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Assessment of Water Contamination by Metallic Trace Elements at Mining Sites: The Case of the Ouham River in the Central African Republic

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Abstract In a context of sustainable management of natural and environmental resources, the preservation of water quality from the adverse effects of anthropogenic activities is of particular interest. Human activities that generate large amounts of waste containing toxic metals are found in the environment as a result of rainfall, atmospheric deposition, etc. Although naturally present at low concentrations in the earth's crust, these toxic trace metal elements accumulate in surface waters, sediments and soils and reach high concentrations, threatening aquatic life and human health. The aim of this work is to identify these elements in the vicinity of the mining zone in the Bozoum region of the Central African Republic in order to propose a depollution process using activated bricks as adsorbent support. The parameters measured at the four mine sites were as follows: the suspended solids were in the range of 276 to 504 mg/l, the water turbidity was in the range of 375 to 630 NTU, iron was present at concentrations in the range of 4.21 to 8.97 mg/l, and the abnormally high mercury was in the range of 4 to 26 µg/l, 26 times above the standard. The order of the degree of contamination of the water is thus established Hg > Fe > As > Zn > Cu > Pb=Cd=Mn. The activated brick filtration depollution process shows a good mercury retension capacity with a rate of abatement of 95.83% compared to sand with a rate of 12.5%.

Keywords: *activated brick, mercury, adsorbent, trace elements, Bozoum*

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1. Introduction

Water is a natural resource essential to the life of all living beings (animals and plants). Sufficient and high-quality consumption helps maintain health. Unfortunately, this vital commodity is often exposed to both natural and anthropogenic pollution. Water pollution is defined as any change in the composition (physical, chemical and biological) of its quality. The categories of chemical pollutants include organic and inorganic pollutants. Inorganic pollutants are mainly highly toxic inorganic compounds and heavy metals, including arsenic and mercury. The greatest public health threat from mercury and arsenic comes from contaminated groundwater. These metalloids are naturally found in trace amounts in soils, springs, wells, rivers to oceans and sediments [1]. Its accumulation may be linked to human activities (agriculture, extraction and mining, wood conservation, etc.) [1]. The mining sector is a mainstay of the Central African Republic's economy, with diamond and gold mining contributing 4% of the country's gross domestic product and nearly half of the country's total export value. The industrial exploitation of diamonds dates back to the 1930s. The country's independence in 1960 was marked by the rapid rise in mining activity and the decline in industrial exploitation. Today artisanal mining dominates the mining sector, producing all the gold and over 98% of the diamonds. If the Central African diamonds come exclusively from alluvial deposits more easily exploitable in a artisanal or semi-mechanized way. Gold, on the other hand, occurs both in alluvial deposits and in some large quartz veins in the form of rock nuggets. As a result, artisanal exploitation can take the form of vertical wells or riverbank construction (along rivers). Since the river bed is potentially mineralized, it can be reached by diverting all or part of the river (using dams or canals) or using mechanized dredges.

The region of Bozoum is marked by artisanal (mainly gold-bearing) exploitation traditionally rooted in the social and community landscape along the Ouham River (photo 1). Mining activities are carried out in parallel with agricultural activities and many villages have opened riverbank sites along their access to the river. Some villages also operate wells, away from the river, during the rainy season. Chinese companies in the Bozoum region seem to use mainly two operating techniques. On the one hand, they set up river diversion canals to explore the river bed with bulldozers and mechanical shovels.

Photo 1. Opportunistic artisanal exploitation on the banks of the canal dug by Chinese mining companies to divert the Ouham River and exploit its dry bed, near Bozoum

On the other hand, they built basins about 10 meters wide and 6 meters deep, perpendicular to the river, on the banks, whose function could not be established with certainty (photo 1). The satellite image shows the same area around Bozoum. The image clearly shows the consequences of mining activities along Ouham. The banks are highly visible due to recent and significant activities, probably made possible by heavy tools such as bulldozers or excavators.

2. Materials and Methods

2.1. Materials

2.1.1. Sample Site Presentation

Our work is defined in time and space. After an initial phase of field reconnaissance by the laboratory team during the campaign, Four sites were identified in the mining area to serve as a water sample for analysis. The geographical coordinates of the points are given in Table 1. The points are divided as follows: one sample in front of the operating area and three at the exit of each operating site. The collection, transport and storage of samples refer to the protocol defined by the French Standardization Agency (AFNOR, NF-T 90-10).

Table 1. Geographical coordinates of selected water points

Picking point	Longitude	Latitude	Altitude
SITE1: TIANG XIAN	016°17'44.3”	06°16'17.9”	648m
SITE 2 AM JIANIN	016°34'72.3”	06°32'31.8”	647m
SITE 3 MENG	016°23'02.4”	06°23'52.5”	636m
SITE 4 MAO	016°23'55.1”	06°25'32.9”	635m

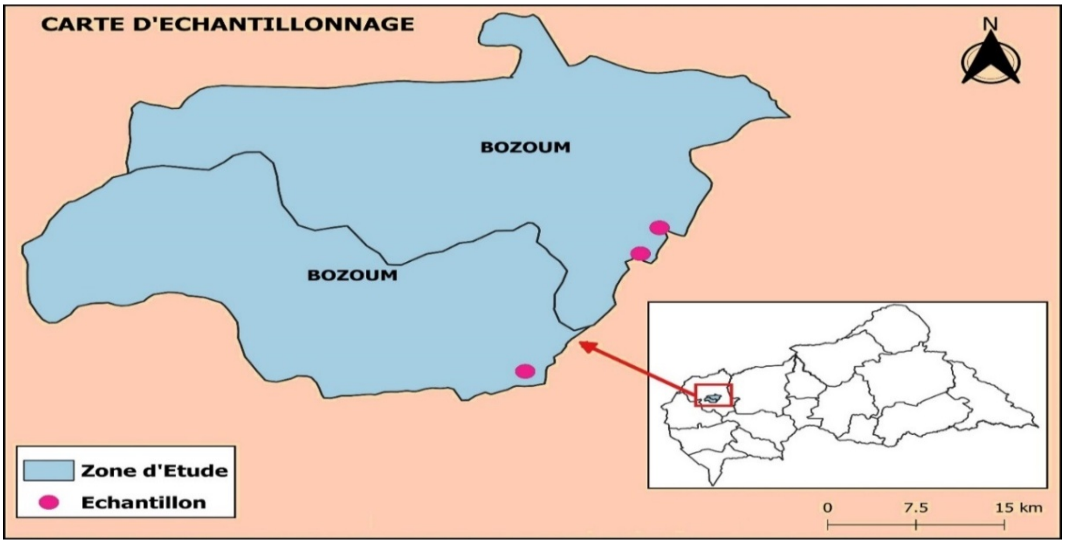


Figure 1. Sample area

2.2. Methods

The following in situ analyzes were conducted on each point water body using the following materials: - pH (pH-meter WTW 340i), - Electrical Conductivity and Temperature (Conductimeter WTW 340i), then at the analysis laboratory the equipment used are - Hach Turbidimeter, Balance Precisa XT 220A, Varian Spectra AA 55 Atomic Absorption Spectrometer (SSA- 55), VGA -77 and a nitrogen generator of ALLIA-1/1 brand. The main parameters measured are: turbidity, suspended matter (MES), pH, conductivity, water temperature, Total Organic Carbon (TOC), and heavy metals that can pollute the watercourse: arsenic, cadmium, mercury, nickel, lead, copper, zinc, total iron.

2.2.1. Determination by Spectrometry

The method chosen for the determination of the elements is Atomic Absorption Spectroscopy at different wavelengths as indicated in Table 2, the mercury was determined after reduction by sodium borohydride without enrichment (Eq (A.1) and Eq (A.2)) and arsenic by hydride generation (Eq (A.3)):

$$\mathrm{NaBH_4} \xrightarrow{H_2O} \mathrm{Na^+ + BH^-_4} \qquad (A.1)$$

$$\mathrm{BH^{4-} + Hg^{2+} + H^+}\left(aq\right) + 2e^- \xrightarrow{H_2O} \mathrm{HgO + BH_3 + H_2} \; (A.2)$$

$$\mathrm{3BH^{-4} + 3H^+ + 4H_3AsO_3 \rightarrow 3H_3BO_3 + 4AsH_3 + 3H_2O} \; (A.3)$$

Table 2. Measurement parameters on SAA-55

Elements	Wavelength, (nm) n	Slope	Concentration range (ppm) for standard solutions	Feasibility Area
Cadmium	228.8	0.5	0,5; 1; 2; 4; 8	0.02˚g/mL to 3˚g/ml
Copper	324.7	0.5	0,5; 1; 2; 4; 8	0.03mg/mL to 10 mg/ml
Iron	248	0.2	0,25; 1; 3; 7; 10	0.06 mg/mL to 15 mg/ml
Zinc	213.9	1	0,25; 0,5; 1; 2	0.01mg/mL to 20mg/ml
Lead	217.0	1	0,5; 1; 2; 4; 8	0.1mg/mL to 30 mg/ml
Arsenic	193.76	0.5	0,1; 0,2; 0,3; 0.4	200 to 400
Mercury	253.7	0.5	0,1; 0,2; 0,3; 0.4	200 to 400

2.3. Calibration for Analyzes

2.3.1. Calibration of VGA-77 for Arsenic Assay

For the determination of arsenic we have prepared different ranges of standard solutions ranging from 0 to 50 μg/L and then from 0 to 100 μg/L in order to determine the fidelity and reliability of the technique. The calibration curve from 0 to 100 µg/L was established first and represented by the following figure:

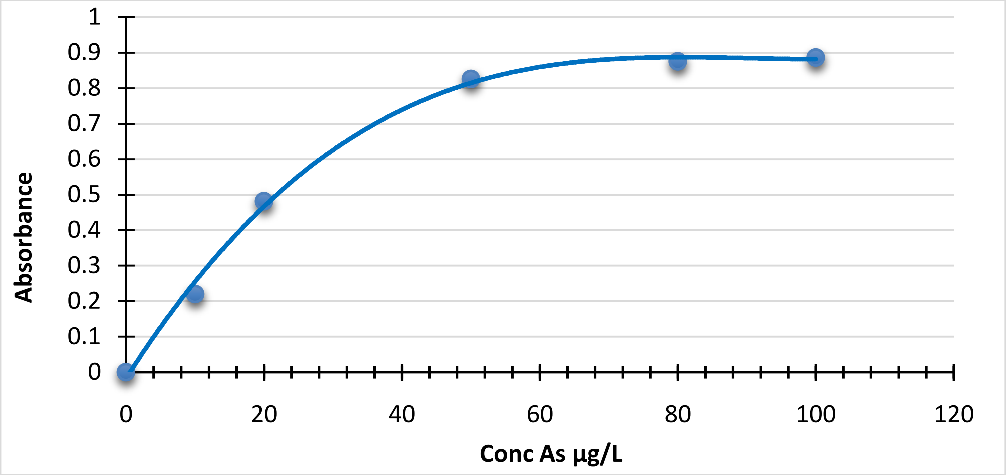


Figure 2. Calibration curve of 0-100 µg/L solutions for arsenic determination

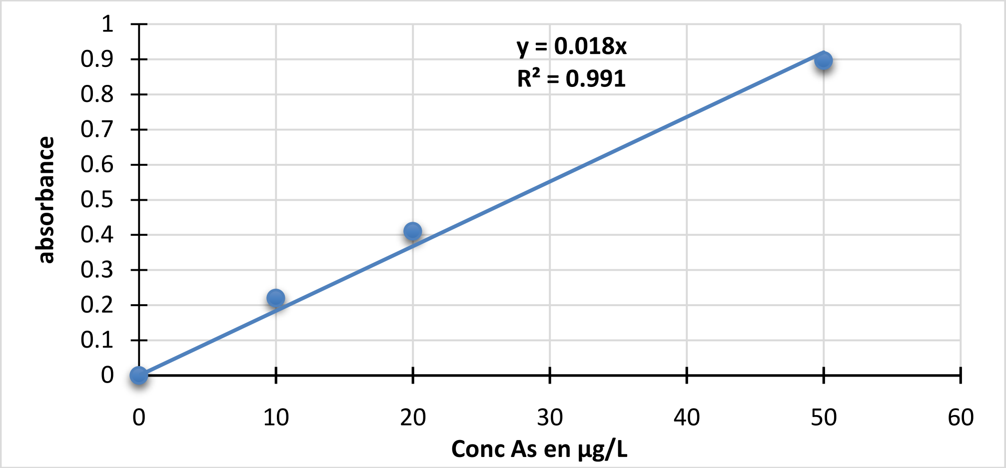


Figure 3. Calibration curve of 0-50 µg/L solutions for arsenic determination

Figure 2 shows that the calibration of the VGA-77 for arsenic determination in a calibration range of 0 to 100 µg/L shows signal saturation from 80 µg/L. This signal saturation shows the upper limit of the technique and requires that samples with higher arsenic concentrations be diluted. These results demonstrate that the fidelity and reliability of the technique is below 80 µg/L arsenic. For this purpose, a new calibration curve in the range of 0 to 50 µg/L is presented in the following figure for greater accuracy.

We note in Figure 3 that in the range from 0 to 50 µg/L, the calibration curve is linear with a regression coefficient close to 1 and equal to 0.9917. These results show the field of fidelity and reliability of the technique of determining arsenic by hydride generation. With this calibration curve we can measure water samples containing arsenic between 0 to 50 µg/L concentration. Since the standard is set at 10 µg/L, this calibration range provides precision around the guide value and even in samples containing arsenic in trace form. In the remainder of this study, all the arsenic dose curves per hydride generation with the VGA-77 in the various samples analyzed are established between 0 and 50 µg/L.

2.3.3. Calibration of VGA-77 for Mercury Assay

For the determination of mercury we have also prepared a range of standard solutions ranging from 0 to 400 µg/L and then from 0 to 1000 µg/L in order to be able to determine the fidelity and reliability of the technique. The different calibration curves obtained are represented by the following figures.

Figure 4 shows the calibration curve for mercury metering in a calibration range from 0 to 1000 µg/L. A signal saturation limit is observed on the curve at about 500 µg/L of mercury. As in the previous case, this signal saturation indicates the upper limit of the technique for the determination of mercury. These results demonstrate that the accuracy and reliability of the mercury-hydride generation technique is below 500 µg/L. For this purpose, a new calibration curve in the range of 0 to 400 µg/L is presented in the following figure for greater accuracy.

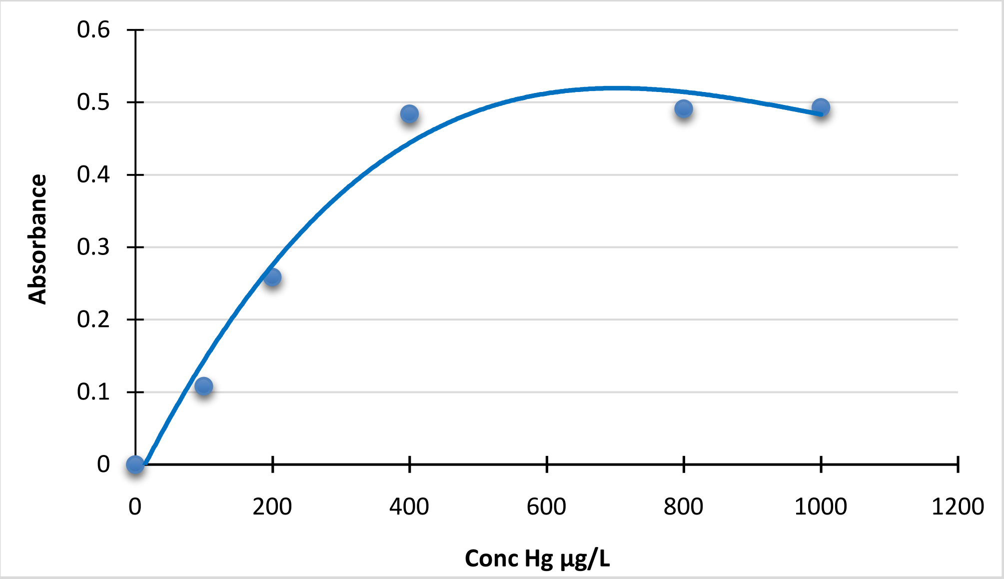


Figure 4. Calibration curve with solutions of 0 to 1000µg/L for the determination of mercury

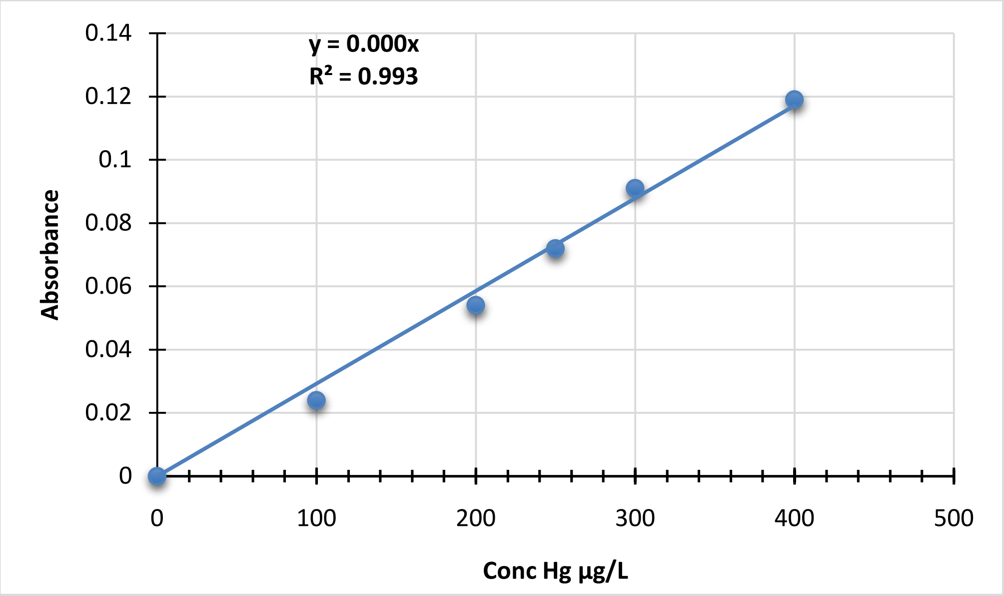


Figure 5. Calibration curve with 0 to 400 µg/L mercury solutions

Figure 5 of the calibration in the range from 0 to 400 µg/L is a linear straight line with a regression coefficient of 0.9933, therefore close to unity. These results show that the mercury-hydride generation technique has a greater range of fidelity and reliability than the arsenic generation technique. For mercury, water samples containing mercury up to 400 µg/L or 0.4 mg/L may be measured. In the remainder of this study, all the curves for the determination of mercury in the various samples analyzed are established between 0 and 400 µg/L.

It should be noted that a range of 0 to 10 was tested and we obtained weak or no signals on AA. We defined a range of 0 to 400 μg/l, which allowed us to have a curve that presents a straight line passing through the origin with a regression coefficient very close to unity, allowing us to obtain accurate and reliable results.

2.4. Method of Assessing Water Quality by TA-M

This part allows the assessment of the quality of the watercourse through the calculation of the pollution indices by metallic trace elements, namely: the contamination factor (FC), the degree of contamination (DC).

The contamination factor (FC) represents the ratio between the reference value of a given heavy metal and its content in the sediment studied. The FC is expressed by the following formula:

$$FC = \mathbf{CxBgx},$$ [2]

If **FC <1** contamination is absent at low;
1≤ FC < **3** contamination is moderate;
3≤ FC< 6, the contamination is considerable; and
6≤ FC, then the contamination is very strong.

Degree of contamination (DC)

The degree of contamination is used as a reference for assessing the level of metal contamination in water [3]. It is defined by:

$$\mathbf{DC} =_{n=1} FCi,$$

where FCi denotes the contamination factor ith parameter and n denotes the normative value of the element analyzed. It is also divided into three categories.

If **DC** < **1**, very low contamination;
If **1**< **DC** < **3**, limit contamination;
If **DC** > **3**, the contamination is high.

3. Results and Discussions

The investigation of four mining sites (Figure 1) in the region led to physical and chemical analyzes of water samples, the results of which are presented as follows (Table 3).

The parameters measured at the 4 sites (Table 3) have the following significant values:

- The suspended solids (MES) are in concentrations ranging from 276 mg/l to 504 mg/l; these values are above the current standard of 25 mg/l.
- The turbidity of the water is between 375 and 630 NTU; these values are significantly higher than the permissible value of 50 NTU.
- For heavy metals, iron and mercury are present in water at abnormally high concentrations. Total iron is found at concentrations between 4.21 and 8.97 mg/l above the 0.3 mg/l standard.
- Mercury is present at concentrations ranging from 4.2 to 26˚g/l (standard = 1˚g/l).

It should be noted that mercury was not detected upstream of the mining site. So this pollution of very toxic heavy metals would come from the mining sites. The use of mercury is dangerous not only for gold miners but also for the ecosystem in general [4]; people and animals living nearby. The potential dangers of mercury vapor are immense, especially as it is odorless and colorless. Mercury released into air and water by artisanal diggers is particularly harmful to the ecosystem in general and to plants in particular because it makes the environment anoxic (low in oxygen) [5,6,7]. Exposure to mercury in humans and animals, in particular, causes significant neurotoxicity, which can lead to side effects on the kidneys, liver, cardiovascular system, lungs, etc [8,9]. Mercury is a very slowly degradable element and accumulates in the food chain, especially in fish and herbivorous species [10], increasing the risk of contamination for humans living more or less far from the pollution site [11,12]. Mercury is particularly dangerous for children and pregnant women (risk of fetal malformation) [13,14,15]. The results of this work have made it possible to implement a treatment process by filtration on a heavy metal support using a simple, inexpensive technique in order to eliminate toxic substances, in particular mercury and Arsenic [16-21].

Table 3. Results of physicochemical analyzes

PARAMETERS	S1, Upstream	S 2 MAO	S 3 MENG	S4 TIANG XIAN	STANDARDS
Temperature, °C	26.1	27.2	27	27.6	20 - 25
pH	7.54	7.49	7.16	7.25	6 <pH< 8.5
Conductivity, µS/cm	26.1	28	26	30	
Turbidity, NTU	8.86	606	375	630	15 - 30 NTU
MES, mg/l	5	470	276	504	25 - 30 mg/l
Iron Total, mg/l	**1.17**	**0.76**	**4.21**	**8.97**	**0.3 mg/l**
Mn, mg/l	0	0	0	0.01	0.5 mg/l
Zn, µg/l	0.12	0.11	0.15	0.1	5 µg/L
Pb, µg/l	0	0	0	0	50 µg/L
Cu, µg/l	0.01	0.01	0.01	0.02	1 µg/l
Cd, µg/L	0	0	0	0	5 µg/L
As, µg/L	2	4	**12**	9	10 µg/L
Hg, µg/L	**0**	**26**	**25.1**	**4.2**	**1 µg/L**
TOC, mg/l	5.107	4.863	8.475	8.128	10 mg/l

3.1. Water Quality Assessment by TA-M

The application of hydride generation analysis technology has also been the subject of heavy metals research in the waters of the UHAM River. These analyzes were carried out upstream and downstream of the gold mining sites in the BOZOUM region. The purpose is to verify possible mercury pollution used by operators and to verify the hypothesis that Arsenic is an indicator of the presence of Gold in the river bed. According to Table 4, the pollution indices of **Zn, Cu, Pb, Cd and Mn** are less than 0.1 indicating a low level of contamination. The waters are moderately contaminated with Arsenic with a **DC** equal to 2.7 less than 3. On the other hand, Iron and mercury present a very high level of contamination in the stream assessed. The order of pollution is thus established **Hg > Fe > As > Zn > Cu > Pb=Cd=Mn**. Mercury contamination is widespread and affects all sites. The metal contamination assessment indices provide sufficient evidence that these elevated Hg levels are primarily anthropogenic. Figure 6 shows the level of pollution of the watercourse by the two major metals compared to the current standard.

Mercury analysis in the OUHAM River upstream and downstream of the various gold mining sites revealed mercury contamination of the river waters downstream of the mining sites. Mercury values at sites 2 to 4 are 4 to 26 times higher than the allowable limit of 1 µg/L. However, upstream of the exploitation sites, i.e. at the site1, no mercury was detected. These results show that gold operators at these sites use mercury to form amalgam with gold and the washwater would be discharged directly into the OUHAM River.

Arsenic levels slightly above 12 µg/L are also noted at site 3 and 9µg/L at gold mining site 4. This presence of arsenic may be indicative of the presence of gold in the area. So to eliminate these very toxic elements for humans and the environment, we have carried out some tests of mercury fixation by adsorption using local materials. In order to protect watercourses from such pollution, it is important to implement a process for the depollution of water leaving mining sites by using low-cost and environmentally friendly techniques. In our case, we will propose adsorption processes using clay-based filter materials.

Table 4. Calculation of the degree of contamination by different metallic elements

	Fe	Mn	Zn	Pb	Cu	Cd	As	Hg
Site 1 (upstream)	1.17	0	0.024	0	0.01	0	0.2	0
Site 2	0.76	0	0.021	0	0.01	0	0.4	26
Site 3	4.21	0	0.03	0	0.01	0	1.2	25.1
Site 4(downstream)	8.97	0.02	0.02	0	0.02	0	0.9	4.2
Degree of contamination	15.11	0.02	0.095	0	0.05	0	2.7	55.3

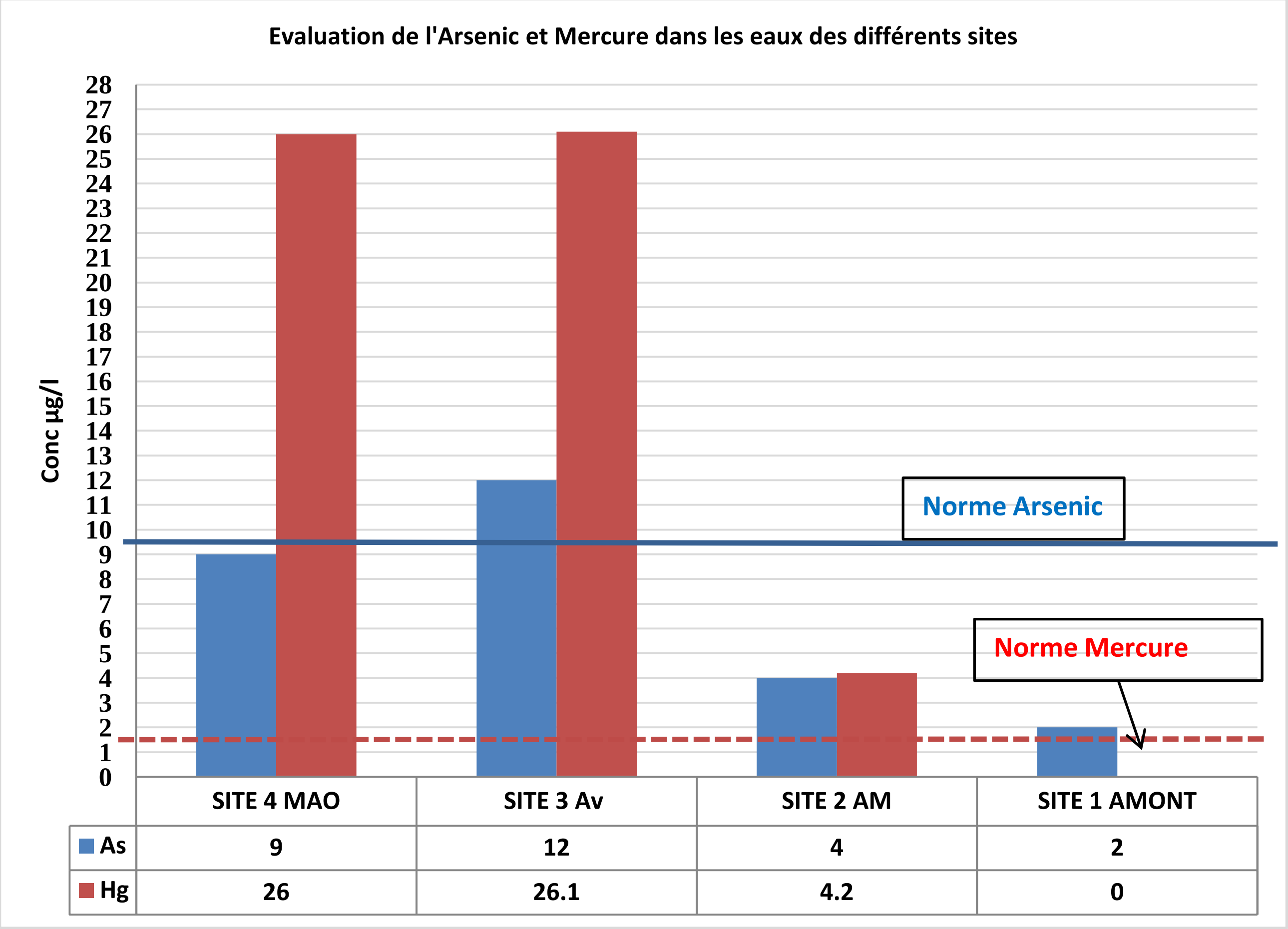


Figure 6. Diagram of mercury and arsenic levels in the OUHAM River upstream and downstream of gold mining sites

3.2. Adsorption Depollution Process

a) Preparing Media

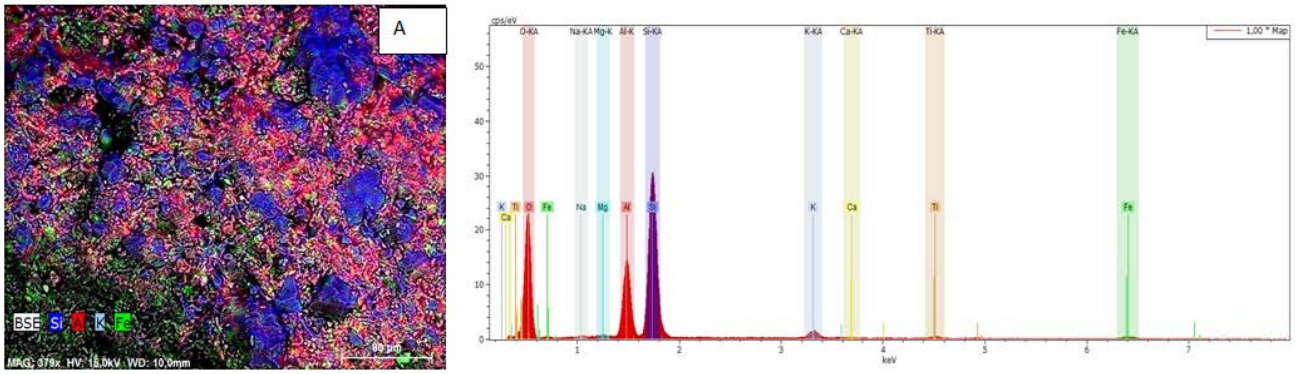
In this work, we explored the use of a brick commonly made by artisans from the Bangui region (Central African Republic). This brick results from the calcination of local soils which are mainly composed of sand, kaolinite and, to a lesser extent, illite, iron oxides, feldspar, mica and biotite [22,23,24]. To obtain this brick, Africans mix local kaolinite-rich soils with river water. Manually shaped and calcined inside a basic oven with dry wood for about three days at temperatures not exceeding 650°C. This temperature allows the transformation of the kaolinite of the brick into metakaolinite, but not into mullite (at 900°C) [26,27,28], as is generally the case in the manufacture of industrial bricks in developed countries. The average mineralogical composition of the Bangui brick is the following: 61 to 65% by weight of quartz, 21 to 25% of metakaolinite, 3 to 4% of illite, 4% of iron, etc. [22,23,24]. However, this brick is often accompanied by a certain amount of unprocessed kaolinite, revealing a temperature defect inside the oven. This natural material has some advantages to be used as an adsorbent because sand ensures good permeability and facilitates the flow of water through brick beds, while clays (such as metakaolinite after chemical and surface modifications) are known to possess good adsorption characteristics. Prior to use, several physical/chemical treatments were performed on the raw brick. First, it was crushed into grains and sieved with sizes ranging from 0.7 to 1 mm.

50 g of these grains were placed in 250 ml of a solution of NaOH of different concentrations 0.8 mol/L (Poumayé et al. 2020) and then placed under slow stirring for 24 hours at room temperature and then the mixture was heated at 90°C. for 6 days.

This treatment promotes the transformation of metakaolinite into zeolite. The latter is known for its high adsorption capacity. The following figures show the crystals.

The MEB image (Figure 7A) confirms that the rough brick grains consist of sand or quartz (blue grains) and metakaolinite (mainly composed of Si and Al), thus showing the abundant presence of Si and Al and the near absence of Na that can be observed in FIG. 7A of the spectrum. The presence of the limited potassium content on the brick grains confirms the presence of illite. ESEM/EDS mapping of Si, Al, Na and Fe elements (Figure 7B) and the spectrum (Figure 7B) of brick grains activated at 0.8M NaOH for 6 days at 90°C show that the surface of these brick grains has two cubic and spherical crystal forms. [26,27] showed that the crystals obtained from the brick grains were similar to those already identified in the literature by [28] for NaA zeolite (cubic crystals) [29] for NaP zeolite (spherical crystals). These zeolitic crystals obtained give our support a great capacity for elimination [30-35].

(A) : Grains de brique brute



(B) : Grains de brique activée par NaOH 0,6M

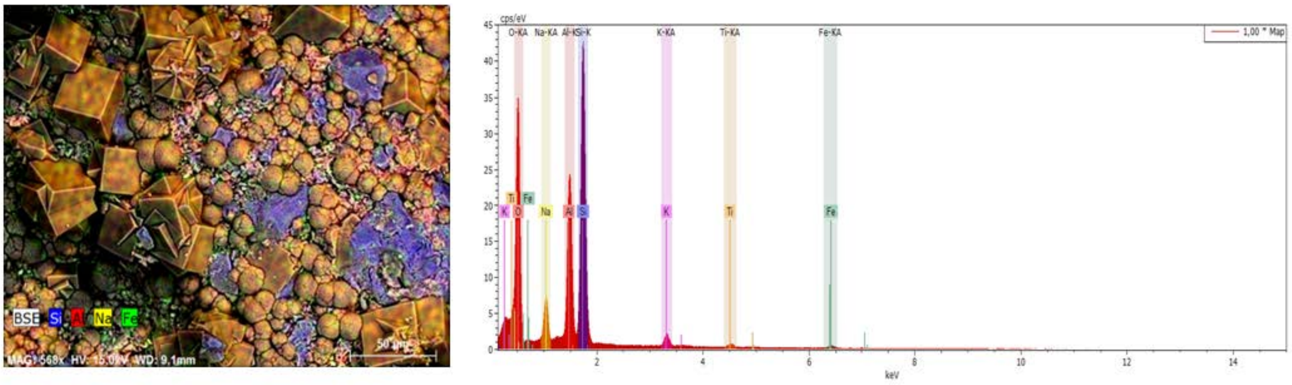


Figure 7. MEB images of the surface of untreated brick grains (A) treated with 0.8 M NaOH at 90°C for 6 days (B)

b) Support filtration treatment device

1→ Mercury or arsenic solution

2 Column (Cooked brick activated zeolite)

3→ Electrodes for pH and Conductivity measurements

4→ Tank

5→ Filtrate

6→ Balance

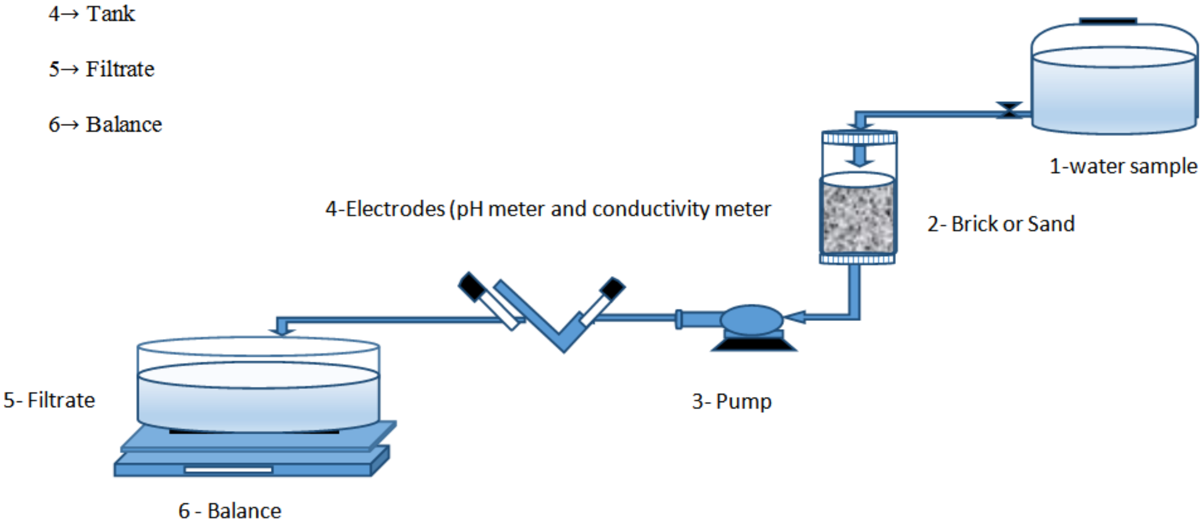


Figure 8. Device for fixing ETMs by fired brick and sand

The purpose of this method is to adsorptively bind arsenic and mercury to meet the requirements of current standards that require a reliable, accurate, repeatable and reproducible analytical technique. It is therefore necessary to periodically calibrate the equipment in order to verify these parameters.

3.3. Mercury Fixation Test Results

Mercury removal tests in the OUHAM River were conducted on two local materials prepared at the laboratory.

Table 5 shows the mercury removal results from the site 4 river water sample (MAO) on both filter media. There is a clear decrease in the mercury concentration at the outlet of each treatment device. A significant decrease in the reduction was observed on the activated fired brick grains ranging from 26 to 0.9 µg/l, i.e. 95.83 compared with 12.5% for sand.

Table 5. Fixation of mercury by cooked bricks and sand grains

	Activated bricks	Sand
C/E	MAO Site-4	MAO Site 4
[Hg] initial µg/L	26	26
[Hg] Final µg/L	0.9	21
Abatement (%)	95.83	12.5

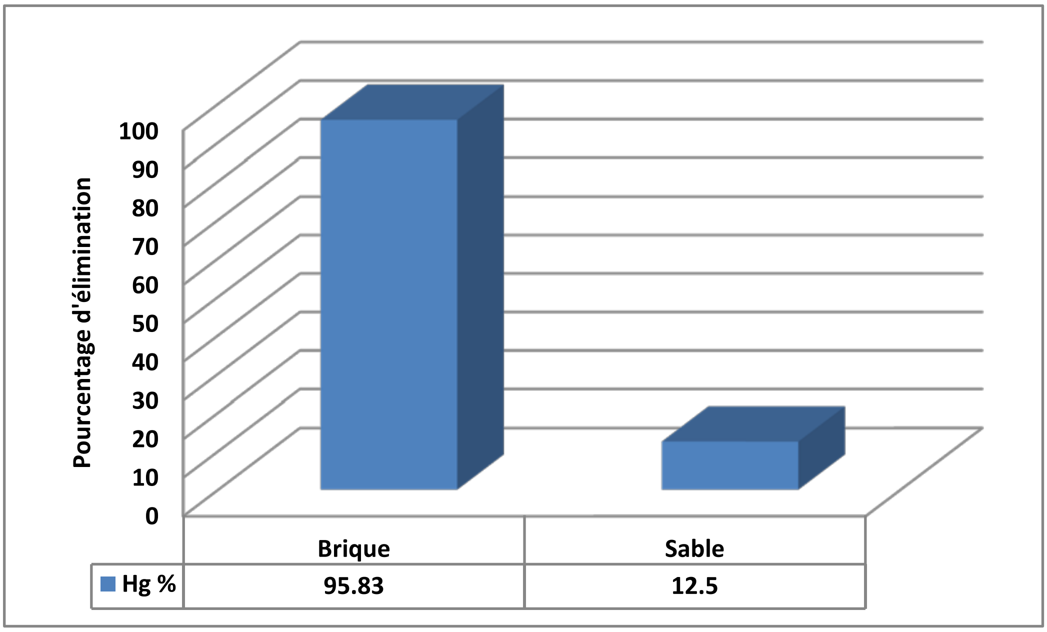


Figure 9. Mercury removal rate from cooked bricks and sand grains

The abatement rate is given by:

$$\textbf{Abatement rate} = \left(\mathbf{C_0} - \mathbf{C_1}\right) / \mathbf{C_0} * \mathbf{100};$$

Or C_0 is the initial concentration in milligrams per liter and C is the final measured concentration.

According to the results of the analyzes, cooked bricks have a good mercury retention capacity with a rate of abatement of 95.83% compared to sand with a low rate of abatement of 12.5%. This difference is due to the fact that activated fired brick has adsorbent sites formed from clay and soda, unlike sand grains which do not. Therefore, activated cooked brick grains could reduce mercury contamination in the environment.

4. Conclusion

Metals are ubiquitous in surface waters, but their concentrations are generally very low, which explains their designation as "trace metals" or "metallic trace elements" (MTEs). These elements are toxic to humans and the environment on a large scale. The objective of this study is to assess the level of contamination of the water of the Ouham River by MTAs impacted by anthropogenic activities in the Bozoum region of the Central African Republic. The results showed that the suspended solids (MES) are in concentrations ranging from 276 mg/l to 504 mg/l; these values are above the current standard of 25 mg/l, The turbidity of the water is between 375 and 630 NTU; These values are well above the allowable value of 50 NTU, For heavy metals, iron and mercury are found in water at abnormally high concentrations. Total iron is found at concentrations ranging from 4.21 to 8.97 mg/l (standard = 1 mg/l) and mercury is found at concentrations ranging from 4.2 to 26˚g/l (standard = 1˚g/l). The degree of mercury contamination is 55.3 and 15.1 for iron, the DC"" 3, shows that the contamination is very high by these two heavy metals. It should be noted that the presence of mercury was not detected upstream of the mining site, so this very toxic heavy metal pollution would come from the mining sites. Gold prospecting is the main source of mercury emissions to water. In contrast, Fe, Pb, Cd, Cu and As are naturally occurring. They come from the geochemistry of the area. The installation of a treatment device from the activated brick shows a good mercury retension capacity with a rate of abatement of 95.83% as sand which has a low rate of abatement of 12.5%.

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