351±0.295	7.203±0.422	6.332±0.274	6.071±0.359a
Mean	4.327±0.091bc	4.555±0.316abc	4.475±0.480abc	-0.667±0.06d	4.708±0.319ab	4.909±0.714a	4.221±0.332c	4.748±0.398ab	4.163±0.238c	3.938±0.328

\* : Non-mining site (reference site); averages with the same letters on the line or column are not statistically different at the significance level  $\alpha = 0.05$ ; Campaign 1: August 2016; campaign 2: December 2016; Campaign 3: August 2017; Campaign 4: December 2017; Igeo: mercury geoaccumulation index

In fact, the Campaign 4 (December 2017) presented average values of the mercury geoaccumulation indices which were highest for all the sites studied ( $6.071 \pm 0.359$ ), followed successively by Campaign 3 (August 2017) for which the average values of mercury geoaccumulation indices were of the order of  $3.786 \pm 0.377$ ; the Campaign 2 (December 2016) with mean values of geoaccumulation indices equal to  $2.994 \pm 0.302$  and finally the Campaign 1 (August 2016) for which the average values of the mercury geoaccumulation indices were of the order of  $2.899 \pm 0.275$ . These results are similar to those of [6] as well as [21] stipulating that the values of the mercury geoaccumulation indices in the soils of the sites studied varied very significantly both according to the sites and the investigation Campaigns. Similarly, they reported that the soils of the non-mining sites (reference sites) close to gold washing sites in Mongolia and Kédougou were not polluted by mercury while the soils of all the gold panning sites Mongolia and Kédougou have been highly polluted.

The results reported in Table 4 show that the average mercury contamination factor values in the soils of the above sites were  $6.699 \pm 0.271$ . It was noted that the mercury contamination factor values in the soils of the study sites varied significantly from site to site ( $P$ -value = 0.000). In fact, the soils of the Lulimba reference site (non-mining site) had the lowest mean value of mercury contamination factor ( $0.071 \pm 0.006$ ) compared to the soils of the other sites studied. In addition, the soils at the Misisi gold panning site had the highest average mercury contamination factor (CF) value compared to the mean values for mercury contamination factors in the soils of other gold mining sites ( $9.243 \pm 0.791$ ) followed successively by the soil of Nyangé gold mining sites ( $8.254 \pm 0.706$ ), Makungu ( $7.816 \pm 0.540$ ), Lubichako I ( $7.501 \pm 0.236$ ), Lubichako II ( $7.340 \pm 0.032$ ), Kuwa ( $6.962 \pm 0.038$ ), Ngalula ( $6.702 \pm 0.014$ ) and Tulongé ( $6.402 \pm 0.079$ ). With respect to the mercury contamination gradient of the sites studied, it should be noted that the mercury contamination gradient was zero in the soils of the Lulimba reference site (non-mining site) because their average value of contamination factor was less than 1. However, the mercury contamination gradient was very high in the soils of all gold mining sites in Misisi, Nyangé, Makungu, Lubichako I, Lubichako II, Kuwa, Ngalula and Tulongé (since  $6 \leq CF$ ).

In addition, the mean values of soil mercury contamination factors in the study sites also varied significantly from one investigation Campaign to another ( $P$ -value = 0.000) while experiencing considerable increases in Campaign 1 (August 2016) to Campaign 4 (December 2017). Indeed, Campaign 4 (December 2017) presented average values of the mercury contamination factors which were highest for all the sites studied ( $10.214 \pm 0.288$ ) followed respectively by Campaign 3 (August 2017) for which the average values of the mercury contamination factors were in the order of  $8.519 \pm 0.278$ , from Campaign 2 (December 2016), whose average values of mercury contamination factors were equal to  $4.725 \pm 0.265$  and finally from Campaign 1 (August 2016) for which the average values of the mercury contamination factors were of  $3.337 \pm 0.254$ . These findings are similar to those of [23] as well as [12] stipulating that the values of the mercury contamination factors in the soils of the studied sites varied very significantly successively according to the sites and the investigation campaigns. These researchers also reported that the mercury contamination gradient was zero in the soils of the non-mining sites (reference sites) surrounding the gold panning sites in the North Central region of Mexico, while the mercury contamination gradient was very strong in the soils of all gold panning sites in the North Central region of Mexico.

In view of the results shown in Table 5, the average values of the potential ecological risk factor relating to mercurial pollution in soils of the sites studied was  $162.548 \pm 0.200$ . It was also found that the values of the ecological potential risk factor for mercury (EPRF) relating to the mercurial pollutions in the soils of the studied sites varied in a highly significant manner from one site to another ( $P$ -value = 0.000).

In fact, the soils of the Lulimba reference site (non-mining site) had on average the lowest ecological potential risk factor for mercury value related to mercury pollution ( $2.822 \pm 0.086$ ) compared to the soils of the other sites studied. In addition, the soils of the Misisi gold mining site had the highest average value of the ecological potential risk factor ( $207.070 \pm 0.054$ ) compared to the average values of the ecological potential risk factor to the mercury pollution of the soils of other gold panning sites followed successively by the soils of the gold panning sites of Nyangé ( $197.258 \pm 0.260$ ), Makungu ( $191.182 \pm 0.149$ ) and Lubichako I ( $185.782 \pm 0.199$ ) whose average EPRF values are statistically identical; and Lubichako II ( $173.267 \pm 0.169$ ), Kuwa ( $170.297 \pm 0.149$ ), Ngalula ( $169.330 \pm 0.150$ ) and Tulongé ( $165.922 \pm 0.120$ ) whose EPRF values are statistically similar.

### 3.4. Mercury contamination factor (CF) in the studied soils

Information on mercury contamination factor (CF) values in the soils studied is shown in Table 4.

**Table 4:** Mercury Contamination Factor (CF) values in the studied soils

Sites / campaigns and Parameter	Kuwa	Lubichako I	Lubichako II	Lulimba*	Makungu	Misisi	Ngalula	Nyangé	Tulonge	Mean
CF	P-value for the Rivers : 0.000				p-value for the Campaigns : 0.000					
Campaign 1	3.288±0.029	3.453±0.225	3.355±0.022	0.068±0.002	3.741±0.513	5.805±0.774	3.202±0.010	3.865±0.687	3.259±0.022	3.337±0.254a
Campaign 2	5.267±0.035	5.338±0.237	5.289±0.030	0.070±0.004	5.436±0.512	5.491±0.776	5.143±0.013	5.459±0.700	5.029±0.080	4.725±0.265b
Campaign 3	8.694±0.042	9.638±0.239	9.408±0.035	0.072±0.007	10.105±0.547	11.891±0.800	8.266±0.015	10.891±0.714	7.708±0.100	8.519±0.278c
Campaign 4	10.599±0.046	11.573±0.242	11.306±0.039	0.075±0.010	11.983±0.587	13.786±0.812	10.196±0.018	12.800±0.725	9.611±0.114	10.214±0.288d
Mean	6.962±0.038cde	7.501±0.236bcd	7.340±0.032bcde	0.071±0.006f	7.816±0.540bc	9.243±0.791a	6.702±0.014de	8.254±0.706b	6.402±0.079e	6.699±0.271

\* : Non-mining site (reference site); averages with the same letters on the line or column are not statistically different at the significance level  $\alpha = 0.05$ ; Campaign 1: August 2016; campaign 2: December 2016; Campaign 3: August 2017; Campaign 4: December 2017; CF: mercury contamination factor

### 3.5. Ecological Risk Potential Risk for Mercury (EPRF) Values

The different values for the ecological potential risk factor for mercury (EPRF) are presented in Table 5.

**Tableau 5 :** Values for the ecological potential risk factor for mercury (EPRF)

Sites / campaigns and Parameter	Kuwa	Lubichako I	Lubichako II	Lulimba*	Makungu	Misisi	Ngalula	Nyangé	Tulonge	Mean
EPRF	P-value for the Rivers : 0.000				p-value for the Campaigns : 0.000					
Campaign 1	91.364±0.133	94.807±0.170	86.805±0.150	2.724±0.041	99.311±0.119	113.189±0.463	83.433±0.162	105.298±0.162	82.869±0.102	84.422±0.167a
Campaign 2	108.909±0.146	113.798±0.200	97.601±0.168	2.807±0.081	120.103±0.124	139.647±0.551	97.680±0.180	128.412±0.227	94.184±0.111	100.349±0.199b
Campaign 3	133.598±0.151	176.962±0.210	166.352±0.172	2.837±0.105	181.488±0.132	196.462±0.563	160.581±0.124	186.462±0.312	160.980±0.127	151.747±0.211c
Campaign 4	347.316±0.166	357.563±0.217	342.308±0.185	2.921±0.118	363.824±0.142	378.982±0.571	335.624±0.127	368.859±0.327	325.653±0.147	313.672±0.222d
Mean	170.297±0.149b	185.782±0.19ab	173.267±0.169b	2.822±0.086c	191.182±0.15ab	207.070±0.54a	169.330±0.15b	197.258±0.26ab	165.922±0.12b	162.548±0.200

\* : Non-mining site (reference site); averages with the same letters on the line or column are not statistically different at the significance level  $\alpha = 0.05$ ; Campaign 1: August 2016; campaign 2: December 2016; Campaign 3: August 2017; Campaign 4: December 2017; EPRF: ecological potential risk factor for mercury

Concerning the assessment of the degree of mercurial pollution of soils of the sites studied based on the average values of potential ecological risk factors, it should be noted that the potential ecological risk relating to mercurial pollution of soils of the Lulimba reference site (non-mining site) was low because the average EPRF of these soils was less than 40. In addition, the level of ecological potential risk factor related to mercurial pollution of the soils of all gold mine sites of Misisi, Nyangé, Makungu, Lubichako I, Lubichako II, Kuwa, Ngalula and Tulongwe were raised (because  $160 \leq \text{EPRF} < 320$ ).

Nevertheless, the average values of the ecological potential risk factor for mercury relating to the mercurial pollutions of the soils of the sites studied also varied in a highly significant manner from one investigation Campaign to another ( $P\text{-value} = 0.000$ ) while undergoing increases from Campaign 1 (August 2016) to Campaign 4 (December 2017). In fact, Campaign 4 (December 2017) presented average values of the ecological potential risk factor for the highest mercury pollution for all the sites studied ( $313.672 \pm 0.222$ ), followed successively by Campaign 3 (August 2017) for which these values were of order  $151.747 \pm 0.211$ ; Campaign 2 (December 2016) with average EPRF values of  $100,349 \pm 0,199$  and finally of Campaign 1 (August 2016) for which the average EPRF values were  $84,422 \pm 0,167$ . These findings are consistent with those [12] and [22] indicating that the values of potential ecological risk factors for mercurial pollutions in the soils of the sites studied varied very significantly respectively according to sites and surveys. These researchers also reported the low level of potential ecological risk related to mercurial pollution of soils in non-mining sites (reference sites) close to gold washing sites in the North Central region of Mexico and those in the Central Region of Mongolia. However, the level of potential ecological risk related to mercurial pollution of the soils of all gold panning sites in the North Central region of Mexico and those in the Central Region of Mongolia has been high.

Based on the results presented in Table 6, it can be seen that the average value of the ecological risk index in relation to the mercurial pollutions in the soils of the studied sites was  $460.258 \pm 2.940$ . It has been observed that the values of the ecological risk indices (ERI) related to the mercurial pollutions in the soils of the studied sites varied in a highly significant way from one site to another ( $P\text{-value} = 0.000$ ). Thus, the soils of the Lulimba reference site (non-mining site) had, on average, the smallest value of the ecological risk index (ERI) for mercury pollution ( $33.240 \pm 1.550$ ) relative to the soils of the other sites studied. In addition to this, the soils of the Misisi gold mining site had the highest average value of the ecological risk index for mercury pollution ( $585.371 \pm 3.580$ ) compared with the average values of the ecological risk indices relating to mercury pollution of soils of other gold washing sites followed successively by the soils of the gold panning sites of Nyangé ( $552.532 \pm 3.600$ ); then Makungu ( $518.296 \pm 3.020$ ) and Lubichako I ( $507.841 \pm 2.940$ ) whose average values of the ecological risk indices are statistically identical; then Lubichako II ( $502.400 \pm 3.070$ ), Kuwa ( $494.086 \pm 2.690$ ) and Ngalula ( $477.618 \pm 3.550$ ) whose average values of ecological risk indices are statistically similar; finally Tulongwe ( $472.942 \pm 2.440$ ).

Concerning the assessment of the level of mercurial pollution of the soil of the sites studied based on average values of the ecological risk indices, it was pointed out that the ecological risk related to mercurial pollution of soils of the Lulimba reference site (site non-mining) was low because the average value of the ecological risk index of these soils was well below 150. Moreover, the level of ecological risk in relation to the mercury pollution of the soils of all gold panning sites Misisi, Nyangé, Makungu, Lubichako I, Lubichako II, Kuwa, Ngalula and Tulongwe was considerable (because  $300 \leq \text{ERI} < 600$ ).

However, the average values of the ecological risk indices for mercurial pollutions of the soils of the studied sites also varied in a highly significant way from one investigation campaign to another ( $P\text{-value} = 0.000$ ) while amply increasing the Campaign 1 (August 2016) to Campaign 4 (December 2017). The Campaign 4 (December 2017) presented average values of the ecological risk indices for the highest mercury pollution for all the studied sites ( $585.440 \pm 3.170$ ) followed alternatively by the Campaign 3 (August 2017) for which these values were about  $538.139 \pm 3.040$ ; the Campaign 2 (December 2016) with average values of ecological risk indices equal to  $383.890 \pm 2.860$  and finally the Campaign 1 (August 2016) for which the average values of ecological risk indices were equal to  $333.564 \pm 2.670$ . These observations are in agreement with those of [21], [12] and [22] which show that the values of ecological risk indices (ERI) in relation to the mercurial pollutions in the soils of the sites studied have varied very significantly successively in depending on the sites and study campaigns.

### 3.6. Values of the ecological risk index of mercury (ERI)

The data that relate to the mercury ecological risk index values are presented in Table 6.

**Table 6:** Values of the ecological risk index of mercury (ERI)

Sites / campaigns and Parameter	Kuwa	Lubichako I	Lubichako II	Lulimba*	Makungu	Misisi	Ngalula	Nyangé	Tulonge	Mean
ERI	P-value for the Rivers : 0.000				p-value for the campaigns : 0.000					
Campaign 1	350.316±2.120	373.199±2.762	360.517±2.870	31.699±1.329	380.292±2.900	369.328±3.140	320.248±3.325	506.251±3.475	310.231±2.112	333.564±2.67b
Campaign 2	420.595±2.280	443.584±2.967	430.730±3.000	32.651±1.423	455.703±2.968	439.728±3.623	390.694±3.589	460.573±3.574	380.754±2.312	383.890±2.86b
Campaign 3	595.477±3.014	592.713±3.007	602.457±3.114	33.797±1.651	611.841±3.100	607.875±3.715	593.327±3.647	615.786±3.652	589.974±2.471	538.139±3.04a
Campaign 4	609.954±3.300	621.867±3.015	615.896±3.315	34.812±1.785	625.349±3.120	924.551±3.829	606.204±3.653	627.519±3.693	602.809±2.852	585.440±3.17a
Mean	350.316±2.120	373.199±2.762	360.517±2.870	31.699±1.329	380.292±2.900	369.328±3.140	320.248±3.325	506.251±3.475	310.231±2.112	333.564±2.67b

\* :Non-mining site (reference site); averages with the same letters on the line or column are not statistically different at the significance level alpha = 0.05; Campaign 1: August 2016; campaign 2: December 2016; Campaign 3: August 2017; Campaign 4: December 2017; ERI: ecological risk index of mercury

They also showed that the ecological risk related to mercurial pollution of soils of non-mining sites (reference sites) near the gold panning sites of the Kédougou, North Central Mexico and Central Mongolia regions was low. While the level of ecological risk related to the mercurial pollution of soils of all Kedougou, North-Central Mexico and Central Mongolia gold mining sites has been considerable.

## Conclusion

With the exception of the soils of the reference site of Lulimba (non-mining site), the soils of all gold panning sites in the Fizi territory are heavily polluted by mercury with regard to their mercury content and the values of their indices of mercurial pollution, namely the mercury enrichment factor (EF), the mercury contamination factor (CF), the mercury geoaccumulation index (Igeo), the ecological potential risk factor for mercury (EPRF) and the ecological risk index for mercury (ERI), which deviates from the standards recommended by the Canadian Council of Ministers of the Environment. The soils of the Misisi gold mining site were heavily polluted by mercury, followed respectively by the soils of the Nyangé, Makungu, Lubichako I, Lubichako II, Kuwa, Ngalula and Tulonge gold panning sites.

Given this high level of mercurial pollution of the studied soils, it is necessary to consider the best solutions likely to protect the terrestrial ecosystems of the artisanal mining sites of the territory of Fizi. Thus, it would be good to regularly educate gold miners on the management and handling of toxic chemicals such as mercury; to raise the awareness of gold miners in the Fizi territory on the promotion of the use of retorts or mercury recovery hoods, etc.

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