

isotopes ($\delta^{18}\text{O}$ and δD) and major cations (Ca^{2+} and Mg^{2+}) in groundwater and adjacent estuarine tidal creek. During the time series measurements, we pumped groundwater for over 5 tidal cycles to induce flow into a test well.

New hydrologic insights for the region: The time-series groundwater level mimicked water level changes in the estuary. However, the temporal salinity in groundwater did not correspond to tidal salinity changes of estuarine water, indicating that estuarine water did not intrude groundwater. The $\delta^{18}\text{O}$ and δD of the groundwater and estuarine samples are collinear and fall along the local meteoric water line of Douala and had d-excesses of >10 , indicating a non-evaporated rain recharge of groundwater or lack of salinization by evapoconcentration. The salinity- $\delta^{18}\text{O}$ relationship showed that the origin of salinity in the coastal groundwater aquifer is not from seawater intrusion. The $\text{Mg}^{2+}/\text{Ca}^{2+}$ ratios of <1 in groundwater compared to 6-8 in estuarine water support the non-seawater origin of groundwater salinity. We conclude that the salinity of coastal groundwater in Douala is not affected by tide-induced salinization from the adjacent Wouri Estuary but by geogenic sources in the aquifer.

Keywords: Coastal groundwater; groundwater salinization; time series analysis; Wouri Estuary; Cameroon

1. Introduction

The quality of coastal groundwater is a crucial factor in water resource management and sustainability (Lee and Song et al., 2007; Werner et al., 2013; Mirzavand et al., 2018; Mirzavand et al., 2020; Ouhamdouch et al., 2021). Salinization is an important process that causes quality deterioration in groundwater in coastal regions. Studies conducted in coastal settings show that

groundwater salinization occurs from seawater intrusion induced by groundwater overdraft (Melloul and Goldenberg, 1997; Demirel, 2004; Pulido-Laboeuf, 2004; Jørgensen et al., 2008; Werner et al., 2013; Ouhamdouch et al., 2017; Kanagaraj et al., 2018), tidal flooding and storm surges (Mao et al., 2001), geochemical processes such as water-rock interaction, cation exchange, carbonate and mineral dissolution, evaporation of freshwater and mixing with saline connate water in the sedimentary formations (Hem, 1985; Aquilina et al., 2002; Amiri et al., 2016; Mirzavand et al., 2020) and from anthropogenic pollution (Cary et al., 2015; Bahir and Ouhamdouch, 2020).

The effective management of coastal groundwater resources depends on the understanding of sources of salinity in the groundwater. However, the multiplicity of mechanisms of salinization confound the determination of the origin of salinity in coastal groundwater (Mirzavand et al., 2020). Hence, many studies have utilized different methods to identify saline coastal groundwater and determined their salinization mechanisms. Geophysical methods (e.g., electrical resistivity) have been used to detect salinity differences between saline and fresh groundwater and their interface (Mirzavand, 2018, 2020). Although the commonly used electrical resistivity method show salinity disparities between saline and fresh groundwater, the method cannot determine the origin and mechanism of groundwater salinization (Mirzavand 2020). Thus, to determine the source(s) of salinity and mechanisms of groundwater salinization, studies have used hydrogeochemical modeling techniques such as PHREEQE and faces evolution diagrams (Giménez-forcada, 2010) and geochemical tracers such as salinity, minor and major ions (Alcalá and Custodio, 2008; Harkness et al., 2017; Isawi et al., 2016; Mirzavand, 2018; Amiri et al., 2016; 2021). Hydrogeochemical modelling allow researchers to simulate mixing and reaction pathways (PHREEQE), classify water (Piper diagrams) and decipher intrusion and freshening

phases (hydrochemical facies evolution diagram: HFE-D). Geochemical tracers such as major ion ratios (Na^+/Cl^- , Br^-/Cl^- , $\text{SO}_4^{2-}/\text{Cl}^-$, $\text{Ca}^{2+}/\text{Cl}^-$, B/Cl^- , $\text{Mg}^{2+}/\text{Cl}^-$) have been used in studies to identify the major sources of salinity in groundwater (Alcalá and Custodio, 2008; Amiri et al., 2016; 2021). However, these ionic ratios can be affected by processes such as water-rock interaction and anthropogenic contamination which may cause problems with source identification in coastal aquifers in urbanized settings. Due to the limitations of the different approaches to identify the origin of salinity in coastal groundwater, researchers combine multiple approaches to elucidate the origin of salinity in groundwater.

Although groundwater salinization in coastal areas have been widely researched using different methods, few of these studies have focused on assessing the effects of salinization of groundwater by estuarine water (Mao et al., 2001; Mao et al., 2006; Lenkopane et al., 2009; Tularam and Singh, 2009; Dieng et al., 2017; Shalem et al., 2019) as a result of tidal forcing. Studies on tide-induced groundwater salinization in estuarine settings requires a unique experimental approach assessing near real time behavior of water and salinity in both groundwater and the estuary. A significant challenge to study tide-induced salinization of coastal groundwater in estuarine environments is to acquire hydrochemical data at a frequency adequate to elucidate the effects of tides in this process. Thus, there is a need to acquire data on water level (tides) and tracers of salinity (e.g., salinity, major cations) and tracers of water origin (stable oxygen ($\delta^{18}\text{O}$) and hydrogen (δD) isotopes) at a frequency adequate to characterize tidal effects. When tracers of salinity and water origin are collected over multiples tidal cycles, the data allows for direct comparison between the near real time behavior of the tracers in estuarine water to those in groundwater (Jones et al., 1999; Mao et al., 2006; Lenkopane et al., 2009; Currell et al., 2015; Shalem et al., 2019).

In this study, we investigated groundwater salinization from the coastal aquifer adjacent to the tropical Wouri Estuary (Douala, Cameroon). Groundwater in the coastal Douala aquifer serves as both a supplementary and a primary source of water supply for domestic and industrial activities (Takem et al., 2010; Takem et al., 2015; Fantong et al., 2016; Ketchemen-Tandia et al., 2017; Wirmvem et al., 2017a). A study of the hydrogeochemical characteristics of surface flows and groundwater in Douala by Fantong et al. (2016) confirmed quality impacts from salinization. The groundwater salinization characterized by high chloride concentrations is attributed to tide-induced flooding by estuarine water (Mafany, 1999; Fantong et al., 2016). Tide-induced salinization adequately explains seawater contamination of coastal groundwater immediately adjacent to the coast, where high tides cause (1) higher heads relative to groundwater causing estuarine water to flow into and salinize groundwater or (2) estuarine water to overtop and flood the land surface, which subsequently infiltrates, recharges and salinize groundwater (Mao et al., 2006; Tularam and Singh, 2009; Shalem et al., 2019).

To test if tide-induced estuarine water head and/or flooding are responsible for estuarine water contamination of the coastal aquifer in Douala (Mafany, 1999; Fantong et al., 2016), we investigate tide induced salinization of groundwater in the Youpwe neighborhood, adjacent to the Wouri Estuary. The Youpwe neighborhood in the coastal city of Douala Cameroon (Fig. 1) is an ideal location to study estuarine salinization of groundwater. A tidal creek (Dr. Creek) forms a linear boundary between groundwater below the Youpwe neighborhood and the estuary, and can be envisioned as a linear source of estuarine water which could potentially impact shallow groundwater. We employed a combination of time series water level and salinity measurements and assessed the $\delta^{18}\text{O}$ and δD and major cation ratios to track estuarine salinization of groundwater. Our objective was to ascertain if saline water from the Wouri Estuary is the cause

of salinization of groundwater. Our findings are important for understanding coastal groundwater salinization and contamination and for management of coastal groundwater resources in this tropical estuarine environment.

2. Study site

2.1 Location of water sampling sites

The groundwater and estuarine water (Dr. Creek) investigated in this study are located in the Youpwe neighborhood near the head of the Wouri Estuary in the city of Douala, Cameroon (Fig. 1). The Wouri Estuary (3°49' - 4°04' N and 9°20' - 9°40' E) is located in the Gulf of Guinea on the Atlantic coast of Cameroon's coastline plain (Fig. 1a). The Wouri Estuary comprises open water of approximately 1200 km² and extensive (1750 km²) mangrove forests (Simon and Raffaelli, 2012; Ndongo et al., 2015; Fossi Fotsi et al., 2019). Freshwater discharge into the Wouri Estuary are from the Wouri River at the head and the Dibamba River and Mungo River about 15 km from the head (Fig. 1a).

2.2. Climate

Douala has a humid equatorial climate characterized by a rainy season that spans from April to October and a dry season that spans from November to March (e.g., Fantong et al., 2016). The mean annual precipitation is 4000 mm and rainfall amounts peak in June and September. During the rainy season, saturated air masses blow from the Gulf of Guinea. During the dry season, the Harmattan winds blow southwestward from the Sahara Desert causing low humidity and high temperature conditions. The daily temperature ranges between 23 and 33 °C.

2.3. Geological setting

The study area is in the Phanerozoic Cretaceous-Quaternary Douala Basin (Dumort, 1968; Regnault, 1986). The Douala Basin consists of up to 5 km thick sedimentary sequence of unconsolidated and semi-consolidated rocks which overlie a Precambrian basement (Regnault, 1986; Tamfu et al., 1995). The vertical succession consists of the Moundeck, Logbaba, Nkappa, Souelaba, Matanda and Wouri Formations (Regnault, 1986). The lithologic composition of the Moundeck Formation varies from shale at the top, through medium sized sand, to basal conglomerate that overlie the Precambrian crystalline basement. The Logbaba Formation is shale that intercalates with fossiliferous limestone and gravelly sand. The Nkappa Formation is primarily shale-rich in channel-filled sands. The Souelaba Formation is composed of marls, sandy shale, calcareous sandstone and ferruginous limestone. The mineral phases in the Nkappa and Souelaba Formations include smectite, halloysite, chlorite, feranhydrite, calcite, ilmenite, muscovite, K-feldspar, gypsum, pyrite, and corundum (Ngon et al., 2016). The Matanda Formation comprises of shale, overlain by coarse-grained sand that intercalate with Tertiary basalts from Mount Cameroon lava flows. The Wouri Formation is comprised of coarse-grained sand and gravel, intercalated with ferruginous clay dominated by quartz and varying proportions of ilmenite, magnetite, muscovite, sillimanite, epidote and sphene (Regnault, 1986).

2.4. Groundwater hydrogeology

The sedimentary sequence in the Douala Basin hosts a deep and a shallow aquifer. The deep aquifer consists of the Basal Sandstones of the Cretaceous Moundeck Formation, the Logbaba Formation and the Paleocene Sands of the Nkappa Formation. The Basal Sandstones aquifer unit is confined by the underlying Precambrian granites and the upper Paleocene Sands unit is

confined by the impermeable marine shales of the Souelaba Formation (Regnault, 1986; Mafany, 1999). The Paleocene sand aquifer is 200 m thick and is exploited by bore holes (Djeuda-Tchapnga et al., 2001). The shallow aquifer unit is unconfined and composed of Pleistocene sands and alluvium belonging to the Matanda Formation and the Wouri Formation (Regnault, 1986). The thickness of the aquifer ranges between 50 and 60 m (Djeuda-Tchapnga et al., 2001).

The static water level in the shallow aquifer varies from 2 to 22 m below ground surface (bgs) and drops a 4 to 16 m range in some areas due to overexploitation (Fantong et al., 2016). The storage coefficient of the shallow aquifer varies from 2.4×10^{-5} to 7.4×10^{-5} , transmissivity ranges from 1.1×10^{-2} to 2×10^{-3} m/s, the hydraulic conductivity is from 1.1×10^{-4} to 1×10^{-3} m/s and the specific yield varies from 2.2 to 4.9 (Fantong et al., 2016). Groundwater flow in the shallow aquifer is multidirectional; from southeast to northwest, northwest to west and from northeast to south-west (Mafany, 1999). In the Youpwe neighborhood, groundwater levels are <1.5 m bgs and groundwater flow is generally to the south, discharging into the estuary.

2.5. Dr. Creek hydrology

Dr. Creek is a tidal creek located in the head of the Wouri Estuary and is below the southern boundary of the eastern part of the city of Douala (Fig. 1a). Dr. Creek opens to the eastern side of the Wouri Estuary, and the nearly 10 km long channel is nearly perpendicular to the estuary. Dr. Creek is flanked to the north by the Youpwe neighborhood built on previously destroyed marshland and to the south by mangrove forests of the estuary (Fig. 1b). The hydrologic regime of the Dr. Creek and the Wouri Estuary is characterized by semidiurnal mixed tides, with an average range of about 3 m (Olivry, 1974; Onguene et al., 2014).

3. Methodology

3.1. Locations for groundwater and estuarine water investigation

We selected three locations to investigate groundwater properties (Fig. 1b and 1c). Well 1 (4°0'27.16"N, 9°42'3.62"E) is a 2.5 cm diameter borehole that is 30 m deep and located 122 m from Dr. Creek. Well 1 was drilled to support commercial activities of a hotel and is not in use because of contamination by salinity. Well 2 (4°0'29.89"N, 9°42'3.20"E) is a hand dug well 3 m deep with a diameter of 1 m and is located 200 m from Dr. Creek. Well 3 (4°0'30.18"N, 9°42'2.64"E) is a hand dug well 2 m deep and 0.5 m in diameter and is located 250 m from Dr. Creek. Well 2 and Well 3 are domestic wells in active use. The time series measurements in Dr. Creek were conducted near an island (4° 0'4.28"N; 9°42'3.45"E). Estuarine water was sampled for chemical and isotopic measurements from Dr. Creek were collected at 10 stations in the creek (from 3°58'37.3"- 4°00'26.6" N to 9°41'40.1"- 9°44'09.7" E) adjacent to the Youpwe neighborhood (Fig. 1b).

3.2. Time series monitoring of air temperature, barometric pressure, water level, water temperature and electrical conductivity

We recorded air temperature (°C) and barometric pressure with a Solinst™ barologger. The Solinst™ barologger was set to record and convert the barometric pressure in the same units as water level (m) by using a conversion factor of 0.101972 m/kPa. The barologger pressure sensor has an accuracy of +0.05 kPa and the air temperature sensor has an accuracy of + 0.05 °C and a resolution of 0.003 °C. The barologger was deployed outside the Aquarius Marina 2000 Hotel (4° 0'26.60"N, 9°42'3.35"E) and set to an altitude of 0 m above sea level (asl).

We used Solinst™ LTC (level, temperature, conductivity) loggers to record water level, temperature and electrical conductivity (EC) in groundwater and estuarine water. The Solinst™ LTC logger records water level with an accuracy of $\pm 0.1\%$ percentage of full scale, and is automatically temperature compensated between 0 to 40 °C. The temperature sensor of the Solinst™ LTC has an accuracy of ± 0.1 °C and a resolution of 0.1 °C. The Solinst™ LTC measures the EC using a 4-electrode conductivity sensor that operates between -20 to 80 °C, has the ability to measure on a full range of 0 to 80,000 $\mu\text{S}/\text{cm}$ with a resolution of 1 $\mu\text{S}/\text{cm}$, and the EC is normalized to 25 °C. The EC values were converted to salinity by multiplying the EC by 0.0006, which was empirically determined from a least squares regression of EC vs. salinity measured in the Douala estuary by us (Bennett, 1976). A Solinst™ LTC logger was deployed in Well 1 from 6/11/2019 at 15 h to 6/14/2019 at 10 h. The level, temperature and conductivity of groundwater in Well 1 was monitored for 19.5 h before it was subsequently pumped at 4.7 m^3/minute for the duration (47.5 h) of the study using a high capacity well pump. Another Solinst™ LTC logger was deployed in Dr. Creek near a small (200 m diameter) island (4° 0'4.28"N; 9°42'3.45"E) in the middle of the tidal channel (Fig. 1b). The logger was deployed from 6/12/2019 at 10 h to 6/14/2019 at 10 h at 5 cm from the channel bottom in a 5.2-cm diameter perforated PVC tube.

The Solinst™ barologger and LTC loggers were programmed to collect data in a linear sampling mode at minute intervals. The water levels recorded by the LTC levelloggers were corrected for barometric pressure effects using the time equivalent barometric pressure values recorded by the barologger. Barometric pressure compensation is necessary because of elevation differences above and below sea level and pressure changes from storms which can introduce errors to LTC levelloggers recorded water levels.

3.3. Water sampling

Groundwater samples from shallow and hand dug wells (Wells 1, 2 and 3) and surface water samples from the estuary were collected using standard sampling procedures (APHA, 1985). Six samples were collected from Well 1 during well pumping at 2 h intervals on 6/13/2019 (8, 10, 12, 14, 16 and 18 h). Groundwater from Well 2 and Well 3 were collected at 25 cm below the surface on 6/21/2019 by the grab technique using clean plastic buckets. Estuarine samples from Dr. Creek were collected from 15 h to 16.7 h on 7/4/2019 at 25 cm below the surface by the grab technique along a 5 km axial transect (Fig. 1b) at high tide.

3.4. Water sample analyses

During the water sampling, the salinity and electrical conductivity (EC) were measured in situ in groundwater and in Dr. Creek using a Hanna (HI98194) multi-parameter (pH/Oxidation-reduction potential (ORP), electrical conductivity (EC), total dissolved solids (TDS), salinity, dissolved oxygen (DO), temperature) probe. For groundwater, the outlet of the pump was directed into a large 20 L plastic bucket where the Hanna multi-parameter probe was immersed and readings were recorded after stabilization of pH, temperature and EC. For water in Dr. Creek, the Hanna multi-parameter probe was lowered to 25 cm below the surface and readings were taken after stabilization of pH, temperature and EC.

Groundwater was collected directly from the outlet of the hose attached to the pump. Surface water from Dr. Creek was collected at 25 cm below the surface by the grab technique. All water samples were filtered through 0.45 μ m nylon filters during collection. The sampling bottles used were rinsed with the filtered sample to be collected and then filled and capped. Water for cations

analysis were collected in 60 mL polypropylene bottles and acidified to a pH <2 with trace metal grade nitric acid. Water samples for isotopic analysis were collected unacidified in 25 mL glass scintillation vials with inverted cone closures. All the water samples were kept cool and transported to the University of Delaware (USA) where they were refrigerated until analyses.

In the laboratory, the water samples were analyzed for calcium and magnesium by Inductively Coupled Plasma Mass Spectrometer (Agilent Tech. ICP-MS:7500cx series). The $\delta^{18}\text{O}$ and δD were measured using a Los Gatos Research Liquid Water Isotope Analyzer (LGR LWIA). The isotopic ratios are reported in delta notation (δ) in per mil (‰):

$$\delta (\text{‰}) = ((R_{\text{sample}} - R_{\text{standard}}) / R_{\text{standard}}) \times 1000$$

where R is the ratio of D/H, or $^{18}\text{O}/^{16}\text{O}$ in the sample and standard (e.g., Brand, 2011). The standard is the Vienna Standard Mean Ocean Water (VSMOW) international standard. The precision (1σ standard deviation) of the LGR LWIA is better than $\pm 0.3 \text{ ‰}$ for δD and $\pm 0.07 \text{ ‰}$ for $\delta^{18}\text{O}$.

4. Results

4.1. Time series water level, salinity and temperature in groundwater and in Dr. Creek

4.1.1. Water level

The groundwater level measured in Well 1 fluctuated by 1.1 m (Table S1). The temporal groundwater table response was sinusoidal, with peak water level or low water level separated by ~ 13 h (Fig. 2a). The sinusoidal behavior and the range in change of groundwater level observed for 19.5 h before pumping was the same as during pumping over 47.5 h, except for the lowering of the water table by ~ 1.5 m from the pumping action. Water level in Dr. Creek varied by 1.2 m

(Table S1). The water level reflects semidiurnal mixed tides with a periodicity of ~13 h. (Fig. 2b).

4.1.2. Salinity

The salinity of groundwater in Well 1 varied between 0.62 and 0.48 psu (Table S1). The temporal salinity in Well 1 for the 19.5 h before pumping decreased only slightly from 0.62 to 0.49 psu, and during pumping, the salinity was nearly constant at 0.48 psu for the 47.5 h (Fig. 2c). The salinity in Dr. Creek varied by 1.4 (Table S1). The temporal changes in the salinity coincide with tidal cycles, with high salinity observed during low tides and lower salinity observed during high tides (Fig. 2d). Additionally, the salinity peaks and troughs continuously decrease slowly over time. The salinity peaks are relatively broad compared to the troughs, which are narrower and show small salinity peaks at peak high tides.

4.1.3. Water temperature and air temperature

The temperature of groundwater in Well 1 varied between 28.2 and 28.6 °C (Table S1). The temporal temperature in Well 1 for 19.5 h before pumping decreased slightly from 28.6 to 28.4 °C, and during the 47.5 h of pumping, the temperature was nearly constant at 28.2 °C (Fig. 2e). The temperature in Dr. Creek ranged from 27.6 to 29.0 °C (Table S1). The temporal temperature showed small fluctuations and a general decrease (Fig. 2f). In addition, during the temporal temperature decreases, the low temperature perturbations show correspondence to the peak tides.

The air temperature in Youpwe varied between 24.3 and 32.8 °C (Table S1). The temperature varied on a diurnal basis with higher temperatures during the day and lower temperatures at night. The daily highs show a peak at ~10:00 h, a low at ~12:00 h and another high at 15:00 h

(Fig. 2g and h). From the afternoon at 15:00 h, the temperature decreases continuously to their lowest values at ~7:00 h in the morning.

4.2. Stable isotopes, salinity, and major cations in groundwater and in Dr. Creek

4.2.1. Stable isotopic composition

The δD of groundwater in Well 1 for all samples was -10 ‰ and the $\delta^{18} O$ ranged between -2.9 and -2.7 ‰, with a mean of -2.8 ± 0.1 ‰ (Table 1). In groundwater from Well 2, δD and $\delta^{18} O$ composition were -10 and -2.7 ‰, respectively. The δD of groundwater in Well 3 was -11 ‰ and the $\delta^{18} O$ was -3.3 ‰. The δD of water in Dr. Creek ranged between -14 and -13 ‰ and averaged -14 ‰ and the $\delta^{18} O$ ranged between -3.4 and -3.2 ‰ and averaged -3.3 ± 0.1 ‰.

4.2.2. Salinity, calcium and magnesium

The salinity in Well 1 ranged from 1.2 to 1.3 psu and averaged 1.2 ± 0.04 psu (Table 1). Well 2 had salinity of 0.2 and Well 3 had a salinity of 1.2 psu. The salinity in Dr. Creek ranged from 0.3 to 0.5 psu, with a mean of 0.4 ± 0.04 psu (Table 1).

The EC in Well 1 ranged from 2320 to 2499 $\mu S/cm$ and averaged 1.2 ± 0.04 $\mu S/cm$ (Table 1). Well 2 had an EC of 399 and Well 3 had a salinity of 825 $\mu S/cm$. The salinity in Dr. Creek ranged from 679 to 924 $\mu S/cm$, with a mean of 822 ± 77 $\mu S/cm$ (Table 1).

The Ca^{2+} concentrations in Well 1 ranged from 3.2 to 3.3 mg/L and averaged 3.2 ± 0.06 mg/L. The Ca^{2+} concentrations were 7.2 mg/L in Well 2 and 9.0 mg/L in Well 3. The Mg^{2+} concentrations in Well 1 varied between 2.0 and 2.3 mg/L and averaged 2.1 ± 0.1 mg/L. The Mg^{2+} concentrations in Well 2 was 6.5 mg/L and in Well 3, the Mg^{2+} concentration was at 7.9 mg/L. The Mg^{2+}/Ca^{2+} ratios varied between 0.6 and 0.9 in groundwater from Well 1, Well 2

and Well 3. The Ca^{2+} concentrations in Dr. Creek varied between 1.4 and 2.4 mg/L and averaged 1.9 ± 0.3 mg/L. The Mg^{2+} concentrations in Dr. Creek ranged of 12.0 to 17.1 mg/L and averaged 14.4 ± 1.7 mg/L. The $\text{Mg}^{2+} / \text{Ca}^{2+}$ ratios in Dr. Creek varied from 6.2 to 8.3.

5. Discussion

5.1. Assessing tide-induced groundwater salinization

The EC in the coastal groundwater sampled in the Youpwe neighborhood in Douala, Cameroon ranged from 399 to 2499 $\mu\text{S}/\text{cm}$ (Table 1). The EC exceeded the WHO drinking water limit of 750 $\mu\text{S}/\text{cm}$ for drinking water (WHO 2004). The salinized groundwater observation is consistent with the results of previous studies that show highly salinized groundwater in the Douala coastal aquifer (Takem et al., 2010; Takem et al., 2015; Fantong et al., 2016; Ketchemen-Tandia et al., 2017; Wirmvem et al., 2017a). We use our time series groundwater and estuarine water salinity and temperature and the stable water isotopes, salinity and ionic ratios to assess if the origin of salinity that impairs the groundwater quality in the coastal aquifer in Douala Cameroon is tidally induced.

5.1.1. Evaluation of tidal-induced groundwater salinization from temporal salinity and temperature variations

Tidal intrusion of estuarine water into coastal aquifers that causes salinization occurs during high tides when estuarine water (1) has a higher head which induces flow of saline water into the groundwater aquifer where groundwater has a lower head and/or (2) overtops the topography and cause flooding and seepage of saline estuarine water into the groundwater aquifer (Mao et al., 2001; Mao et al., 2006; Lenkopane et al., 2009; Tularam and Singh, 2009; Shalem et al., 2019).

Tidal intrusion of saline water into coastal aquifers will induce concomitant or rhythmic cyclicity in groundwater levels and chemistry (Wang and Tsay, 2001; Lenkopane et al., 2009; Mitra et al., 2011). To track groundwater salinization due to tidal forcing, we compare the time series variations in water level (tides), salinity and temperature in estuarine water from Dr. Creek and the adjacent groundwater aquifer in Youpwe (Fig. 2). Similar to water in Dr. Creek, groundwater level increases and decreases coincide with the semidiurnal mixed tidal forcing in the Wouri Estuary (Fig. 2a vs. 2b). The variations in salinity in Dr. Creek (Fig. 2d) corresponds to tidal cycles (Fig. 2b), while the temporal salinity in groundwater (Fig. 2c) does not correspond to tidal cycles (Fig. 2a). This disparity between the behavior of tides and the temporal salinity in groundwater allows us to argue that shallow coastal groundwater at a distance of 120 m from the estuary is not affected by tide-induced salinization. Because tides are rhythmic, over time, the effect of tidal salinization should show a rhythmic behavior in the nearshore groundwater (Shalem et al., 2019).

It is clear that the temperature in Dr. Creek (Fig. 2f) only slightly mimics the sinusoidal patterns of tides (Fig 2b), with slight temperature decreases at high tide. On the other hand, groundwater temperature (Fig. 2e) mimics neither the behavior of the air temperature (Fig. 2g) nor Dr. Creek water temperature (Fig. 2f). The nearly constant temperature (temporal difference of 0.1 ° C; Table S1) in groundwater indicates that groundwater temperature is not affected by mixing with warmer surface water (temperature range 1.4 ° C; Table S1) from Dr. Creek. We posit that groundwater level fluctuations (Fig. 2a) which coincided with tidal cycles in Dr. Creek (Fig. 2b) is due to the effects of tidal energy, and not from the intrusion or infiltration of estuarine water into the groundwater (e.g., Wang and Tsay, 2001).

5.1.2. Cause of groundwater salinization from isotopic and hydrochemical tracers

The $\delta^{18}\text{O}$ and δD and salinity are conservative, and are therefore good indicators of salinization by evapoconcentration and tracers of mixing between fresh groundwater and saline estuarine water (Kim et al., 2003; Bouchaou et al., 2008; Mongelli et al., 2013; Cary et al., 2015; Bahir et al., 2018; Carreira et al., 2018; Amiri et al., 2016). The $\delta^{18}\text{O}$ and δD of water from Dr. Creek, groundwater in the Youpwe neighborhood and the Wouri River water (Wirmvem et al., 2017a) co-vary (Fig. 3a), cluster along the LMWL of Douala (Wirmvem et al., 2017b), and plot above the GMWL (Craig, 1961). The clustering of groundwater samples along the LMWL indicates meteoric origin of the groundwater (Ketchemen-Tandia et al., 2007; Wirmvem et al., 2017b).

Evidence for evapoconcentration in increasing solute concentrations, and therefore responsible for groundwater salinization by evapoconcentration can be assessed from the relationship between salinity and d-excess (Fig. 3b). Lower d-excess values indicate greater extent of evaporation (e.g., Dansgaard, 1964) which can be linked to salinization (e.g., Fröhlich et al., 2002; Huang and Pang, 2012; Krishan et al., 2020). The d-excess in groundwater, estuarine water in Dr. Creek and the Wouri River are between 11 and 15 and are in the range of 10 to 17 observed in Douala precipitation (Wirmvem et al., 2017b) indicating a meteoric origin. The wide range in d-excess with a relatively limited variation in salinity is an indication of minimal effect of evaporation, consistent with unevaporated rain recharge of groundwater, short residence time of water in the creek and river and/or low evaporation rates from high humidity (Fantong et al., 2016).

We can use the relationship between salinity and $\delta^{18}\text{O}$ (Fig. 4a) to assess saline estuarine water intrusion and mixing with groundwater (e.g., Gonfiantini and Araguás, 1988; Bahir et al.,

2018). In Figure 4a, we show a model line of freshwater (salinity = 0; $\delta^{18}\text{O} = -2.9\text{‰}$) and seawater (salinity = 35; $\delta^{18}\text{O} = 0\text{‰}$) end member mixing. We also show a line that depicts dissolution and leaching in freshwater with a $\delta^{18}\text{O} = -2.9\text{‰}$. The arrow showing increasing salinity indicates that the dissolution and leaching processes do not modify the $\delta^{18}\text{O}$ composition of water (Gonfiantini and Araguás, 1988). Both model lines have a $\delta^{18}\text{O}$ end member value of -2.9‰ , which represents the average $\delta^{18}\text{O}$ composition of groundwater in Douala. On the salinity vs. $\delta^{18}\text{O}$ plot, the data points from Dr. Creek, the Wouri River (Wirmvem et al., 2017a), coastal groundwater from Youpwe and the rest of the Douala coastal aquifer (Fantong et al., 2016) do not fall along the freshwater seawater mixing line (Fig. 4a), indicating no seawater intrusion. The groundwater samples that were analyzed from Well 1 were collected between low and high tide regimes (Fig. 2a), and yet, the data forms a cluster in the salinity vs. $\delta^{18}\text{O}$ plot. This similarity of groundwater $\delta^{18}\text{O}$ and salinity despite variations in collection times relative to tides indicates no systematic increases or decreases from tide-induced intrusion of estuarine water into groundwater. Similarly, estuarine water, Wouri River and groundwater are not affected by dissolution and/or leaching as the samples do not plot in the direction of the model line indicating dissolution and leaching (Fig. 4a).

The lack of seawater intrusion into the coastal aquifer is further supported by the $\text{Mg}^{2+}/\text{Ca}^{2+}$ vs. the electrical conductivity relationship (Fig. 4b). The $\text{Mg}^{2+}/\text{Ca}^{2+}$ ratios should increase in groundwater with increased proportion of seawater intrusion where higher ratios (>1) depict seawater intrusion, and lower ratios (<1) indicate no seawater intrusion (Pulido-Leboeuf et al., 2003; Salem and Osman, 2017). The $\text{Mg}^{2+}/\text{Ca}^{2+}$ of estuarine water from Dr. Creek range from 6-8 and are much higher compared to groundwater (Fig. 4b). Groundwater from the coastal aquifer in Youpwe, as well as from the rest of the Douala coastal aquifer (Takem et al., 2010,

2015; Fantong et al., 2016) show low Mg^{2+}/Ca^{2+} ratios of <1 , indicating no tide-induced salinization of groundwater (Fig. 4b).

The finding of no tide-induced groundwater salinization is inconsistent with the previous suggestion of possible seawater source for the high chloride concentrations observed in about 30% of the shallow groundwater in the Douala aquifer located 0.44-3 km from the shoreline by Fantong et al. (2016). Fantong et al. (2016) argued that during high tides, seawater that flows through the Dibamba River and Wouri River cause bank overflow and flooding. The saline water from bank overflow and flooding recharges the unconfined shallow aquifer causing the high chloride concentrations observed. Although estuarine water overflow and flooding is able to salinize groundwater in close proximity to the rivers, Takem (2012) established from Na^+/Cl^- ratios in groundwater in the Douala aquifer that less than 20% of samples had Na/Cl ratio lower than that of seawater, and argued against seawater intrusion as the primary source of the chloride. The widely distributed high chloride concentrations in groundwater of the Douala coastal aquifer was then attributed to sea spray aerosol and atmospheric sea salt deposition, as an alternative explanation to seawater intrusion (Takem et al., 2015). The salts deposited on the surface from the sea spray and atmospheric deposition are flushed by rain recharge into groundwater (Takem et al., 2015).

During our study, we observed tidal flooding at the surface adjacent to the groundwater sampling location, which can potentially induce saline water flow and/or recharge into the shallow groundwater. Moreover, the pumping of Well 1 at $4.7\text{ m}^3/\text{s}$ should have further promoted lower groundwater head (which was decreased by 1.5 m) and facilitated salinization from inflow of estuarine water with a higher head. However, the Mg^{2+}/Ca^{2+} ratios of <1 in groundwater is consistent with results from Takem (2012) that indicated that the high chloride

concentrations in the Douala coastal aquifer are not from modern seawater from the Gulf of Guinea. Our results do not support the Takem et al. (2015) idea of a sea spray origin for the high chloride concentrations in groundwater either. We make this argument because the ratio of the ions (e.g., $\text{Mg}^{2+}/\text{Ca}^{2+}$) in sea spray or sea salt deposition will be the same as in the seawater. If the sea spray or salt deposition was responsible for the high chloride concentrations in groundwater, we should observe $\text{Mg}^{2+}/\text{Ca}^{2+}$ ratios $\gg 1$, which is not the case for groundwater in Youpwe neighborhood and the rest of the Douala coastal aquifer (Fig. 4b). Wirmvem et al. (2017a) posit that the main source of chloride in groundwater in the Douala coastal aquifer is from seepage from the numerous pit latrines in the city. Although our current study is unable to test this idea of anthropogenic pollution, we do observe higher salinity values (~ 1 psu) in groundwater (Well 1 and Well 3) (Fig. 3b) which can be attributed to either aquifer source of contamination or anthropogenic pollution.

6. Conclusions

We assessed tide-induced salinization in coastal groundwater in the Youpwe neighborhood in Douala which is adjacent to the Wouri Estuary in Cameroon. We employed a combination of time series investigations of water level (tides), salinity and temperature, and assessments of major cations and $\delta^{18}\text{O}$ and δD in estuarine water and in groundwater. During our experiment, we pumped groundwater to decrease its head and to induce flow from estuarine water with higher head. Groundwater table showed tidal induced fluctuations similar to flood and tide ebb in the estuary. The salinity, $\delta^{18}\text{O}$ and δD and $\text{Mg}^{2+}/\text{Ca}^{2+}$ used as tracers showed salinity in groundwater did not originate from seawater and that there was no evidence of mixing between saline water in the estuary and groundwater. Thus, our results show that shallow groundwater

120-250 m from the estuary coast and groundwater in the broader Douala coastal aquifer in general is not affected by tidal-induced salinization. Our findings indicate that in estuarine settings where tidal salinization is not a major process controlling groundwater quality, other processes such as anthropogenic pollution and/or contamination from the aquifer formation (connate water in the aquifer units) that may affect groundwater quality should not be ignored.

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CRedit authorship contribution statement

Goabaone J. Ramatlapeng: Writing- original draft, Data Analysis, Methodology, Visualization. **Eliot A. Atekwana:** Writing-reviewing and editing, Supervision, Funding Acquisition, Conceptualization, Methodology. **Hendratta N Ali:** Writing-reviewing and editing, Funding Acquisition. **Isaac K. Njilah:** Writing-reviewing and editing. **Gustave R. N. Ndondo:** Writing-reviewing and editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Disclosure statement

No potential conflict of interest was reported by the authors

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Table Captions

Table 1: Station type, station location, stable isotopes of hydrogen (δD) and oxygen ($\delta^{18} O$), salinity, electrical conductivity (EC), Mg^{2+} , Ca^{2+} , total iron, nitrate, phosphate and ammonia measured in shallow coastal groundwater (Well 1, 2, 3) in Youpwe, Douala and in Dr. Creek in the Wouri Estuary.

Figure Captions

Figure 1: Figure 1: (a) Insert showing the map of the Wouri Estuary in Douala Cameroon, Africa and the sampling location (yellow square), (b) Google earth image showing the sampling locations along Dr. Creek in the Wouri Estuary near the Youpwe neighborhood in Douala. (c) a zoomed in insert showing groundwater sampling locations in Youpwe.

Figure 2: Temporal relative water level (a, b), salinity (c, d), and temperature (e, f) in groundwater (Well 1) in Youpwe and in Dr. Creek in the Wouri estuary and air temperature (g, h) in Youpwe.

Figure 3: Figure 3: Cross plot of (a) $\delta^{18}\text{O}$ vs. δD and (b) salinity vs. d-excess for water samples from Dr. Creek and coastal groundwater (Well 1, 2, 3) from Youpwe and Wouri River water samples (Wirmvem et al., 2017a). Also shown on the plot are the Global Meteoric Water Line (GMWL; Craig, 1961) and Local Meteoric Water Line (LMWL) of Douala (Wirmvem et al., 2017b).

Figure 4: Cross plot of (a) Salinity vs. $\delta^{18}\text{O}$ and (b) $\text{Mg}^{2+}/\text{Ca}^{2+}$ ratios vs. Electrical conductivity (EC) for samples from Dr. Creek and groundwater (Well 1, 2, 3). Also shown are groundwater samples from the Fantong et al. (2016) study and river water samples from Wouri River (Wirmvem et al., 2017ba. The freshwater-seawater mixing line and change in salinity from the dissolution and leaching process are shown. Adapted from Gonfiantini and Araguás-Araguás (1988).

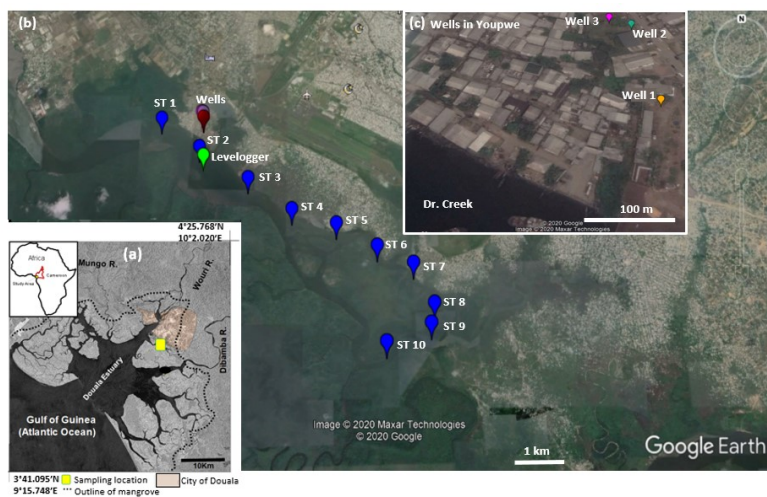


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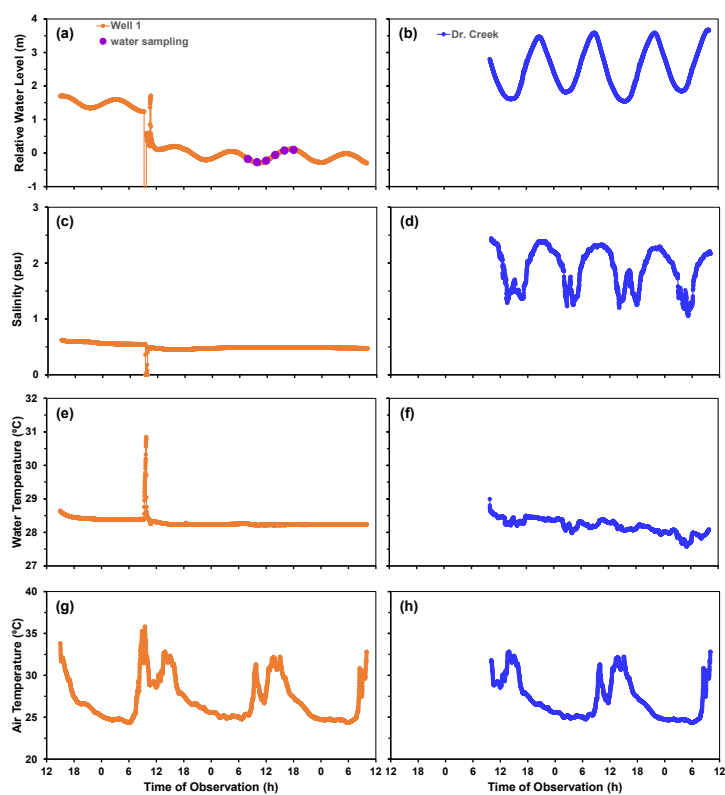


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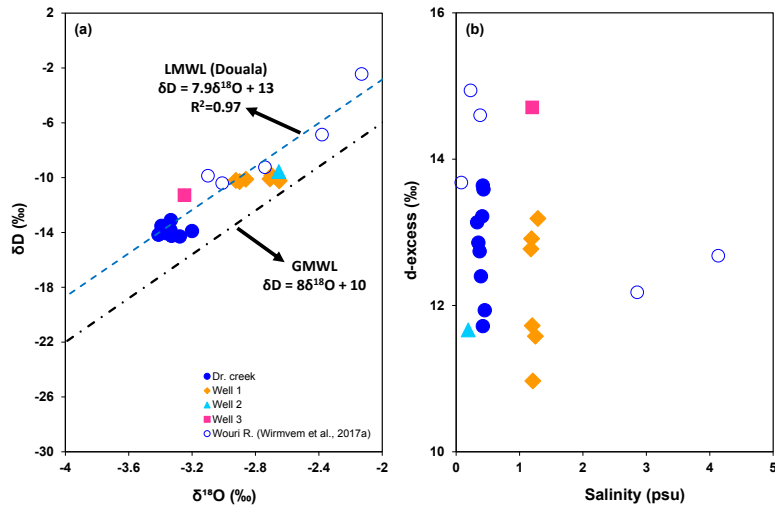


Figure 3: Cross plot of (a) $\delta^{18}O$ vs. δD and (b) salinity vs. d-excess for water samples from Dr. Creek and coastal groundwater (Well 1, 2, 3) from Youpwe and Wouri River water samples (Wirmvem et al., 2017a). Also shown on the plot are the Global Meteoric Water Line (GMWL; Craig, 1961) and Local Meteoric Water Line (LMWL) of Douala (Wirmvem et al., 2017b).

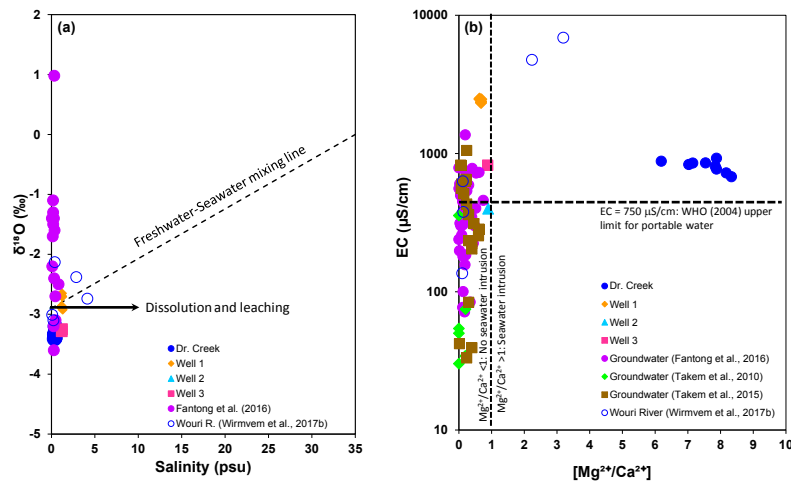


Figure 4: Cross plot of (a) Salinity vs. $\delta^{18}\text{O}$ and (b) $\text{Mg}^{2+}/\text{Ca}^{2+}$ ratios vs. Electrical conductivity (EC) for samples from Dr. Creek and groundwater (Well 1, 2, 3). Also shown are groundwater samples from the Fantong et al. (2016) study and river water samples from Wouri River (Wirmvem et al., 2017b). The freshwater-seawater mixing line and change in salinity from the dissolution and leaching process are shown. Adapted from Gonfiantini and Araguás-Araguás (1988).