Mercury Pollution Linked To Gold Panning In DR Congo: Contamination Of Aquatic Systems And Health Impact On Residents

Jean-Noël Mputu Kanyinda, Nsambu Mukondwa Pascal, and Musibono Dieudonné

***Abstract* — The interactions between selenium (Se) and mercury (Hg) were assessed on fish, crabs, and molluscs to assess the risks to public health associated with dietary exposure to mercury from their consumption. To this end, mass concentrations of mercury and selenium have been determined in the edible tissues of three species of fish, in crabs and molluscs taken from the rivers of the gold zones of Fizi in South Kivu in the Democratic Republic of Congo. We found values greater than 1 µg/g Hg in all fish samples regardless of the river, but also that the Kimbi River was the most polluted with an average mercury content of about 5 µg/g. Crabs and molluscs also had Hg values greater than 1.**

***Index Terms* — Aquatic systems, Health impact, mercury, selenium, Se/Hg molar ratio.**

I. INTRODUCTION

Crabs, fish and shellfish are important sources of protein, essential amino acids, polyunsaturated fatty acids (omega-3 and omega-6), carbohydrates, glutamic acids, vitamins A, B, C and D and minerals (phosphorus, potassium, sodium, calcium, magnesium, zinc, copper, selenium, iron and iodine) in food [1]-[9]. They are involved in the prevention of cardiovascular disease, as well as in the development and functioning of the retina, brain and nervous system [10]. Aquatic organisms such as fish can accumulate amounts of mercury from their direct environment in their tissues [11]. This accumulation occurs either directly by adsorption through the skin and gill membranes during their respiration, or indirectly by ingestion of already contaminated prey [11]-[13]. Despite their benefits for human health, crabs, fish and molluscs can contain varying amounts of potentially toxic substances, especially heavy metals caused by water pollution [14]-[17].

The presence and dispersion of pollutants in the environment have been a concern for many years. Among the main environmental contaminants, metallic trace elements (ETM) pose serious ecological problems due to their ubiquitous nature, their toxicity and their bioaccumulation power in several animal species, notably aquatic ones (crabs, fish, molluscs, etc.). At low concentrations, mercury and its derivatives have no destructive effect on fish. High exposure causes damage to gill tissue, death occurs by asphyxiation. Aquatic flora can also concentrate the mercury present in its environment. The methylmercury ion and other organomercury compounds are known to be responsible for inhibiting the growth of phytoplankton at concentrations of the order of 0.1 µg/g [18].

Mercury (Hg) is one of the most dangerous contaminants that directly threaten our environment and our health. Methylmercury, formed from inorganic Hg in aquatic systems is toxic and bioaccumulative, excessive consumption of fish from its waters can be harmful to human health, especially on the brain and nervous system [6], [7], [19].

In the various gold panning sites of the Fizi territory in South Kivu, the waste is directly discharged into the rivers draining the sites, causing unprecedented mercury pollution. The work of Pascal et al. [20] were able to demonstrate the mercury contamination of the various rivers draining the gold panning sites. They also established a link between the various contaminations of rivers, soils and sediments by the activity of gold washers and that the residents had high mercury levels [10], [20]-[22].

Another trace element is selenium (Se), it is an essential micronutrient for mammals, it is naturally produced in the environment. Selenium is present as an active component of the 21st genetically encoded amino acid, which is necessary for the functioning of enzymes which are important for preventing and reversing oxidative damage, as well as regulating the thyroid hormone and calcium metabolism [23].

Excessive consumption of selenium (Se) can cause toxicity in the body, but in general, it is lower than the recommended daily dose, and its deficiency causes health problems. Selenium is also known to have the potential to prevent disease in humans, so low selenium consumption is essential for life. Selenium enters the aquatic food chain through the particles of selenium consumed by invertebrates and small fish. It is known as an effective agent to detoxify mercury (Hg) and to offer some protection against the toxicity of mercury (Hg) by reducing the accumulation of Hg in mammals and fish. The presence of selenium in the environment and food is considered to be a bioindicator of the harmful effects of [14], [17], [23], [24].

The Hg/Se molar ratios must be evaluated in order to

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interpret the effects of exposure to Hg in terms of food safety and evaluation of the risks of consumption of crabs, fish and molluscs for human health. Se/Hg molar ratios greater than 1 provide protection against Hg toxicity and reduce the harmful effects of Hg [23].

At present, there is no information on the concentration of total mercury (HgT), no data on the level of mercury (Hg) intoxication of these aquatic organisms (crabs, fish and molluscs) in rivers draining gold panning sites. In view of the above, it appears that the crabs, fish, and molluscs from different rivers draining the gold panning sites in the Fizi territory would be exposed to mercury contamination.

II. MATERIALS AND METHODS

A. Study zone

The samples of the fish studied were caught in nine rivers (Kimbi, Mandje, Misisi, Kimuti, Kacumvi, Kuwa, Lubichako, Etó and Makungu) which drain the gold panning sites of the Fizi territory. Fizi is one of the eight territories of the province of South Kivu in the Democratic Republic of the Congo. Geographically, this territory is located between 3 ° 30 'and 4 ° 51'32' south latitude, 27 ° 45 'and 29 ° 14' 10 " east longitude. This territory administratively shares borders with the territory of Uvira in the north, the territory of Mwenga and Shabunda in the west, the territory of Kalemie (Tanganyika) in the south and Lake Tanganyika in the east.

B. Materials

The choice of sampling sites for benthic macroinvertebrates and fish was based on the existence or not of sources of water pollution from the various rivers coming from the gold panning sites. Three fish species (Silurus sp, Oreochromis sp and Haplochromis sp), one species of crab and mollusk were sampled in 2018 from nine rivers in the territory of Fizi (Makungu, Misisi, Kuwa, Kimbi, Lubichako, Etó, Kimuti, Kacumvi and Mandje) in South Kivu in the Democratic Republic of the Congo. The crab, fish and mollusc samples were transported to the laboratory in polyurethane pots containing 10% formalin in a cooler at 4 °C. For fish specimens, length (cm) and weight (g) were measured in the laboratory. 5 g meat of crab, fish and mollusc flesh were stored separately in the freezer at -20 ° C in plastic jars.

C. Methods

First, the sample is digested in the presence of hydrochloric acid (HCl) and potassium permanganate ($KMnO_4$). This step breaks down organic matter and transforms mercury into Hg^{2+} form. Hydrochloric acid promotes the rapid decomposition of cinnabar (HgS), resistant to the attack of nitric acid and sulfuric acid. Potassium permanganate provides complete oxidation of organic compounds refractory to acid decomposition. During this stage, manganese is reduced from the Mn^{7+} form to the Mn^{4+} form. After digestion, a hydroxylamine chloride solution ($NH_2OH.HCl$) reduces MnO_2 and excess $KMnO_4$ without reducing Hg^{2+}. In the second step, the mercuric ions reduced to elemental mercury, by a solution of stannous chloride ($SnCl_2$) are brought in gaseous form into a cell by air bubbling. Mercury in the cell is measured by atomic absorption spectrophotometry SAA at a wavelength of 253.7 nm [10].

After thawing at room temperature, 0.5 g of sample (fish, crab and mollusk) are placed in different digestion tubes (100 mL) to which are successively added 4 ml of 65% pure HNO_3, 2 ml of pure H_2SO_4 98% and 1 mL of 37% pure HCl. The tubes are then placed in a water bath at 90±5 °C for 70 minutes. After the mineralization has cooled, 5 ml of the 6% $KMnO_4$ solution are added to it. After 30 minutes of reaction, 5 mL of 20% hydroxylamine hydrochloride are added to neutralize the excess of $KMnO_4$. Transfer it to 50 mL flasks and make up to the mark with distilled water. 5 ml of the filtrates of each solution prepared are introduced into separate vials and 1 ml of the stannous chloride dihydrate solution ($SnCl_2$. $2H_2O$) at 10% is added to each bottle. Atomic absorption spectroscopy type AA 500 equipped with a specific mercury lamp, with a wavelength of 253.7 nm was used for this purpose for the determination of mercury [10], [22].

D. Data analysis

All analyses were triplicated. Mean values and standard deviations were calculated and the results were presented in µg/g (ppm) wet weight. The molar concentrations (µmol/kg) of Hg and Se were calculated by dividing the concentration (µg/g) by the molecular weight (200.59 g/mol for Hg and 78.9 g/mol for Se). The molar ratio of Se to Hg was calculated by the following equation [10], [23]: Molar ratio (Se/Hg)=molar concentration of Se (µmol/kg) / molar concentration of Hg (µmol/kg).

III. RESULTS AND DISCUSSION

The information relating to the fish samples (*Silurus sp, Oreochromis sp* and *Haplochromis sp*) such as the length, the weight, the place of capture and the number of fish samples used are given in Table I. The mercury contents and in selenium (µg/g) as well as the Se/Hg molar ratio are presented in Table II. In Table III we have presented the mercury and selenium concentrations of the crabs and molluscs with the Se/Hg molar ratios. We observe that the fishes of the Kimbi and Lubichako rivers presented sizes and weights largely superior to the other rivers.

In relation to the results presented in Table I relating to the mass and size of the different fish (Silurus sp, Oreochromis sp and *Haplochromis sp*), we observe some differences by river. These different species of fish (*Silurus sp, Oreochromis sp* and *Haplochromissp*) captured in the Kimbi river had relatively high masses compared to the other rivers with respective mean values of 134.7±2.1 g, 58.8±0.7 g and 13.7±0.6 g, followed by the rivers Lubichako (120.9±2.3 g; 53.1±0.6 g; 11.6±0.5 g), Kuwa (93.7 g ± 1.3 g; 47.4±0,7 g; 10.5±0.6 g), Mandje (82.7±1.9 g; 43.3±0.9 g; 6.4±0.4 g), Misisi (80.5±1.8 g; 36.1±0.8 g; 8.1±0.8 g), Makungu (80.2±1.6 g; 44.4±0.6 g; 9.4±0, 5 g), Etó (61.1±0.7 g; 35.1±0.4 g; 7.4±0.3 g), Kacumvi (46.5±0.4 g; 39.2±0,6 g; 5.9±0.2 g) and Kimuti (37.9±0.4 g; 31.7±0.5 g; 4.1±0.1 g). The same observations are made for the sizes of these different species of fish. These variations in masses and sizes would be due to the diversity of the hydrological regimes of the coasts and to the effects of anthropic activities in the lands crossed by these rivers on the other hand.

TABLE I: RESULTS RELATING TO THE WEIGHT (G) AND THE SIZE (CM) OF THE FISH SAMPLES

Rivers	Sample (n)	*Silurus sp*		*Oreochromis sp*		*Haplochromis sp*	
		Weight (g)	Length (cm)	Weight (g)	Length (cm)	Weight (kg)	Length (cm)
Eto '	16	61.1 ± 0.7	5.9± 0.1	35.1 ± 0.4	4.9±0.5	7.4 ± 0.3	3.4±0.2
Kacumvi	22	46.5 ± 0.4	7.6± 0.1	39.2 ± 0.6	5.9±0.7	5.9 ± 0.2	4.4±0.2
Kimbi	17	134.7 ± 2.1	29.8± 1.1	58.8 ± 0.7	18.5±1.6	13.7 ± 0.6	9.7±0.8
Kimuti	17	37.9 ± 0.4	5.2± 0.2	31.7 ± 0.5	3.8±0.1	4.1 ± 0.1	2.9±0.1
Kuwa	18	93.7 ± 1.3	16.7± 1.4	47.4 ± 0.7	11.9±0.9	10.5 ± 0.6	8.6±0.6
Lubichako	23	120.9 ± 2.2	15.7± 1.2	53.1 ± 0.6	11.2±0.8	11.6 ± 0.5	7.7±0.6
Makungu	15	80.2 ± 1.6	13.0± 1.0	44.4 ± 0.6	9.3±0.9	9.4 ± 0.5	7.4±0.5
Mandje	19	82.7 ± 1.9	9.4± 1.1	43.3 ± 0.9	7.8±0.7	6.4 ± 0.4	5.3±0.6
Misisi	21	80.5 ± 1.7	9.5± 1.2	36.1 ± 0.8	6.2±0.6	8.1 ± 0.8	4.7±0.7

TABLE II: MASS CONCENTRATION AND MOLAR RATIOS OF MERCURY AND SELENIUM OF DIFFERENT FISH SPECIES

	Silurus sp			*Oreochromis sp*			*Haplochromis sp*		
	Mercury content (µg/g)	Selenium content (µg/g)	Molar ratios Se/Hg	Mercury content (µg/g)	Selenium content (µg/g)	Molar ratios Se/Hg	Mercury content (µg/g)	Selenium content (µg/g)	Molar ratios Se/Hg
Eto '	3.35 ± 0.13	3.77 ± 0.35	2.86	2.42 ± 0.18	2.84 ± 0.65	2.98	3.02 ± 0.13	3.44 ± 0.06	2.90
Kacumvi	2.61 ± 0.02	3.03 ± 0.16	2.95	1.88 ± 0.06	2.30 ± 0.07	3.11	2.34 ± 0.08	2.77 ± 0.05	3.01
Kimbi	8.28 ± 1.06	8.69 ± 0.34	2.67	5.95 ± 0.44	6.31 ± 0.53	2.70	7.46 ± 0.32	7.87 ± 0.43	2.68
Kimuti	1.57 ± 0.03	1.99 ± 0.04	3.22	1.14 ± 0.07	1.56 ± 0.02	3.48	1.42 ± 0.01	1.83 ± 0.03	3.28
Kuwa	2.16 ± 0.12	2.58 ± 0.07	3.04	1.55 ± 0.07	1.96 ± 0.08	3.21	1.93 ± 0.03	2.35 ± 0.03	3.10
Lubichako	3.04 ± 0.03	3.46 ± 0.37	2.89	2.19 ± 0.33	2.61 ± 0.65	3.03	2.74 ± 0.04	3.15 ± 0.06	2.92
Makungu	3.63 ± 0.25	4.05 ± 0.42	2.84	2.62 ± 0.20	3.05 ± 0.66	2.96	3.29 ± 0.07	3.68 ± 0.09	2.84
Mandje	1.89 ± 0.02	2.31 ± 0.06	3.11	1.36 ± 0.08	1.78 ± 0.08	3.33	1.69 ± 0.02	2.11 ± 0.03	3.17
Misisi	3.81 ± 0.27	4.22 ± 0.13	2.82	2.75 ± 0.17	3.17 ± 0.66	2.93	3.43 ± 0.07	3.84 ± 0.09	2.85

The average value of the total mercury content in fish Silurus sp caught in rivers (Makungu, Misisi, Kuwa, Kimbi, Lubichako, Etó, Kimuti, Kacumvi and Mandje) from gold panning sites in Fizi is greater than 1 with a maximum of approximately 8.3 µg/g of fish. As shown in Table II, fish in the Kimbi River have the highest concentration of mercury of all the other rivers. With an average of 8.28 µg/g followed respectively by fish from the rivers Misisi 3.81 µg/g, Makungu 3.65 µg/g, Eto' 3.05 µg/g, Lubichako 3.04 µg/g, Kacumvi 2.61 µg/g, Kuwa 2.16 µg/g, Mandje 1.89 µg/g and Kimuti with a concentration of 1.57 µg/g.

Concerning the species *Haplochromis sp* captured in our rivers, it also contains a high concentration of mercury as shown in Table II. It appears from our results that the fish of the Kimbi river are those which present the highest concentration in Hg than the fish of the other rivers studied. With an average of around 7.46 µg/g followed respectively by the Misisi rivers with an average concentration of 3.43 µg/g, Makungu 3.29 µg/g, Eto' 3.02 µg/g, Lubichako 2, 74 µg/g, Kacumvi 2.34 µg/g, Mandje with a content of 1.69 µg/g and the Kimuti river which has the highest concentration of the series with 1.42 µg/g. Similar work in Brazil, Turkey and Argentina has shown that benthic species can accumulate high concentrations of mercury [23], [25], [26].

As for the species Oreochromis sp, observations made on species *Silurus sp* and *Haplochromis* sp also seem identical to it. The Kumbi river remains the one with the highest Hg concentration in fish (5.95 µg/g) followed respectively by the Misisi rivers 2.75 µg/g, Makungu 2.62 µg/g, Eto' 2.42 µg/g, Lubichako 2.19 µg/g, Kacumvi 1.88 µg/g, Kuwa 1.55 µg/g, Mandje 1.36 µg/g and the Kimuti river remains the least polluted of all with a concentration of 1.14 µg/g. Although it contains high concentrations of mercury (Hg), this species has lower values than the two previous species, namely *Silurus sp* and *Haplochromis sp*. In 2006, Koffi et al showed that fish species from the Ivorian coast also contain mercury [17].

Referring to the quality standards of fishery products, it turns out that all the fish caught in the various rivers of the Fizi gold panning zones have a total concentration of Hg considerably exceeding the standards according to which recommends that the concentration in total mercury in fish cannot exceed 0.5 µg/g. Similar results were found by Nsambu et al. [10] who had studied the mercury contamination of fish from the Fizi gold panning areas [10].

Fish from the Kimbi River have the highest selenium content of all rivers with averages of 8.63±0.34 µg/g, 6.31±0.53 µg/g and 7.87±0,43 µg/g respectively for the species *Silurus sp*, *Oreochromis sp* and *Haplochromis sp*. As for the mercury content in the different rivers, the selenium (Se) content follows the same variations, with the Kimuti river which has the lowest selenium content with averages of 1.99±0.04 µg/g, 1.56±0.02 µg/g and 1.83±0.03 µg/g respectively for the species *Silurus sp*, *Oreochromis sp* and *Haplochromis sp*. Selenium being an indicator to lower the level of mercury in the body, we note that its content is relatively higher than that of mercury in our samples. It is a good thing to minimize the risk of mercury contamination. Similar results from high concentrations of selenium relative to mercury in fish have been found in tropical estuaries in southeast Brazil [25], in Turkish waters [23], in the Colorado River basin in the southwest of the United States [14].

It has been proven that an excess of Se over Hg confers a protective effect on fish. Selenium has a high binding affinity for Hg and MeHg, which leads to the formation of biologically unavailable Se-Hg precipitates. Glove et al. [27] suggested that a Se / Hg molar ratio greater than 1 largely protects the harmful effects of Hg and that risk assessments should consider Se/Hg ratios rather than tissue concentrations of Hg alone.

The results relating to the molar ratios of total selenium / total mercury (SeT/HgT) for the different fish species as a function of the rivers are presented in Table II. We have shown that not all the fish species caught in these different rivers constitute not a threat to human health, i.e. they are very safe for human consumption because their SeT/HgT molar ratios are greater than 1 as recommended by the standard [15], [27], [28]. With a ratio (SeT/HgT) greater than 1, the antagonistic effect occurs between mercury and selenium, the latter thus protecting aquatic organisms against the toxicity of mercury. Similar results have been found in previous work [14], [23]-[26].

TABLE III. MASS CONCENTRATION AND MOLAR RATIOS OF MERCURY AND SELENIUM IN CRABS AND MOLLUSCS

Sample		*Crabs*			*MolluscsLimnaea sp*		
		Mercury content (µg/g)	Selenium content (µg/g)	Molar ratios Se/Hg	Mercury content (µg/g)	Selenium content (µg/g)	Molar ratios Se/Hg
Eto '	12	1.93 ± 0.27	2.34 ± 0.27	3.22	1.54 ± 0.12	1.96 ± 0.12	3.24
Kacumvi	11	1.45 ± 0.15	1.92 ± 0.12	3.43	1.12 ± 0.15	1.62 ± 0.04	3.68
Kimbi	13	4.76 ± 0.90	5.18 ± 0.23	2.83	3.81 ± 0.88	4.23 ± 0.02	2.82
Kimuti	11	0.90 ± 0.19	1.32 ± 0.03	4.00	0.72 ± 0.01	1.14 ± 0.01	4.03
Kuwa	14	1.23 ± 0.14	1.65 ± 0.05	3.33	0.99 ± 0.03	1.40 ± 0.05	3.60
Lubichako	14	1.74 ± 0.19	2.16 ± 0.25	3.38	1.34 ± 0.11	1.83 ± 0.02	3.47
Makungu	15	2.08 ± 0.28	2.50 ± 0.28	3.10	1.67 ± 0.18	2.09 ± 0.01	3.18
Mandje	12	1.07 ± 0.14	1.49 ± 0.05	3.61	0.88 ± 0.02	1.30 ± 0.02	3.76
Misisi	16	2.20 ± 0.26	2.28 ± 0.24	2.79	1.76 ± 0.11	2.31 ± 0.03	3.34

In addition to the three species of fish studied previously, we also measured in parallel the selenium and the mercury in the crabs and molluscs *Limnaea sp* populating the various rivers because being part of the food habits of the neighboring populations. Regarding its results, we observe that all rivers are all polluted and that the Kimbi river remains the most polluted with an average of 4.76±0.90 µg/g of Hg in crabs and 3.81±0,88 µg/g in molluscs; as for the Kimuti river, it remains the least polluted with averages of 0.90 µg/g±0.01 µg/g of crabs and 0.72±0.01 µg/g of molluscs *Limnaea sp*. The Se content remains higher compared to that of Hg in the various samples (5.18±0.23 µg/g for the crabs and 4.23±0.02 µg/g for the molluscs of the Kimbi river). Our previous results had shown that the Kimbi river was the most polluted because it is the confluence of eight other rivers. Consequently, it turns out that all *Limnaea sp* crabs and molluscs populating the rivers of the Fizi gold panning regions do not pose any risk for human consumption, especially since their Se / Hg molar ratios are greater than 1 well. that the total mercury content greatly exceeds the normative limits.

IV. Conclusion

In the present study, crabs, fish species and molluscs are considerably important food sources, commonly consumed by local populations. It has been found that crabs, fish (*Silurus sp*, *Oreochromis sp* and *Haplochromis sp*) and *Limnaea sp* molluscs caught in different rivers of the Fizi panning areas have concentrations of total mercury considerably exceeding the standard according to which the value acceptable in fish and shellfish should not exceed 0.5 µg / g. In addition to this, *Silurus sp* fish had the highest total mercury levels compared to other fish followed by *Haplochromis sp* and *Oreochromis sp* fish respectively. However, not all of these fish species studied were found to pose a threat to human health because their molar ratios of total selenium and total mercury (TSe: THg) are at least greater than 1. In such circumstances, the antagonistic effect between mercury and selenium occurs, in that selenium protects aquatic organisms from mercury toxicity. In view of the above, it is necessary to combat mercury pollution of aquatic ecosystems in gold mining sites in the territory of in order to preserve both the health of aquatic organisms and the health of consumers of the above-mentioned fishery products. It would be important to train gold miners in the Fizi territory in the proper management of gold mining effluents, the promotion of modern artisanal gold mining techniques that significantly reduce environmental pollution, etc.

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