

Full Length Research Paper

Nitrate in drinking water: A major polluting component of groundwater in gulf region aquifers, south of Togo

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Hydrochemical investigations of groundwater have been carried out on objectives of evaluating groundwater quality. Groundwater quality contaminations have emerged in many geographical areas due to natural environmental processes and human intervention in the geosystems. Geochemical evaluation of nitrate contaminated groundwater in Gulf region was investigated. Results showed that most of the samples contained high concentrations of nitrate. More than 70% of samples in central area contained high concentration of nitrate. Furthermore, results show that nitrate has a negative relationship with calcium and sodium, whereas relationship with magnesium, potassium, alkalinity, sulphate, chloride and electrical conductivity (EC) are positive. Factor analysis was used to identify key parameters that described the groundwater quality. Two factors were extracted as follows: Factor 1 explained 60% of the total variance, reflected the importance of the intrusion of salt water into the groundwater and factor 2 explained 35% of the total variance, and reflected impact of human activities and acid-rain recharge of the aquifers in the area. The geochemical modeling reveals that the high nitrate groundwater behaves like two kinds of the mechanism of hydrogeochemical evolution, namely the intervention of human activities, and the evaporation effect of the shallow groundwater.

Key words: Groundwater, geochemical, factor analysis, nitrate, drinking water.

INTRODUCTION

Contamination of groundwater by nitrate (NO_3^-) from various sources continues to be a problem in much of the Togo. Nitrate contamination can be a severe problem in some Gulf region and vicinity aquifers where the drinking water standard of 10 mg/L as nitrogen (10 mg/L NO_3^{2-}N [WHO, 1996]) in supply wells is often exceeded. The sources and fate of nitrate can be complicated and are not well understood due to the mixing of nitrate from different sources and the variation in the occurrence and amount of denitrification within an aquifer. Nitrate is naturally present in groundwater at very low concentrations, and its source is human activities such as

domestic or industrial waste or agricultural. However, pollution of groundwater from increased utilization for human needs and increased industrial activity is a major concern (Mande et al., 2011a). Differences in concentrations of dissolved ions in groundwater are generally governed by lithology, groundwater flow, geochemical reactions, solubility of salts, and human activities (Bhatt and Salakani, 1996; Mande et al., 2011b). The quality of groundwater for human consumption is dictated by its quantitative and qualitative composition of suspended solids and dissolved minerals or organic compounds (Jain et al., 2005). Water quality can be compromised when toxic substances come into contact with the aquifer. This can make the water unsafe for human consumption. The study of groundwater chemistry aims to identify the chemical species in the water, and its quality for irrigation and drinking. Drinking water quality can also be monitored using the spatial variation of

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physicochemical parameters, and examine sources by correlating data with the geology and piezometry.

Methemoglobinemia is the primary adverse health effect associated with human exposure to nitrate or nitrite. To cause methemoglobinemia, nitrate must be converted to nitrite. Nitrite causes the oxidation of normal hemoglobin to methemoglobin which is unable to transport oxygen from lungs to tissues. Bacteria in the gastrointestinal system mediate the conversion of nitrate to nitrite.

Consequently, the risk of methemoglobinemia from ingestion of nitrate depends not only on the dose of nitrate but also on the number and type of enteric bacteria. Low levels of methemoglobin occur in individuals with typical values ranging from 0.5 to 2.0% (NRC, 1981). While methemoglobinemia can be clinically diagnosed at levels of 1%, methemoglobin levels up to 10% are generally not considered adverse. At levels above 10%, methemoglobinemia causes cyanosis, and at higher concentrations, asphyxia (WHO, 1996; EPA US, 1997).

Infants are generally recognized as the subpopulation most susceptible to nitrate induced methemoglobinemia (EPA US, 1990, 1997). Several factors make infants particularly sensitive to methemoglobinemia including: (1) Infants have a greater total fluid intake per unit body weight than adults; (2) The gastrointestinal system in infants normally has a high pH that allows bacterial proliferation and nitrate catalysis to nitrite; (3) Frequent occurrences of infant gastroenteritis may favor the development of methemoglobin when the upper gastrointestinal tract becomes colonized with bacteria; (4) Fetal hemoglobin is more readily oxidized than adult hemoglobin, and (5) Infants have half the methemoglobin reductase activity of older children and adults and are therefore less capable of metabolizing excess methemoglobin (Winton et al., 1971; NRC, 1981; Kross et al., 1992).

There are other individuals who may be predisposed to the development of nitrate-induced methemoglobinemia due to altered physiological states. These include pregnant women and possibly others with glucose-6-phosphate dehydrogenase deficiency, adults with reduced gastric acidity (for example, from diseases including achlorohydria or atrophic gastritis) and those with a lack of methemoglobin reductase (EPA US, 1997; NRC, 1981).

Since the mid 1950's, more than 2,000 cases of infantile methemoglobinemia have been reported worldwide. Most cases are associated with exposure concentrations greater than 20 mg nitrate nitrogen/L in drinking water that was used to prepare infant formula. Cases of infantile methemoglobinemia associated with nitrate exposure concentrations of 11 to 20 mg nitrate nitrogen/ L are usually associated with concomitant exposure to water contaminated with bacteria. Thorough reviews of the literature on the occurrence of methemoglobinemia in infants are provided by EPA US (1990), Fan et al. (1987),

(1987), and Fan and Steinberg (1996).

Concern has been raised about a possible increased risk of cancer in humans from the endogenous and exogenous formation of N-nitroso compounds from nitrite, many of which are carcinogenic in animals. However, recent epidemiological studies have not supported an association between nitrate or nitrite exposure from drinking water and increased cancer rates in humans. In experimental animals, nitrate and nitrite have not been shown to be carcinogenic. Nitrite has only been shown to be carcinogenic in animals when administered concurrently with nitrosable amines, apparently as a result of the endogenous formation of carcinogenic amines (WHO, 1996; NRC, 1995; Fan and Steinberg, 1996).

In addition to the cancer studies evaluated by the World Health Organization (WHO, 1996), the National Research Council (NRC, 1995) and EPA US (1991), three recent epidemiological studies were identified examining the effects of nitrate exposure on human cancer risk. The results of one case-control study from Germany (Steindorf et al., 1994) suggest the absence of an association between low nitrate levels in drinking water (16 mg nitrate-nitrogen/L) and the risk of brain tumors. Two studies by Morales-Suarez-Varela were published on the impact of elevated nitrate concentrations in drinking water (greater than 50 ppm) on cancer incidence and mortality in Valencia, Spain (Morales-Suarez-Varela et al., 1993, 1995). The authors suggest a possible association between elevated nitrate concentrations in drinking water and cancer of the bladder and stomach. However, a definite cause-and-effect relationship was not shown. Another case-control study conducted in Nebraska examined nitrate in drinking water and the risk of non-Hodgkin's lymphoma (Ward et al., 1996). Authors reported that long-term exposure to elevated nitrate in drinking water (greater than 50 ppm) may contribute to the risk of non-Hodgkin's lymphoma, but again, no cause-and-effect relationship was shown.

The work aims at a hydrogeochemical characterization of the groundwater bodies in the area, with special attention to nitrate contamination. To date, a complete analysis of the region's waters has not been completed. The physicochemical characteristics, including major and minor chemical elements were determined and analyzed by factor analysis. This technique can be used to characterize and plan monitoring of the groundwater quality (Zeng and Rasmussen, 2005). Factor analysis can be used to determine the structure in the relationships between water quality parameters and identify the most important factors contributing to this structure.

Geological and hydrogeological aspects

The region of and around Gulf, Togo (Figure 1) has two rainy seasons, which are from April to July and early

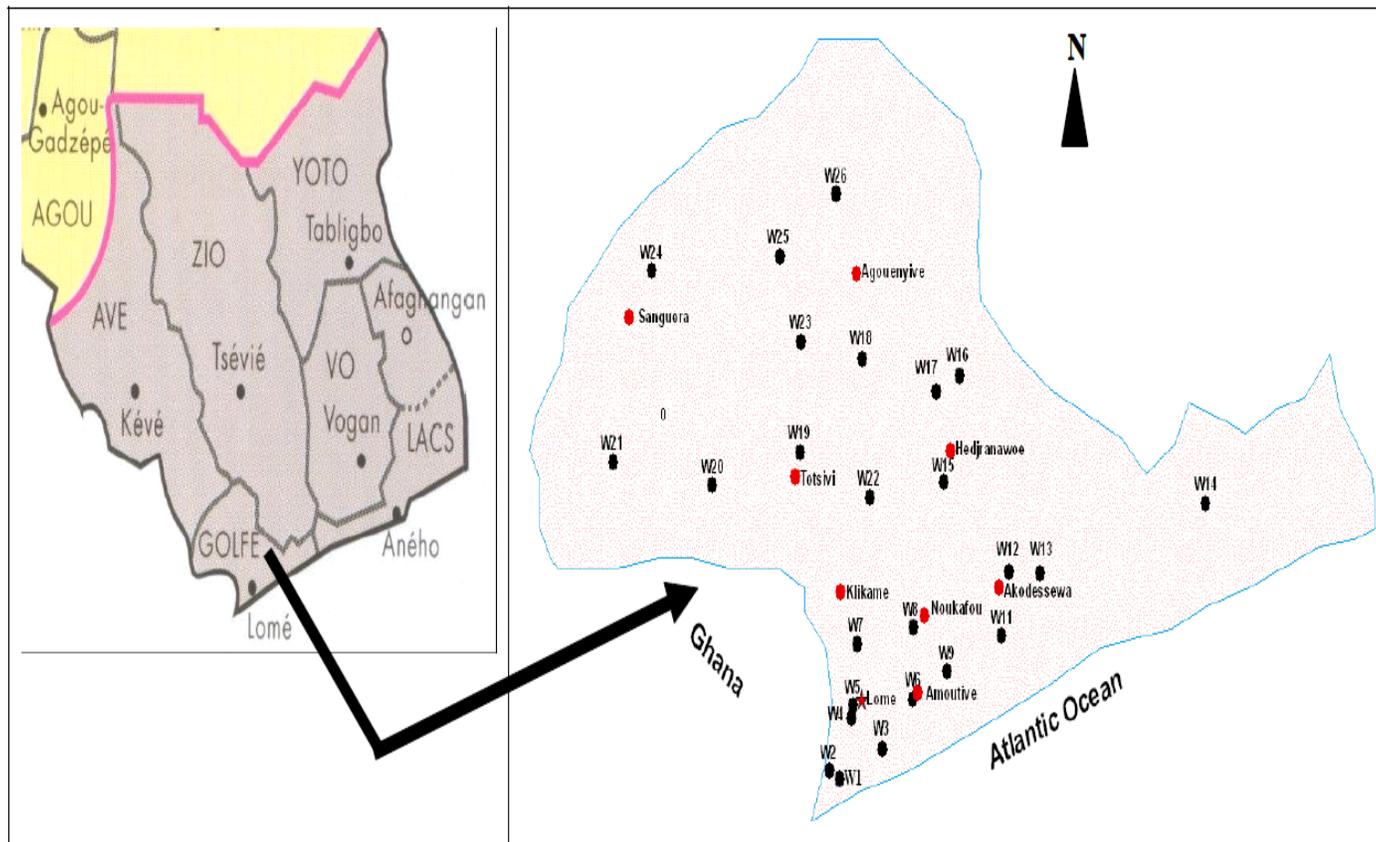


Figure 1. The study area in Gulf Region, Togo.

September to late November. However, the rainfall is relatively low (800 mm per year) for the latitude of the city. The climate is stabilized by the ocean and sea breezes, and the average maximum and minimum temperatures are 30 and 23°C, respectively. The study area was located between the latitudes 6°15'20.00" and 6°6'48.83"N and the longitudes 1°12'30.00" and 1°25'56.00"E. It covered an area of 455 km². The region and its vicinity are located on coastal sedimentary rock mainly of sand and alluvium (Figure 2).

MATERIALS AND METHODS

Groundwater quality monitoring was carried out for 26 well locations in the study area during June to August 2010, which were selected to provide a uniform distribution over most of the drinking water supply area. Physicochemical parameters, including temperature, pH, electrical conductivity (EC) and total dissolved solid (TDS), were conducted in the field. Other samples were transported to the Laboratory. Samples were stored in the dark in a cold room at 4°C. Analysis was carried out according to established methods as follows: Volumetric analysis for HCO₃⁻ molecular spectroscopy for NO₃⁻, SO₄²⁻, NH₄⁺, Mn²⁺ and Fe_{tot}; atomic absorption spectroscopy for Na⁺ and K⁺; and complexometric EDTA titration for Ca²⁺ and Mg²⁺. The analytical methods were based mainly on the normalized AFNOR (AFNOR, 1996) methods. The results were analyzed using

AquaChem 5.1, which is a software package developed specifically for graphical and numerical analysis and modeling of water quality data (Waterloo Hydrogeologic Inc., 2009). All the data were statistically analyzed by factor analysis. Before analysis, the data were normalized to a distribution with a mean of zero and standard deviation of one. Factor extraction was performed using principal component analysis. Three factors with eigenvalues above one were retained following the Kaiser criterion (Kaiser, 1958).

RESULTS AND DISCUSSION

Hydrogeochemical modeling

Physico-chemical parameters were recorded from a selection of monitoring bores in and around Gulf when measuring water levels and collecting groundwater samples. A summary of the recorded physico-chemical parameters is listed in Table 1.

The EC results for the wells ranged from 566 to 8685 $\mu\text{S}\cdot\text{cm}^{-1}$. The World Health Organization (WHO) standard for EC for drinking water is between 500 and 1500 $\mu\text{S}\cdot\text{cm}^{-1}$ (Rodier, 1996). However, 30% of the wells could be classed as moderately mineralized (EC = 500 to 1000 $\mu\text{S}\cdot\text{cm}^{-1}$) and 25% as highly mineralized (EC = 1000 to 1500 $\mu\text{S}\cdot\text{cm}^{-1}$). Many of the wells (45%) could be classed

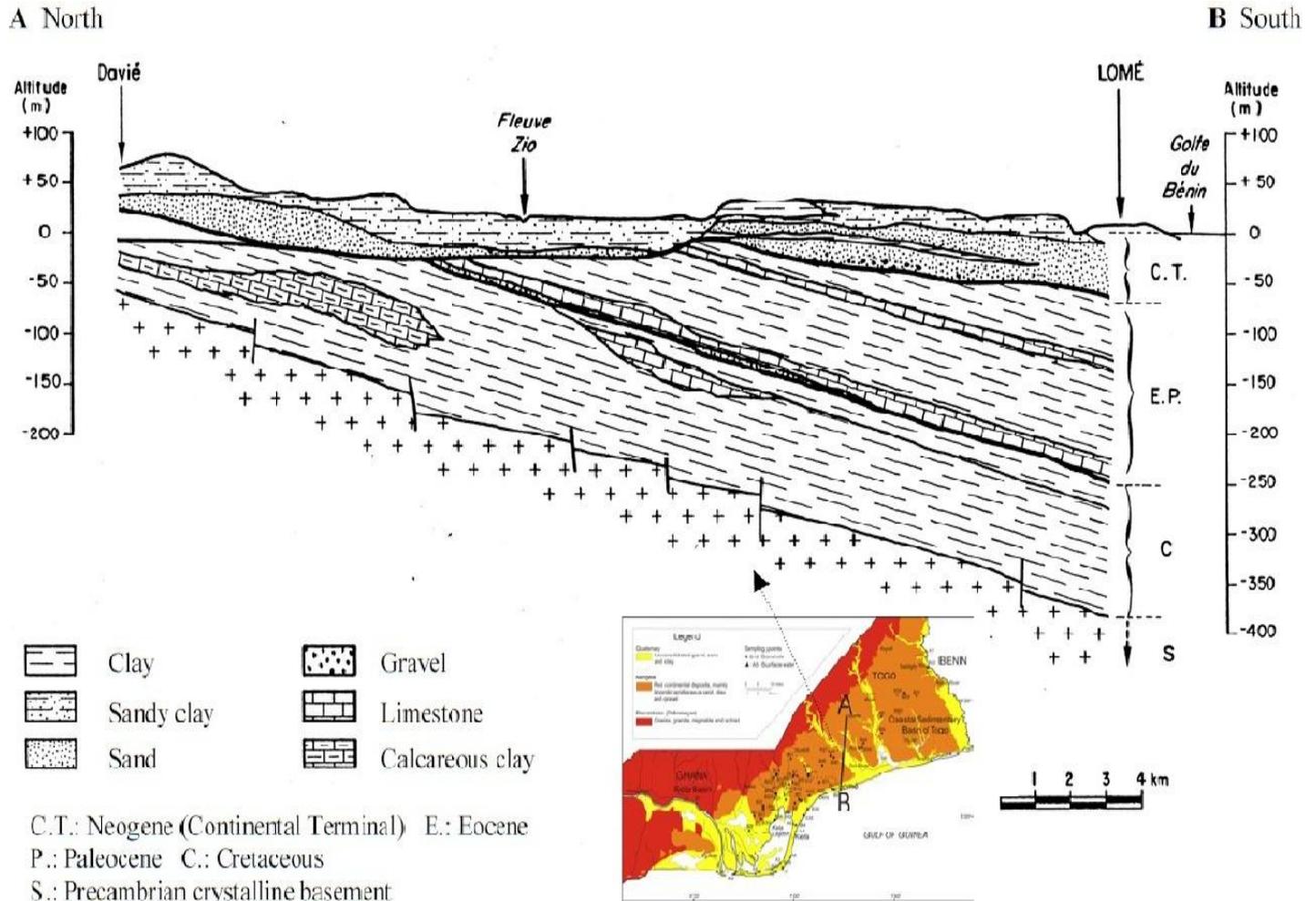


Figure 2. Geological north - south cross-section of cretaceous-quaternary sediments from the western part of the Coastal Sedimentary Basin of Togo.

Table 1. Hydrochemical parameters for (June to August, 2010) in and around Gulf, Togo.

Parameter	Unit	Max	Min	AM
pH		7.45	5.07	6.214
Temp	°C	31	24.4	27.093
EC	uS/cm	8680	327	1677
TDS	mg/L	4570	210	992.79
K	meq/L	1.969	0.051	0.772
Na	meq/L	62.202	1.559	10.618
Ca	meq/L	9.581	0.349	2.339
Mg	meq/L	8.475	0.131	2.182
Cl	meq/L	75.677	0.801	12.503
SO ₄	meq/L	14.094	0.416	2.341
HCO ₃	meq/L	7.597	0.219	2.574
Fe	meq/L	0.922	0	0.182
NH ₄	meq/L	0.018	0	0.0036
NO ₃	meq/L	6.080	0	0.982
SiO ₂	meq/L	42.1	17.5	32.9

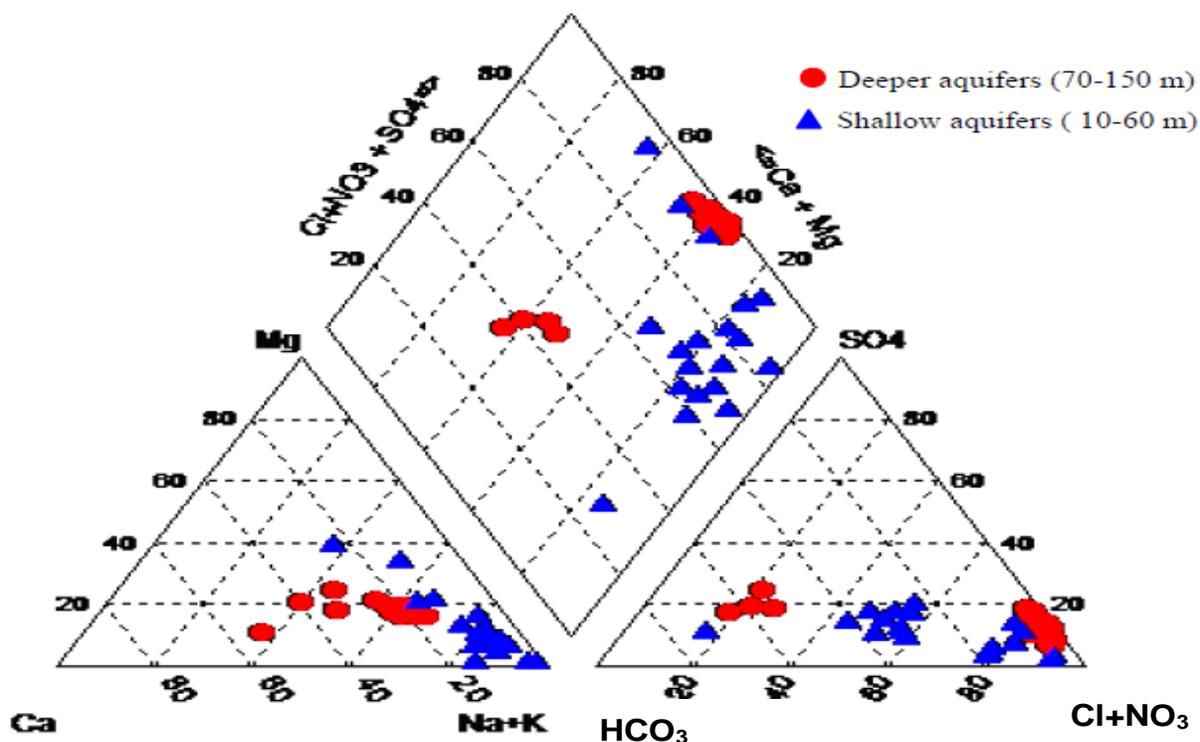


Figure 3. Piper diagram of groundwater wells in the study area.

as excessively mineralized ($EC > 1500 \mu\text{S}\cdot\text{cm}^{-1}$). Groundwater monitoring bores were found to have an increased EC from north to south. The average pH value of groundwater within aquifer was slightly acidic.

The major ions of groundwater from monitoring bores within the in and around Gulf are plotted on a Trilinear diagram shown in Figure 3. The trilinear plots display data based on the percentage of major cations and anions of water sample. This plot can reveal useful properties and relationships of different groundwater groups. Trilinear diagrams can indicate samples with similar chemical compositions, via the clustering of data points. The trilinear plot of groundwater samples in and around Gulf shows that the dominant water type in the area is Na-Cl type water.

The most abundant cation in the groundwater was Na^+ , which was at levels ranging from 1.559 and 62.202 meq/L. This was followed by Ca^{2+} (0.349 to 9.581 meq/L), Mg^{2+} (0.131 to 8.475 meq/L), and K^+ (0.051 to 1.969 meq/L). The dominant anion in the groundwater was Cl^- (0.801 to 75.677 meq/L). Over half of the wells (77%) had chloride levels exceeding the WHO standard for drinking water (250 mg/L). The source of Cl^- is probably marine infiltration, and it may also originate from human waste, particularly urine and some cleaning products. The groundwater concentration ranges for HCO_3^- , SO_4^{2-} , and NO_3^- were 0.219 to 7.597, 0.416 to 14.094, and 0 to 6.080 meq/L, respectively. The NO_3^- concentrations at some locations (70% of the wells) exceeded the 50 mg/L

limit specified by the WHO drinking water standard. High nitrate concentration in the groundwater is observed in the central part of Lome. The correlation coefficients among various groundwater quality parameters were obtained to investigate their interdependence. This shows that nitrate has a negative relationship with calcium and sodium, whereas relationship with magnesium, potassium, bicarbonate alkalinity, sulphate, chloride and EC are positive (Table 2).

Multivariate analysis

Result of factor analysis of the groundwater chemistry data indicates two factors that can be related to various controlling processes presumed to have produced the different water species. Table 3 shows the varimax rotated factor matrix consisting of the component factors, the loading of variables on each factor and percentage of data variance explained by each factor. These two factors account for 95% of the total variance in the dataset.

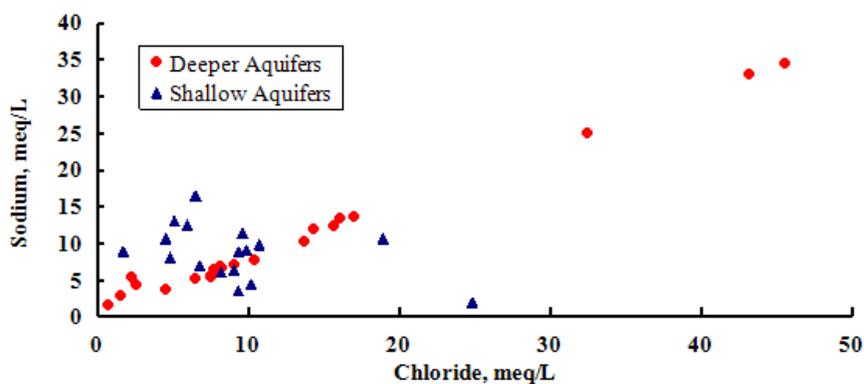
Factor 1 has a high loading of EC, Na, Cl, Ca, SO_4 and Mg and explains 60% of the total variance (Table 3). The concentration of Na and Cl in seawater is much greater than in continental water. This factor can be ascribed to the intrusion of seawater into the aquifer system which increases the concentrations of these ions and hence values of the dissolved solids. Correlation coefficients (r) between EC and each of the ions were calculated using

Table 2. Correlation coefficients matrix for June-august 2010 hydrochemical data.

Parameter	TDS	Conductivity	pH	K	Na	Ca	Mg	Cl	SO ₄	HCO ₃	NO ₃
TDS	1	0.983	0.047	0.259	-0.072	0.879	0.968	0.961	0.944	0.399	0.662
Conductivity		1	-0.034	0.245	0.281	0.46	0.797	0.976	0.918	0.113	0.486
pH			1	-0.063	-0.097	-0.219	-0.215	-0.158	0.17	0.879	0.075
K				1	-0.129	0.248	0.239	0.277	0.191	-0.019	0.15
Na					1	0.422	0.422	0.328	0.256	-0.163	-0.087
Ca						1	0.88	0.551	0.515	-0.32	-0.29
Mg							1	0.857	0.793	-0.216	0.033
Cl								1	0.875	-0.047	0.357
SO ₄									1	0.256	0.4
HCO ₃										1	0.34
NO ₃											1

Table 3. Varimax rotated factor loading matrix for groundwater chemistry data in Gulf region and its environs.

Parameter	Factor 1	Factor 2
Mg	0.995	
Na	0.994	
EC	0.993	
Cl	0.981	
Ca	0.979	
SO ₄	0.957	
HCO ₃		0.961
SiO ₂		0.953
pH		0.950
NO ₃		0.793

**Figure 4.** Relationship between chloride and sodium concentration.

data from all the wells. These correlation coefficients show that the EC is strongly influenced by Cl⁻ and SO₄²⁻ (Table 2). The correlation between Cl⁻ and Na⁺ showed that the Cl⁻ is largely present in the form of NaCl (Figure 4).

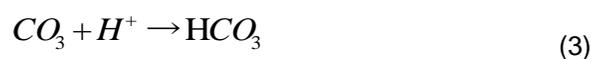
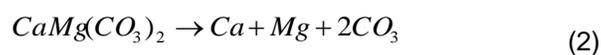
Factor 2, which explains 35% of the total variance, includes SiO₂, NO₃, pH and HCO₃ (Table 3). This factor reflects the signatures of natural water recharge and water-soil/ rock interaction. Surface water charged with atmospheric and biogenic CO₂ infiltrates into the

subsurface and aggressively attack calcite, dolomite, aluminosilicates including feldspars and micas present in the formation (SI values are <0) liberating cations such as Ca and Mg into the water and leaving residues of clay minerals. A consequence of this incongruent dissolution is a rise in pH and in HCO_3^- concentration of the water (Freeze and Cherry, 1979). HCO_3^- has good correlation with pH (Table 2). Nitrate is naturally present in groundwater at very low concentrations, and its source is human activities such as domestic or industrial waste or agricultural. Consequently, F2 also reflects the anthropogenic impact of domestic sewage, industrial waste, uncontrolled landfill waste, fertilizers, and manure on the study area.

Together these two factors accounted for 95% of the variability of groundwater quality. Their parameters include EC, Na, Cl, Ca, SO_4 and Mg (F1), and SiO_2 , NO_3^- , pH and HCO_3^- (F2). However, measurement of only three parameters (HCO_3^- , NO_3^- , and EC) is sufficient for regular monitoring of the quality of well water. The other parameters can be determined by simple linear regression, because of their correlation to these three parameters.

Geochemical reactions

The actual changes in concentrations of chemical species as a function of sulphate concentration help in obtaining the information on the possible geochemical reactions that may be occurring in the study area. To investigate this, analysis was carried out using the hydrochemical data. The increase in calcium, magnesium and pH as sulphate increases are shown in Figure 5a to c. The groundwater in the area of study is undersaturated with calcite and dolomite having a saturation index of -3.70 to 0.22 and -7.32 to 0.40, respectively. Calcite dissolution causes a pH increase due to consumption of H^+ by carbonate during the dissolution process. The decrease in the CO_2 in the solution leads to the dissolution of calcite and dolomite and CO_2 dissolution, thereby increasing the magnesium and calcium concentration in the solution. The geochemical equations for possible reactions in the aquifer are as follows:



where (g) in (4) refers to the gaseous phase and (aq) to the aqueous phase.

Although the trends in calcium, magnesium and pH with sulphate are evident in the groundwater of the northern part of Gulf region, there is variation in the temperature and other reactions in addition to dedolomitization or dissolution of carbonates. Dedolomitization is a specific geochemical process and has been reported by Plummer et al. (1990) and Kloss and Goebelbecker (1992). The plot of bicarbonate alkalinity as a function of dissolved sulphate concentration (Figure 5d) shows that bicarbonate alkalinity is increasing with decrease in sulphate, and is possibly indicative of dedolomitization or carbonate dissolution. Cation exchange with calcium and magnesium cations could contribute to additional bicarbonate on the flow path with uptake of calcium and magnesium and release of sodium from exchange sites on clay minerals causing dissolution of carbonate minerals. The molar concentrations of sodium plus potassium are slightly more than the concentration of chloride. This is indicative of presence of evaporites. These waters have high bicarbonate concentration. Thus, there is a tendency to form sodium-bicarbonate waters. For the groundwater in the study area, the sodium chloride-bicarbonate water is derived from the dissolution of carbonate minerals and sea water intrusion.

Conclusion

High nitrate concentration in groundwater, ranging in between 0 and 6.08 meq/L, is observed in large parts of Lome urban area, Togo, and is a matter of concern for drinking water supply since it exceeds the maximum permissible nitrate concentration of 50 mg/l for public water supply systems. Dissolved nitrate is usually less than 0.774 meq/L in the northern part of Gulf region, whereas the central and southern regions are the most affected areas, having nitrate concentrations of more than 1.61 meq/L in some areas. The result of the factor analysis, as applied to the chemical data set of groundwater in the Coastal Sedimentary Basin of Togo provides an insight into the underlying controlling hydrochemical processes in the area. The interpretation of the results due to the factor analysis depends mainly on the investigator, his experience and the knowledge about researched area. Multivariate statistical techniques used in this study were to enable identifying the key parameters which describe groundwater quality and to characterize the spatial variability of groundwater quality data. The variability in the water quality depended on the following three factors: Alkalinity (HCO_3^-), the impact of human activities (domestic or industrial discharge), and

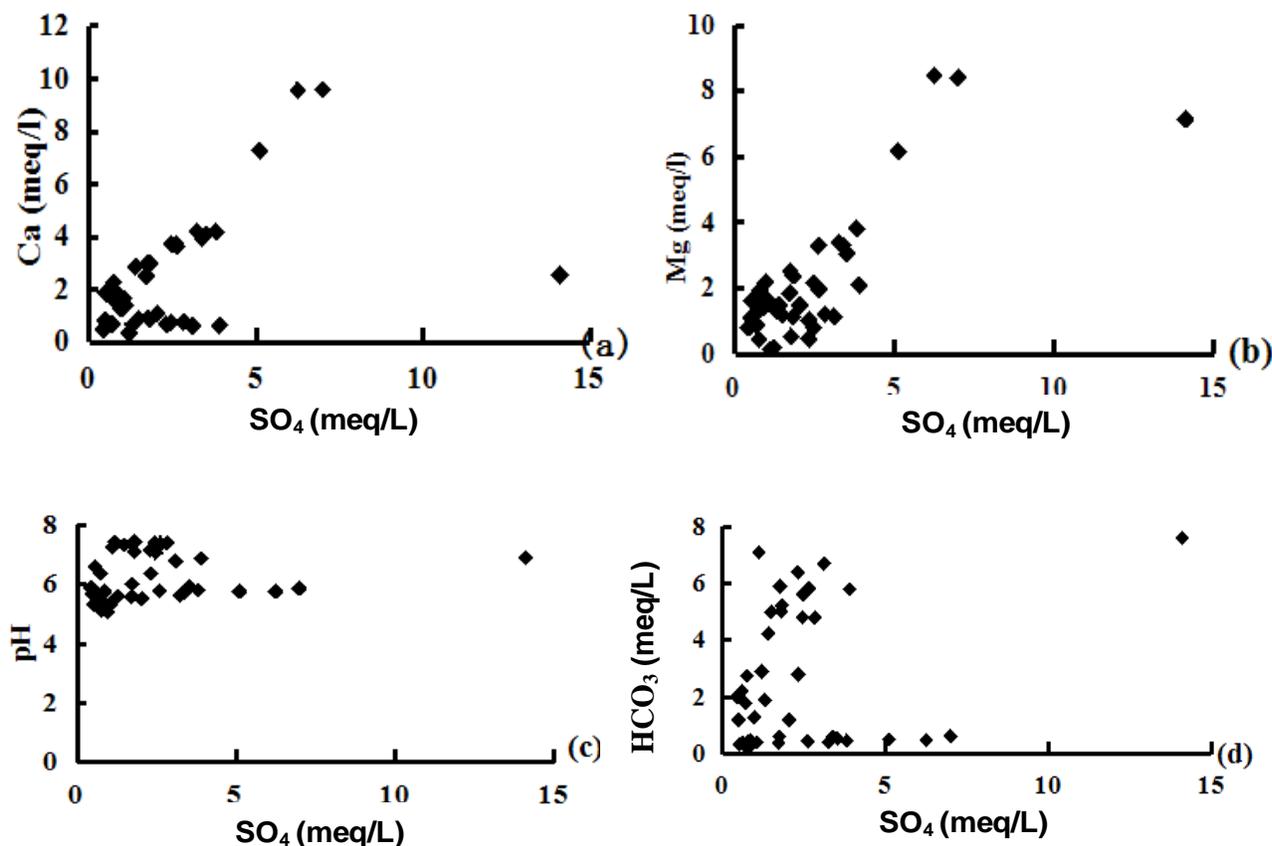


Figure 5. Plot of calcium, magnesium, pH and bicarbonate alkalinity as a function of sulphate.

salinity. The plausible geochemical reactions in the area of study are dissolution of calcite and dolomite, carbon dioxide and sulphate minerals with ion exchange.

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