

# Use Hydrochemistry and Environmental Isotopes for the Assessment Mineralization of Groundwater in Miopliocene Aquifers in Douala 3 (Cameroon)

Tatou Rel Dechangue<sup>1,\*</sup>, Kamgang Kabeyene Veronique<sup>2</sup>

<sup>1</sup>Department of Earth Sciences, University of Maroua, Cameroon

<sup>2</sup>Department of Earth Sciences, University of Dschang, Cameroon

\*Corresponding author: [rtatou@yahoo.fr](mailto:rtatou@yahoo.fr)

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**Abstract** Linked hydrochemical and isotopic studies were carried out to identify and explain the processes that control the mineralization in miopliocene aquifers in Douala basin. According to the results of the chemical analysis, the water is acidic and poor mineralized. The investigation highlights that groundwater mineralization is mainly influenced by ion exchange process, carbonate dissolution and silicate weathering. Based on isotopic signatures, shallow aquifer groundwater samples were classified into waters with depleted  $\delta^{18}\text{O}$  contents, highlighting the result of a rapid infiltration of local meteoric waters which has undergone a low evaporation process. Deep aquifer and some points of shallow aquifer are more enriched in  $^{18}\text{O}$  than the local rains. This reflects a certain level of evaporation which the rainwater would have undergone before reaching the aquifer or the leaching of heavy isotopes accumulated in the unsaturated zone during the dry season. The relatively recent age of the waters that recharge the aquifer explains the weak action of the water-rock interaction hence the weak mineralization of the waters overall.

**Keywords:** hydrochemistry, isotopes, mineralization, groundwater, miopliocene aquifers, Douala

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## 1. Introduction

The drinking water supply of populations in many cities in coastal areas is dependent on groundwater resources [1]. This is the case in the city of Douala where the water used is captured in the various aquifers encountered. On the outskirts of this area, mainly in the district of Douala 3, one of the aquifers captured for water needs is the Miopliocene aquifers. The final chemistry is the result of the inputs by the different environments crossed, according to the physico-chemical conditions prevailing there and also of the residence time of the waters [2,3,4]. Coupled analysis of hydrochemical and isotopic groundwater data is one of the essential elements for the study of hydrogeological potentialities and the management of water resources. Chemical and isotopic studies may therefore help to better understand the functioning of the aquifer system. Several such studies have already been conducted to date, but they are still insufficient to understand the miopliocene aquifers in the Douala Basin. The objective of this study is to use the

physicochemical and isotopic characteristics of the groundwater of the miopliocene aquifers to understand the origin of ions in water as well as the groundwater recharge form.

## 2. Presentation of the Study Area

Study area is located in the Douala sedimentary basin. It is situated lies between North latitudes  $4^{\circ}00'29''$  and  $4^{\circ}02'08''$ ; East longitudes  $9^{\circ}46'11''$  and  $9^{\circ}48'06''$  with a total drainage of about five  $\text{km}^2$  (Figure 1). The study area is located in Douala city subject to a sub-equatorial climate influenced by altitude, with an average annual rainfall of 3664.86 mm, and average annual temperatures of  $24.9^{\circ}\text{C}$ . The main watercourse (Kambo) and some tributaries have little water supply to the system. The soils encountered are ferrallitic soils that derive from sedimentary rocks [5]. Aquifer formations in the study area are the miopliocene sediments [6,7]. These are coarse sands at the base and fine at the top, alternating with pockets of coloured clays, sometimes interstratified with basalts (Figure 2, [41]).

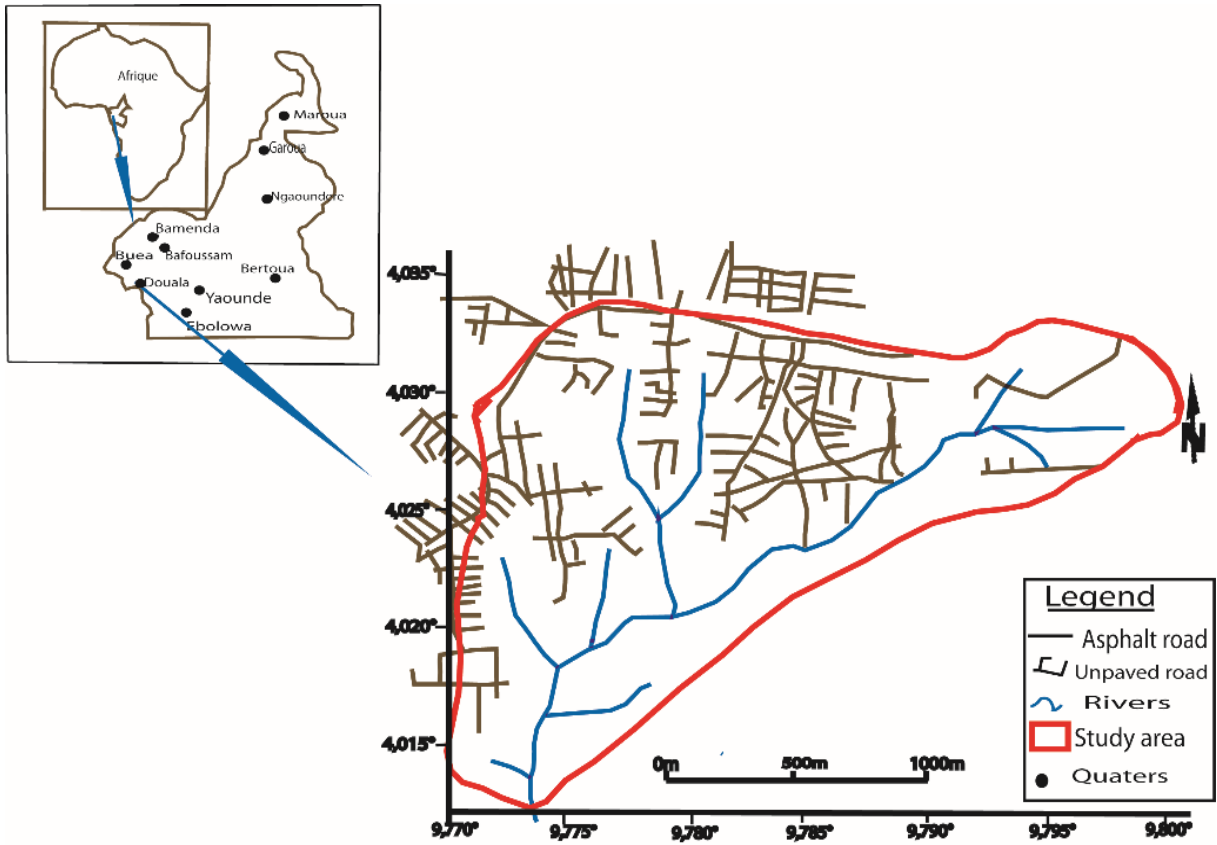


Figure 1. Location of study area

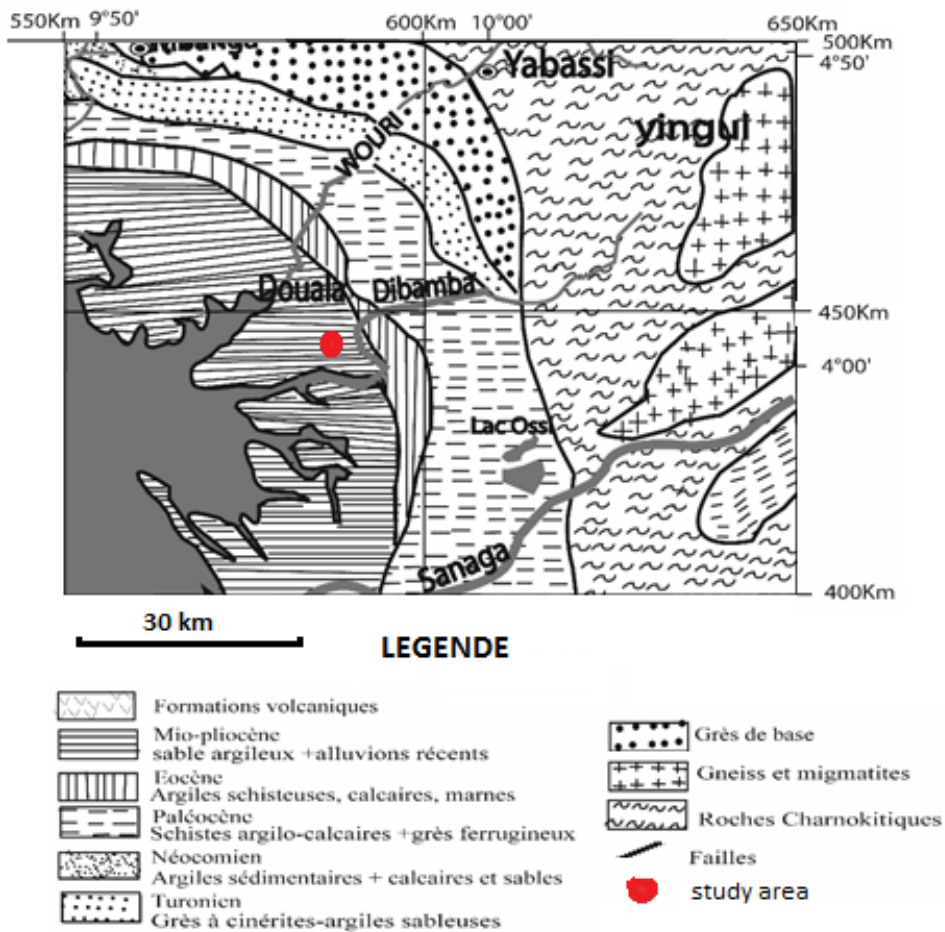


Figure 2. Location of the study area in the geological map in the Douala basin [8]

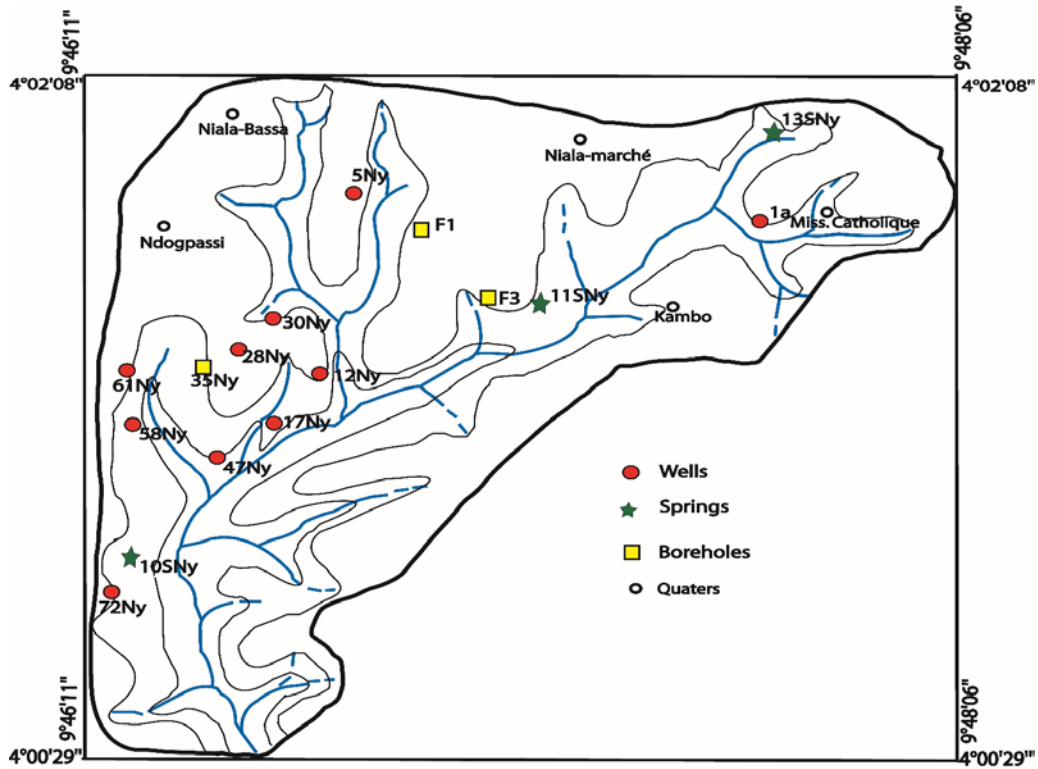


Figure 3. Sampling points

### 3. Materials and Methods

A field campaign was conducted in the study area for a sampling network (Figure 3): 13 samples in shallow aquifer (10 dug-wells and 3 springs) and 3 in deep aquifer (boreholes). Water samples for chemical analysis were taken using the previously sterilized 500ml PVC bottles and kept in coolers at 4°C before being sent to the laboratory. Physico-chemical parameters such as pH, electrical conductivity, and TDS are measured through the WTW multi-parameter. Another set of samples was collected for chemical and isotopic analyses ( $\delta^2\text{H}$ ,  $\delta^{18}\text{O}$ ). Sixteen groundwater points were sampled in August 2013 corresponding to the dry season. Major ions analyses were carried out in the soil,

plant, fertilizer and water analysis laboratory.

### 4. Results and Discussions

#### 4.1. Hydrochemical Composition of Groundwater

Physical and chemical data are reported as raw data in Table 1 from the Institute for Agronomic Research for Development (IRAD) in Yaoundé. Cations (Ca, K, Na, Mg) were analyzed using the flame atomic absorption spectrophotometer, chlorides and bicarbonates by titrimetry, nitrate and sulfates by colorimetry.

Table 1. Physical and chemical data of the study area

code of samples		pH	C.E (µS/Cm)	TDS (mg/l)	Ca <sup>2+</sup> (mg/l)	Mg <sup>2+</sup> (mg/l)	Na <sup>+</sup> (mg/l)	K <sup>+</sup> (mg/l)	NO <sub>3</sub> <sup>-</sup> (mg/l)	Cl <sup>-</sup> (mg/l)	SO <sub>4</sub> <sup>2-</sup> (mg/l)	HCO <sub>3</sub> <sup>-</sup> (mg/l)	SiO <sub>2</sub> (mg/l)
5Ny	S	6.12	466	172	14.07	10.65	12.99	16.16	2.65	13.85	5.13	96.5	4.2
12Ny	S	5.48	218	86	4.79	5.83	15.09	23.03	2.06	12.43	5.83	15.25	1.16
17Ny	S	6.21	143.1	158	4.61	18.42	2.81	3.54	0.31	0.46	0.74	122	3.29
28Ny	S	6.43	263	171	23.85	7.79	3.39	2.74	0.83	0	2.73	124.18	3.21
30Ny	S	4.5	236	325	8.93	11.29	22.11	13.4	3.92	2.3	0.7	256.2	3.37
35Ny	D	6.25	396	82	13.07	8.38	9.16	1.63	12.09	9.6	9.92	14.05	2.49
47Ny	S	5.15	44.3	23	0.2	1.66	2.43	0.6	1.13	0.67	0.74	12.2	2.3
58ND	S	4.08	305	85	3.16	7.57	15.38	0.82	13.36	25.85	5.74	8.1	2.17
61ND	S	4.48	215	98	4.28	6.57	13.19	3.76	18.82	10.84	9.69	21.58	5.3
72ND	S	5.91	197.2	166	7.88	6.59	11.99	6.29	2.93	3.49	1.44	111.3	8.9
1a	S	4.42	198.4	42	1.84	5.67	3.27	0.67	4.51	17.75	0.41	2.1	3
10SNy	S	4.95	46.5	19	1.26	1.59	1.89	0.64	1.24	1.8	1.25	6.1	2
11SNy	S	4.33	121.9	60	6.58	3.42	6.28	2.07	20.4	3.3	7.8	4.23	2.98
13SNy	S	4.7	89.6	69	1.39	4.22	6.79	11.34	25.78	4.75	8.99	0.97	2.94
F1	D	4.9	29	17	0.31	0.43	2.09	0.024	1.96	0.11	4.98	0.03	4.62
F4	D	5.66	92.8	68	1.39	3.64	6.4	2.89	1.65	0	4.83	39.65	4.58
Min		4.08	29	17	0.2	0.43	1.89	0.024	0.31	0	0.41	0.03	1.16
Max		6.43	466	325	23.85	18.42	22.11	23.03	25.78	25.85	9.92	256.2	8.9
Mean		5.22	191.36	102.56	6.11	6.48	8.45	5.60	7.10	6.7	4.4	52.15	3.48

S=shallow aquifer D= deep aquifer.

Silica was analyzed by ultraviolet atomic absorption spectrophotometer at the Waste Water Research Unit (WWRU) laboratory of the University of Yaoundé I. Isotopic analyses were performed at the Laboratory of Water Geochemistry (LAGE) of MINRESI (Nkolbisson-Yaounde) using Picarro Cavity Ring-Down Spectrometer, following the procedures described by [9] and [10].

In the groundwater samples, pH values ranges from 4.08 to 6.43 (mean= 5.22) then waters are acidic. Lower values of pH, characterizing shallow groundwater samples, are related to the oxidation of organic matter [11]. Total dissolved solids (TDS) contents of almost all the samples are relatively low (mean= 102.56 mg/l), represented fresh water. The electrical conductivity values range from 29 to 466.0  $\mu\text{s}/\text{cm}$  (mean= 191.36). Concentrations of Mg, K, Cl,  $\text{SO}_4$ , and  $\text{NO}_3$  are relatively low. Calcium is the most abundant cation in groundwater, it varies from 0.2 to 23.85 mg/l with an average of 6.1 mg/l. Sodium varies in concentration ranging from 1.89 to 22.11 mg/l, with average of 8.45 mg/l. Magnesium concentration ranges from 0.43 to 18.42 mg/l with an average of 6.48 mg/l. Potassium ranges from 0.024 to 23.03 mg/l with an average of 5.6 mg/l. The cations in the groundwater are in the following order:  $\text{Ca} > \text{Na} > \text{K} > \text{Mg}$

The chloride in groundwater ranges from 0.0 to 25.85 mg/l, with average value of 6.7 mg/l. Bicarbonates ranges from 256.2 to 0.03 mg/l ( mean:52.15 ). Nitrate ion in the study area ranges from 0.31 to 25.78 mg/l with an average of 7.1 mg/l. Sulphate ion ranges from 0.41 to 9.92 mg/l with an average of 4.43 mg/l. Silica in the groundwater ranges from 1.16 to 8.9 mg/l with an average of 3.48 mg/l. The anions in the groundwater are in the following order:  $\text{HCO}_3 > \text{Cl} > \text{NO}_3 > \text{SO}_4$ .

### 4.2. Water Types

According to the Piper chart (Figure 4) and the Chadah chart (Figure 5), an average of the data from two sampling campaigns distinguishes three major facies: Cl-Ca-Mg (5 samples),  $\text{HCO}_3$  - Ca (8 samples), Cl - Na (3 samples).

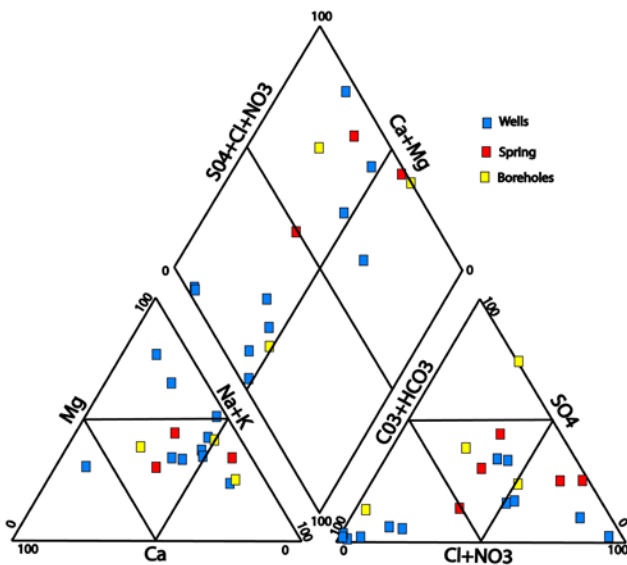


Figure 4. Piper diagram

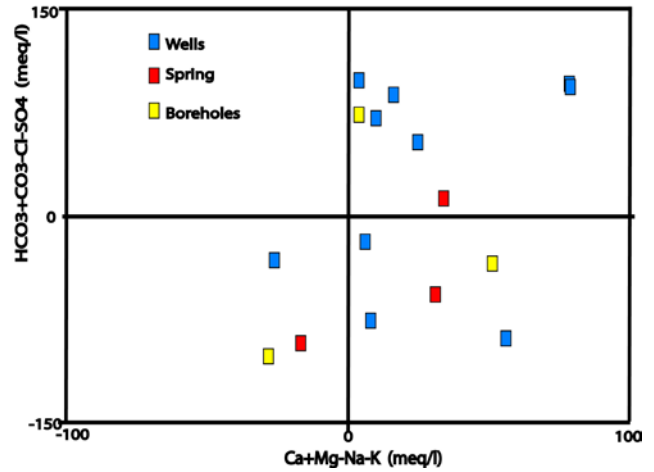


Figure 5. Chadah diagram

### 4.3. Mechanisms of Mineralization Acquisition

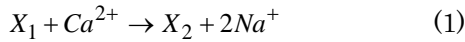
#### 4.3.1. Highlighting Basic Exchanges

Table 2. Base exchange index (BEI) and saturation indices (SI)

code	BEI	SI <sub>Calcite</sub>	SI <sub>Aragonite</sub>	SI <sub>Dolomite</sub>
5NY	-1.50	-2.11	-2.25	-3.99
12NY	-2.55	-3.99	-4.13	-7.55
17NY	-15.39	-2.40	-2.54	-3.85
28NY		-1.46	-1.60	-3.06
30NY	-19.11	-3.52	-3.67	-6.59
35NY	-0.63	-2.83	-2.97	-5.50
47NY	-5.41	-5.74	-5.89	-10.22
58ND	0.054	-5.85	-5.99	-10.97
61ND	-1.19	-4.89	-5.03	-9.24
72ND	-5.93	-2.49	-2.64	-4.72
1a	0.68	-6.3	-6.45	-11.77
10SNY	-0.94	-5.45	-5.59	-10.44
11SNY	-2.50	-5.54	-5.69	-11.02
13SN	-3.37	-6.49	-6.63	-12.14
F1	-28.49	-8.40	-8.55	-16.32
F4		-3.91	-4.05	-7.05

The Base exchange index is negative for all samples (Table 2). This indicates that, there were exchanges between the alkaline of the clays and the alkaline earths of the water. The increase in Na+K concentration by cation exchange is assessed by considering that water from precipitation or recharge was in ionic balance with Cl and Na+K [12]. In this case, enrichment of water in Na + K compared to Cl is due to an exchange of ions between (Ca + Mg) of water and (Na + K) of clays [13,14,15]. The points affected by the ion exchange are located below the seawater dilution line (Figure 6). In addition, the base exchange phenomenon was also verified through the diagram  $(\text{Ca}^{2+} + \text{Mg}^{2+}) - (\text{HCO}_3^- + \text{SO}_4^{2-})$  Vs  $\text{Na} + \text{K} - \text{Cl}$  (Figure 7). The operation of the parameter subtraction allows to get rid of origins mainly due to other dissolution reactions of carbonated and evaporitic minerals [16]. When ion exchange is a major process in the acquisition

of mineralization, the points must line up on a line [17,18]. However, in this study, all points are not aligned but, the decrease of Ca+Mg is accompanied by an increase of Na+K [19]. Montmorillonite (smectite) is one of the constituents of the miopliocene aquifers [20]. In this clay, cationic bond intensity evolves as follows:  $Na^+ < H^+ < K^+ < NH_4^+ < Mg^{2+} < Ca^{2+}$  [21]. In this sequence, the  $Na^+$  ion easily passes into the aqueous phase and  $Ca^{2+}$  ion is trapped in the mineral during the ion exchange process (equation 1). It is therefore likely that the base exchange is carried out between water and this clay.



$X_1$  =Montmorillonite- $Na_2$

$X_2$  =Montmorillonite-Ca

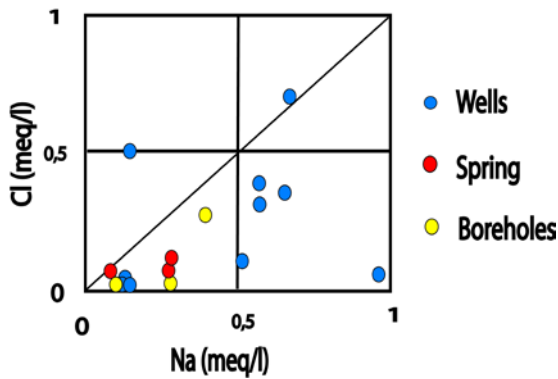


Figure 6. Plot Cl vs Na

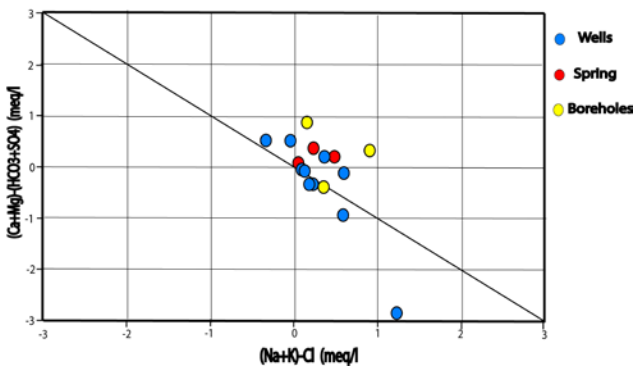


Figure 7. Plot (Na+K) –Cl versus (Ca+Mg) – (HCO<sub>3</sub> +SO<sub>4</sub>)

### 4.3.2. Dissolution of Carbonates

Saturation indices of all calculated minerals (calcite, aragonite, dolomite) are negative (Table 2). This suggests the subsaturation of the water in the Kambo watershed in all these elements. The negative correlations between calcium and sulphates demonstrate a non-existence of evaporitic minerals in the studied waters. A significant correlation is noted between  $Ca^{2+}$  and  $Mg^{2+}$  and between  $Ca^{2+}$  and  $HCO_3^-$  [22]. This indicates the presence of carbonate minerals (calcite and dolomite) in the aquifer. The Ca/Mg ratio (Figure 8) in the waters makes it possible to distinguish the points influenced by the alteration of silicates, by the dissolution of calcite and dolomite [23,24]. Points with a Ca/Mg ratio =1 are controlled by dolomite dissolution. Ratio between 1 and 2 indicates the dissolution of calcite. For this study, the majority of points are concerned with the dissolution of dolomite. This

dissolution is also explained through the diagram (Ca+Mg) versus (HCO<sub>3</sub>+SO<sub>4</sub>) (Figure 9) where most points lie above the line of slope [25]. Although calcite and dolomite were not reported in Douala weathering materials. They were nevertheless revealed at significant depths in the Douala basin [27].

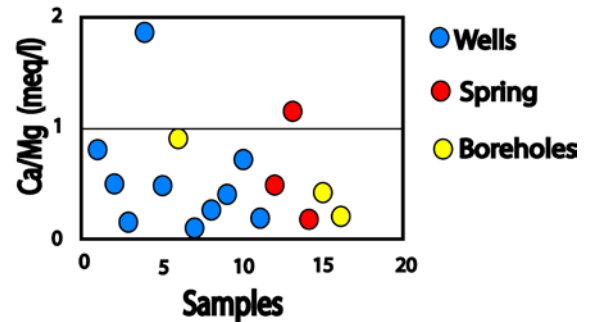


Figure 8. Plot Ca/Mg versus samples

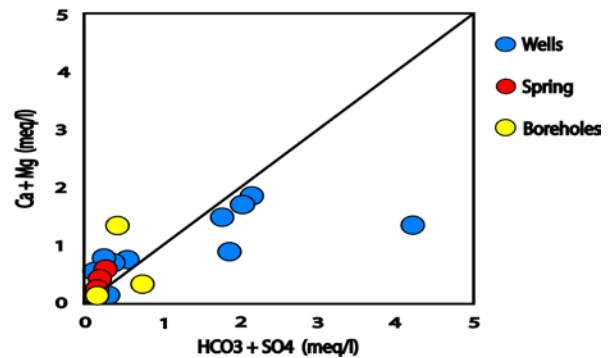


Figure 9. Ca + Mg versus HCO<sub>3</sub> + SO<sub>4</sub>

### 4.2.3. Silicate Weathering

Hydrolysis of silicates is highlighted through (Ca + Mg) versus  $TZ^+$  (Figure 10), Na+K versus  $TZ^+$  (Figure 11) [3,27]. However, Ca+Mg Versus HCO<sub>3</sub> diagram (Figure 12) shows that, most of the points are above the equilibrium line. This reflects a low contribution of silicate weathering in the acquisition of the ions [3,28]. Therefore, the major cations are derived from the alteration of plagioclases, pyroxenes, amphiboles and montmorillonite [29]. The sandy formations that constitute the miopliocene aquifers captured by the sampled wells contain a set of silicate minerals. The hydrolysis of silicates is facilitated by infiltration waters rich in dissolved CO<sub>2</sub>. In the presence of very aggressive acidic waters, silicate minerals will undergo hydrolysis and release cations [13,30,31]. Depending on the degree of rainfall and the base content, the products newly formed after destruction of the primary minerals could be gibbsite, kaolinite, montmorillonite or l'illite [32]. The presence of the minerals in the study area which have contributed to the setting up of those clays are derived from the calc-magnesium complex of Ntem [6]. These clays reflect the supergene alteration of rocks rich in quartz, feldspath and mica [33].

Silica contents are low in all samples (Table 1). However, there are points with values close to unity (1mg/l) suggesting that this mineral phase is present in the aquifer material. Silica in groundwater results mainly from the weathering of silicate minerals in the aquifer matrix. Indeed, analysis of sediments or soils in the Douala basin

show that they contain significant levels of quartz [34,35]. The low levels of silica found in the waters tested may be due to several reasons: the Mio-Pliocene aquifers are sedimentary and contain mainly clay minerals and sands [6,20]. These neofomed clays are very resistant to weathering. These waters with a relatively short residence time in the aquifer [36] did not have the necessary time for intense weathering of rocks.

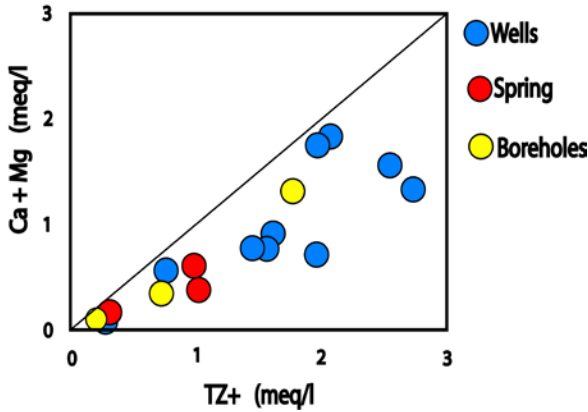


Figure 10. Plot Ca + Mg vs TZ<sup>+</sup>

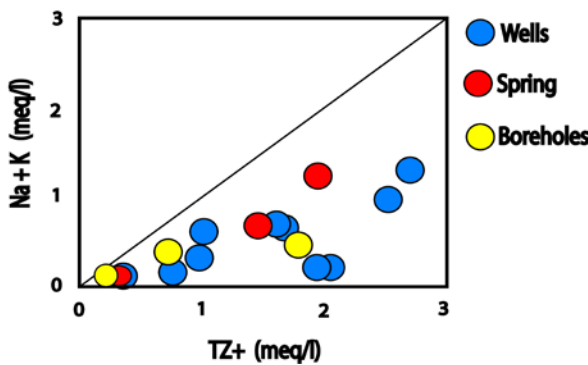


Figure 11. Na + K vs TZ<sup>+</sup>

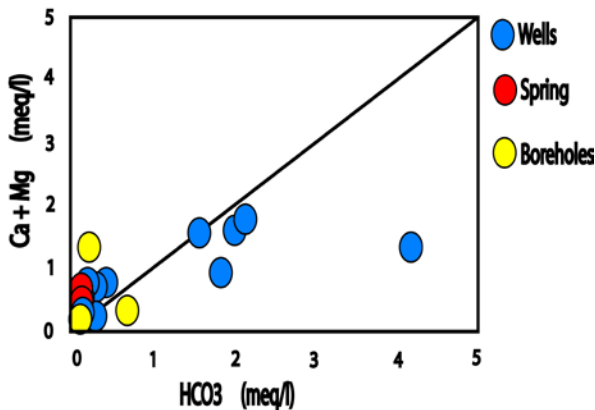


Figure 12. Ca + Mg vs HCO<sub>3</sub>

### 4.4. Stable Isotopes of Water

#### 4.4.1. Isotopes Contents

Global Statistics (Table 3) show levels oxygen18 (<sup>18</sup>O) ranging from -4.02‰ to -3.12‰ for an average of -3.55‰.

Deuterium (<sup>2</sup>H) contents range from -16.66‰ to -8.05‰ for an average of -13.53‰. On the diagram  $\delta^{18}O$  vs  $\delta^2H$  (Figure 13), all points are located above the world meteoric line (WML), these are characterized by

<sup>18</sup>O depletion compared to world meteoric waters. Eleven (11) points are located above the local meteoric line (LML) and two are below this line. The water below this line is enriched with oxygen 18.

#### 4.4.2. Groundwater Origin

The  $\delta^{18}O$  and  $\delta^2H$  values obtained (between -2.10‰ and -6.10‰; -11.90 ‰ and -26.19 ‰) are close to those obtained by [37] in deep aquifers in Douala. Precipitation being the main source of groundwater recharge in most continental hydrogeological systems [38], the isotopic comparison between precipitation and groundwater is used to determine groundwater recharge [39,40,41]. In non-evaporated waters above the local meteoric line (Figure 13), <sup>18</sup>O levels are very low. These are therefore the result of a rapid infiltration of local meteoric waters which have hardly undergone evaporation. This corroborates with the sandy nature of the shallow aquifer of the miopliocene, which is essentially made up of sand characterized by a high permeability [20]. The low slope correlation line (3.15) different from that of the local meteoric waters would be a consequence of the isotopic enrichment processes of the water that occur during its infiltration [4,42].

The evaporated water below the local meteoric line (Figure 13) is relatively more enriched in <sup>18</sup>O than the local rains. There are several reasons for the enrichment: (1) The rainfall should have evaporated to some level before reaching the aquifer; (2) Groundwater would not be recharged directly by current rains but by old water or a distant source; (3) After recharge, heavy isotopes accumulated in the unsaturated area may have leached during the dry season [43,44,45]. Comparing the mean of the isotopic oxygen-18 contents of the spring water (<sup>18</sup>O=-3.61) and the drill water (<sup>18</sup>O=-3.29 with the mean rainfall weighted rainfall (<sup>18</sup>O=-3.64) obtained by [46]. The results obtained in samples and rainwater are very close to each other, this reflects the existence of a communication or direct relationship between rainwater and ground water. This also suggests that, groundwater recharge is due to recent local precipitation which has undergone a low evaporation process [4].

Unlike the waters of the shallow aquifer of the Cameroonian Sahelian zone (plain of Diamare and Grand Yaere), which are highly vaporized [11,46], the unconfined aquifer water is characterized by relatively low evaporation. This suggests that the climatic conditions prevailing in the coastal zone would influence the isotopic fractionation of the waters. The low evaporation of groundwater would be the fact that the meteoric waters percolate rapidly to the water table without undergoing significant evaporation phenomena, due to, the sandy nature of the miopliocene aquifers.

For deep aquifer, however, the water crosses significant depths before the accumulation zone the similarity between the isotope contents in the waters of the shallow aquifer and deep aquifer may be justified by hydrogeological properties which are favorable to a rapid infiltration of the waters. The rapid percolation of reported rainwater shows that the efficiency of water-rock interactions and isotopic fractionation in these samples are very low in the different levels of the miopliocene aquifers [47].

Table 3.  $\delta^{18}\text{O}$  and  $\delta^2\text{H}$  values

code		depth(m)	$\delta^{18}\text{O}\text{‰}$	$\delta\text{D}\text{‰}$
58ND	S	10.08	-3.73	-14.21
12Ny	S	2.57	-3.88	-16.58
13SN	S		-3.91	-10.08
28NY	S	11.12	-3.17	-12.57
17Ny	S	16.49	-3.2	-8.05
61ND	S	3.51	-3.22	-13.82
1a	S	8.00	-3.38	-13.56
30Ny	S	7.13	-3.52	-14.41
47Ny	S	4.6	-4.02	-16.03
10SNy	S		-3.78	-13.46
11SNy	S		-3.83	-13.28
72ND	S	4.6	-3.51	-11.16
5NY	S	2.38	-3.75	-11.52
35NY	d	22	-3.34	-16.01
F4	d	31	-3.43	-16.66
F1	d	28	-3.12	-15.05
min			-4.02	-16.66
max			-3.12	-8.05
mean			-3.55	-13.53

S=shallow aquifer, D=deep aquifer.

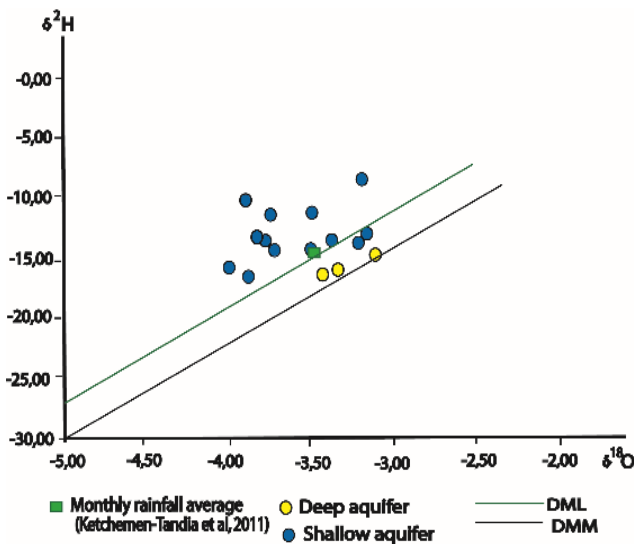


Figure 13. Plot  $\delta^{18}\text{O}$  Vs  $\delta^2\text{H}$

#### 4.4.3. Isotope-depth Relationship

The diagram Oxygen 18 Vs depth (Figure 14) shows an increase in  $^{18}\text{O}$  content with depth. This increase is clearly observed in the wells of the surface layer. This increase is clearly observed in the waters of the shallow aquifer. The waters of spring have the lowest oxygen levels 18. Waters are marked by depletion of this isotope. These waters, although directly charged by local rains, are exposed to evaporation due to their direct contact with the atmosphere. Progression in the depths is accompanied by enrichment in isotopes. Rapid infiltration of the water would have facilitated a low evaporation of the water. Borehole waters are also enriched with isotopes but there is no net limit between the isotopic contents of wells and boreholes. Isotopic resemblance between the two aquifers expresses homogeneity between waters of the miopliocene aquifers.

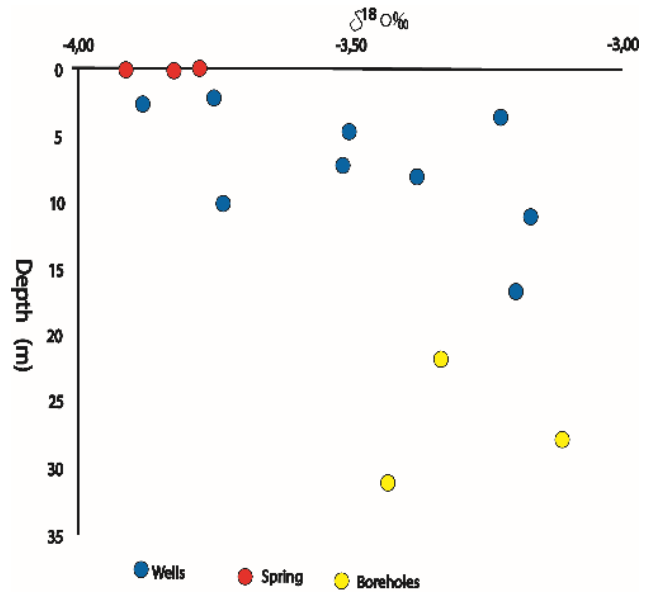


Figure 14. Plot Oxygen 18 Vs depth

## 5. Conclusion

Hydrochemistry data of the study area revealed that water is acidic and poor mineralized. The abundance of the major cations and anions in the groundwater are of the following order:  $\text{Ca} > \text{Na} > \text{K} > \text{Mg}$  and  $\text{HCO}_3 > \text{Cl} > \text{NO}_3 > \text{SO}_4$ . The groundwater showed paths of hydrochemical evolution, from  $\text{Ca}-\text{HCO}_3$  type via  $\text{Ca}-\text{Cl}$  type to  $\text{Na}-\text{Cl}$ . Mineralization into the groundwater is controlled by ion exchange, dissolution and leaching from weathering silicate. The groundwater is generally undersaturated with respect to major carbonate species namely calcite and dolomite. The  $\delta^2\text{H}$  versus  $\delta^{18}\text{O}$  diagram shows that the groundwater data are characterized by  $^{18}\text{O}$  depletion. The low level of isotopes revealed that groundwater recharge is due to recent local precipitation which has undergone a low evaporation process. The similarity between the water of shallow and deep aquifer reflects the existence of a communication or direct relationship between rainwater and ground water. The low evaporation of groundwater is due to the rapid percolation meteoric waters to the water table without undergoing significant evaporation phenomena. The diagram Oxygen 18 Vs depth shows an increase in  $^{18}\text{O}$  content with depth. The rapid percolation of reported rainwater shows that the efficiency of water-rock interactions and isotopic fractionation in the samples are very low in the different levels of the mio pliocene aquifers.

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