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Contribution to the Speciation of Some ETM in the Drinking Waters of the City of Daloa (Ivory Coast)

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ABSTRACT

Our study aims to determine the different chemical forms of metallic trace elements (MTE) in the groundwater of the city of Daloa. To do this, we took ten water samples, six of which were from peasant wells, two from boreholes and springs. The concentrations of the MTEs studied were determined before and after filtration in addition to the statistical processing of the data, in particular the study of their correlations and the principal component analysis. At the end of our study, it emerges that the metallic trace elements studied are present in the drinking water of the city of Daloa. Their consumption could cause health risks due in particular to the presence of arsenic, which has effects without thresholds.

Also, there are inorganic salts of calcium $CaCl_2$, zinc ($ZnCl_2$), cadmium ($CdCl_2$) and arsenic ($AsCl_3$) in the drinking waters of the city of Daloa. Moreover, after filtration, we have more or significant reductions in concentrations for the various trace elements studied. This reveals the presence of MTE in the dissolved phase. Hence the presence of bioavailable forms of arsenic As (s), cadmium (Cd^{2+}) zinc (Zn^{2+}), nickel (Ni^{2+}) and lead (Pb^{2+}) in the drinking waters of the town of Daloa.

KEYWORDS : groundwater, filtration, speciation, bioavailability, health risks,

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INTRODUCTION

Access to so-called "potable" water is a universal right which is one of the United Nations' sustainable development objectives adopted in September 2015. To date, the United Nations estimate that nearly a billion people do not have access to drinking water. Populations without access to good quality water are mostly found in developing countries[1]. In Ivory Coast, SODECI, in agreement with the state, must provide the population with treated water that is bacteriological, physicochemical and organoleptic in conformity with the consumption of drinking water, a determining factor in the prevention of water-related diseases should receive special attention[2]. The water produced and distributed by SODECI-Daloa (taken from the tap) is microbiologically drinkable, that is to say that it meets WHO guidelines[3]. As a result, it does not represent any danger for the populations. However, its yellowish color, the presence of deposits, the smell of bleach and its bad taste do not leave consumers indifferent who question its quality. This leads them to consume water from peasant wells, springs and boreholes without prior treatment.

In addition, MTEs are present in all compartments of the environment[4,5], especially in the groundwater of the city of Daloa. They are naturally present there (natural sources) and numerous human activities favor their dispersion. In addition, unlike organic pollutants, MTEs are hardly subject to biological or chemical degradation reactions and can accumulate in food chains[6,7].

Furthermore, the toxicity of a metal depends not only on its concentration but also on its speciation[8]. Also, it is accepted that the mobility, bioavailability and toxicity of MTEs depend more on their chemical forms rather than on their total concentration[9,10,11]. In fact, only a fraction of the total content of trace elements in water is available for living organisms. Speciation, i.e. the chemical forms of an MTE, is thus intimately linked to its bioavailability and is therefore of fundamental importance in estimating the environmental risks associated with its presence.

In Daloa, drinking water could contain bioavailable chemical forms toxic to human health. It is with this in mind that this study attempts to specifically determine not only the levels of arsenic, cadmium and chromium in groundwater (drinking water) in the city of Daloa but also to identify the bioavailable chemical forms. toxic of these MTEs.

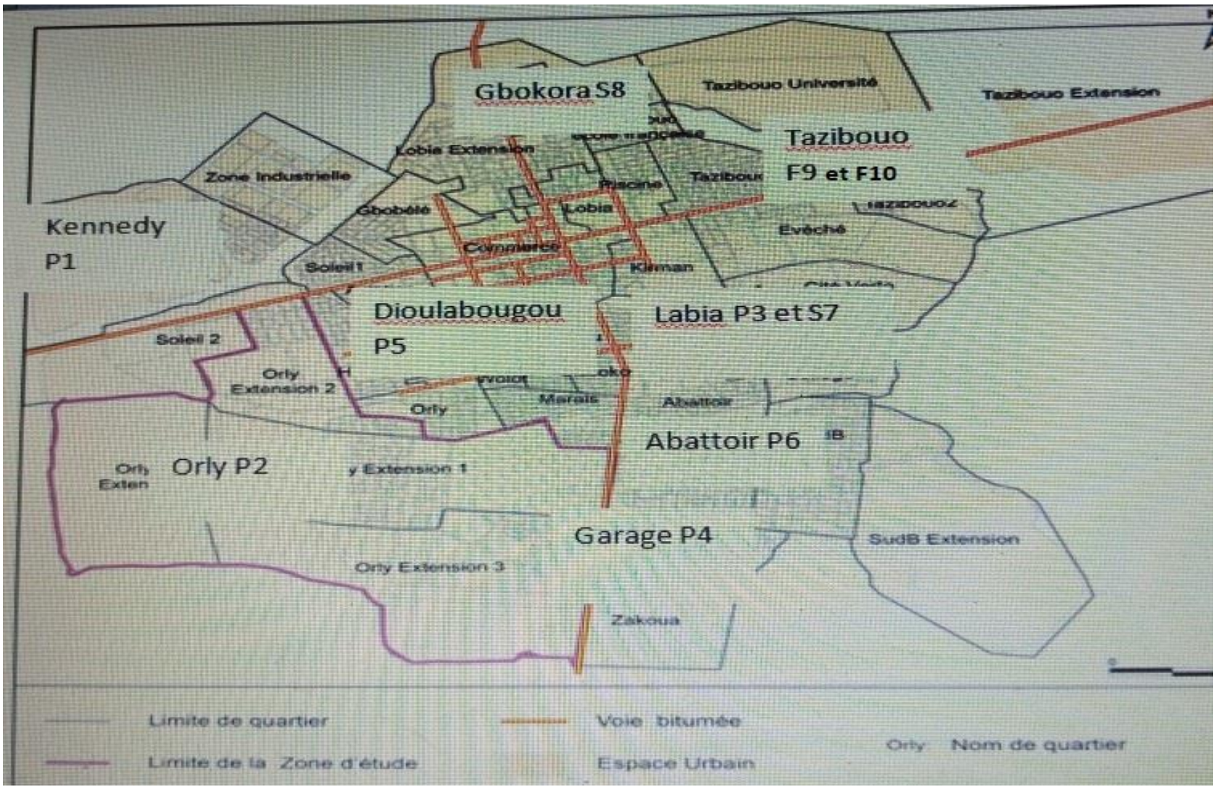
MATERIALS AND METHOD

Presentation of the study area

Daloa is a town located in the center-west of the Ivory Coast between 6 ° 53’ North latitude and 6 ° 27’ West longitude. The capital of the Haut-Sassandra region, the city is the main urban center of the Center-West. It covers an area of 3,802 km^2. Its population was estimated at 243,048 inhabitants[12] with an annual growth rate of 3.5 %. Today this population is estimated at 251,555 inhabitants. Figure 1 gives the map of the city of Daloa with its districts.

Sampling

The sampling stations were chosen according to specific criteria, in particular those used for consumption and their situations, in particular the stations located in the most disadvantaged districts.



Graph.1: Geographical location of the different sources (BNETD / CCT, 2016, modified)

Bushner filtration method

After filtration on bushner using porosity of 0.48 µm the calculation of the reduction percentage was carried out according to the followind formula :

% R = (Cx-Cx')/ Cx

Cx : Concentration of the metallic trace element before filtration

Cx ' : Concentration of the metallic trace element after filtration

% R : Percentage reduction of the trace element after filtration

Methods of analysis of physicochemical parameters

Temperature, pH, conductivity, TDS in groundwater

The physicochemical parameters, namely temperature, pH, electrical conductivity, salinity and TDS were measured in situ at each water point sampled using the HANNA portable multi-parameter. For these various parameters, the previously calibrated measuring devices are switched on a few minutes before any manipulation. The probe is then immersed in the water sample taken from a small weighted bucket and rinsed three times, selecting the desired parameter function gives the value of the latter on the display screen.

Chemical parameters determined in the laboratory

Flame method (Fe, Zn, Cl^-, Ca^{2+})[13]

Each water sample is introduced into the device through the capillary tube which aspirates approximately 2 mL / min of each sample which will be sprayed on the optical path of the device while respecting the operating conditions mentioned below in accordance with the method analytical used. The sample is sprayed into the flame (atomic excitation source) while forming an atomic cloud which is crossed by the monochromatic light characteristic of the element to be quantified. The excited atoms of the element absorb (case of absorption) or emit (case of emission) an amount of energy to go from the excited state to the ground state. This absorption or emission is proportional to the concentration of atoms which absorb or emit.

Furnace method (As, Cd, Cr, Ni and Pb)[13]

Between 10 and 20 µL / min of water samples are taken by the automatic injector to be injected into the graphics tube. Once deposited, this drop is dried and then sprayed onto the optical path of the device, forming an atomic cloud, which is crossed by the monochromatic light characteristic of the element to be quantified. Atoms of this element absorb a characteristic amount of light at a specific wavelength to go from the excited state (atomic cloud) to the ground state. The amount of absorbance is proportional to the concentration of absorbing atoms.

Hydride Method (Hg)[13]

The water or mineralized sample is aspirated and mixed with tin chloride in a gas-liquid separator where the mercuric ion is reduced to gaseous elemental mercury according to the following reduction reaction :

$$Hg^{2+} + Sn^{2+} \rightarrow Hg^{\circ} + Sn^{4+}$$

The generated cold mercury vapors are entrained by a stream of nitrogen directly to the detector for quantification.

Statistic study

In our study, we carried out a principal component analysis (PCA), with a view to highlighting the relationships between variables on the one hand, the distribution of individuals taking into account all of their physicochemical characteristics[14] and to determine the possible vectors of pollution. PCA is a technique for representing data under certain algebraic and geometric criteria, its objective is to extract most of the information contained in the data tables and to provide a graphical representation that is easy to interpret given the data correlations. The statistical processing of the data is carried out by the STATISTICA version 7 software.

RESULTS AND DISCUSSION

Results

Temp, pH, Cond, STD, Sal, Ca^{2+} and Cl^- in groundwater

The results of Table I show that the temperature of the groundwater is between 26.85 ° C and 28.1 ° C with an average value of 27.3 ° C. In-situ measurements show that the pH of the water studied varies from 3.95 (P_2) to 6.11 (P_1). There is a relatively high mineralization of drinking water in stations P_2 (580 µS.Cm^{-1}), P_3 (922 µS.Cm^{-1}) and P_5 (927 µS.Cm^{-1}). On the other hand, the waters of springs and boreholes, respectively S_7, S_8 and F_9, F_{10}, are weakly mineralized. Indeed, their conductivities do not exceed 100 µS.Cm^{-1}. The results obtained for total dissolved solids and salinity are in agreement with those obtained for these two parameters. Indeed, during our study, we noted the highest values in stations P_2 (STD = 289 ppm, Sal = 1.1), P_3 (STD = 461 ppm, Sal = 1.8) and P_5 (STD = 464 ppm, Sal = 1.8)

Table: 1 Values of physical parameters in groundwater.

Samples	T°(C)	pH	Cond (µS.cm^{-1})	Sal (‰)	STD (ppm)	Ca^{2+} (mg/L)	Cl^- (µg/L)
P_1	27.5	6.11	355	0.7	177	6.62	56.80
P_2	28.1	3.95	580	1.1	289	5.14	127.80
P_3	27.1	5.66	922	1.8	461	10.26	173.95
P_4	27.45	5.4	144.5	0.3	72,7	4.70	31.95
P_5	26.9	5.23	927	1.8	464	11.17	166.85
P_6	27.1	4.95	209.4	0.4	1051	5.3	46.15
S_7	28.1	5.19	62.8	0.1	31.4	3.91	28.40
S_8	27.3	5.22	73.6	0.1	36.8	4.51	21.30
F_9	26.85	5.89	98.9	0.2	49.3	4.98	28.40
F_{10}	27.11	5.6	97.3	0.2	50.7	10.36	42.60

MTE in groundwater

The results of Table 2 show that MTEs are present in the groundwater (drinking water) of the city of Daloa. From these results, relatively low levels of arsenic, cadmium, chromium, iron, mercury, nickel and lead are noted. Indeed, the concentrations determined in all the drinking waters studied do not exceed 20 µg / L. However, the maximum levels of mercury, nickel and lead were obtained in the spring waters of the Labia district (S_7). While those of arsenic, cadmium and chromium were determined respectively in the districts of Abattoir (P_6), Tazibouo (F_9), Labia (water from peasant wells P_3).

In addition, our results show that the iron and zinc contents are the most important during our study. In fact, they are all greater than 100 µg/L with maximum concentrations obtained in the drinking water of the Abattoir (P_6) and Gbokora (S_8) districts.

In addition, after filtration, we have more or less significant reductions in the concentrations of MTE in drinking water (Table 2). In fact, for arsenic, we have relatively low abatement rates for all the groundwater studied, with a greater reduction for water from peasant wells in the Dioulabougou district (P_5).

For cadmium, filtration leads to very significant reductions in concentrations for all water from peasant wells. Also, those in the Abattoir district have a 95.12 % decrease in cadmium levels. On the other hand, for spring water and boreholes, the reduction rates are very low or even zero.

The results obtained for chromium after filtration show very high reduction rates for all the drinking waters studied. Indeed, these rates are greater than 50% with a maximum reduction rate for the water from peasant wells in the Labia P_5 district (% R = 91.01 %). Also, for zinc, we have reduction rates greater than 50 % in the majority of the drinking waters studied except those of the Kenedy (P_1) and Orly (P_2) districts where we respectively 42.44 and 17.44 % (Table 2).

For iron, nickel and lead, filtration leads to a reduction in the levels of these MTEs which do not exceed 50 % for the majority of groundwater studied except for the water from peasant wells in the Orly and Labia districts where we have a rate of reduction of 58.67 and 54.19 % respectively for iron and lead (Table 2).

The results obtained for mercury after filtration give us very disparate abatement rates. However, there is a very little or no reduction in mercury content after filtration for the spring waters of the Gbokora district (S_8). On the other hand, for the spring waters of the Labia district (S_8), the reduction in mercury concentrations is very significant after filtration (86, 36 %).

Table: 2 MTE content and reduction rate after filtration in groundwater

	Arsenic		Cadmium		Chromium		iron		Mercury		Nickel		lead	
	C_{As} (µg/L)	%R	C_{Cd} (µg/L)	%R	C_{Cr} (µg/L)	%R	C_{Fe} (mg/L)	%R	C_{Hg} (µg/L)	%R	C_{Ni} (µg/L)	%R	C_{Pb} (µg/L)	%R
	9.29	13.25	1.52	50.36	5.22	55.34	1.59	58.67	0.38	6.68	16.07	47.20	1.28	25.95
P_2	9.93	16.84	1.51	44.02	6.89	62.74	1.46	50.99	0.546	7.69	17.64	54.19	1.21	22.43
	9.85	7.46	1.19	63.50	8.50	91.01	1.33	41.75	0.89	76.19	17.34	40.97	1.32	32.09
	8.98	10.96	1.32	81.69	8.20	50.59	1.48	41.53	0.34	75	16.34	39.39	1.25	22.64
gou P_5	10.06	28.24	1.19	79.46	5.14	56.69	1.34	37.60	0.84	5	15.69	25.06	1.24	5.34
6	10.11	11.93	1.64	95.12	8.33	52.40	1.70	46.42	1.01	0.83	15.37	40.08	1.24	28.79
	9.09	1.78	1.51	0.1	5.36	69.81	1.10	20.09	0.92	86.36	18.48	49.02	1.35	29.95
S_8	8.26	9.52	1.64	0.1	6.63	84.23	1.31	35.14	0.87	0.1	18.34	36.91	1.32	31.31
ouo F_9	9.97	9.24	1.65	0.1	5.18	87.98	1.46	27.79	0.68	4.85	17.71	49.53	1.3	22.02
ance F_{10}	9.54	10.21	1.51	0.1	7.2	52.21	1.57	37.94	0.42	20	16.46	35.27	1.23	35.34

Study of the correlation between the different parameters studied

This matrix highlights important associations between the parameters (Table 3). Indeed, we have correlations between the physicochemical parameters measured in situ, in particular Temp-pH (0.56) (average correlation) ; Sal-Cond (0.99) and Cond-TDS (0.9) ; TDS-Sal (0.9) which have strong correlations respectively.

In addition, we noted associations between parameters measured in situ and those determined in the laboratory.

- Strong correlations between Ca^{2+} -Sal (0.74), Ca^{2+} - Cond (0.74), Ca^{2+} -TDS (0.74), Cl^- Cond (0.99), Cl^- Sal (0.90) and Cl^- -TDS (0.99) ;
- Average correlations between As-Sal (0.55), As-Cond (0.54), As-TDS (0.54), Cl^- - As (0.56), Cl^- - Ca^{2+} (0.68), Cl^-- Zn (0.57) and Pb-Ni (0.65), Cd-Zn (0.50)

Table: 3 Correlation matrix between the different parameters studied

Temp	**pH**	**Cond**	**Sal**	**TDS**	**As**	**Ca^{2+}**	**Cd**	**Cr**	**Fe^{2+}**	**Hg**	**Ni**	**Pb**	**Zn**
1	0.56	0.01	-0.01	0.02	-0.20	-0.37	-0.01	-0.21	-0.38	-0.12	0.33	-0.02	-0.07
	1	-0.16	-0.14	-0.16	-0.15	0.44	-0.07	-0.19	0.16	-0.32	-0.20	0.25	-0.36
		1	**0.99**	**0.9**	**0.54**	**0.74**	-0.79	0.10	-0.11	0.17	-0.27	-0.23	-0.57
			1	**0.9**	**0.55**	**0.74**	-0.79	0.09	-0.09	0.15	-0.29	-0.24	-0.57
				1	**0.54**	**0.74**	-0.79	0.10	-0.10	0.17	-0.28	-0.24	-0.57
					1	0.43	-0.22	0.04	0.36	0.15	-0.46	-0.47	-0.40
						1	-0.66	0.07	0.08	-0.02	-0.44	-0.27	-0.65
							1	-0.18	0.29	0.04	0.27	0.08	**0.48**
								1	0.34	0.04	-0.21	-0.25	0.02
									1	-0.40	-0.72	-0.68	0.16
										1	0.20	0.44	0.08
											1	**0.65**	-0.06
												1	-0.010
													1

Principal Component Analysis

The PCR performed resulted in a circle of correlation. The factor extraction was performed by the principal components method. Three factors whose eigenvalues are greater than 1 were retained according to the criterion of Kaiser[15]. They correspond to 65.93 % of the total variance. Factor F1 is the most important with an expressed variance of 30.80 %. The factor F_2 has a variance of 18.94 % and F_3 of 16.18 %.

Table 4 gives us the eigenvalues and percentages of variance of the various factors.

Table: 4 Eigenvalues and percentage of cumulative expressed variance.

	Own values	% total variance Expressed	Cumulation of eigenvalues	Cumulative %
F_1	3.08	30.81	3.08	30.81
F_2	1.89	18.94	4.97	49.75
F_3	1.62	16.8	6.59	65.93

In Table 5, we noted the coordinates of the various physicochemical parameters studied as a function of each factor (main component) F_1, F_2 and F_3.

Table: 5 Coordinates of the different variables.

	F_1	F_2	F_3
As	-0.61	-0.31	0.40
Cd	0.16	**0.62**	-0.50
Cr	-0.37	0.16	0.26
Fe	-0.86	0.26	-0.31
Hg	0.35	-0.16	0.19
Ni	**0.88**	0.12	0.11
Pb	**0.83**	-0.36	-0.26
Zn	0.02	**0.68**	-0.45
pH	0.003	-0.65	-0.71
Temp	**0.50**	**0.51**	0.47
Cl^-	0.40	**0.45**	-0.70
Ca^{2+}	0.43	**0.71**	-0.23
Sal	0.27	**0.54**	-0.59
Cond	0.25	**0.53**	-0.60
STD	0.25	**0.53**	-0.60

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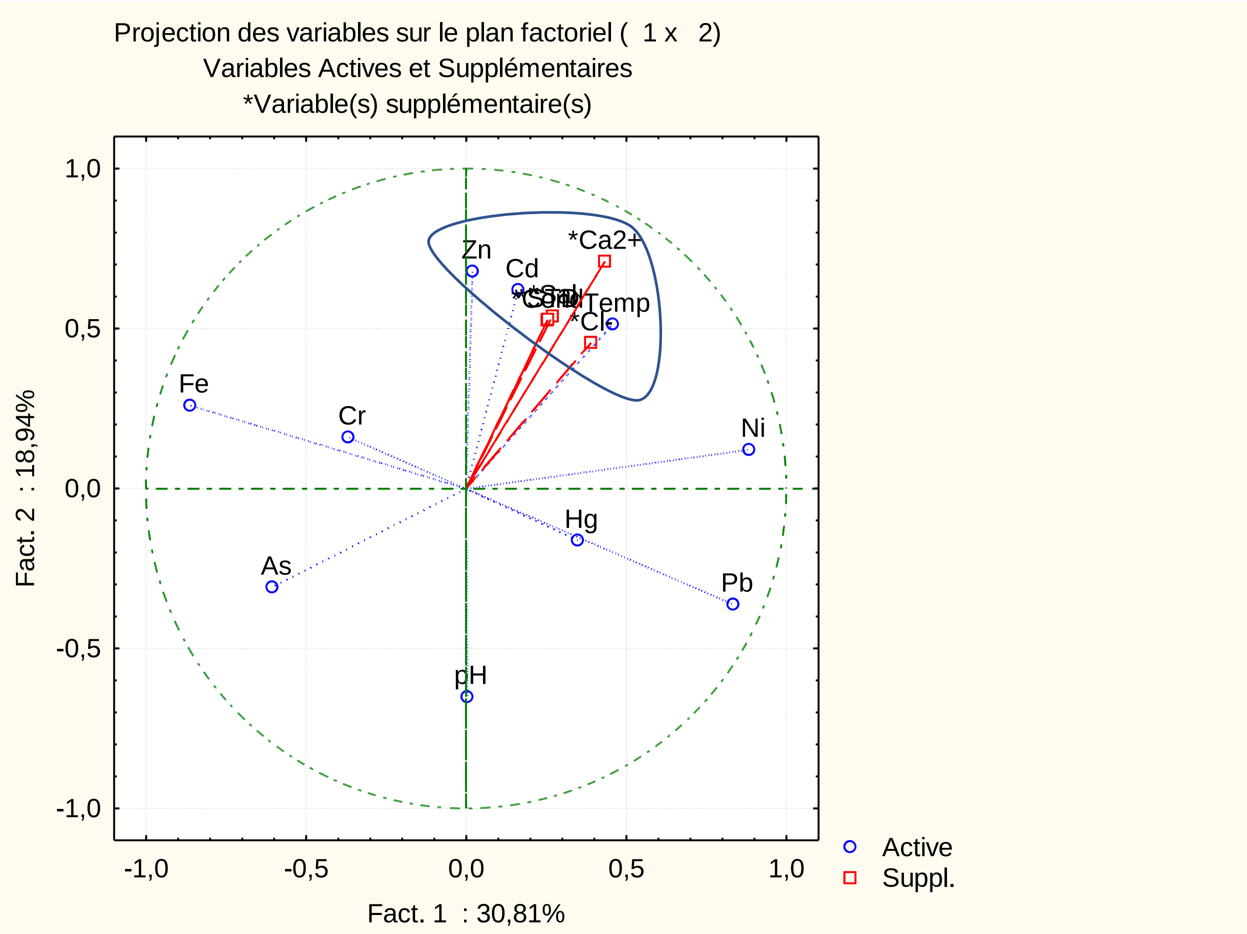
Factor F_1 is in association with nickel, lead and temperature as for factor F_2, it is correlated with STD, Cond, Sal, Ca^{2+}, Cl^-, Temperature, Zn and Cd.

The following figure shows the parameters which have an important correlation between them.



Graph.2: Correlation circle of the parameters studied in the factorial plane (F_1 - F_2).

Graph 2 shows us that the projection of the variables in the factorial plane (F_1 - F_2) reveals a single group of parameters which contain in particular Ca^{2+}, Cl^-, Zn, Cond, Temp, STD, Cd, Sal. These parameters are therefore decisive and have a very good correlation between them.

Discussion

The groundwater temperature is 27.5 ° C. Compared to the WHO guideline, Daloa's water appears warm. As this region is located in a tropical country, the average ambient temperature very rarely drops below 25 ° C. This is why, according to Rodier[13], 30 ° C should be considered an average groundwater temperature in West Africa due to climatic conditions.

The water from wells, boreholes and spring from the city of Daloa has an acidic character with pH between 4 and 6. This result is in agreement with those obtained by Ohou[16] and Gone[17], who worked on the well water of Buyo and Agboville, respectively. Moreover, according to Ahoussi[18], the acidity of water is one of the key characteristics of groundwater in Côte d'Ivoire. This acidity is linked to the nature of the soil. Indeed, the waters subjected to our study have an average pH = 5,4 ; this result is similar to that of Oga[19] in Tiassalé. In fact, in a humid tropical zone, this acidity comes mainly from the decomposition of plant organic matter, with the production of CO_2 in the first layers of the soil.

The waters studied have dissolved elements and the results obtained by stations P_3 and P_5 show that they have a large amount of fine particles. These elements in suspension could consist of mineral salts. The measured conductivity values allow them to be confirmed. In fact, we note that the underground water stations in the Labia (P_3) and Dioulabougou (P_5) districts have the highest conductivity values. Also, the values obtained for total dissolved solids and salinity are the most important in these same stations. In addition, during our study, the drinking waters of the Labia (P_3) and Diouloubougou (P_5) districts all have relatively high concentrations of chloride (Cl^-) and calcium (Ca^{2+}) ions compared to the other stations covered by our study. study. These results obtained are due not only to the geochemical characteristics specific to these stations, to the poor maintenance of peasant wells and to the acidity of the water which would cause the minerals to dissolve.

In addition, the results of our study show that MTEs are present in the drinking water studied. However, only the arsenic, cadmium and mercury contents respectively of the groundwater of the Dioulabougou P_5 and Abattoir P_6 districts and of the groundwater of all the stations are higher than the WHO drinking standards of 10µg / L and 1µg / L. Also, the concentrations obtained in groundwater are much lower than those obtained by Yapi[20] during the evaluation of the chemical quality of drinking water in the sub-prefecture of Hiré. Miramond[21] state that the presence of arsenic and mercury may be due to the dissolution of sulphurous minerals such as arsenopyrite generally present in soils and to the geochemical characteristics specific to the different stations. These geochemical characteristics would be responsible for the pollution of natural origin of the groundwater of the city of Daloa. To this pollution of natural origin must be added the poor maintenance of traditional wells, in particular the P_1 (kennedy) and P_5 (Dioulabougou) stations. In fact, the water flowing on the inner wall of these wells causes the minerals present there to dissolve[21].

Moreover, on the basis of the WHO drinking water standards, it could be concluded that only the groundwater in the Dioulabougou and Abattoir districts is unfit for human consumption.

During the statistical processing of the data, the correlations observed between the physicochemical parameters measured in situ, in particular between Cond-TDS (0.9), suggest that certain elements that make up the total dissolved solids strongly influence the conductivity of the groundwater studied. In addition, the associations between chloride ions and total dissolved solids (Cl^-- TDS (0.99)) and conductivity (Cl^- - Cond (0.99)) allow us to conclude that total dissolved solids would be composed of inorganic minerals composed in particular of chloride ions Cl^- which would intervene in the mineralization of water.

In addition, the correlations observed between chloride ions and calcium, cadmium and zinc suggest the presence of inorganic minerals such as calcium chloride $CaCl_2$, cadmium $CdCl_2$ and zinc $ZnCl_2$ in the drinking waters studied. This results in a high rate of reduction of cadmium and zinc filtration, especially in stations P_4, P_5 and P_6. These results were confirmed by factor analysis. Indeed, from this statistical analysis the factor F_1 reflects a pollution of natural origin of the groundwater in the city of Daloa. The mineralization of water due to the presence of inorganic salts is reflected by the factor F_2. In addition, the projections of the variables in the factorial plane (F_1 - F_2) lead to a single group of parameters which contain these same elements including STD, Cond, Sal, Ca^{2+}, Cl^-, Temp, Zn and Cd.

It should be noted that the presence of inorganic zinc and cadmium salts does not exclude the presence of bioavailable free species (Cd^{2+}, Zn^{2+})[22] in the drinking waters studied. Indeed, the results obtained after filtration, in particular in spring water (S_7, S_8) and boreholes (F_9, F_{10}) where we have very low or even zero reduction rates of cadmium.
In addition, from this factorial plane, iron, chromium, arsenic, lead, mercury and nickel are not part of this group, which suggests that these MTEs could exist in other forms, notably hydroxides and colloidals in groundwater.

The associations between arsenic, conductivity, chloride ions and TDS (Table III) suggest the presence of arsenites (+III) and arsines (-III) in the groundwater of the city of Daloa, in particular a fraction of Arsenic chloride $AsCl_3$[23]. It is necessary to add a larger fraction of free arsenic (As (s)). In fact, the low rates of reduction in arsenic content after filtration reflect this state.

Chromium III would be present in the groundwater of the city of Daloa and largely under fine particles. In fact, after filtration, very significant reductions in chromium contents ranging from 50 to 91% were observed. The physicochemical parameters in particular the acidic pH of the studied waters suggests a result. It is a very stable compound which forms hydroxyl complexes in natural waters, in particular $Cr(OH)^{2+}$, $Cr(OH)_2^{+}$, $Cr(OH)_3$ and $Cr(OH)_4^{-}$[24]. Trivalent chromium is less mobile and hydroxylated species can precipitate at pH between 4 and 5, total precipitation can occur at pH 5.5[24].

As for mercury, the low reduction rates in certain stations, in particular P_1, P_2, P_5, P_6, P_8, P_9, suggests the presence of the free bioavailable species, the mercuric ion Hg^{2+} since the mercury ion Hg^{+} is unstable due to disproportionation.

In aqueous media, nickel only exists at the divalent oxidation state Ni (II)[24]. Due to its basic nature, hydrolysis of nickel is poor under acidic or near neutral conditions. Consequently, the free nickel species, in particular Ni^{2+}, would be present in the groundwater studied[23]. However, the precipitated forms of this compound could exist in the drinking waters studied, in particular in the forms of hydroxides ($Ni(OH)_2$) and carbonates ($NiCO_3$). The reduction rates of the nickel concentration after filtration allow us to make this observation.

The aqueous chemistry of lead is mainly controlled by carbonate and hydrolyzed species, in particular $PbCO_3$, $Pb(OH)^{+}$, and free Pb^{2+} species[24]. Our studies have shown that these species could exist in the drinking waters of the city of Daloa. Indeed, the results obtained after filtration show that at least 50 % of this MTE would be in the form of bioavailable free species.

CONCLUSION

At the end of our study, it emerges that the metallic trace elements studied are present in the drinking water of the city of Daloa. From the results of our study, we note that the arsenic contents in the drinking water of the Dioulabougou P_5 and Abattoire P_6 districts are higher than the WHO standards for drinking water. These results then reflect the evidence of the poor quality of this drinking water and the risks it could cause.

In addition, statistical techniques show the presence of inorganic salts of calcium $CaCl_2$, zinc ($ZnCl_2$), cadmium ($CdCl_2$) and arsenic ($AsCl_3$) in the drinking waters of the city of Daloa.

Moreover, after filtration, we have more or significant reductions in concentrations for the various trace elements studied. Also, it reveals the presence of MTE in the dissolved phase. Hence the presence of bioavailable forms of arsenic As (s), cadmium (Cd^{2+}) zinc (Zn^{2+}), nickel (Ni^{2+}) and lead (Pb^{2+})

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