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Distribution and Health Risk Assessment of Trace Metals in Surface Waters and Groundwater Around Artisanal Gold Mining Areas in Central-western Côte d'Ivoire, West Africa

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Abstract: The present study aimed to evaluate Ni, Cr, Zn, Cd, Hg, and Pb distribution and the health risks in surface waters and groundwater around artisanal mining areas in the central-western Côte d'Ivoire. Trace metals spatial distributions were assessed using ArGIS method. Waters pollution status were ascertained through water pollution indices. Human health risk was investigated using non-carcinogenic and carcinogenic risks indices. In surface waters, the same distribution trends were observed for Ni and Cd, and, for Hg and Pb. While, the distribution patterns of Zn and Cr differed from those of Ni, Cd, Hg, and Pb. Trace metals (Ni, Cr, Cd, Zn, Hg and Pb) concentrations in groundwater were within their guidelines values. While, the averages total of Ni, Cd, Hg, and Pb concentrations for the surface waters exceeded their guidelines values. The pollution status revealed high and low pollution levels of surface waters and groundwater, respectively. Averages of the total non-carcinogenic risk for Hg, Cd, Ni and Pb in surface waters varied between 1.529±1.162 and 80.507±104.615, indicating adverse effects to human health. The averages total carcinogenic risks for Ni, Cd, Pb, and Cr indicated that the population around the artisanal gold mining area could develop cancer. Therefore, it is important to treat waters for removing trace metals before using for domestic purposes.

Keywords: Trace Metals, Pollution Assessment, Risks Assessment, Surface Waters, Groundwater

1. Introduction

Water is an important element for human survival [1]. However, its quality is threatened by many activities like agricultural practices and gold extraction [1]. In many developing countries, mining activities contribute to economic growth and poverty reduction via job creations [2]. But, the intensification of these mining activities can impact negatively the water ressources and rural population through the release of toxic chemical pollutants [3, 4]. Among these contaminants, heavy metals have attracted global attention due to their toxicity, persistence and bioaccumulation [5–7]. Therefore, environmental studies of mining areas are important.

Cr, Ni and Zn are considered to be essential for humans [8]. While, Cd, Pb, and Hg are not essential for humans [9]. But they may cause deleterious effects on the environment and human health beyond their tolerance values [10]. Trace metals Cr and Zn exposure can cause lung cancer [11]. Cd exposure may lead to prostatic, kidney problems, hypertension and lung cancer [11-13].

Chronic exposure to Hg can have adverse effects such as neurological disorders and renal dysfunction [2, 14, 15]. Ni toxic effects include respiratory disorders, lungs and even cancers [16]. Excess Pb can damage the neurological system, cause renal disfonctionment, skeletal weakness, mutations and cancer [8, 11, 17]. Therefore, monitoring of Zn, Cr, Ni, Cd, Hg and Pb concentrations in environment are essential.

High concentrations of Ni, Zn, Cr, Cd, Pb and Hg in groundwater and surface waters due to mining activities were reported in many countries such as China [3, 4, 18], Colombia [19, 20], Brazil [21], South Korea [22] and India [23]. In addition, the negative impacts of trace metals on human health in the mining regions of Europe, South America, and Central Africa have been reported in the literature. For example, high concentrations of Cd and Hg were observed in urine of children living near mining area in Andalusia (Spain) [24]. Miners from a mining area in Colombia accumulated high concentrations of Hg in blood, urine and hair [25]. High levels of Pb, Cd and Ni were observed in hair of older children (10 years) living around Migori gold mining area in Kenya, Central Africa [26]. Countries in West Africa, are not left out of this mining contamination. For example, high concentrations of Pb and Zn were reported in the urine of population living near a small-scale gold mining area in Ghana [27]. Many investigations have documented the contamination of surface waters and groundwater around mining areas in West Africa, with studies focused in Nigeria [2, 28, 29], Ghana [14, 30-32], Senegal [33] and Burkina Faso [34], but in Côte d'Ivoire, studies are in their infancy. Recently, Kone et *al* [35] showed that surface waters around Tongon industrial mining area in northern Côte d'Ivoire were contaminated by Pb, As and Cr. These studies are not focused on the assessment of potential non-carcinogenic and carcinogenic risks. In addition, studies on waters quality assessment around artisanal gold mining areas in Côte d'Ivoire are not previously investigated. Therefore, it is important to establish first data on the human health risks associated with Ni, Zn, Cd, Pb, Cr, and Hg in waters near artisanal mining areas in Côte d'Ivoire. This study aimed to assess: (i) Zn, Cr, Ni, Cd, Hg and Pb distribution in surface waters and groundwater; (ii) the quality and the pollution levels of the surface waters and groundwater; (iii) the health risks from Zn, Cr, Ni, Cd, Hg and Pb. The health risk evaluation is important since waters are mostly utilized without any treatment by population living near artisanal gold mining areas.

2. Material and Methods

2.1. Study Area and Sampling

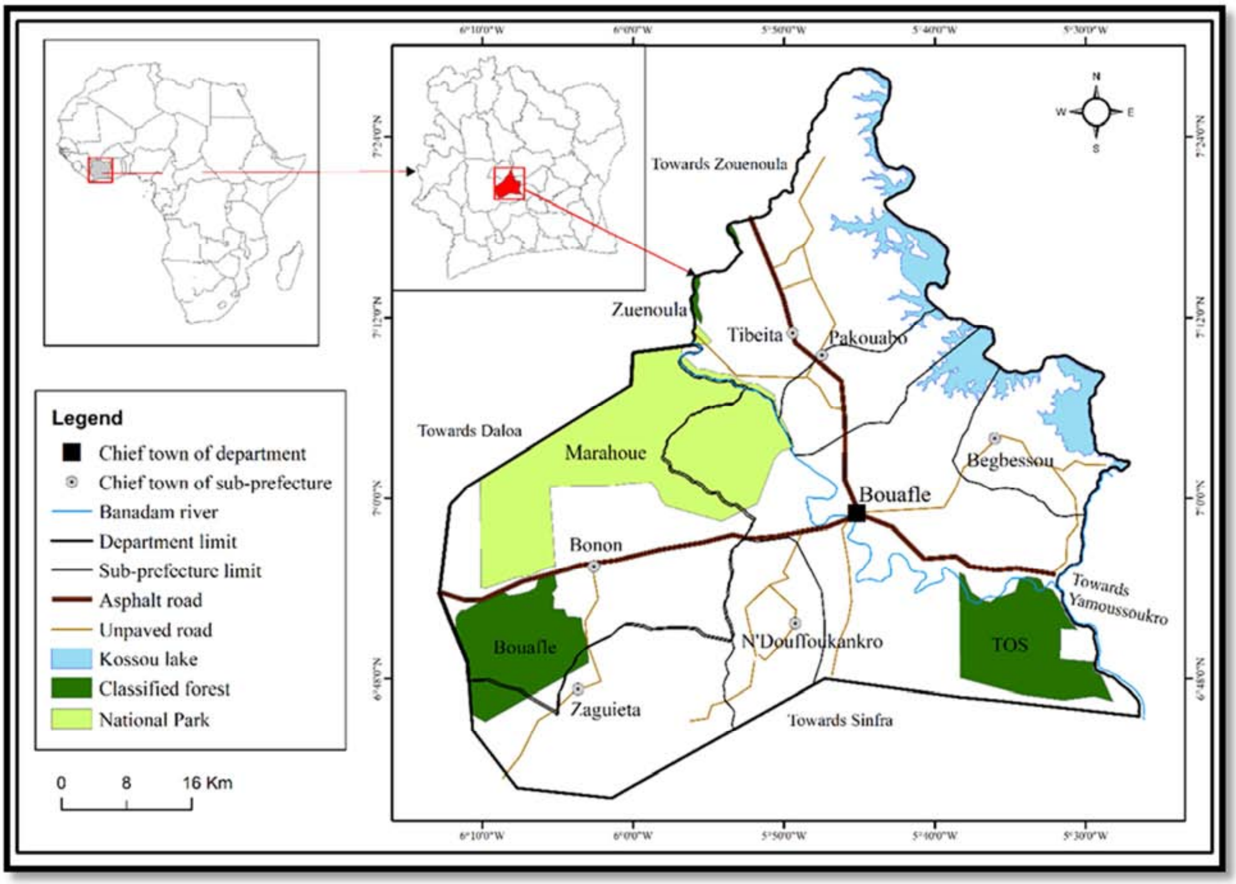
The department of Bouaflé is located in the Central Western Côte d'Ivoire, West Africa (Figure 1). It has an area of 4,222.48 km². The departement is well is well known through activities such as livestock, cocoa, banana, palm oil, rice, yam, vegetables, food and mining activities. Bouafle lies in the Bandama river basin which is the longest and the largest river in Côte d'Ivoire [36]. It takes its source in the northern Côte d'Ivoire, between Korhogo and Boundiali at an altitude of 480 m and flows into Grand-Lahou Lagoon and the Gulf of Guinea in the south. The Bandama river length and area are 1050 km and 97,500 km^2, respectively [37, 38]. The Kossou Lake is largest lake in Côte d'Ivoire. It has an area of 1700 km², a reservoir of 30 billion m^3 of water and a length of 150 km. The Kossou lake plays an important role in the Ivorian economy. Indeed, Kossou represents the biggest hydroelectric plant which provides power for Côte d'Ivoire, it allows the creation of large areas of irrigated agriculture (50,000 hectares of land) and it is one of the main places of continental fishing activities [39]. It is also the largest dam in Cote d'Ivoire. The selected sites are mining villages of Angovia, Allahou-Bazi, Kouakou Gnanou, Pangba Kouamekro, Allekran, Kaviessou and Duonfla in Bouafle departement. The stations are located in the Bandama river (SP11, SP12, SP13 and SP14), in the Kossou lake (SP15, SP16, SP17 and SP18), in the pond water (SP19, SP20, SP21 and SP22) and in the groundwater (SP23, SP24, SP25 and SP26) (Figure 1). The water from the pond is used for washing gold tailings and the wastewaters after washing processes are discharged again into the pond.

The waters sampling was carried out in December 2019 and August 2020 at 16 stations from the Bandama river, the Kossou lake, the pond water, and the groundwater. The water sampling method described by APHA was used in this study [40]. Surface waters were sampled with a Niskin bottle (5 L) at 50 cm depht. To prevent metal precipitation, water samples were acidified with 1 mL of nitric acid (65% suprapur, E. Merck, Germany) and stored at 4°C until analysis [36]. Previously, all glassware and plastic bottle were rinced with de-ionized water, cleaned with 20% of HNO_3 and rinsed three times with de-ionized water before use [36, 41].

2.2. Analytical Methods

A HANNA multiparameter HI.9828 was used for *in situ* determination of the pH and electrical conductivity. The anoins (SO_4^{2-}, Cl^-, F^- and NO_3^-) and the cations (Mg^{2+}, Na^+ and Ca^{2+}) were determined using a spectrophometer HACH DR 6000. Ni, Cr, Cd, Zn, Hg and Pb total concentrations were determined using the Inductively Couple Plasma Mass Spectroscopy (ICP-MS). The detection limit for trace metals were 0.001 mg/L for Ni and Cr, 0.0001 mg/L for Cd, 0.005 mg/L for Zn, 0.0005 mg/L for Hg and Pb. The experiment was performed in triplicate.

The spatial distributions of Ni, Cr, Cd, Zn, Hg and Pb were assessed using ArcGIS V (10.2).


Legend
Chief town of department
Chief town of sub-prefecture
Banadam river
Department limit
Sub-prefecture limit
Asphalt road
Unpaved road
Kossou lake
Classified forest
National Park
0
8
16 Km
Towards Zouenoula
Zuenoula
Tibeita
Pakouabo
Towards Daloa
Marahoue
Begbessou
Bouafle
Bonon
Towards Yamoussoukro
Bouafle
N'Douffoukankro
TOS
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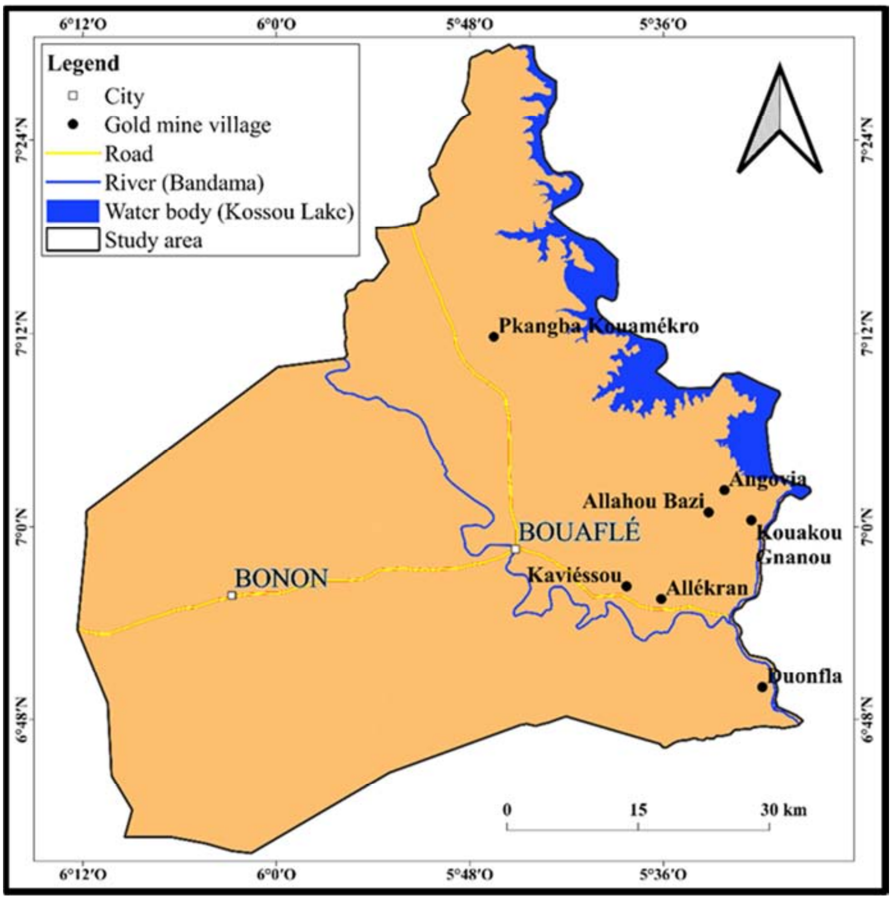

Legend
City
Gold mine village
Road
River (Bandama)
Water body (Kossou Lake)
Study area
6°12′O
6°0′O
5°48′O
5°36′O
7°24′N
7°12′N
7°0′N
6°48′N
Pkangba Kouamékro
Allahou Bazi
Angovia
Kouakou Gnanou
BOUAFLÉ
BONON
Kaviéssou
Allékran
Ouonfla
0
15
30 km

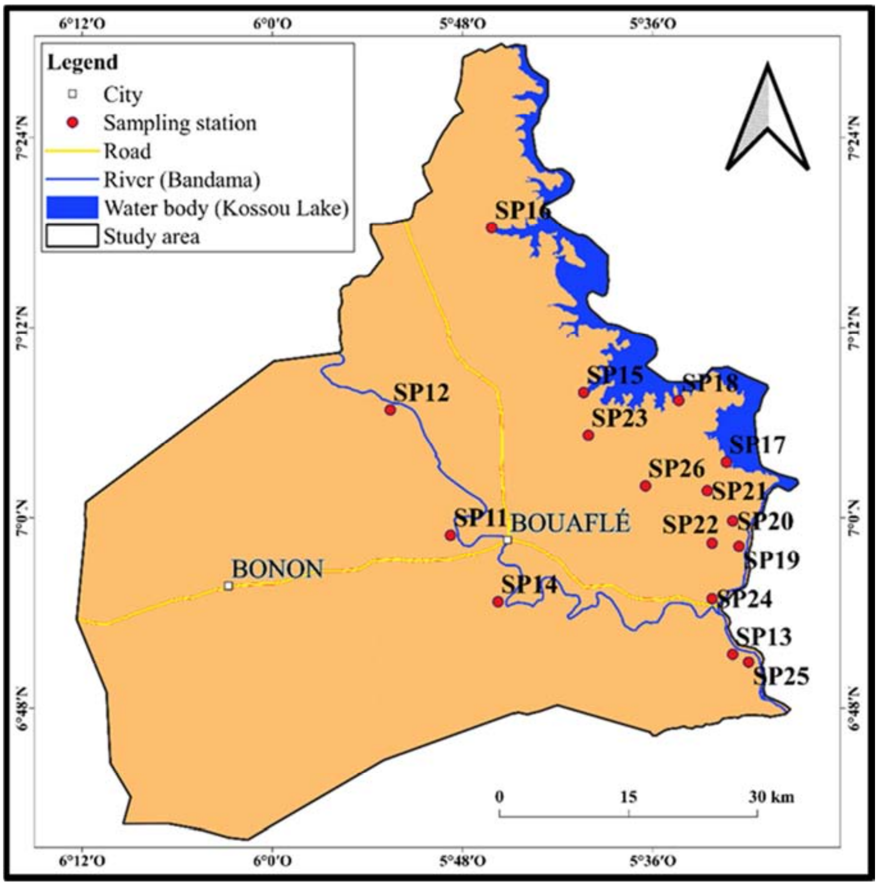


Figure 1. *Study area and sampling stations.*

2.3. Pollution Level Assessment

2.3.1. Heavy Metal Pollution Index (HPI)

The HPI index is calculated using the following equation [42]:

$$HPI = \frac{\sum_{i=1}^{n} W_i \times Q_i}{\sum_{i=1}^{n} W_i} \quad (1)$$

Where n is the number of metals analysed. The unit weighting of ith metal is defined by wi. Qi is the sub-index for ith trace metal. The unit weighting (wi) was calculated by the relation below [43]:

$$w_i = \frac{1}{MAC_i} \quad (2)$$

Where MACi is the maximum admissible concentration of ith pollutant.

Qi is computed using the relationship below:

$$Q_i = \sum_{i=1}^{n} \frac{C_i(-)L_i}{M_i - L_i} \quad (3)$$

Where C_i was the determined concentration of the pollutant i. The maximum permissible and lower desirable values of metal i were given by Mi and Li, respectively. The algebraic sign (−) indicates the numerical differences between the two values. The algebraic sign is ignored. Mi and Li values were taken from WHO [44] and USEPA [45]. HPI < 100 indicates low pollution of heavy metal. HPI =100 indicates that harmful health effects are probable. HPI > 100 suggests that the water is not suitable for drinking [46].

2.3.2. Heavy Metal Evaluation Index (HEI)

The overall surface water quality with respect to heavy metal concentration is evaluated by HEI. HEI index is calculated as follows [46, 47]:

$$HEI = \sum_{i=1}^{n} \frac{C_i}{MAC_i} \quad (4)$$

Where C_i is the measured concentration of the ith heavy metal. MAC_i is the maximum admissible concentration of metal i. The HEI value below 10 indicates low pollution level. The water is moderately polluted when HEI is between 10 and 20. HEI higher than 20 indicates high pollution level [46]. Table 1 gives the values of Mi, Li and MAC_i for heavy metals.

Table 1. *Mi, Li, MACi and wi values for heavy metals [44, 45].*

Metals	M_i (µg/L)	L_i (µg/L)	MAC (µg/L)	w_i
Ni	70	20	20	0.05
Cr	100	50	50	0.02
Cd	5	3	3	0.3
Zn	5000	3000	5000	0.0002
Hg	2	1	2	0.50
Pb	100	10	1.5	0.7

2.4. Water Quality Assessment

The water quality is assessed using the water quality index (WQI). The following equation (5) is used to calculate WQI [48, 49]:

$$\text{WQI} = \sum\left[W_i \times \left(\frac{C_i}{S_i}\right) \times 100\right] \quad (5)$$

$$W_i = \frac{w_i}{\sum w_i} \quad (6)$$

Where C_i was the determined value of ith parameter. S_i was the standard value according to WHO. Wi and wi were the relative weight and the weight attributed to the element i, respectively.

$\sum w_i$ was the sum of w_i. A WQI value below 50 indicates excellent quality; water quality is good when $50 \leq \text{WQI} < 1$; when $100 \leq \text{WQI} < 200$, water quality is poor; if $200 \leq \text{WQI} < 300$, water quality is very poor; $\text{WQI} \geq 300$ indicates that water is not drinkable [50]. The values of S_i and the relative weights (Wi) are given in Table 2.

Table 2. *Relative weight and standard S_i values.*

Parameters	WHO standard (2017))	Weight (w_i)	Relative weight (W_i)
pH	6.5-8.5	4	0.0816
SO_4^{2-}(mg/L)	250	4	0.0816
Cl^- (mg/L)	250	3	0.0612
NO_3^- (mg/L)	50	5	0.1020
Ca^{2+} (mg/L)	300	2	0.0408
Mg^{2+} (mg/L)	30	2	0.0408
Na^+ (mg/L)	200	2	0.0408
F^- (mg/L)	1.5	5	0.1020
Ni (mg/L)	0.07	1	0.0204
Cr (mg/L)	0.05	5	0.1020
Cd (mg/L)	0.003	5	0.1020
Zn (mg/L)	3	1	0.0204
Hg (mg/L)	0.006	5	0.1020
Pb (mg/L)	0.01	5	0.1020
		$\sum w_i$= 49	$\sum W_i$= 1

2.5. Health Risk Assessment

2.5.1. Non-carcinogenic Risks

The following equations are used to assess the exposure [51, 52]:

$$\text{CDI}_{\text{ing}} = \frac{C_i \times \text{IR} \times \text{EF} \times \text{ED}}{\text{BW} \times \text{AT}_{\text{nc}}} \quad (7)$$

$$\text{CDI}_{\text{der}} = \frac{C_i \times \text{SA} \times K_p \times \text{ET} \times \text{EF} \times \text{ED} \times \text{CF}}{\text{BW} \times \text{AT}_{\text{nc}}} \quad (8)$$

Where CDI_{ing} (µg/Kg.day) indicates the chronic daily intake through ingestion. CDI_{der} (µg/Kg.day) expresses the chronic daily intake through dermal contact. The other exposure parameters were reported in Table 3.

The hazard quotient (HQ) was used to assess the non-carcinogenic risk

The hazard index (HI) expresses the total non-carcinogenic risk. HI is computed as follows [51]

$$\text{HQ}_{\text{ing}} = \frac{\text{CDI}_{\text{ing}}}{\text{RfD}_{\text{ing}}} \quad (9)$$

$$\text{HQ}_{\text{der}} = \frac{\text{CDI}_{\text{der}}}{\text{RfD}_{\text{der}}} \quad (10)$$

$$\text{RfD}_{\text{der}} = \text{RfD}_{\text{ing}} \times \text{ABS}_{\text{g}} \quad (11)$$

$$\text{HI} = \sum(\text{HQ}_{\text{ing}} + \text{HQ}_{\text{der}}) \quad (12)$$

The HI value below 1 indicates low adverse effects. When $\text{HI} \geq 1$, adverse effects can occur on human health [51].

2.5.2. Carcinogenic Risks

Trace metals Ni, Cd, Cr, and Pb were used to assess the carcinogenic risk. The carcinogenic risk is calculated as follows [52, 53, 56, 57]:

$$\text{CR} = \sum \text{CDI}_i \times \text{SF}_i \quad (13)$$

Where SF_i is a slope factor.

$$\text{CR}_{\text{ing}} = \text{CDI}_{\text{ing}} \times \text{SF}_{\text{ing}} \quad (14)$$

$$\text{CR}_{\text{der}} = \text{CDI}_{\text{der}} \times \text{SF}_{\text{der}} \quad (15)$$

$$\text{SF}_{\text{der}} = \frac{\text{SF}_{\text{ing}}}{\text{ABS}_{\text{g}}} \quad (16)$$

$$\text{CR} = \sum(\text{CR}_{\text{ing}} + \text{CR}_{\text{der}}) \quad (17)$$

CR value $\leq 10^{-6}$ indicates no significant risk. When CR value $\geq 10^{-4}$, humans can develop a cancer. The range of acceptable carcinogenic risk is 10^{-6} to 10^{-4} [56].

Table 3. *Exposure assessment parameters.*

Parameter	Meaning	Value	Unit	Reference
IR	Ingestion rate	2.2	L/day	[53]
EF	Exposure frequency	365	day	[53]
ED	Exposure duration	58	year	[54]
BW	Body weight	60	Kg	[53]
AT_{nc}	Average time for non-carcinogenic	365×58	day	
SA	Skin-surface area	18,000	Cm^2	[53]
K_p	Permeability coefficient	Ni: 0.0002; Cr:0.002; Zn: 0.0006; Cd, Hg and Pb: 0.001	cm/h	[53]
ET	Exposure time	0.58	h/day	[53]
CF	Conversion factor	0.001	L/cm^2	[53]
RfD_{ing}	Reference dose of heavy metals through ingestion	Ni: 20; Cr:3; Cd:0.5; Zn:300; Hg:0.3; Pb: 1.4	µg/Kg.day	[53]
SF_{ing}	Slop factor of metal through ingestion	Ni: 0.91; Cr: 0.42; Cd: 0.38; Pb: 0.0085	µg/Kg.day	[53]
ABS_{g}	Gastrointestinal absorption factor	Ni (0.27); Cr (0.025); Cd (0.05); Zn (0.02); Hg (0.07); Pb (0.3)		[55]

3. Results and Discussion

3.1. Surface Waters and Groundwater Physicochemical Parameters

The physicochemical parameters mean values of all the waters were given by Table 4. pH average values were 7.33±0.45, 6.70±0.99 and 6.90±0.62 in Bandaman, Kossou and groundwater, respectively. In the pond water the pH average was 3.87±0.94, which indicates that the water is highly acidic and may be explained by the release of mine wastes [58]. The mean values of the Badaman river, the Kossou lake and the groundwater are in the permissible range (6.5 - 8.5) of WHO, while that of pond water was lower than the lower limit (< 6.5) set by WHO. These results indicated that pond waters are problematic for drinking usage, while groundwater could be used for drinking. In addition, the Badaman river and the Kossou lake could be used for diverse uses such as irrigation, domestic purposes, and recreational purposes.

Electrical conductivity mean values were 371±178.09 µS/cm in the Bandama river, 765.5±193.35 µS/cm in the Kossou lake, 1,446±571.57 µS/cm in pond water and 72.5±28.84 µS/cm in groudwater. According to Şener et *al* [49], water conductivity is correlated with dissolved solids. High waters EC values may be caused by contaminants [49]. Therefore, the maximun values observed in the Kossou lake and the pond water can be explained by the discharges product of grinding operation, amalgamation of the material and sewage [20].

The average concentrations of SO_4^{2-} were 11.00±2.68 mg/L, 10.08±2.29 mg/L, 339.00±127.30 mg/L, and 3.65±0.51 mg/L in the Bandama river, the Kossou lake, the pond water, and the groundwater, respectively. The mean value obtained in the pond water exceeded the WHO limit. The organic substances of weathered soils breakdouwn, the leaching of sulfate from fertilizers and the wastewater might explain the high value of sulfate in the pond water [59].

Chloride (Cl^-) concentrations obtained in this study ranged from 10.08±2.29 mg/L to 331.85±182.53 mg/L. The average values were 33.33±10.46 mg/L in the Bandama river, 10.08±2.29 mg/L in the Kossou lake, 331.85±182.53 mg/L in the pond water and 26.70±1.01 mg/L in the groundwater. The mean value of chloride in the pond water exceeded its standards value of 250 mg/L [44] due to sewages product during the gold extraction process.

The mean nitrate (NO_3^-) concentrations were 36.80±9.68 mg/L in the Bandama river, 29.58±8.46 mg/L in the Kossou lake, 48.88±3.66 mg/L in the pond water and 4.87±0.98 mg/L in the groundwater. The mean values of nitrate in all waters were below the WHO limit (50 mg/L). However, the maximun value obtained in the pond water may be attributed to fertilizer used in agricultural activities.

Calcium (Ca^{2+}) average concentrations recorded for the all water-bodies did not exceed the limit 300 mg/L [45]. The values were 17.83±2.47 mg/L, 20.38±3.73 mg/L, 30.70±12.66 mg/L and 60.88±1.59 mg/L in the Bandama river, the Kossou lake, the pond water and the groundwater, respectively. However, the dissolution of calcite and dolomite may explain the maximum value obtained in groundwater [49].

Magnesium (Mg^{2+}) concentrations of water samples varied between 4.27±0.46 mg/L and 99.07±46.28 mg/L. The averages were 35.75±8.63 mg/L in the Bandama river, 35.93±13.82 mg/L in the Kossou Lake, 99.08±46.28 mg/L in the pond water, and 4.28±0.46 mg/L in the groundwater. The values obtained in the surface waters exceeded the desirable limit of 30 mg/L [44]. Higher Mg^{2+} concentrations may be due to the decomposition of the ferromagnesia, minerals, the metamorphic rock and the magnesium carbonate in sedimentary rock [60].

The averages content of Na^+ in surface waters samples were 3746.51±718.76 mg/L, 4112.22±469.21 mg/L, and 5071.04±197.78 mg/L in the Bandama river, the Kossou lake and the pond water, respectively. The mean values of Na^+ in all surface waters exceeded the acceptable limit of 200 mg/L which may be explained by the decomposition of igneous rocks and the wastes discharges [60, 61]. While, Na^+ mean concentration in groundwater (23.37±2.91 mg/L) was below its WHO limit (200 mg/L).

The fluoride (F^-) mean concentrations (mg/L) obtained were 0.17±0.16, 0.31±0.16, 0.28±0.06, and 0.03±0.02 in the Bandama river, the Kossou lake, the pond water and the groundwater, respectively. These mean values did not exceed the acceptable limit (1.5 mg/L) according to WHO [44].

Our findings showed that, all the physicochemical parameters in the groundwater samples were found to be within the WHO guideline values, indicating good quality. Therefore, to better understand the quality of surface waters and groundwater around artisanal mining areas in Bouafle, water pollution indices assessment was performed.

Table 4. *Average values of the physical and chemical parameters.*

	Bandama	Kossou	Pond water	Groundwater
pH	7.33±0.45	6.70±0.99	3.87±0.94	6.90±0.62
Conductivity (µS/Cm)	371.75±178.97	765.5±193.34	1446.00±571.57	72.25±28.84
SO_4^{2-} (mg/L)	11.00±2.69	10.07±2.29	339.00±127.30	3.65±0.51
Cl^- (mg/L)	33.32±10.46	29.02±8.59	331.85±182.52	26.7±1.01
NO_3^- (mg/L)	36.8±9.68	29.57±8.46	48.87±3.66	4.86±0.97
Ca^{2+} (mg/L)	17.82±2.47	20.37±3.73	30.70±12.66	60.87±1.59
Mg^{2+} (mg/L)	35.75±8.63	35.92±13.82	99.07±46.28	4.27±0.46
Na^+ (mg/L)	3746.50±718.76	4112.21±469.21	5071.04±197.78	23.37±2.91
F^-(mg/L)	0.17±0.15	0.30±0.16	0.28±0.06	0.03±0.02

3.2. Trace Metals Distribution in Surface Waters and Groundwater

The spatial distributions of total Ni, Cr, Cd, Zn, Pb and Hg in surface waters and groundwater are shown in Figure 2. Ni concentrations varied between 3.36×10^{-3} and 3.86 mg/L. The averages of total Ni were 1.47±0.45 mg/L, 1.22±0.29 mg/L, 2.36±1.09 mg/L and $4.7\times10^{-3}\pm1.42\times10^{-3}$ mg/L, in the Bandama river, the Kossou lake, the pond water and the groundwater, respectively. Cr concentrations varied between < LD and 0.08 mg/L. The mean values were 0.02±0.01 mg/L in the Bandama river, 0.02±0.03 mg/L in the Kossou lake, 0.03±0.14 mg/L in the pond water and $1.83\times10^{-3}\pm1.42\times10^{-3}$ mg/L in the groundwater. As shown in Figure 1, Cd concentrations varied between 8.16×10^{-4} and 0.32 mg/L. The averages Cd concentrations were 0.05±0.03 mg/L, 0.04±0.03 mg/L, 0.21±0.14 mg/L and $9.75\times10^{-4}\pm3.23\times10^{-4}$ mg/L in the Bandama river, the Kossou lake, the pond water and the groundwater, respectively. Trace metal Zn concentrations ranged from 5.45×10^{-3} to 0.93 mg/L. The means values were 0.71±0.69 mg/L in the Bandama river, 0.61±0.48 mg/L in the Kossou lake, 0.47±0.37 mg/L in the pond water and $5.65\times10^{-3}\pm5.81\times10^{-4}$ mg/L in the groundwater. Hg total concentrations range was 1.01×10^{-3} - 0.97 mg/L. The averages of total Hg were 0.06±0.04 mg/L, 0.62±0.8 mg/L, 0.33±0.46 mg/L and $1.33\times10^{-3}\pm8.6\times10^{-4}$ mg/L, in the Bandama river, the Kossou lake, the pond water and the groundwater, respectively. Pb concentrations varied between 5.4×10^{-4} and 0.25 mg/L. The averages Pb concentrations were 0.06±0.04 mg/L, 0.07±0.05 mg/L, 0.1±0.09 mg/L and $7.34\times10^{-4}\pm4.03\times10^{-4}$ mg/L in the Bandama river, the Kossou lake, the pond water and the groundwater, respectively.

In the surface waters, the averages of total Ni, Cd, Hg, and Pb concentrations exceeded their guidelines values (0.07 mg/L for Ni, 0.003 mg/L for Cd, 0.006 mg/L for Hg, and 0.01 mg/L for Pb) indicating water pollution by these trace metals. While, Cr and Zn average concentrations were below the limit of WHO (0.05mg/L for Cr and 3 mg/L for Zn). In the groundwater, the concentrations of Ni, Zn, Pb, Cd, Cr, and Hg were lower than their WHO guidelines values. The results also showed that surface waters exhibited the most contaminated by Ni, Zn, Pb, Cd, Cr, and Hg than groundwater. This is because average of total Ni, Zn, Pb, Cd, Cr, and Hg concentrations in all surface waters were higher than their WHO guidelines values.

Ni, Zn, Pb, Cd, Cr, and Hg spatial distribution trend in surface waters was as follow:

Ni: pond water > Bandaman > Kossou; Cr: pond water > Bandaman = Kossou; Cd: pond water > Bandaman > Kossou; Zn: Bandaman > Kossou > pond water; Hg: pond water > Kossou >Bandaman; Pb: pond water > Kossou >Bandaman. Ni and Cd showed the same distribution trend, and, Hg and Pb showed the same distribution trend. The distribution patterns of Zn and Cr differed from those of Ni, Cd, Hg and Pb. The same metals distribution trends could indicate the same sources of trace metals. Based on the total concentrations, trace metals Ni, Cd, Hg, and Pb in surface waters might be derived from anthropogenic activities. While, Zn and Cr may be introduced in surface waters through natural processes. Trace metals (Ni, Pb, Cd, and Hg) in surface waters may be due to the discharge from urban, agricultural and mining wastes. For example, during gold production, mercury was used to amalgamate and concentrate the gold [14]. After the washing process, the mercury was released into the water. In addition, the study area is dominated by agricultural activities. Therefore, Ni, Cd, Hg, and Pb in the pond water, the Bandama river and the Kossou lake may also derive from the use of pesticides and fertilizers as mentioned by Lü et *al* [62] and by Ouattara et *al* [36]. Lü et *al* [62] and Luo et *al* [63] have reported that vehicular exhausts from leaded gasoline are a source of Pb in the environment. In the study area vehiculars were used by the miners to collect water for the washing process. Therefore, the use of these vehicles may also explain the high concentrations of Pb. The low concentrations of all trace metals in groundwater may be explained by natural process. [11, 64].

For comparison purposes, the trace metals concentrations in this study and in some other mining areas are summarized in Table 5. Trace metals (Ni, Pb, Cd, Cr, and Hg) concentrations obtained in surface waters around Bouafle mining areas were higher than those reported in surface waters from Ceraína, Brazil [21], Anka, Nigeria [28], Smalong river, China [3], Suárez Cauca, Colombia [20]. However, Zn concentrations were lower than those from Sambo, South Korea [22] and Tributary, China [3]. In groundwater, trace metals concentrations were lower than those from Singhbhum, India [65], Tongon (except Cd), Côte d'Ivoire [35], Buddeun, South Korea [22], SW Ashanti, Ghana [32]. However, the averages concentrations of Cd and Zn in groundwater were higher than those from Samalong, China [3] and Tongon, Côte d'Ivoire [35], respectively.

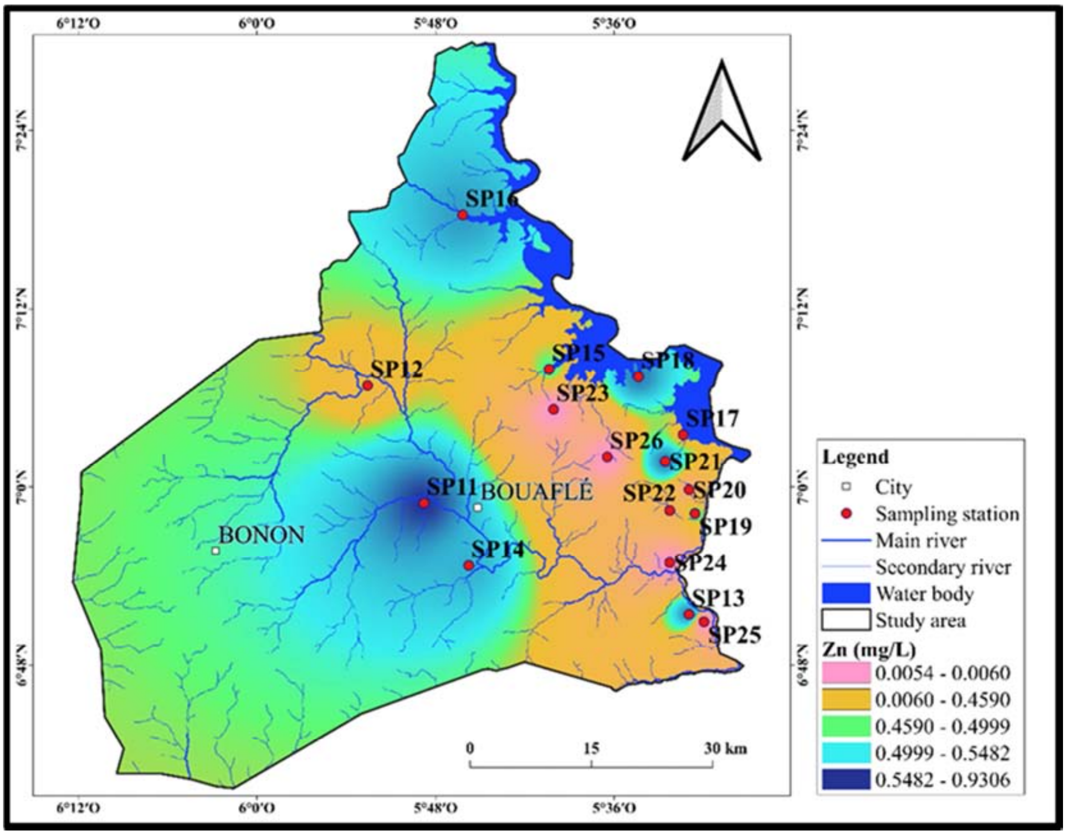
Table 5. *Trace metals concentrations in this study and in some other mining areas.*

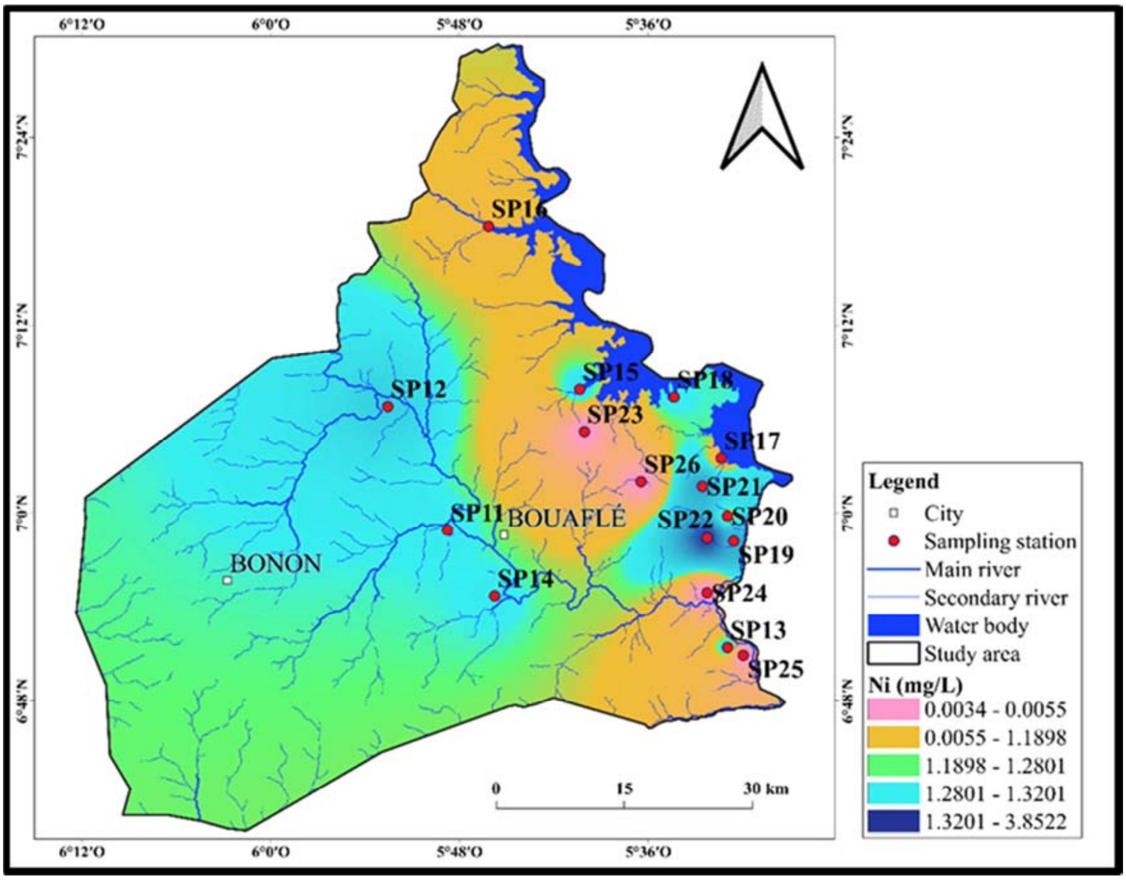
Sample		Country	Ni (µg/L)	Cr (µg/L)	Cd (µg/L)
Surface water	Bandama River	Côte d'Ivoire	1471.2±447.0	22.5±18.3	51.7±32.9
	Kossou lake	Côte d'Ivoire	1220.0±294.5	22.5±31.4	41.5±30.6
	Pond water	Côte d'Ivoire	2360±1088	27.7±34.3	208.0±145.6
	Tongon	Côte d'Ivoire		6±1 – 2505±2711	0.001±0.001 – 4.3±5.96
	Smalong River	China		5.3±0.6	0.009±0.002

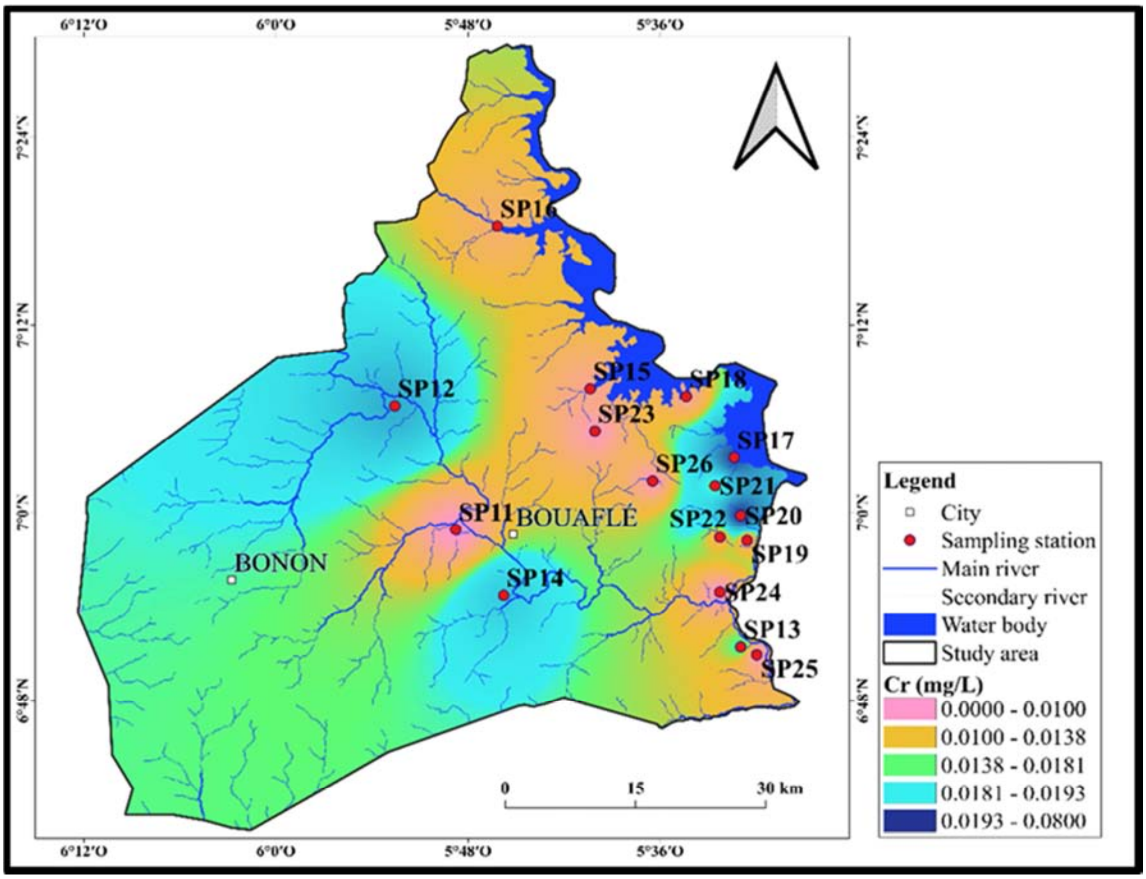
Sample		Country	Ni (µg/L)	Cr (µg/L)	Cd (µg/L)
	Tributary River	China		2.8±1.1	1.04±0.04
	Ceraíma	Brazil	2.1±0.5	2.2±0.4	1.3±0.2
	Suárez Cauca	Colombia			
	Sambo	South Korea			14.0±4.2
	Anka	Nigeria	0.02±0.0001	0.02±0.0001	0.006±0.01
	Ankobra	Ghana			0 -0.004
	Manyera	Nigeria			
Groundwater	Bouafle	Côte d'Ivoire	4.71±1.43	1.83±0.56	0.98±0.32
	Tongon	Côte d'Ivoire		17±17 – 40±77	0.01±0.01 – 0.19±0.54
	Samalong	China	-	3.76±1.61	-
	Buddeun	South Korea			170.2±142.2
	Singhbhum	India	50±30	150±50	20±20
	SW Ashanti	Ghana			

***Table 5.** Continued.*

Sample	Zn (µg/L)	Hg (µg/L)	Pb (µg/L)	Reference
Surface water	701.0±694.5	39.1±37.3	57.5±43.7	This study
	610.755±475.0	616.9±801.6	71.2±52.5	This study
	466.5±368.7	327.0±458.8	97.7±97.9	This study
	5±4 – 625±817		1±1 – 228±455	[35]
	4.46±2.82		0.5±0.16	[3]
	1402±0.04		1.87±2.55	[3]
	38±3	-	19±2	[21]
		0.29±0.36		[20]
	16655.0±14660.9		3.0±2.8	[22]
	0.217±0.004	0.205±0.006	0.510±0.005	[28]
		0.006 – 0.0093	0.006 – 0.0387	[31]
		21.0±4.0		[29]
Groundwater	5.65±0.58	1.33±0.86	0.70±0.40	This study
	19±16 – 71±13.8		1±1 – 34±78	[20]
	2.70±1.58			[3]
	22678.0±26830.7	-	8.25±8.34	[22]
	250±2 00		290±90	[65]
		135.3±86.9		[32]




6°12'O
6°0'O
5°48'O
5°36'O
7°24'N
7°12'N
7°0'N
6°48'N
SP16
SP15
SP18
SP12
SP23
SP17
SP26
SP21
SP11
BOUAFLÉ
SP22
SP20
BONON
SP19
SP14
SP24
SP13
SP25
0
15
30 km
Legend
City
Sampling station
Main river
Secondary river
Water body
Study area
Ni (mg/L)
0.0034 - 0.0055
0.0055 - 1.1898
1.1898 - 1.2801
1.2801 - 1.3201
1.3201 - 3.8522


6°12'O
6°0'O
5°48'O
5°36'O
7°24'N
7°12'N
7°0'N
6°48'N
SP16
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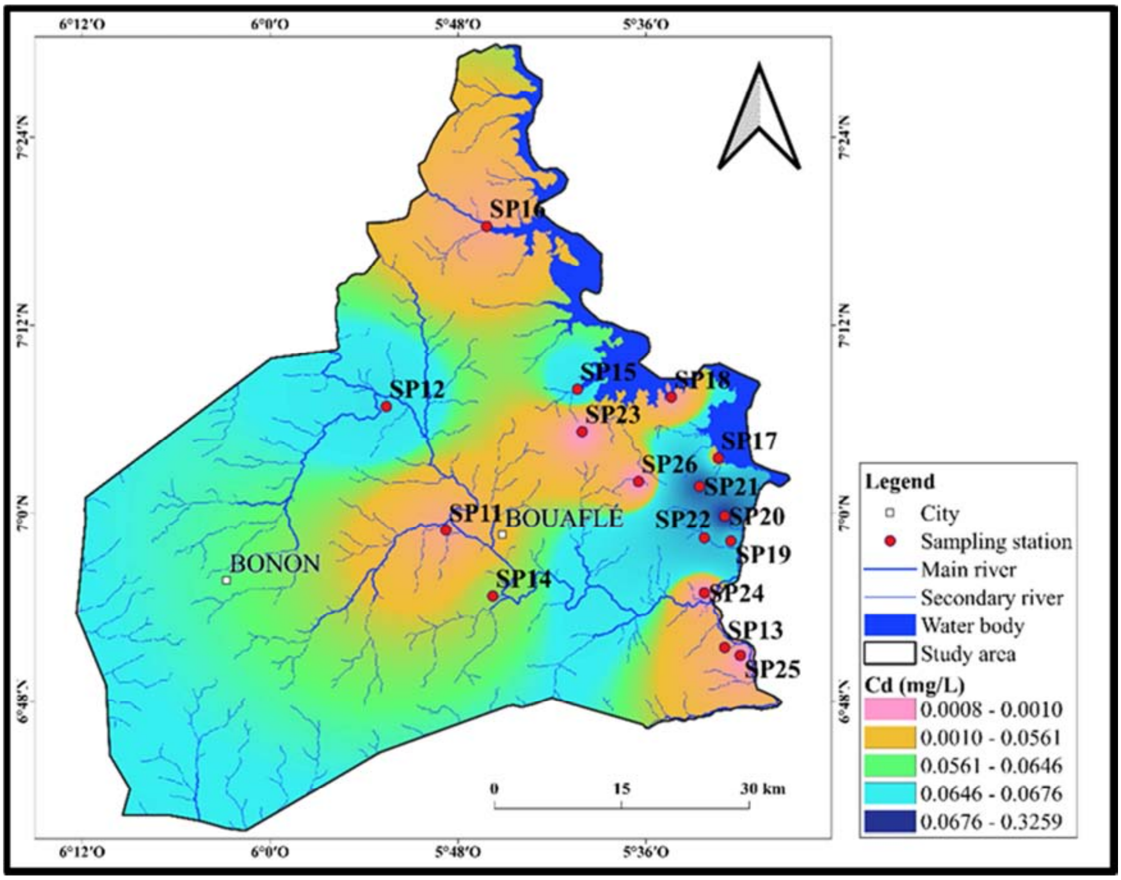
SP18
SP12
SP23
SP17
SP26
SP21
SP11
BOUAFLÉ
SP22
SP20
BONON
SP19
SP14
SP24
SP13
SP25
0
15
30 km
Legend
City
Sampling station
Main river
Secondary river
Water body
Study area
Cr (mg/L)
0.0000 - 0.0100
0.0100 - 0.0138
0.0138 - 0.0181
0.0181 - 0.0193
0.0193 - 0.0800



6°12'O
6°0'O
5°48'O
5°36'O
7°24'N
7°12'N
7°0'N
6°48'N
SP16
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SP18
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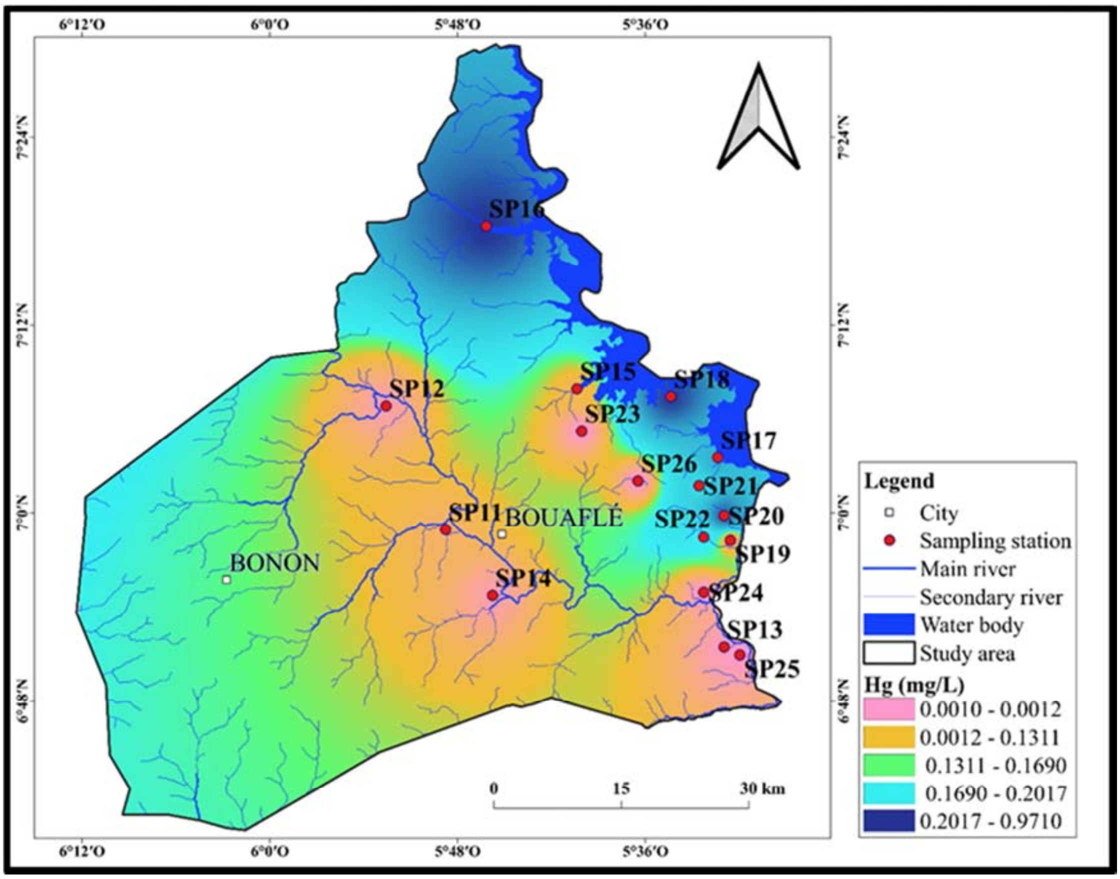
SP23
SP17
SP26
SP21
SP11
BOUAFLÉ
SP22
SP20
SP19
BONON
SP14
SP24
SP13
SP25
0
15
30 km
Legend
City
Sampling station
Main river
Secondary river
Water body
Study area
Cd (mg/L)
0.0008 - 0.0010
0.0010 - 0.0561
0.0561 - 0.0646
0.0646 - 0.0676
0.0676 - 0.3259



6°12'O
6°0'O
5°48'O
5°36'O
7°24'N
7°12'N
7°0'N
6°48'N
SP16
SP15
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SP23
SP17
SP26
SP21
SP11
BOUAFLÉ
SP22
SP20
SP19
BONON
SP14
SP24
SP13
SP25
0
15
30 km
Legend
City
Sampling station
Main river
Secondary river
Water body
Study area
Hg (mg/L)
0.0010 - 0.0012
0.0012 - 0.1311
0.1311 - 0.1690
0.1690 - 0.2017
0.2017 - 0.9710

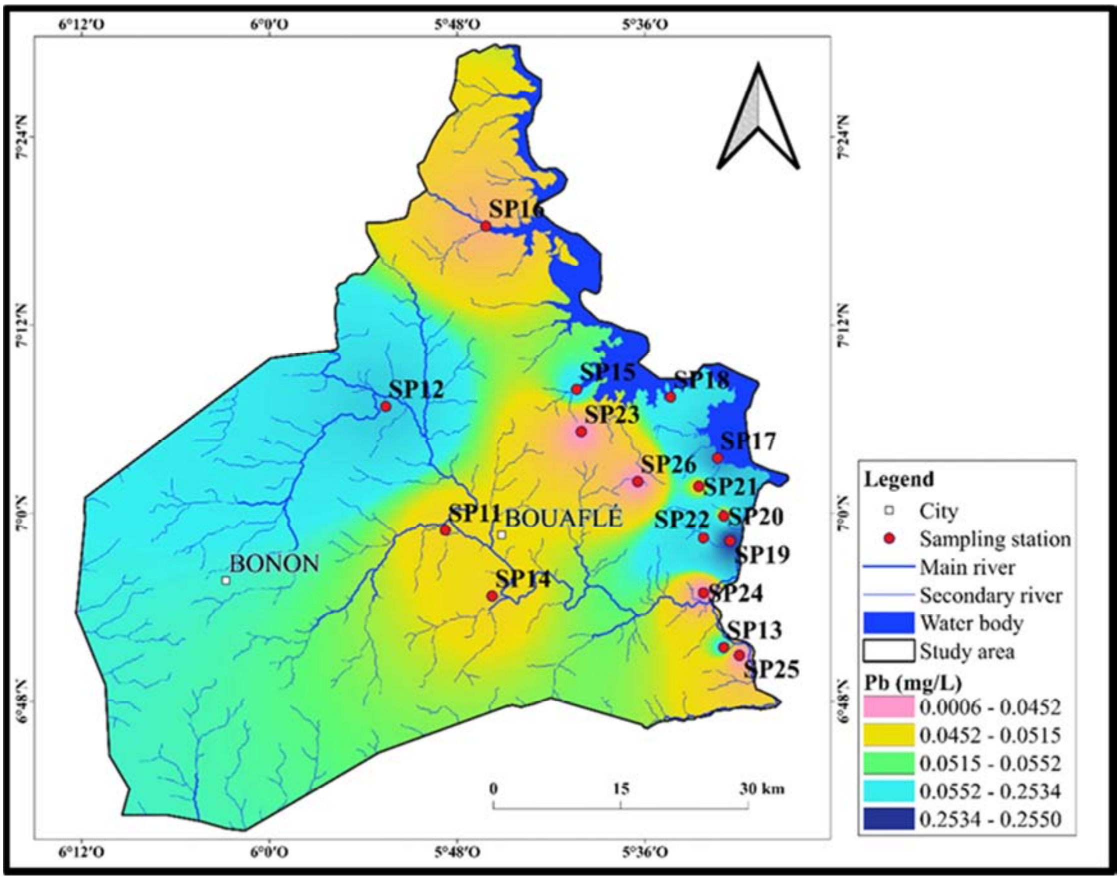

Figure 2. *Distribution patterns of Zn, Ni, Cr, Cd, Hg, and Pb in surface waters and groundwater.*

3.3. Pollution Indices and Water Quality Assessment

The pollution indices HPI and HEI values are shown in Table 6. Based on HPI scale, HPI mean values (1796.39±1258.04 ≤ HPI ≤ 20086.88±25433.87) obtained in surface waters showed that all waters samples were unsuitable for consumption. While, groundwater represented low pollution of heavy metal (HPI = 50.12±12.86). The mean values of HEI varied between 149.30±65.99 and 431.34±405.43 in surface water, indicating high pollution of trace metals in surface waters (HEI > 20). In groundwater, the HEI mean value (1.75±0.77) was lower than 10 showing low pollution of trace metals.

The results of water quality index WQI are reported in Table 5. The WQI mean values of surface waters varied between 372.09±91.54 and 1487.28±800.85. In groundwater, the mean values of WQI were 17.88±1.37. These results showed that groundwater quality is excellent. While, all surface waters are undrinkable.

The high pollution level of surface waters may be due to mining wastes and waste residues from agricultural activities discharge in the surface waters. Therefore, environmental management and reduced surface waters pollution by trace metals are highly crucial.

Table 6. *Averages values of WQI, HPI, and HEI..*

	WQI	HPI	HEI
Bandama	372.09±91.54	1796.39±1258.04	149.30±65.99
Kossou	1325.12±590.19	20086.88±25433.87	431.34±405.43
Pond Water	1487.28±800.85	12532.65±14590.87	416.65±182.69
Groundwater	17.88±1.37	50.12±12.86	1.75±0.77

3.4. Assessment of Human Health Risk

3.4.1. Non-carcinogenic Risk

The averages of HQ_{der}, HQ_{ing}, and HI were reported in Table 7. The results showed that for groundwater, the HQ_{der}, HQ_{ing} and HI values were below 1 for all trace metals, showing low adverse effects.

Trace metals Zn and Cr did not cause any adverse effects in surface waters (HQder, HQing, and HI values were less than one). Ni, Hg, Cd and Pb HQ_{ing} and HI mean values in all the surface waters varied between 1.505±1.144 and 80.507±104.615. Therefore, adverse effects can occur on human health through the ingestion of Ni, Hg, Cd, and Pb. While, HQ_{der} mean values for Ni and Pb were lower than 1 (varying between 0.009±0.003 and 0.04±0.041), showing low adverse effects through dermal contact. In addition, potential adverse effects can occur through Hg by dermal contact with the waters of the Kossou Lake (HQ_{der} = 5.116±6.642) and of the pond water (HQ_{der} = 2.709±3.802). While, for the Bandama river no effects can be caused. Indeed, the HQ_{der} average was 0.324±0.309. Cd concentration in the pond waters can cause adverse effects by dermal contact due to the HQ_{der} average of 1.447±1.013. Surface waters around Bouafle mining areas can cause potential adverse effects related to metals through ingestion due to HQ_{ing} higher than HQ_{der}.

Table 7. *Calculated HQ_{der} HQ_{ing} and HI with trace metals..*

		Bandama	Kossou	Pond water	Groundwater
	HQ_{ing}	2.697±0.819	2.236±0.539	4.326±1.994	0.008±0.003
Ni	HQ_{der}	0.009±0.003	0.008±0.002	0.015±0.007	$3.03\times10^{-5}\pm9.2\times10^{-6}$
	HI	2.707±0.822	2.244±0.542	4.342±2.001	0.008±0.003
	HQ_{ing}	0.275±0.224	0.275±0.224	0.339±0.419	0.022±0.007
Cr	HQ_{der}	0.104±0.085	0.104±0.085	0.128±0.159	0.008±0.002
	HI	0.379±0.309	0.379±0.309	0.468±0.579	0.031±0.009
	HQ_{ing}	3.795±2.414	3.043±2.241	15.253±10.680	0,071±0.024
Cd	HQ_{der}	0.360±0.229	0.288±0.212	1.447±1.013	0.007±0.002
	HI	4.155±2.644	3.332±2.455	16.701±11.694	0.078±0.026
	HQ_{ing}	0.085±0.084	0.074±0.058	0.057±0.045	$6.9\times10^{-4}\pm7.1\times10^{-5}$
Zn	HQ_{der}	0.012±0.012	0.011±0.008	0.008±0.006	$9.84\times10^{-5\pm}1.01\times10^{-5}$
	HI	0.097±0.096	0.085±0.066	0.065±0.051	$7.9\times10^{-4}\pm8.1\times10^{-5}$
	HQ_{ing}	4.781±4.569	75.396±97.973	39.966±56.081	2.560±0.105
Hg	HQ_{der}	0.324±0.309	5.116±6.642	2.709±3.802	0.040±0.007
	HI	5.106±4.879	80.507±104.615	42.676±59.882	0.173±0.112
	HQ_{ing}	1.505±1.144	1.866±1.375	2.560±2.566	0.019±0.010
Pb	HQ_{der}	0.024±0.018	0.029±0.021	0.040±0.041	$3\times10^{-4}\pm1.7\times10^{-4}$
	HI	1.529±1.162	1.895±1.396	2.601±2.607	0.019±0.011

3.4.2. Carcinogenic Risk

Trace metals Pb, Cr, Ni and Cd are recognized as carcinogens for human [66]. Therefore, the carcinogenic risk was assessed using these trace metals. The values of CR_{ing}, CR_{der} and total risk (CR) are reported in Table 8.

Dermal contact of groundwater would have no carcinogenic effect due to all trace metals as, the dermal carcinogenic risk values of Cd, Cr and Ni were below 10^{-6} ($8.2\times10^{-9} \leq CR_{der} \leq 7.5\times10^{-8}$) and those of Pb ($3.1\times10^{-6} \leq CR_{der} \leq 1\times10^{-5}$) were within the tolerable range ($10^{-6} - 10^{-4}$). With surface waters, dermal contact would pose great risk due to Pb ($6.1\times10^{-4} \leq CR_{der} \leq 0.002$), while no significant risk ($2.08\times10^{-8} \leq CR_{der} \leq 1.1\times10^{-5}$) would be pose due to Ni, Cd and Cr.

The CR_{ing} values of Ni, Cr, Cd, Zn, Hg, and Pb for the surface waters exceeded 10^{-4} ($8.7\times10^{-4} \leq CR_{ing} \leq 1.165$), indicating potentially great carcinogenic effects. In groundwater CR_{ing} average for Cd was $9.3\times10^{-5\pm}3.3\times10^{-5}$. The CR_{ing} averages of Ni, Cr and Pb in groundwater were $2\times10^{-4\pm}5.8\times10^{-5}$, $1.6\times10^{-4\pm}5.2\times10^{-5}$ and 0.003±0.002, respectively, indicating possible great carcinogenic effects through ingestion.

The total carcinogenic risk (CR) values of Ni, Pb, Cd, and Cr for the surface waters ranged between 0.002±0.001 and 0.422±0.423. In groundwater, the CR averages of Ni, Pb and Cr varied between $1.6\times10^{-4\pm}5.2\times10^{-5}$ and 0.003±0.002. These results showed that possible carcinogenic effects can occur. In this study, CR_{ing} averages were higher than those of CR_{der}. Therefore, it can be inferred that the population around the artisanal gold mining areas in Bouafle could develop cancer related to Ni, Pb, and Cr through waters ingestion. It is therefore important to treat waters for removing trace metals before using them for irrigation and domestic purposes.

Table 8. *Calculated carcinogenic risk through ingestion (CR_{ing}) and dermal contact (CR_{der}), and total carcinogenic risk (CR) with trace metals.*

		Bandama	Kossou	Pond water	Groundwater
	CR_{ing}	0.059±0.018	0.049±0.012	0.095±0.044	$2\times10^{-4\pm}5.8\times10^{-5}$
Ni	CR_{der}	$1.5\times10^{-5\pm}4.6\times10^{-5}$	$1.2\times10^{-5\pm}3\times10^{-6}$	$2.4\times10^{-5\pm}1.1\times10^{-5}$	$4.7\times10^{-8\pm}1.5\times10^{-8}$
	CR	0.059±0.018	0.049±0.012	0.095±0.044	$1.8\times10^{-4\pm}5.8\times10^{-5}$
	CR_{ing}	0.002±0.001	0.002±0.003	0.002±0.003	$1.6\times10^{-4\pm}5.2\times10^{-5}$
Cr	CR_{der}	$4.6\times10^{-7\pm}3.8\times10^{-7}$	$4.6\times10^{-7\pm}6.5\times10^{-6}$	$5.7\times10^{-7\pm}7.1\times10^{-7}$	$3.7\times10^{-8\pm}1.2\times10^{-8}$
	CR	0.002±0.001	0.002±0.003	0.002±0.003	$1.6\times10^{-4\pm}5.2\times10^{-5}$
	CR_{ing}	0.005±0.003	0.004±0.003	0.020±0.014	$9.3\times10^{-5\pm}3.3\times10^{-5}$
Cd	CR_{der}	$1.2\times10^{-6\pm}7.5\times10^{-7}$	$9.5\times10^{-7\pm}6.9\times10^{-7}$	$4.7\times10^{-6\pm}3.3\times10^{-6}$	$2.2\times10^{-8\pm}7.9\times10^{-9}$
	CR	0.005±0.003	0.004±0.003	0.020±0.014	$9.3\times10^{-5\pm}3.3\times10^{-5}$
	CR_{ing}	0.248±0.188	0.307±0.226	0.422±0.422	0.003±0.002
Pb	CR_{der}	$3.5\times10^{-4\pm}2.6\times10^{-4}$	$4.3\times10^{-4\pm}3.2\times10^{-4}$	$6\times10^{-4\pm}6\times10^{-4}$	$4.7\times10^{-6\pm}2.6\times10^{-6}$
	CR	0.248±0.188	0.307±0.226	0.422±0.423	0.003±0.002

4. Conclusion

Trace metals (Ni, Cr, Zn, Cd, Hg and Pb) distribution and human health risk assessment in surface waters and groundwater around artisanal gold mining areas in the central western of Côte d'Ivoire were investigated. The results showed that Ni, Cd, Pb, and Hg concentrations in groundwater were below the WHO guidelines. While in the surface waters, the averages of total Ni, Cd, Hg, and Pb concentrations exceeded their WHO guidelines values. The spatial maps of trace metals indicated same distribution trend for Ni and Cd, and for Hg and Pb, while those of Zn and Cr differed from those of Ni, Cd, Hg and Pb. The pollution indices HPI and HEI indicated low and high pollution of groundwater and surface waters, respectively. In addition, the WQI index indicated that groundwater quality was excellent. While, all surface waters were undrinkable. The results of non-carcinogenic risk showed that for groundwater, the HQ_{der}, HQ_{ing} and HI values were below 1 for all trace metals, showing low adverse effects. With the surface waters, trace metals Zn and Cr did not cause any threats to human health. The ingestion hazard quotient (HQ_{ing}) and total index (HI) of Cd, Ni, Hg, and Pb exceeded 1 in surface waters, showing adverse effects on humans. Total carcinogenic (CR) of Cd for surface waters showed potential great carcinogenic effects. In contrast, with groundwater, humans are not exposed to cancer effects related to Cd. The total carcinogenic risks (CR) of Cr, Pb, and Ni for surface waters and groundwater exceeded 10^{-4}, indicating a possible carcinogenic effect on human health. It is therefore important to treat waters for removing trace metals before using them for irrigation and domestic purposes. Complementary studies including Ni, Zn, Cd, Hg, Cr, and Pb accumulation in the blood, urine, and hair of residents should be investigated to better understand the risks related to Ni, Zn, Cd, Hg, Cr, and Pb.

Conflict to Interest

The authors declare no competing interests.

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