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Assessing Levels and Health Risks of Fluoride and Heavy Metal Contamination in Drinking Water

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Abstract

A systematic study was carried out to assess the level of contamination with fluorides and heavy metals in the drinking water of the city of Daloa as well as the risks to the health of consumers. The waters of 11.11% of the sites sampled exceeded the fluoride limit for drinking water with a contamination index (CI) greater than 0. All the waters recorded concentrations of cadmium (Cd), copper (Cu), iron (Fe), manganese (Mn) and lead (Pb) above the recommended values with CI > 0. However, 22.22% of the sites recorded concentrations below the standard for zinc (Zn) with IC < 0. The assessment of adverse effects on human health showed that the chronic daily intake (CDI) of fluorine and metals was less than 1 (CDI < 1) for both adults and children except for Zn where the CDI > 1 for children in 22.22% of drinking water studied. HQs have an average of less than 1 for fluorine and greater than 1 for all metals. Moreover, the danger indices have values greater than 1. The incremental lifetime cancer risk (ILCR) and the total ILCR are above the recommended values. These results showed that the drinking water sampled is of poor quality due to higher levels of heavy metals, which can constitute a danger to human health. Long-term use of one of these poor quality waters can lead to cancer in consumers. It is therefore necessary to treat this water in order to eliminate the metals before using it for drinking. This study can help decision-makers and competent authorities in charge of water management.

Keywords

Physico-Chemical Parameters, Fluoride, Heavy Metals, Drinking Water, Sanitary Risks

1. Introduction

Water is a limited and irreplaceable resource (Zakir et al., 2020). In addition to this fact, it is essential not only for the survival of all living beings but also for their well-being (Touré et al., 2019). Access to drinking water, can be a requirement for health, a fundamental right and a key element of effective health protection policies (Touré et al., 2019). Indeed, better access to safe drinking water is of great importance since it leads to tangible health benefits. The supply of safe drinking water is therefore necessary for human life (Mohammadi et al., 2019).

However, the water problem is globally acute more specifically in countries with low water resources (Udhayakumar et al., 2016; Ugwu et al., 2022). Furthermore, access to drinking water and sanitation is a daily challenge for hundreds of thousands of people who live mainly in developing countries (Ohou-Yao et al., 2014). In sub-Saharan Africa, 135 million people, twice as many as in 2000, have limited access to water and sanitation (OMS/UNICEF, JMP, 2020). According to the World Health Organization (WHO), 80% of diseases in developing countries are water-related. In addition, waterborne diseases caused by unsafe water kill 2.6 million people each year (WHO, 2017). Water with an adequate chemical composition is therefore essential for the health and flourishing of human life (Dippong et al., 2021).

Fluoride and heavy metals are common inorganic contaminants in drinking water and, depending on their concentrations in drinking water, they can be hazardous to consumers (Golaki et al., 2022; Ugwu et al., 2022). Fluoride is present in natural waters in the form of free ions and is generally formed by the dissolution of minerals (Li et al., 2014). It is an essential element for the human body, for skeletal and dental growth. The presence of fluoride in drinking water promotes the maintenance of good health, the growth of teeth and bones, but it can be dangerous if the concentration exceeds the maximum limit of 1.5 mg/L (WHO, 2017; Chen et al., 2017). Recently, side effects such as degradation of tooth enamel, paralysis of major joints and spine, nausea, diarrhea, damage to blood cells caused by calcification, neurological problems, bone fractures, impaired thyroid function, reduced birth rate, kidney stones and decreased intelligence in children due to high fluoride intake through drinking water have been reported (Rajmohan, 2022).

Heavy metals are present in the environment in trace amounts. These low concentrations of metals in natural waters have no effects on the aquatic environment and humans. However, human industrial and agricultural activities increase the content of heavy metals in the aquatic environment resulting in the destruction of the aquatic ecosystem and affecting human health (Coulibaly, 2013; Sanou, 2018). Heavy metals are known to harm humans (Sanou et al., 2021a), even at low concentrations (Sanou et al., 2021b). Indeed, these metals are not biodegradable; they are carcinogenic and accumulate in living organisms to cause a wide range of diseases and disorders (Ravikumar & Udayakumar, 2020). Human health hazards related to heavy metal pollution are numerous and some include headache, liver disease, renal, hematopoietic, gastrointestinal system, nerve toxicity and carcinogenic effects (Sanou et al., 2020). Previous studies carried out in Côte d'Ivoire on water resources by Amon et al. (2017), Ohou-Yao et al. (2017), Konan et al. (2018), Kone et al. (2019), Kouyaté et al. (2021), Mangoua-Allali et al. (2021) and Yapo et al. (2021) reported the presence of contaminants such as nutrients, heavy metals and fluorine. This situation remains desperate in many towns such as Daloa. It is therefore important that in-depth studies be carried out, in order to prevent the risks associated with the pollution of drinking water.

This study aims at determining the level of contamination of fluorine and heavy metals in the drinking water of the city of Daloa in order to assess the health risks incurred by the local population.

2. Materials and Methods

2.1. Presentation of the Study Area

The study area (**Figure 1**) is located in the region of "Haut Sassandra" (Centre: West of Côte d'Ivoire) between latitudes 6˚20 and 5˚45 North and longitudes 7˚15 and 6˚30 West. The climate is of the subequatorial type (hot and humid) characterized by four seasons including the long rainy season with off-seasons and storms (April to mid-July), the short dry season (mid-July to mid-September), the short rainy season (mid-September to November) and the long dry season (December to March). However, the duration of the dry seasons clearly gains ground on that of the rainy seasons because of the destruction of the forests by anarchic and abusive exploitation. The average rainfall which fluctuated between 1500 and 1700 mm is only 1200 mm currently. The average annual temperature is 25˚C (Die, 2006).

2.2. Collection and Storage of Water Samples

The study was conducted in six districts of the city of Daloa and a village located about 25 km from the city. A total of ten (10) water points including nine (9) drinking water points and one river water were sampled. The drinking water sampled is of four types: traditional well water, spring water, borehole water and tap water. These different types of water are the main sources of drinking water in the study area. As for the Lobo River, it is used as raw water for the production of drinking water from the taps. The sampling sites and their characteristics are given in Table 1. A total of 11 water quality parameters were analyzed. Water samples were taken in 1.5 liter polyethylene bottles for physico-chemical parameters and 0.5 liter for heavy metals. Each bottle is rinsed with the water to

Table 1. References and geographical coordinates of the sampling sites.

Types of water	Localities	Geographical coordinates		
		Latitude (DMS)*	Longitude (DMS)*	Altitude (m)
Well (P1)	Lobia	6˚54'06.04"N	6˚27'09.81"W	267 m
Well (P2)	Sud C Collège	6˚52'32.08"N	6˚26'06.26"W	242 m
Spring (S1)	Gbokora	6˚54'22.33"N	6˚27'26.21"W	262 m
Spring (S2)	Soleil 1	6˚52'48.88"N	6˚28'48.38"W	218 m
Borehole (F1)	Tazibouo-Université	6˚54'20.68"N	6˚26'71.03"W	275 m
Borehole(F2)	Marais	6˚26'49.25"N	6˚52'06.03"W	245 m
Tap (R1)	Tazibouo-Université	6˚54'20.31"N	6˚26'04.37"W	237 m
Tap (R2)	Nibeigbeu	6˚56'42.99"N	6˚36'04.38"W	235 m
Tap (R3)	Nibeigbeu	6˚56'41.65"N	6˚26'04.37"W	231 m
Lobo River (L)	Nibeigbeu	6˚57'02.01"N	6˚36'15.93"W	240 m

*DMS (Degrees Minutes Seconds).

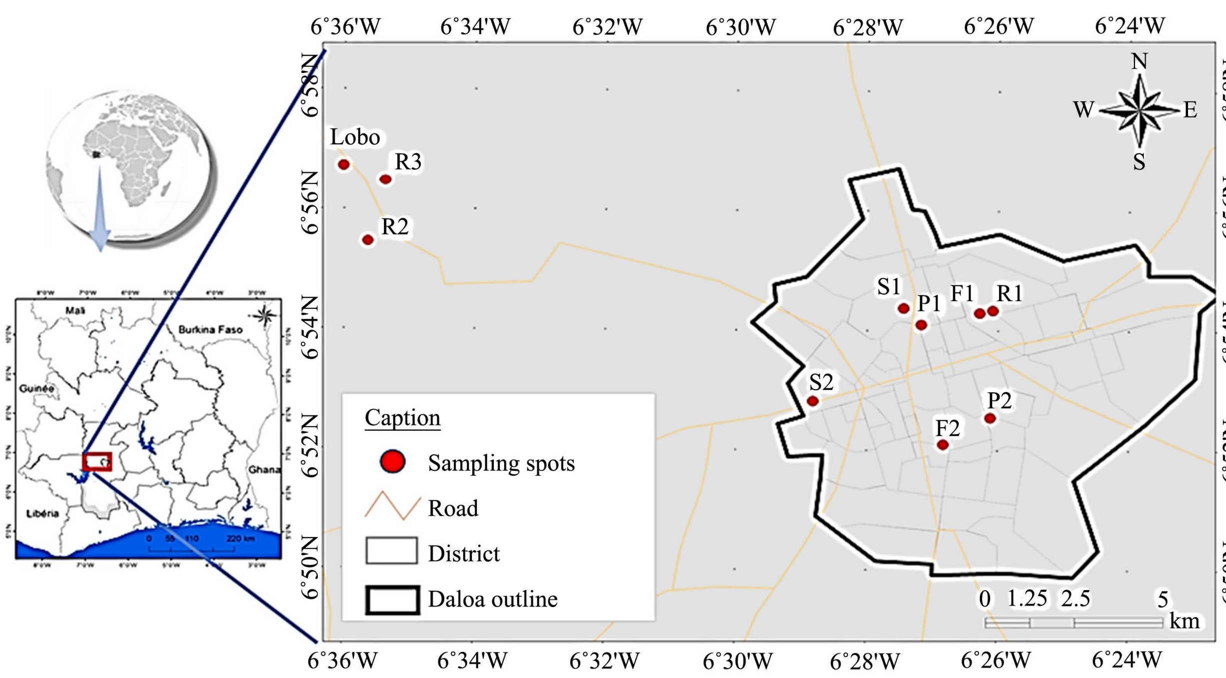

Figure 1. Map of the study area showing the sampling sites.

be collected then filled to be stored in a cooler containing ice. However, a few drops of concentrated hydrochloric acid were added to the 0.5 L samples intended for the analysis of heavy metals.

2.3. Determination of Parameters

The pH was determined in situ using a Horiba LAQUAtwin PH-11 portable pH meter. Magnesium (Mg^{2+}) and calcium (Ca^{2+}) ions were determined by complexometric assay using EDTA with NET (Black Eriochrome T) and Patton and Reeder as indicators. Total hardness (TH) in water samples was determined using Equation (1) in which Ca^{2+} and Mg^{2+} concentrations were expressed in mg/L (Zakir et al., 2020). The fluoride (F^-) content in the water samples was determined by potentiometry using a pH/Ionometer XL250 as described by Yapo et al. (2021). The heavy metals copper (Cu), zinc (Zn), cadmium (Cd), iron (Fe), manganese (Mn) and lead (Pb) were determined using an atomic adsorption spectrometer in flame mode air-acetylene (Varian SpectrAA 20).

$$\mathrm{HT} = \left(2.5 \times \mathrm{Ca}^{2+}\right) + \left(4.1 \times \mathrm{Mg}^{2+}\right) \quad (1)$$

2.4. Metal Load

Metal loading (ML) is defined as the arithmetic sum of the concentration of all metals assayed in the sample. The ML (mg/L) for each water sample was calculated with the mathematical formula given by Equation (2) (Zakir et al., 2020):

$$\mathrm{ML} = \sum_{i=1}^{n} C_m = C_{\mathrm{Cd}} + C_{\mathrm{Cu}} + C_{\mathrm{Fe}} + C_{\mathrm{Mn}} + C_{\mathrm{Pb}} + C_{\mathrm{Zn}} \quad (2)$$

2.5. Metal Pollution Index

The Metal Pollution Index (MPI) is suggested as a reliable and accurate method for monitoring metal pollution (Liu et al., 2019). It makes it possible to compare the total heavy metal content between the different study sites (Sanou et al., 2021b; Sanou et al., 2022). The MPI was calculated with the equation proposed by Usero et al. (1997):

$$\mathrm{MPI} = \left(C_1 \times C_2 \times \cdots \times C_n\right)^{1/n} \quad (3)$$

where C represents the concentration of the metal in the sample and n the total number of metals assayed in the water sample.

2.6. Contamination Index

The Contamination Index (CI) is generally used to assess the cumulative effect of metal content in waters against recommended international standards for drinking water (Backman et al., 1998; Rajmohan, 2022). In the present study, CI is used to explore fluoride and metal contamination. The CI is calculated using Equation (4) (Rajmohan, 2022):

$$\mathrm{CI} = \frac{C_{Ai}}{C_{Ni}} - 1 \quad (4)$$

2.7. Comprehensive Pollution Index

The Comprehensive Pollution Index (CPI) is a comprehensive pollution index method applied to assess qualitatively overall water quality. CPI has been used to classify water quality status by several authors (Zhao et al., 2012; Mishra et al., 2015; Kaioua et al., 2022). The global pollution index is evaluated by Equation (5) (Zhao et al., 2012):

$$\text{CPI} = \frac{1}{n}\sum_{i=1}^{n}\frac{C_i}{S_i} \tag{5}$$

where C_i is the concentration of contaminant i, S_i is the standard norm of contaminant i, n is the number of parameters and CPI is the global pollution index. The permissible standard concentrations of each parameter considered in the study were obtained from WHO standards (WHO, 2017). The CPI varies from 0 to 2 and is used to classify the level of water quality as shown in **Table 2**.

2.8. Health Risk Assessment

Calculation models have been recommended by the United States Environmental Protection Agency (USEPA) to assess the hazards to humans of various contaminants in air, food and water. Among the contaminant exposure pathways, oral ingestion of contaminants such as drinking water consumption is a major concern (Rajmohan, 2022). In the present study, the health risk through the consumption of water contaminated with fluoride and heavy metals was calculated for adults and children.

2.8.1. Non-Carcinogenic Health Risk

The chronic daily intake (CDI) of the different contaminants was calculated using Equation (6) (Alimohammadi et al., 2018; Golaki et al., 2022):

$$\text{CDI} = \frac{C \times \text{IR}}{\text{BW}} \tag{6}$$

where C represents the concentration of the contaminant in drinking water (mg/L), IR is the ingestion rate (L/day), BW is the average body weight (kg) as indicated in **Table 3**.

The non-carcinogenic effects of each contaminant (fluoride and heavy metals) for humans in drinking water are evaluated by the hazard quotient (HQ). The

Table 2. Standard of water quality classification (Mishra et al., 2015).

Comprehensive pollution index (CPI)	Class	Water quality level
≤0.20	I	Cleanness
0.21 - 0.40	II	Sub-cleanness
0.41 - 1.00	II	Slight pollution
1.01 - 2.0	IV	Moderate pollution
≥2.01	V	Severe pollution

Table 3. Parameters used in assessing the health risk of drinking water.

Parameters	Units	Values		References
		Adults	Children	
Concentration of contaminants	mg/L	-	-	**Present study**
Ingestion rate (IR)	L/day	2.5	0.78	USEPA, 2014
Body weight (BW)	kg	65	15	Rajmohan, 2022

HQ is the ratio of the chronic daily dose to the reference dose (Wei et al., 2015). HQ values are estimated in reference to Equation (7) (Qasemi et al., 2018; Rajmohan, 2022):

$$\mathrm{HQ} = \frac{\mathrm{CDI}}{\mathrm{RfD}} \tag{7}$$

where RfD is the oral reference dose (mg/kg/day) of contaminant is recorded in **Table 4**.

When HQ ≤ 1, there are no adverse health effects, however values of HQ > 1 indicate that there are likely adverse health effects (Wei et al., 2015).

The effects due to all the heavy metals on the one hand and to all the contaminants (metals and fluoride) on the other hand were assessed using the hazard index (HI). The HI is the total of the potential non-carcinogenic risks to human health caused by the various contaminants present in drinking water (Zakir et al., 2020). HIs were calculated using Equations (8) and (9).

$$\mathrm{HI}_{\mathrm{m}} = \sum_{i=1}^{n} \mathrm{HQ}_{\mathrm{m}} = \mathrm{HQ}_{\mathrm{Cd}} + \mathrm{HQ}_{\mathrm{Cu}} + \mathrm{HQ}_{\mathrm{Fe}} + \mathrm{HQ}_{\mathrm{Mn}} + \mathrm{HQ}_{\mathrm{Pb}} \tag{8}$$

$$\mathrm{HI} = \sum_{i=1}^{n} \mathrm{HQ}_{\mathrm{m}} + \mathrm{HQ}_{\mathrm{F}} = \mathrm{HQ}_{\mathrm{d}} + \mathrm{HQ}_{\mathrm{Cu}} + \mathrm{HQ}_{\mathrm{Fe}} + \mathrm{HQ}_{\mathrm{Mn}} + \mathrm{HQ}_{\mathrm{Pb}} + \mathrm{HQ}_{\mathrm{Zn}} + \mathrm{HQ}_{\mathrm{F}} \tag{9}$$

If the value of HI ≤ 1, no significant risk of non-carcinogenic effects is expected to occur. However, if HI > 1, there is a possibility of non-carcinogenic effects, and this probability increases with increasing HI value (USEPA, 2001a, 2001b; Wei et al., 2015).

2.8.2. Carcinogenic Health Risk

The carcinogenic risk refers to the probability of occurrence of any type of cancer during the entire lifetime in the event of exposure to a carcinogenic element (Tepanosyan et al., 2017). Incremental lifetime cancer risk (ILCR) was assessed due to exposure to potential carcinogens (Cd and Pb). The possibilities of potential carcinogenic risk that an individual may develop cancer over a lifetime of exposure are calculated by multiplying the CDI by the cancer slope factor (CSF) (USEPA, 2004; Wei et al., 2015; Zakir et al., 2020). The ILCR for each carcinogen was calculated using Equation (10):

$$\mathrm{ILCR} = \mathrm{CDI} \times \mathrm{CSF} \tag{10}$$

where CSF is the cancer slope factor (mg/kg/day). CSF values for the above carcinogens are given in **Table 4**.

Table 4. Reference dose (RfD) and cancer slope factor (CSF) for fluoride and different metals.

Contaminants	RfD (mg/kg/day)	CSF (mg/kg/day)	References
F^-	0.06		USEPA, 1993
Cd	0.001	0.5	
Cu	0.04		
Fe	0.3		USEPA, 2003 USEPA IRIS, 2011
Mn	0.02		
Pb	0.0014	0.0085	
Zn	0.3		

Acceptable risk limits are defined as 1.0×10^{-6} to 1.0×10^{-4} (USEPA, 2010; RAIS, 2017; Mohammadi et al., 2019). However, in the case of the single-element carcinogenic risk, the admissible risk limit is assumed to be 10^{-6}, while for the multi-element carcinogenic risk (ΣILCR, total ILCR), the admissible limit is $<10^{-4}$ (RAIS, 2017).

2.9. Statistical Analysis

To establish a relationship between the physico-chemical parameters, fluoride ions and heavy metals, the Bravais-Pearson correlation test was performed using STATISTICA software (Version 7.1). The linear correlation coefficient r of Bravais-Pearson varies from −1 to +1. The value −1 indicates a perfect negative correlation and the value +1 represents a perfect positive correlation while the value 0 shows an absence of correlation between the parameters (Kouyaté et al., 2021).

3. Results and Discussion

3.1. Physico-Chemical Parameters of Water

Table 5 presents the values of the physico-chemical parameters of the drinking water sampled. Drinking water in the study area is acidic in nature with pH values ranging from 4.4 to 6.53. Only tap water has a pH within the pH range (6.5 - 8.5) recommended for drinking water (WHO, 2017). Raw water from the Lobo River is alkaline in nature (7.18) and falls within the allowable range for water for human consumption. Non-compliant values observed at groundwater levels could lead to irritation of eyes, skin and mucous membranes (Kalankesh et al., 2022). The concentrations of calcium and magnesium in drinking water vary from 2.40 to 28.05 mg/L and from 0.07 to 0.68 mg/L respectively. These values are below the maximum limit authorized by the WHO in drinking water, which is 75 mg/L for Ca^{2+} and 50 mg/L for Mg^{2+} (Salam et al., 2021). The total hardness recorded values that oscillate between 3.29 mg/L and 74.72 mg/L. According to Udhayakumar et al. (2016), the maximum value of total hardness allowed for consumption purposes is 600 mg/L. However, hardness above 300 mg/L can lead

Table 5. Values of the physico-chemical parameters of the sampled waters.

	Types of drinking water									
	P1	P2	S1	S2	F1	F2	R1	R2	R3	L
pH	4.4	5.52	5.34	5.15	5.43	5.75	6.53	6.52	6.42	7.18
Ca^{2+} (mg/L)	5.21	5.21	2.40	1.20	2.81	15.23	28.05	23.25	15.23	8.82
Mg^{2+} (mg/L)	0.17	0.10	0.22	0.07	0.10	0.68	1.12	0.46	0.63	1.61
TH (mg/L)	13.72	13.44	6.90	3.29	7.44	40.86	74.72	60.01	40.66	28.65

to heart and kidney problems (Udhayakumar et al., 2016). With regard to the results of total hardness, all the water samples were identified as being suitable for human consumption. All tap water samples (100%) comply with the potability standard, indicating that the raw water treatment technique used by the Ivorian water distribution company is adequate for these parameters.

3.2. Concentration of Fluorine in Waters

The fluoride concentrations in the sampled waters oscillate between 0 and 2.4 mg/L (**Figure 2**). The P1, S1 and F1 sites do not contain fluoride (0 mg/L). According to He et al. (2020), a low concentration of fluoride (F < 0.5 mg/L) in drinking water causes dental caries in infants and children, as well as osteoporosis.

The P2, S2, F2, R2 and R3 sites have contents which fluctuate between 0.8 and 1.3 mg/L. These concentrations remain below the standard (<1.5 mg/L) suggested by the WHO (WHO, 2017) and could be beneficial for consumers. Indeed, fluoride concentrations between 0.4 and 1.0 mg/L in drinking water has beneficial effects on teeth, especially in young children, as it promotes calcification of dental enamel and protects teeth against dental caries (Loganathan et al., 2013; Yapo et al., 2021). The R1 tap is not suitable for consumption. Indeed, this water has a fluoride content (2.4 mg/L) higher than the standard suggested by the WHO. Consumption of water with a high fluoride concentration (F > 1.5 mg/L) for a long period can lead to dental fluorosis (Adewole et al., 2021). Concentrations above the standard have already been encountered in borehole water at Boguédia in the department of Daloa (2.20 ± 0.01 mg/L) by Yapo et al. (2021).

3.3. Heavy Metal Content, Metal Load and Metal Pollution Indices in Water

The heavy metal contents in the drinking water samples taken varied from 2.56 to 4.47 for Cd, from 3.14 to 4.74 for Cu, from 4.93 to 10.70 for Fe, from 2.74 to 5.91 for Mn, from 4.42 to 11.66 for Pb and from 1.93 to 20.91 for Zn. Taking into account the standard norms (**Table 6**) recommended for drinking water by

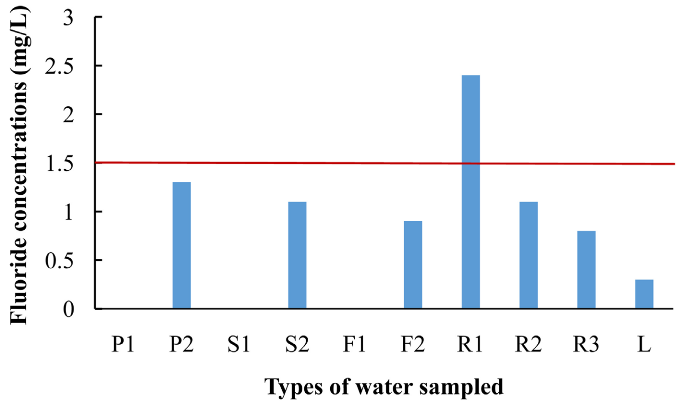

Figure 2. Concentration of fluoride in water.

Table 6. WHO standards for fluorides and heavy metals.

	Contaminants (mg/L)						
	F^-	Cd	Cu	Fe	Mn	Pb	Zn
WHO (2017)	1.5	0.003	2	0.3	0.4	0.01	3

the WHO, the results recorded show that all the water samples are inappropriate for consumption except the well P2 and the borehole F1 in the case of Zn. These high concentrations can create adverse health effects such as cancer, hypertension, lung disease, gastrointestinal bleeding, kidney disease, neurological disorders and reproductive effects (Bhan & Sarkar, 2005; Zakir et al., 2020). The results indicate that the treatment of the distribution company effectively reduces the content of the metals Cd, Cu and Pb. However, there is an increase in the content of Fe, Mn and Zn in all the tap waters in comparison with that of the raw water (**Table 7**). This presence of metals at a high rate is probably due to the aging of the equipment of the supply or connection network pipes on the one hand (Douard & Lebental, 2013) and on the other hand to the inefficiency of the treatment. Metal loading (ML) reflects the total amount of metals assayed in a sample. The metal load is higher in tap R3 (50.19 mg/L) and lower in borehole F1 (26.09 mg/L). The order of accumulation of metal load in drinking water is: R3 > R1 > S1 > R2 > P1 > S2 > F2 > P2 > F1 (**Table 7**). The Metal Pollution Index (MPI) was used to compare the overall content of heavy metals in the sampled waters. The values recorded indicate overall contamination in the following decreasing order: R3 > S1 > R2 > R1 > P1 > S2 > F2 > P2 > F1 (**Table 7**).

However, these values are greater than 1 (MPI > 1); thus reflecting that all the metals contained in drinking water are not without danger for human consumption (Abdel Ghani, 2015; Sanou et al., 2021b). In general, the treatment provided by the water distribution company reduces overall metal pollution. Indeed, the MPI of the Lobo River, source of raw water for the production of drinking water, is higher than the MPI of all the taps (**Table 7**).

Table 7. Concentrations of heavy metals, ML and MPI in water.

	Heavy metals (mg/L)						ML	MPI
	Cd	**Cu**	**Fe**	**Mn**	**Pb**	**Zn**		
P1	3.79	4.74	4.93	4.99	11.02	7.17	**36.64**	**5.72**
P2	3.88	3.93	7.04	4.66	6.33	2.66	**28.50**	**4.51**
S1	3.57	3.14	10.40	4.42	7.08	14.90	**43.51**	**6.15**
S2	3.30	3.48	8.57	3.91	5.52	11.53	**36.31**	**5.39**
F1	4.47	3.76	5.78	3.07	7.08	1.93	**26.09**	**4.00**
F2	3.77	4.00	8.74	2.74	5.39	6.12	**30.76**	**4.78**
R1	2.86	4.13	10.39	4.29	4.42	20.91	**47.00**	**6.04**
R2	2.88	4.46	9.46	5.91	5.11	13.88	**41.70**	**6.09**
R3	2.56	3.25	10.70	5.46	8.42	19.80	**50.19**	**6.58**
L	4.35	4.55	7.15	5.18	11.66	10.18	**43.07**	**6.66**

3.4. Contamination and Comprehensive Pollution Indices

In the waters sampled, the values of the IC vary from −1 to 0.6 with an average of −0.47 ± 0.51 for F^-; from 852.33 to 1489 with an average of 1180 ± 212.82 for Cd; from 0.57 to 1.37 with an average of 0.97 ± 0.27 for Cu; from 15.43 to 34.67 with an average of 26.72 ± 6.74 for Fe; from 5.85 to 13.78 with an average of 10.16 ± 2.52 for Mn; from 441 to 1165 with an average of 719.3 ± 247.10 for Pb; from −0.36 to 5.97 with an average of −2.67 ± 2.19 for Zn. According to Rajmohan (2022), IC value ≤ 0 indicates drinking while water samples with IC value > 0 indicate unpleasant drinking water as shown in **Table 8**.

The CI for fluoride is less than zero in 88.88% of samples whose consumption is advised. Regarding heavy metals, all CIs are greater than zero in the samples studied. The consumption of these waters is therefore not recommended. However, 37.5% of the samples have IC < 0 for Zn which makes them acceptable as drinking water. The complete pollution index (CPI) has values ranging between 241.89 and 443.11 with an average of 324.39 ± 63.97. These values are well above 2 (CPI > 2); indicating severe pollution of the waters studied (Mishra et al., 2015) which makes them unfit for human consumption. The high CPI values could be attributed to the high concentration of iron, lead and zinc recorded during the study.

3.5. Correlation between Variables

The Bravais-Pearson correlation matrix was created to elucidate the relationships between the variables measured in the different water samples. The correlation coefficients are presented in **Table 9**. The coefficients in bold are significant at $p < 0.05$. Examination of the matrix indicates the existence of positive and significant correlations between the physico-chemical parameters, fluorides

Table 8. Values of contamination index and comprehensive pollution.

	Contamination Index (CI)							CPI
	F^-	Cd	Cu	Fe	Mn	Pb	Zn	
P1	−1	1262.33	1.37	15.43	11.48	1101	1.39	**399.83**
P2	−0.13	1292.33	0.97	22.47	10.65	632	−0.11	**327.53**
S1	−1	1189	0.57	33.67	10.05	707	3.97	**325.04**
S2	−0.27	1099	0.74	27.57	8.78	551	2.84	**282.78**
F1	−1	1489	0.88	18.27	6.68	707	−0.36	**371.24**
F2	−0.40	1255.67	1	28.13	5.85	538	1.04	**306.05**
R1	0.60	952.33	1.07	33.63	9.73	441	5.97	**241.89**
R2	−0.27	959	1.23	30.53	13.78	510	3.63	**254.15**
R3	−0.47	852.33	0.63	34.67	12.65	841	5.6	**292.23**
L	−0.80	1449	1.28	22.83	11.95	1165	2.39	**443.11**
Min	−1	852.33	0.57	15.43	5.85	441	−0.36	**241.89**
Mean	−0.47	1180	0.97	26.72	10.16	719.3	2.64	**324.39**
Max	0.6	1489	1.37	34.67	13.78	1165	5.97	**443.11**
SD	0.51	212.82	0.27	6.74	2.52	247.10	2.19	**63.97**

Table 9. Bravais-Pearson correlation coefficients between water variables.

	pH	Ca^{2+}	Mg^{2+}	HT	F^-	Cd	Cu	Fe	Mn	Pb	Zn
pH	1	**0.63**	**0.82**	**0.67**	0.38	−0.24	0.13	0.47	0.36	−0.02	0.47
Ca^{2+}		1	**0.52**	**0.99**	**0.70**	**−0.62**	0.30	**0.53**	0.27	−0.40	**0.59**
Mg^{2+}			1	**0.58**	0.26	0.01	0.39	0.22	0.19	0.25	0.37
HT				1	**0.68**	**−0.59**	0.32	**0.53**	0.27	−0.36	**0.59**
F^-					1	**−0.58**	0.04	0.49	0.04	**−0.66**	0.44
Cd						1	0.26	**−0.77**	−0.44	0.43	**−0.82**
Cu							1	**−0.77**	−0.44	0.43	**−0.82**
Fe								1	0.18	**−0.52**	**0.82**
Mn									1	0.35	0.46
Pb										1	−0.17
Zn											1

*The coefficients in bold are significant correlation at $p < 0.05$.

and heavy metals: pH - Ca^{2+} (r = 0.63), pH - Mg^{2+} (r = 0.82), pH - TH (r = 0.67), Ca^{2+} - Mg^{2+} (r = 0.52), Ca^{2+} - TH (r = 0.99), Ca^{2+} - F^- (r = 0.70), Ca^{2+} - Fe (r = 0.53), Ca^{2+} - Zn (r = 0.59), Mg^{2+} - TH (r = 0.58), TH - F^- (r = 0.68), TH - Fe (r = 0.53), TH - Zn (r = 0.59), Fe - Zn (r = 0.82). These observed significantly positive correlations between the variables indicate the mutual dependence of the parameters on each other, a common source or similar behavior during transport (Shetaia et al., 2020; Sanou et al., 2021a, 2021b). Furthermore, significant negative correlations were found between Ca^{2+} - Cd (r = −0.62), TH - Cd (r = −0.59), F^- - Cd (r = −0.58), Cd - Fe (r = −0.77), Cd - Zn (r = −0.82), Cu - Fe (r = −0.77), Cu - Zn (r = −0.82). These results show an inverse dependence between these variables, thus reflecting the decrease in one of the parameters with the increase in the other (Sanou et al., 2020; Kouyaté et al., 2021; Sanou et al., 2021b). Non-significant correlations (r < 0.5) were observed between certain variables. These low correlations indicate that the presence or absence of one of these parameters has little effect on the content of the other (Kam et al., 2019).

3.6. Health Risk Assessment

3.6.1. Chronic Daily Intake (CDI)

The chronic ingestion of heavy metals, beyond the tolerance threshold in humans, has harmful effects and can cause neurological damage, headaches, diseases of the liver, renal, hematopoietic and gastrointestinal systems (Sanou et al., 2021b). The chronic daily intake (CDI) of fluoride and heavy metals from ingestion of waters was calculated as reported by the United States Environmental Protection Agency (USEPA).

The CDI values obtained for fluoride vary from 0 to 0.092 with an average of 0.030 mg/kg/day for adults and they oscillate between 0 and 0.125 with an average of 0.041 for children (**Table 10**). According to the USEPA guidelines for fluoride, 11.11% and 22.22% of the samples show CDIs above the oral reference dose for adults and children respectively (USEPA, 1993). However, the CDI values for fluoride are all less than 1 (CDI < 1). These results suggest that the effects of fluoride related to the consumption of these waters are very unlikely among consumers. Indeed, according to Rajmohan (2022), only CDIs greater than 1 (CDI > 1) can pose a threat to human health.

The metals studied have CDIs which vary from 0.098 to 0.448 mg/kg/day for adults and from 0.100 to 1.087 for children as presented in **Table 10**. The CDIs

Table 10. Minimum, mean, and maximum values of chronic daily intake (CDI) for fluoride and heavy metals in the water of the study area.

CDI (mg/kg/day)	Adults			Children		
	Min	Mean	Max	Min	Mean	Max
F^-	0	0.030	0.092	0	0.041	0.125
Cd	0.098	0.136	0.172	0.133	0.184	0.232
Cu	0.121	0.152	0.182	0.163	0.205	0.246
Fe	0.190	0.320	0.412	0.256	0.432	0.541
Mn	0.105	0.172	0.199	0.142	0.232	0.307
Pb	0.170	0.277	0.448	0.280	0.375	0.573
Zn	0.102	0.420	0.762	0.100	0.567	1.087

of Cd, Cu, Mn and Pb are above the reference dose for all water samples. However, 33.33% and 11.11% of the samples had Fe CDIs below the oral reference dose for adults and children respectively. Furthermore, Zn has CDIs lower than the oral reference dose in 44.44% of the samples for adults and 22.22% for children. However, only 22.22% have CDIs above 1 (CDI > 1) for zinc. These results suggest that Zn may have health effects on consumers of R1 and R2 tap water.

3.6.2. Non-Carcinogenic Risks

Table 11 presents the results of the hazard quotients (HQ) and the hazard indices (HI). The values of HQ(F) vary from 0 to 1.538 and from 0 to 2.08 for adults and children respectively. The mean value of HQ is less than 1 for adults (0.506) and for children (0.685). However, according to USEPA guidelines, 11.11% and 22.22% of the water samples studied indicate a non-carcinogenic threat to adults and children respectively. The non-cancer risks are higher in children than in adults. This difference would be due to their very small body weight. A recent study indicated that children were more vulnerable to fluoride contamination than adults (Rajmohan, 2022).

In general, all the heavy metals measured in the waters had HQs > 1 for children and adults. Therefore, the latter present harmful non-carcinogenic health risks in all samples collected in the study area except for Fe in three samples for adults and one for children and for Zn in four samples for adults and two for children. The contribution of the metals studied to the non-carcinogenic health risk was of the following order: Pb > Cd > Mn > Cu > Zn > Fe for both adults and children.

The danger index was calculated to estimate the total potential non-carcinogenic effects induced by all the contaminants. The mean values of HIm and HI are very well above 1 (HIm > 1) for adults and children as shown in Table 11.

Table 11. Minimum, mean, and maximum values of non-carcinogenic human health risks posed by fluoride and heavy metals in the water of the study area.

HQ	Adults			Children		
	Min	Mean	Max	Min	Mean	Max
F^-	0	0.506	1.538	0	0.685	2.08
Cd	98.462	136.269	171.923	133.120	184.236	232.440
Cu	3.019	3.792	4.558	4.082	5.127	6.192
Fe	0.632	1.066	1.372	0.855	1.441	1.855
Mn	5.269	8.583	11.365	7.124	11.604	15.366
Pb	121.429	197.885	320.330	164.171	267.54	433.086
Zn	0.274	1.398	2.681	0.335	1.891	2.583
HIm	247.663	348.993	504.196	334.840	471.839	681.673
HI	249.201	349.450	504.388	340.544	474.415	683.697

Table 12. Minimum, mean, and maximum values of the incremental lifetime cancer risk (ILCR) values of carcinogenic human health risks via ingestion exposure to the drinking water of the study area for adults.

Metal	ILCR					
	Adults			Children		
	Min	Mean	Max	Min	Mean	Max
Cd	4.92E−02	6.81E−02	8.37E−02	6.66E−02	9.21E−02	1.16E−01
Pb	1.45E−03	2.35E−03	3.81E−03	1.95E−03	3.18E−03	5.15E−03
Σ ILCR	5.19E−02	7.05E−02	8.83E−02	7.03E−02	9.53E−02	1.19E−01

These results indicate probable non-carcinogenic health risks to consumers. Pb and Cd are the main contributors to the non-carcinogenic risk due to metals with 56.70% and 39.05% respectively, for a total of 95.75%. The high values of HI are due to the HQ values of heavy metals which are very high.

3.6.3. Carcinogenic Risks

Among heavy metals, some are carcinogenic and can increase the risk of cancer in humans (Cao et al., 2014). Long-term exposure to low amounts of toxic metals could therefore lead to many types of cancers (Mohammadi et al., 2019). In the present study, Cd and Pb were used as carcinogens to assess the carcinogenic risk for adults and children. The results obtained are shown in **Table 12**. The ILCR values vary from 4.92E−02 to 8.37E−02 with an average of 6.81E−02 (Cd) and 1.45E−03 to 3.81E−03 with an average of 2.35E−03 (Pb) for adults while for children the ILCR varied from 6.6E−02 to 1.16E−01 with an average of 9.21E−02 (Cd) and 1.95E−03 to 5.15E−03 with an average of 3.18E−03 (Pb). These ILCR values for adults and children are all greater than 1×10^{-6}, suggesting that exposure to carcinogenic risk of Cd and Pb in drinking water is not negligible (RAIS, 2017; Mohammadi et al., 2019). The total ILCR for children and adults are above the allowable limit of 1×10^{-4} (RAIS, 2017). These results suggest a risk of cancer due to all metals, both for adults and for children. Furthermore, the present study indicates that Cd contributed more than 96.60% of the overall ILCR for adults and more than 96.64% for children. Therefore, the carcinogenic risk of Cd and Pb requires attention for the control of pollution by managers and more particularly that of Cd.

4. Conclusion

This study was carried out with the aim of evaluating the health risks of the population of the study area due to the consumption of water with a high content of fluoride and heavy metals. The study showed that the waters of the study area had concentrations of heavy metals above the authorized limits for drinking water except zinc which recorded concentrations below the standard for 22.22% of the waters sampled. The health risk assessment showed that the chronic daily intake of fluorine and metals was less than 1 (CDI < 0) for children and adults except for Zn where the CDI > 1 for children in 22.22% drinking water studied. HQs have an average of less than 1 for fluorine and greater than 1 for all metals. In addition, the HI is greater than 1 for both children and adults. These results suggest that the waters studied are not of good quality and constitute a danger to human health. The ILCR and total ILCR for adults and children are above the recommended values which respectively indicate that long-term consumption of these waters may cause cancer in consumers. Furthermore, cadmium has the highest risk of cancer for adults (96.60%) and for children (96.64%). Therefore, the carcinogenic risk of Cd and Pb requires attention for pollution control by managers and more particularly that of Cd. The present study can help decision makers and competent authorities in charge of drinking water management.

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Conflicts of Interest

The authors declare that they have no conflict of interest.

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