## DEGRADATION DE LA QUALITE DES EAUX SOUTERRAINES (Article 1)

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Research papers

# Degradation of groundwater quality in expanding cities in West Africa. A case study of the unregulated shallow aquifer in Cotonou

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#### ABSTRACT

In Cotonou, as in many expanding West African cities, major population growth and infrastructural development has not kept up with informal settlement development onto floodable plains and marshes. The population of the slum, which makes up about 60% of the city's inhabitants, is highly disadvantaged and vulnerable to rising sea levels, flooding, sanitation and waste management issues. However, the risks associated with the use of contaminated shallow groundwater for domestic purposes are less recognised. Our study demonstrates that, in many instances, the cheaper option of the use of shallow groundwater from the coastal Quaternary aquifer for domestic purposes represents a significant risk for the poorer residents of Cotonou through the voluntary (drinking) or non-volontary (dish washing, cooking) consumption of this unmonitored and untreated water resource. In the 3 neighbourhoods surveyed, environmental tracers (major ions, Cl/Br molar ratios and stables isotopes) showed that this shallow aquifer is degraded by seawater intrusion as well as septic and sewerage contamination. In particular, the higher NOx concentrations correspond to ranges associated with sewerage and septic tank effluent pollution and the major ion concentrations and  $\delta^2$ H– $\delta^{18}$ O signatures showed that high salinity values are where groundwater mixes with saline Lake Nokoue water. The population using this resource from local wells should be made aware of seasonal changes in groundwater contamination and potential health risks associated with sewerage and septic tank contamination.

1. Introduction

With cities in developing countries expanding at unprecedented rates, ensuring clean water supplies for all inhabitants is becoming increasingly more challenging. This is particularly the case in more impoverished urban areas, where infrastructure development often lags behind population growth (Lapworth et al., 2017). In addition, there often exist two types of water supplies: firstly, there is the official water supply that is monitored, treated and comes at a cost for consumers; and then there is the second unregulated water supply that is often sourced from wells accessing shallow groundwater that is unmonitored, untreated but free. The health risks in using the unregulated groundwater resource for domestic purposes, including drinking water supplies, is high due to water quality issues in many urban areas (e.g. Ouedraogo et al., 2016; Hassane, 2010). The same factors driving the demand for water supplies, accelerated urban growth and the expansion of informal settlements, are also significant drivers of groundwater pollution (UNESCO, 2017). One of the many threats to shallow groundwater quality in cities is from sanitary wastewater and solid waste disposal (Lu et al., 2015). Uncontrolled seepage of wastewater from septic tanks and human activities as well as infiltration of urban stormwater lead to groundwater contamination (Dhanasekarapandian et al., 2016). Many parts of the world are now reporting groundwater and surface water nitrate

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pollution issues (Roy et al., 2007; Stuart et al., 2007; Zhang et al., 2014; Ogrinc et al., 2019). For example, in the Coimbatore city, India, population growth, pit latrines and septic tanks, industrial effluents, and irrigation water return flows are the main sources of groundwater contamination (Selvakumar et al., 2017). In Florida, the proximity of wells to septic tanks contributes to increasing fecal coliform, nitrate and phosphate concentrations during wet season compared with the dry season (Arnade, 1999). Many studies have identified sewerage and latrine contamination as a significant public health issue due to the resultant presence of faecal bacteria in waters such as Escherichia coli, faecal Streptococci, Salmonella and Shigella (Boukari, 1998; Odoulami and Gbesso, 2013; Yadouléton, 2015). During this study, for the first time, the seasonal variation in the contamination of waters by sewerage and latrine contaminantion is investigated using environmental tracers.

In addition, shallow groundwater resources in coastal cities are particularly vulnerable to salinity problems due to mixing with saline surface waters and seawater intrusion (Barker et al., 1998; Cary et al., 2015; Petelet-Giraud et al., 2016; Najib et al., 2017; Liu et al., 2017). For example, in Ho Chi Minh city, Vietnam, the groundwater resources are under threat due to saltwater intrusion in the shallow aquifer (Ngo et al., 2015).

In this study, we use the example from Cotonou, Benin, to identify processes resulting in nitrate and salinity contamination of a city's unmonitored and untreated shallow groundwater resource. The complexity of contamination processes in evolving urban environments makes it difficult to study the risks of shallow groundwater quality. This can be due to both natural seasonal shifts from climatic and environmental influences, and significant anthropogenic influences on the hydrogeological system. In this study, we analyse the distribution and concentration of major ions and stable isotopes ( $\delta$ 2H and  $\delta$ <sup>18</sup>O) to investigate the temporal variation in groundwater quality across 3 sites in Cotonou. These 3 neighbourhoods were selected to represent contrasting hydrogeological environments: (i) a neighbourhood (St Jean) where there is no surface inundation; (ii) a neighbourhood (Ladii) bordering Lake Nokoué which overflows towards the end of the wet season; and (iii) a neighbourhood (Agla) located in a swamp low land and is subject to inundation during both the small and large wet seasons. In doing so, we identify hydrological conditions and seasons when groundwater is at its greatest vulnerability in terms of salinization and latrine contamination. At each site, there are wells where local inhabitants have access to unmonitored and untreated shallow groundwater for domestic consumption.

#### 2. Study area

#### 2.1. Location and climate

The city of Cotonou is bordered by Lake Nokoué to the north, and the Atlantic Ocean to the south. The location of Cotonou and the average monthly interannual rainfall at Cotonou station from 1971 to 2016 are shown in Figs. 1 and 2. The topography of the city is relatively flat with an altitude varying between 0 m and 6 m (Boukari, 1998). The average annual rainfall for Cotonou is 1,300 mm (Yabi and Afouda, 2012). The city's climate is characterised by 4 seasons; main dry season (average rainfall is 25 mm from mid-November to mid-March), main wet season (average rainfall is 152 mm from mid-March to mid-July), small dry season (average rainfall is 55 mm in mid-July to mid-September), and small wet season (average rainfall is 75 mm from mid-September to mid-Novembre).

#### 2.2. Aquifer geology

Cotonou is located in the coastal sedimentary basin comprised of Quaternary (Holocene) sediments, which include facies of the littoral plain (sands) and alluvial deposits, underlain by sediments from the Mio-Pliocene (Continental Tenninal), Paleocene and Upper Cretaceous (Maliki, 1993). Previous studies by Oyédé (1991), Maliki (1993), Alidou et al. (1994) and Boukari (1998) have described in detail the different sedimentary units of this coastal sedimentary basin and the distribution are presented in Fig. 1. The shallow Quaternary sediments consist predominantly of fine to medium sands (85.5%), silts (5.5%) and clays (9%) (Oyédé, 1991). The low clay content in these sediments results in the high permeability of the shallow sandy soils (Maliki, 1993), and therefore a high vulnerability of the aquifers to the transfer of surficial pollutants. The clays in Quaternary sediments are constituted by smectite and a kaolinite content, which increases with depth. The traditional domestic wells were sampled in this study so that we could analyse the water consumed and used by local residents. However, due to the intense pumping of these wells, the fluctuations in hydraulic head are heavily influenced by pumping, therefore we were unable to use the hydraulic head data to infer subsurface flow directions. There has however been previous work that has addressed this question, the work of Maliki (1993) and Boukari (1998) focused on the piezometry of the Cotonou water table during the flood and low water periods, and the interactions with the Lake. It appears from these studies that during the flood period, there is a flow direction of groundwater from the center of Cotonou (piezometric dome) to outlets such as Lake Nokoué, the lagoon, the Atlantic Ocean and swamps. In comparison, during the low-water period it was observed that water from the Lake Nokoué flows into the groundwater aquifer.

#### 2.3. Sewerage and waste

The city of Cotonou has about 166,433 households and the population evaluated at 679 012 inhabitants in 2013 (INSAE, 2015) and is subject to increasing urbanization. The poorer dwellings are located in the neighborhoods along the coastline, along the edges of the Cotonou Channel and Lake Nokoué, and in the swamp areas. In these neighbourhoods, solid and liquid wastes are released into the immediate environment without treatment as illustrated in the Fig. 1(c). In addition, they are also scattered in various places of the city where garbage heaps have formed. 78.5% of household wastewater and 33% of solid wastes are ejected in gardens, streets, gutters, unused wells and empty blocks (INSAE, 2016). These poorer neighborhoods are also without adequate sanitation systems. In Cotonou 64.9% of households use latrines that are reportedly leak-proof, whereas 13.5% adopt unsafe and non-hygienic practices such as stilt latrines and open defecation. Only 20.8% of the population use septic tanks, which are generally evacuated by drainage structures (INSAE, 2016). Since there are few market gardens in this heavily urbanised city, the potential sources of nitrate in the study area are predominantly from wastewater, solid waste and septic tanks. Therefore, the risk of pollutant transfers to the shallow aquifers is high in the poorer neighbourhoods because they are exposed to both (i) frequent episodes of inundation and (ii) high levels of pollutants from poorly constructed landfills and latrines.

#### 2.4. Groundwater quality and use

Several studies have evaluated the impact of human activities on the quality of the shallow groundwater in the Cotonou region and have reported relatively high levels of nitrates (e.g. up to 100 mg/L; Maliki, 1993; Boukari, 1998; Odoulami and Gbesso, 2013; Totin et al., 2013). In addition, the vulnerability of this groundwater resource is also related to its proximity to the Atlantic Ocean and the saltwater or brackish lakes (Boukari, 1998; Totin et al., 2013). Data from previous studies have shown an increase in salinity levels in groundwater near Lake Nokoué. The salinity of the Lake Nokoué also exhibits a marked seasonal cycle (0 psu during the small wet season in October and 25–30 psu at the end of the dry season) (Stephen et al., 2010; McInnis et al., 2013; Totin et al., 2013). The coastal groundwater system in Cotonou has relatively high chloride (23.6 to 160 mg/L) and sulphate (6.4 to 25.7 mg/L) concentrations (Alassane et al., 2015; Nlend et al., 2018).



Fig. 1. (a) Location of Cotonou and sampling sites in the three neighbourhoods; (b) Sedimentary unit cross section (A-B) of marginal-littoral area of Benin (modified from Boukari, 1998); and (c) Environmental situation at Agla (A9 site).

Lake Nokoué and the Atlantic Ocean are therefore a major concern in terms of the source of salinization of groundwater resources in the coastal zone (Alassane et al., 2015).

Due to the groundwater pollution problems, shallow groundwater is excluded from the official supply of drinking water to the city of Cotonou. However, the low rate of services to the public water supply network in the neighborhoods of *peri*-urban areas (51%) leads the population to use the groundwater from the shallow Quaternary aquifer for their various domestic uses (INSAE, 2016). This is despite the fact that the Quaternary aquifer is not part of the official monitoring services of the National Water Company of Benin, the only legal water distribution structure.

Groundwater from the shallow Quaternary aquifer, is accessed by

the residents of Cotonou via large-diameter wells installed on private household property. These large-diameter wells are shallow and are designed to pump groundwater from the water table surface so as not to pump deeper saline groundwater (Maliki, 1993). The Quaternary aquifer sands have a high permeability (in the order of  $10^{-2}$  to  $10^{-4}$  m/s) and locally contains fresh to brackish groundwater whose exploitation is related to the position of the lake water and seawater instrusions, and to the replenishment of freshwater lenses.

In Cotonou, 81% of the neighbourhoods have wells, and whilst 9% have not any water supply (no wells nor stand pipe for drinking water supply) for domestic purposes, they sometimes buy water for drinking purposes from neighbours (Hounkpe et al., 2014). Furthermore, the groundwater from the large-diameter private wells is used in



Fig. 2. Average monthly interannual rainfall at Cotonou station from 1971 to 2016 (ASECNA station).

households for drinking, cooking, laundry, bathing and washing dishes (Yadouléton, 2015; EAA, 2018).

In this study, we expand on previous work to elucidate the seasonality of hydrogeological processes resulting in the contamination of groundwater from latrine waste (notably nitrate and nitrite concentrations, herein referred to as NOx) and from salinization.

#### 3. Methods

Three neighbourhoods in Cotonou were selected for sampling based on differences in their hydrogeological environments: (i) St Jean, where there is no long-term surface inundation and the heavy rainfall infiltrates the soils or leaves the site via overland flow; (ii) Ladji, which is located at the shore of Lake Nokoué and is inundated towards the small wet season; and (iii) Agla, which is located in a flood plain and is inundated early during both the small and large wet seasons. The locations of these three neighbourhoods and sampling sites are presented in Fig. 1. In each neighbourhood, groundwater wells were selected for sampling and, where/when possible, surface water samples (permanent and temporary pools, as well as Lake Nokoué) were also included. The depths to the water table and the electrical conductivity (EC) of groundwater in the Quaternary aquifer were monitored monthly at 9 wells and 3 piezometers from each of the 3 neighbourhoods from June 2017 to June 2018. The wells are separated by an average distance of  $\sim 200$  m within the same neighbourhood probe.

The hydrochemistry of waters were analysed 6 times between November 2016 to June 2018 during the months of February (main dry season), June (main wet season) and November (small wet season), resulting in a total of 127 groundwater, and 60 surface water samples. In addition, for the period from June 2017 to June 2018, 13 rainfall samples were analysed to determine water stable isotopes ( $\delta^2$ H and  $\delta^{18}$ O). Lastly, 2 water samples from atlantic ocean (AO) were collected of the periods from January 2019 (main dry season). Groundwater was sampled from the Quaternary aquifer via the large-diameter wells. Groundwater and surface water temperature, EC, and dissolved oxygen (DO) were measured *in-situ* using a WTW 3430i portable digital multiparameter.

Water samples were collected for the analyses of cations (filtered at 0.45  $\mu$ m and acidified with HNO<sub>3</sub>), anions and stable isotopes, that were conducted at the Laboratory of Hydrogeology of the University of Avignon, France. Major ions were analysed using ion chromatography (Dionex; ICS1100 and autosampler AS-AP). Ion analysis uncertainty is in the order of 3%, and all ionic balances were  $\leq$  5%. The alkalinity was measured using a HACH digital titrator, and stable isotopes were

analysed using a Picarro Analyser L 2130-I. For the stable isotopes of waters, the error is  $\pm$  0.1‰ for  $\delta^{18}O$  and  $\pm$  1‰  $\delta^{2}H$ . The results are presented in Tables 1 and 2. The mixing ratio between lake and shallow groundwater is calculated by using stable isotopes values according to Paran et al. (2015):

$$\delta^{18}O_m = f \cdot \delta^{18}O_L + (1 - f) \cdot \delta^{18}O_{GW}$$

The same equation is used with  $\delta^2 H$  to compute the  $\delta^2 H_m$  mixing ratio.  $O_m$  is water sample isotope composition where groundwater (GW) and Lake (L) mixing is supposed.  $O_L$  is Lake isotope composition.  $O_{GW}$  is groundwater isotope composition and F is mixing fraction.

One questionnaire in each sampling site was administered, 10 per study area (Ladji, Agla and St. John). A total of thirty (30) questionnaires were administered through an interview with households in each study area. Main information such as the the source of drinking water supply, groundwater use, waste and wastewater management are mentioned in the questionnaire (supplementary data).

#### 4. Results

#### 4.1. Seasonal variations of the water table and groundwater EC

The monthly monitoring of the depth to water table and EC over a one-year period highlights the variations in seasonal fluctuations of the aquifers between the sites. Typical of unconfined shallow aquifers, the Quaternary aquifer shows marked spatial and seasonal variations and is in phase with the monthly rainfall. The seasonal variations of the water table and groundwater EC in each of the neighbourhoods are illustrated in Figs. 3 and 5.

Between the neighbourhoods, St Jean has a maximum depth to water table (1.5 m at site J5) corresponding with the dry season (February) that is relatively deep compared with the depths of 1.4 m and 1.0 m observed in Ladji (site L7) and Agla (site A4), respectively (Fig. 5). In addition, at 2 of the 4 sites in Agla, the negative depth values indicate that the water table is above the land surface during the wet season (thus contributing to the floods), whereas groundwater at St Jean and Ladji remains below the surface. The seasonal fluctuation amplitudes also vary. In St Jean, the maximum change between wet and dry season water table depth is of 1.2 m. Here the water table fluctuations also vary between wells. Well J5 strongly reacts to the increased rainfall in October (small wet season) whereas the deepening of the water table in wells J1 and J2 remain relatively weak (Fig. 5). These relatively low water table values can be attributed to the use of wells by households (Boukari, 1998; Kadjangaba et al., 2018). Similar to St Jean,

Table 1	
Chemical parameters and isotopic values in shallow groundwater.	

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ID	Sampling month	Origin	Depth	T°C	pН	DO	EC	$NO_{\rm x}$	$\mathrm{HCO_3}^-$	$Cl^{-}$	$\mathrm{NO_2}^-$	$\mathrm{Br}^-$	$NO_3^-$	$PO_4^{}$	$\mathrm{SO_4}^{}$	Na <sup>+</sup>	$\mathrm{NH_4}^+$	$K^+$	$Mg^{++}$	Ca <sup>++</sup>	DOC	d <sup>18</sup> O	d <sup>2</sup> H
Units			m			mg/L	µS∕cm	meq/L	mg/L													‰	
J1	Nov-16	well		29.50	6.97		590	0.67	221.49	27.59	DBL	DBL	37.68	8.69	3.90	47.20	DBL	19.57	5.42	58.13		-3.07	-14.04
	Mar-17	well		29.70	7.24	2.59	998	1.84	244.00	74.90	0.80	0.97	66.12	8.26	85.49	86.41	0.25	29.43	9.19	93.52		-2.85	-12.48
	Jun-17	well	0.80	28.20	7.45	3.60	1002		318.12	77.18	DBL	DBL	DBL	6.99	115.78	94.37	DBL	3.86	9.99	89.42	10.85	-2.94	-12.61
	Oct-17	well	0.90	27.90	7.57	3.93	799	0.44	314.76	28.52	7.13	DBL	17.67	5.68	4.87	44.68	DBL	22.27	9.69	92.45	5.89	-3.89	-21.22
	Feb-18	well	1.23	29.10	7.16	2.35	1285	1.18	384.30	85.78	DBL	DBL	73.45	8.34	8.62	14.51	13.65	35.65	8.76	79.28	6.80	-3.36	-16.10
	Jun-18	well	0.88	29.20	7.40	3.24	731	1.16	152.50	57.74	1.19	DBL	7.37	4.97	54.63	68.32	0.58	19.86	5.16	53.95		-3.22	-13.37
J2	Nov-16	well		29.00	6.92		502	0.76	99.56	34.52	0.12	0.28	46.95	6.93	36.23	45.47	0.28	12.67	2.80	44.77		-2.81	-11.64
	Mar-17	well		29.90	6.93	2.42	552	0.70	113.46	42.75	0.17	0.65	37.70	7.13	31.75	41.98	0.30	18.60	3.24	49.58		-3.35	-15.40
	Jun-17	well	0.59	28.60	7.04	3.13	347		128.10	25.67	DBL	DBL	DBL	5.27	17.90	31.66	DBL	7.32	1.94	33.24	8.30	-3.65	-16.31
	Oct-17	well	0.77	28.60	7.36	5.13	297		121.60	15.94	DBL	DBL	DBL	6.52	13.00	22.97	DBL	7.66	1.56	28.22	6.31	-4.17	-21.87
	Feb-18	well	1.04	29.70	6.88	2.56	586	0.50	119.56	52.64	0.43	DBL	3.49	6.40	49.37	37.97	0.62	13.28	3.14	46.95	5.55	-3.39	-15.90
	Jun-18	well	0.75	29.03	7.06	3.39	341	0.24	86.93	2.19	DBL	DBL	12.43	3.77	26.87	22.87	0.22	8.45	1.79	31.72		-3.54	-15.48
J3	Nov-16	well		29.70	7.02		658	0.74	125.58	46.97	0.53	0.61	45.39	9.82	54.88	62.97	1.64	17.99	3.46	53.96		-3.01	-13.16
	Mar-17	well		29.20	7.15	4.61	891	1.59	191.54	81.73	0.31	0.13	98.13	6.28	53.17	86.30	2.25	21.71	4.96	68.47		-3.02	-12.99
	Jun-17	well		27.60	7.35	7.85	822	0.63	254.68	75.52	DBL	0.18	0.39	5.25	52.61	84.25	DBL	22.82	4.27	63.45	1.17	-3.19	-13.76
	Oct-17	well		29.00	5.91	3.55	630		29.84	56.65	DBL	DBL	DBL	5.19	4.84	51.28	DBL	13.35	4.54	6.62	4.53	-3.56	-16.94
	Feb-18	well		29.20	7.00	3.61	791	1.18	156.16	74.76	0.29	DBL	73.98	4.26	61.19	62.85	0.33	15.11	6.18	79.89	5.37	-3.19	-13.90
	Jun-18	well		29.40	6.90	3.90	792	1.12	158.60	7.88	DBL	DBL	69.65	3.83	41.29	66.47	0.38	14.90	5.79	63.11		-3.38	-15.19
J4	Nov-16	well		29.80	7.11		500	0.22	18.45	27.63	0.77	0.45	13.48	2.47	36.46	37.84	0.31	17.42	3.77	51.35		-2.37	-8.98
	Mar-17	well		29.90	7.15	4.95	572	0.31	195.20	33.68	0.11	0.79	18.69	2.36	37.86	41.44	0.12	13.45	6.22	57.91		-2.58	-9.93
	Jun-17	well		27.90	7.63	3.98	368	0.33	132.37	2.34	0.15	DBL	DBL	1.99	23.22	29.65	DBL	15.53	2.62	31.66	14.96	-3.23	-14.08
	Oct-17	well		28.70	8.06	5.35	342		81.74	29.18	DBL	DBL	DBL	2.45	24.79	25.31	DBL	7.95	2.97	29.79	4.24		
	Feb-18	well		29.20	7.15	3.33	434	0.22	96.38	47.54	DBL	DBL	13.85	2.27	27.26	28.87	0.81	3.52	3.64	36.94	3.79	-3.07	-13.61
	Jun-18	well		28.80	7.41	4.08	379	0.33	122.00	25.24	DBL	DBL	18.82	2.37	28.23	24.75	0.48	16.77	3.00	36.36		-3.52	-15.23
J5	Nov-16	well		29.70	6.83		642	0.45	28.68	38.17	DBL	0.82	0.28	0.82	3.38	46.38	DBL	43.42	6.31	56.38		-3.22	-14.19
	Mar-17	well		29.70	6.89	3.25	715	0.69	17.80	57.97	0.43	0.96	42.66	0.21	59.64	72.32	0.34	37.83	3.69	49.65		-3.06	-13.09
	Jun-17	well	1.05	28.50	7.03	1.96	798	0.48	193.22	79.80	0.22	DBL	DBL	DBL	89.97	72.19	0.58	35.18	6.18	65.25	11.57	-3.17	-13.43
	Oct-17	well	0.43	27.10	7.03	1.31	630		272.60	37.12	DBL	0.19	DBL	DBL	26.37	43.66	DBL	19.78	5.82	65.35	4.93	-3.61	-17.56
	Feb-18	well	1.53	28.60	6.96	2.34	558	0.46	164.70	38.37	DBL	DBL	2.89	DBL	44.12	34.22	0.50	16.46	3.98	46.12	8.59	-2.89	-12.01
	Jun-18	well	0.66	27.80	6.97	2.03	639	0.25	265.35	35.12	DBL	DBL	1.53	DBL	31.40	44.85	0.62	19.72	5.49	62.37		-3.46	-15.23
J6	Nov-16	well		29.10	6.76		548	1.15	66.49	41.99	DBL	0.49	71.59	5.27	51.16	47.46	0.65	21.98	3.87	4.27		-3.24	-15.12
	Mar-17	well		29.40	6.99	3.60	1326	5.67	85.40	111.13	1.95	0.18	311.33	2.68	91.61	115.00	0.73	37.66	13.46	17.85		-2.85	-11.68
	Jun-17	well		29.20	7.06	5.12	1129	3.80	154.79	99.84	54.79	DBL	157.26	2.57	87.74	99.79	0.39	4.98	1.37	87.46	16.65	-2.60	-10.19
	Oct-17	well		28.50	6.99	4.40	637	1.94	54.90	44.28	2.28	DBL	117.19	2.33	53.18	46.00	DBL	21.69	4.32	48.44	8.31	-3.28	-15.11
	Feb-18	well		29.00	6.97	4.35	961	3.37	439.20	51.14	0.17	DBL	28.87	2.62	47.94	93.79	0.16	29.53	15.13	114.74	8.63	-3.32	-15.88
	Jun-18	well		29.00	7.17	5.96	346	0.43	54.90	29.35	DBL	DBL	24.95	4.54	2.49	26.22	0.27	11.87	2.27	24.73		-3.23	-13.82
J7	Nov-16	well		30.10	6.87		431	0.74	93.25	26.42	0.19	0.40	45.76	3.19	27.42	34.79	0.39	13.95	2.89	36.26		-2.85	-12.03
	Mar-17	well		29.90	7.05	5.12	695	0.87	184.22	44.50	0.22	0.11	53.78	0.79	44.46	53.47	0.18	2.82	4.77	55.50		-3.08	-12.98
	Jun-17	well		30.20	7.00	3.78	742	0.56	22.36	61.44	0.26	DBL	DBL	1.14	68.21	63.46	DBL	22.43	5.45	63.31	6.80	-2.61	-9.98
	Oct-17	well		28.90	7.14	4.20	652	1.29	16.14	38.98	13.15	DBL	57.27	0.73	59.96	4.94	DBL	15.58	5.28	62.60	4.59	-3.13	-14.66
	Feb-18	well		29.00	6.90	4.19	647	1.55	113.46	3.99	0.49	DBL	95.59	1.18	59.66	38.17	0.23	24.95	4.69	48.66	7.08	-3.04	-13.22
	Jun-18	well		29.40	7.08	4.19	616	0.66	14.30	37.00	DBL	DBL	4.89	1.87	56.59	47.58	0.24	31.95	4.20	43.35		-3.38	-14.29
J8	Nov-16	well		29.40	6.99		701	2.21	126.41	4.12	0.58	0.66	136.47	4.61	32.89	38.52	0.32	18.17	5.39	78.67		-3.35	-15.92
	Mar-17	well		29.70	6.78	3.62	885	2.58	115.90	72.18	0.48	0.16	159.26	5.87	36.16	74.78	2.78	29.87	4.94	61.28		-2.77	-11.16
	Jun-17	well		29.20	7.27	4.67	894	2.56	128.86	79.23	93.70	DBL	32.54	4.43	54.90	8.34	0.71	25.57	5.65	72.48	5.82	-2.92	-11.84
	Oct-17	well		27.80	7.49	4.75	564	0.99	128.10	22.92	45.57	DBL	DBL	5.67	3.47	36.54	DBL	19.17	3.18	53.58	5.54	-3.64	-17.99
	Feb-18	well		28.50	6.91	3.30	840	2.60	118.34	56.71	0.46	DBL	16.63	6.20	6.15	59.92	0.32	25.37	4.63	59.75	4.09	-3.12	-13.91
	Jun-18	well		28.40	7.65	4.56	377	0.62	128.10	14.14	DBL	DBL	38.59	4.18	18.97	19.89	0.46	1.39	2.17	38.72		-3.55	-13.45
J9	Nov-16	well		29.50	7.19		448	0.34	213.58	25.43	0.23	0.47	0.18	DBL	16.48	18.58	4.60	2.88	3.11	49.79		-2.96	-12.98
	Mar-17	well		30.10	6.98	4.82	461	0.88	82.96	42.48	0.69	0.19	54.42	DBL	21.57	36.85	0.33	11.14	2.94	41.27		-2.64	-9.55
	Jun-17	well		29.10	7.20	4.06	437	0.49	126.58	24.64	8.35	DBL	19.20	DBL	26.35	29.73	0.26	1.53	3.20	46.72	8.89	-3.06	-12.08
	Oct-17	well		28.10	7.21	4.04	323	0.46	97.60	27.93	1.56	DBL	0.42	DBL	11.57	17.19	0.58	5.12	2.22	36.84	0.41	-3.44	-15.80
	Feb-18	well		28.60	7.00	4.63	242	0.13	56.12	28.38	DBL	DBL	8.23	DBL	11.53	14.34	0.60	7.64	1.82	26.23	4.58	-3.74	-18.62

(continued on next page)

Table 1 (continued)

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ID	Sampling month	Origin	Depth	T°C	pН	DO	EC	NO <sub>x</sub>	$HCO_3^-$	C1-	$NO_2^-$	$\mathrm{Br}^-$	$NO_3^-$	PO4 <sup></sup>	SO4 <sup></sup>	Na <sup>+</sup>	$\mathrm{NH_4}^+$	$K^+$	Mg <sup>++</sup>	Ca <sup>++</sup>	DOC	d <sup>18</sup> O	d <sup>2</sup> H
	Jun-18	well		28.60	7.04	3.67	251	0.15	122.00	14.44	DBL	DBL	9.45	DBL	15.59	11.91	0.43	5.34	1.62	38.59		-3.43	-13.35
J10	Nov-16	well		29.20	6.95		572	0.75	13.63	41.52	0.32	0.49	43.32	2.64	38.84	56.63	0.45	26.72	2.86	42.15		-3.44	-16.28
	Mar-17	well		28.50	6.89	2.37	615	0.39	173.24	42.31	0.86	0.96	23.27	DBL	48.34	42.65	1.74	37.39	4.64	52.68		-3.17	-14.39
	Jun-17	well		29.50	7.13	3.43	770	1.12	163.18	65.67	DBL	DBL	69.32	DBL	61.25	72.53	1.46	33.84	4.70	57.83	11.67	-3.17	-13.29
	Oct-17	well		29.30	8.03	4.94	481		98.82	39.22	DBL	DBL	DBL	2.51	47.65	28.13	DBL	13.14	3.82	47.88	3.81	-3.17	-14.61
	Feb-18	well		29.60	6.90	2.77	943	0.99	229.36	8.94	0.56	DBL	6.44	0.52	47.17	6.59	14.36	28.62	5.34	56.68	7.19	-3.14	-13.91
	Jun-18	well		30.50	6.75	0.73	878	0.90	25.10	65.43	DBL	DBL	55.54	1.52	47.69	66.77	5.41	27.62	6.57	65.57		-3.28	-14.91
J11	Feb-18	Piezometer	1.40	31.30	7.21		497	0.89	92.72	57.13	DBL	0.60	55.30	4.48	27.92	42.63	2.29	11.97	4.67	46.88	13.50	-3.09	-15.03
	Jun-18	Piezometer	0.88	29.40	7.33	6.79	444	0.65	141.52	13.61	DBL	DBL	4.26	6.47	22.82	13.44	0.15	12.46	4.81	49.92		-3.73	-17.66
A2	Nov-16	well		28.60	6.60		1164	0.72	432.95	118.77	DBL	0.38	0.44	DBL	33.32	15.85	0.25	35.77	1.85	8.73		-1.81	-5.39
	Mar-17	well		29.90	6.28	3.32	990	0.87	13.70	123.64	0.39	0.22	53.39	DBL	136.56	125.35	0.23	28.66	7.45	49.37		-2.72	-10.94
	Jun-17	well	-0.01	27.30	7.21	2.42	1273		435.85	16.53	DBL	0.35	DBL	DBL	74.85	147.93	DBL	35.17	17.46	78.28	22.27	-2.32	-9.17
	Oct-17	well	0.04	26.50	7.02	2.33	1150		441.64	14.00	DBL	0.53	DBL	DBL	1.45	111.34	DBL	29.87	15.27	81.80	25.62	-2.02	-7.51
	Feb-18	well	0.69	28.30	6.96	2.83	733	0.83	22.52	97.21	DBL	0.40	0.51	DBL	32.23	86.88	0.32	15.55	14.75	41.54	14.65	-2.90	-12.08
	Jun-18	well	0.04	28.90	7.02	1.61	959		292.80	111.26	DBL	DBL	DBL	DBL	3.56	93.69	DBL	24.14	1.24	68.46		-2.75	-11.56
A4	Nov-16	well		29.60	6.02		326	0.27	31.95	17.78	0.35	0.12	1.63	DBL	78.71	29.22	0.16	6.91	3.73	16.65		-3.22	-14.01
	Mar-17	well		30.90	5.85	2.95	269	0.39	27.00	27.70	0.57	0.17	2.32	DBL	44.29	29.25	0.38	6.53	3.67	13.12		-3.12	-14.05
	Jun-17	well	0.47	30.70	6.01	5.34	353	0.43	37.82	56.98	DBL	0.23	2.52	DBL	38.15	32.98	0.54	8.24	5.30	2.99	9.86	-2.96	-12.84
	Oct-17	well	0.29	28.00	5.95	4.53	497	0.18	54.90	69.62	DBL	0.35	1.95	DBL	61.86	46.93	DBL	1.46	6.77	27.46	8.79	-2.68	-10.44
	Feb-18	well	0.99	29.50	5.84	3.97	548	0.42	36.60	86.61	DBL	0.46	25.86	DBL	5.46	51.32	0.76	1.49	7.16	2.14	10.38	-2.89	-12.28
	Jun-18	well	0.25	29.00	5.76	3.32	666	0.66	36.60	97.38	DBL	DBL	37.26	DBL	59.26	6.43	0.10	12.96	1.98	32.71		-2.81	-11.54
A5	Nov-16	well		30.70	6.00		1037	0.27	13.62	158.50	DBL	0.25	16.97	DBL	135.17	131.15	0.90	17.93	6.68	37.45		-2.77	-11.89
	Oct-17	well		27.20	7.01	2.75	500		15.60	5.92	DBL	0.13	DBL	DBL	19.18	51.31	DBL	9.32	6.26	33.27	10.06	-3.52	-17.61
	Feb-18	well		29.00	6.55	2.39	652	0.39	154.94	6.87	0.20	0.12	23.85	DBL	58.67	67.19	2.58	13.73	9.77	47.38	8.85	-3.18	-14.14
	Jun-18	well		29.20	6.74	4.33	652	0.17	155.55	48.84	DBL	DBL	1.53	DBL	52.19	52.72	0.12	11.19	8.15	4.83		-3.46	-16.07
A6	Nov-16	well		30.30	6.16		983	0.86	155.55	1.56	1.67	0.17	51.84	1.38	125.27	113.75	DBL	22.17	5.98	45.76		-2.99	-12.98
	Jun-17	well		29.00	7.12	8.45	1338	0.90	245.53	175.75	8.61	0.17	38.63	DBL	216.32	158.45	DBL	38.76	1.17	96.61	14.60	-2.02	-8.14
	Oct-17	well		27.60	6.50	5.40	1095	0.37	162.26	128.13	16.70	0.26	0.14	DBL	172.62	13.59	DBL	28.97	6.95	62.43	7.43	-2.49	-10.75
	Feb-18	well		30.30	6.22	4.25	967	0.77	87.84	137.65	DBL	0.33	47.68	DBL	165.24	128.49	2.42	26.57	7.92	47.19	7.08	-2.62	-10.75
	Jun-18	well		29.60	6.72	8.31	935	0.59	144.88	91.65	DBL	DBL	31.57	DBL	19.87	13.93	0.24	25.35	6.24	51.88		-2.66	-11.51
A7	Nov-16	well		34.00	7.51		1302	0.36	125.58	44.84	DBL	0.12	0.23	DBL	43.42	58.32	1.63	13.37	3.69	21.27		-3.57	-18.22
	Mar-17	well		30.20	6.21	2.01	631	0.75	97.60	84.84	DBL	0.26	0.46	DBL	72.31	76.93	0.91	21.42	8.34	25.66		-2.96	-13.00
	Jun-17	well		30.70	6.31	2.52	763	0.42	95.75	11.86	0.52	0.28	0.19	DBL	131.16	93.31	0.68	19.39	1.26	34.31	6.80	-2.92	-12.16
	Oct-17	well		29.10	6.28	2.12	687	0.35	112.24	13.59	DBL	0.29	0.19	DBL	74.26	84.18	0.36	16.73	6.42	28.45	6.19	-2.89	-13.15
	Feb-18	well		29.30	6.20	2.12	689	0.56	79.30	12.42	DBL	0.38	0.35	DBL	74.59	8.83	1.77	21.11	8.15	29.82	7.79	- 2.99	-13.11
	Jun-18	well		28.30	6.38	2.83	816	0.63	155.55	18.42	DBL	DBL	0.39	DBL	67.64	89.20	0.89	27.33	9.20	32.96		- 3.02	-13.62
A8	Nov-16	well		29.50	6.20	0.10	551	0.25	75.41	7.68	DBL	0.13	0.16	DBL	59.96	66.77	0.12	6.86	1.90	26.92		-2.58	-10.57
	Mar-17	well	0.00	29.10	6.03	2.13	560	0.28	/8.80	/3.69	0.19	0.24	1.20	DBL	69.90	84.10	1.28	7.44	2.28	22.49	0.05	- 2.90	-12.29
	Jun-17	well	0.02	29.70	6.37	3.98	842	0.27	98.67	11.1/	0.19	0.24	1.39	DBL	1/1.18	10.8/	2.49	13.32	6.34	44.49	9.95	- 2.87	-12.41
	Oct-17	well	-0.12	27.80	6.03	4.51	645	0.10	64.23 72.20	70.00	DBL	0.26	1 1 4	DBL	149.72	92.14	0.22	5.3/	4.96	2.22	/.31	- 2.67	- 10.51
	FeD-18	well	0.48	28.90	6.15	2.93	042 770	0.18	142.25	79.04	DBL	0.30	1.14 DPI	DBL	120.61	82.51 05 70	0.88	5.91	4.66	2.34	0.69	- 2.69	-10.74
4.0	Juli-16	well	-0.20	29.10	7.04	7.49	666		246.67	/0.94 67 77	DBL	0.22	DBL	1 46	0.02	93.79 67.04	0.91	10 00	9.10	24.20		- 2.04	- 10.29
A9	Mor 17	well		29.00	6.85	2 24	765	0.84	174.46	07.77	0.52	0.25	0.45	0.20	02.92	07.54	0.34	1 21	9.32	52 77		-0.55	-11.02
	Oct 17	well		29.70	7 79	2.34	021	0.04	274.92	77.29	0.55	0.20	0.45	4.26	93.83 9.17	7 46	2.09	71.04	9.13 15.94	10 57	20.70	- 2.32	-10.20
	Eeb 18	well		20.90	7.70	4.22	1475	0.27	475.80	155.00	DBL	0.29	0.22	5.15	02.17	107.40	5.78	55.06	10.57	97.97	20.79	- 2.29	2 78
	Jup 19	well		20.40	7.04	9.22	1200	0.37	475.00	04 55	DBL	DRI	DRI	2 10	66 50	86.06	1 / 9	52.02	2.64	07.00	20.91	-0.03	2.78
A10	Nov-16	well		27.40	6.56	0.50	700	0.96	35.84	34 77	0.62	0.74	5 14	3.69	25 72	30.50	0.44	16 32	5.00	64 30		- 2.2/	-14.09
A10	Mar-17	well		20.00	7 55	2 4 8	1023	0.90	381 86	96.20	0.02	0.74	0.50	DBI	23.72 5.60	92.09	2.44 2.52	10.33 31 14	1.53	8 56		- 3.29	-14.90
	Jun-17	well		30.20	7.55	6 36	837	0.90	193.69	84 72	DRI	DRI	DBI	DBI	136 60	85 33	DBI	22 51	7.26	75 40	14 76	-1.85	-6.87
	Oct-17	well		29.20	7.00	6.43	835	0.17	245 22	110 70	DRI	0.20	0.66	DBI	4 98	11 00	DBI	26.01	7.20 8.75	44 20	22 53	-0.20	-1.64
	Feb-18	well		29.20	7 77	0.43	1870	0.12	2-13.22	112.79	DRI	0.20	0.00	DBI	37 49	121.90	88 40	44 49	935	45.65	22.33	-0.29	1 72
	Jun-18	well		20.70	7 33	3 71	654	0.12	262.30	6.83	DRI	DRI	DBI	DBI	4 68	67.80	DRI	14 47	6.75	47 49	20.7/	-2.02	-6.96
A11	Feb-18	Piezometer	0.40	20.00	6.61	1 19	1212	0.14	346.48	12 41	0.56	0.69	0.94	DBL	84 86	85.96	7 45	43.27	9.63	8 93	23.4	- 2.03	-13.02
1111	Jun-18	Piezometer	-0.26	28 20	7 22	0.06	1468	0.42	491 66	148 36	DRI	DRI	24 94	DBI	54 75	114 51	5 11	64 12	13.05	91 52	20.7	- 2.00 - 2.46	-10.21
	Jul-10	i iczometel	-0.20	20.20	1.43	0.00	1400	0.44	491.00	140.00	DDL	DDL	44.24		54.75	114.51	5.11	04.13	13.90	91.54		- 2.40	- 10.21

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Fable 1 (continued)

d²H	1 -8.43	5 -8.75	0 -7.84	8 -8.85	8 -6.20	3 1.60	0 -9.74	2 -8.77	4.27	4 -4.37	6 -10.42	96.96	2 -8.25	0 -12.96	1 -7.32	6 - 14.50	8 -9.83	1 - 9.59	6 -6.72
d <sup>18</sup> O	- 2.2	-2.2	-2.2	-2.3	-1.9	-0.3	-2.3	-1.9	0.35	-1.3	- 2.6	-1.9	-2.3	-2.7	-2.1	- 3.3	-2.3	-2.4	-1.8
DOC		18.69	15.16				24.63	15.12	18.10				7.03	5.93	13.96		11.85		8.45
Ca <sup>++</sup>	54.18	61.73	43.73	64.83	71.64	76.22	76.80	56.54	64.46	39.00	81.14	81.31	58.85	66.91	66.38	48.68	78.17	97.83	13.00
Mg <sup>++</sup>	32.94	29.42	26.65	31.20	23.14	23.73	21.14	17.52	17.66	9.42	24.86	42.15	18.83	19.40	39.95	15.60	17.98	19.18	63.48
$\mathbf{K}^+$	53.61	55.56	46.93	62.13	43.28	45.50	38.96	38.28	48.46	26.70	48.74	39.34	44.73	47.78	31.75	44.64	59.67	7.81	13.91
$\mathrm{NH_4}^+$	2.52	0.48	2.56	0.23	2.75	1.97	4.32	DBL	5.76	0.15	0.65	1.26	0.19	DBL	2.51	0.33	2.69	0.76	7.33
Na <sup>+</sup>	317.38	315.83	295.92	327.83	213.45	244.35	22.77	172.98	192.75	8.56	197.25	365.33	177.48	152.12	374.26	14.17	236.26	229.91	832.90
$\mathrm{SO_4}^{}$	17.50	254.40	221.44	294.16	78.11	1.91	43.27	7.87	4.19	1.46	121.78	134.38	135.36	119.67	151.54	117.46	273.55	248.81	136.77
PO4 <sup></sup>	DBL	DBL	DBL	DBL	DBL	2.17	DBL	2.54	1.84	1.83	4.45	2.38	4.15	3.49	0.83	4.45	2.16	4.33	DBL
NO <sub>3</sub> <sup>-</sup>	0.62	5.57	2.40	8.59	2.57	0.19	0.59	DBL	1.59	DBL	12.69	14.25	89.49	48.44	DBL	75.72	57.98	13.25	0.54
Br <sup>-</sup>	0.14	0.11	0.39	0.15	0.41	0.35	0.96	DBL	0.18	DBL	1.66	0.23	1.44	06.0	DBL	1.21	0.94	1.72	0.87
$NO_2^-$	1.26	1.63	1.22	1.35	0.65	0.89	0.85	0.63	0.91	0.24	0.55	1.29	0.56	0.41	1.68	DBL	0.62	DBL	5.28
<u>-</u> 1	385.88	428.57	355.74	431.66	255.80	31.17	261.68	219.13	232.88	35.77	21.94	414.29	18.85	16.51	446.98	13.48	253.78	292.14	1561.35
HCO <sub>3</sub> <sup>-</sup>	247.66	323.30	337.94	366.61	41.99	512.40	47.50	386.74	529.48	266.57	323.22	585.60	279.69	263.52	622.20	248.27	35.00	266.57	4.16
NOx	0.17	1.67		0.63							0.13	0.17	0.39	5.46			0.15	2.51	
EC	2090	2180	2090	2040	1608	1660	1543	1352	1669	750	1623	2350	1357	1378	2600	1210	1945	2004	5340
DO	1.69	3.10	3.75	2.52		2.37	2.29	1.61	1.63	0.56		1.70	7.32	4.32	1.98	4.87	3.60	3.32	0.10
Ηd	6.80	6.73	6.93	6.92	6.87	7.12	7.73	7.69	7.84	7.39	7.13	7.13	7.69	7.39	7.17	7.68	7.00	6.89	6.84
T°C	29.30	26.20	28.00	28.00	28.50	29.50	28.70	26.30	28.50	28.60	31.90	30.70	29.40	27.00	29.10	29.70	30.00	29.70	33.40
Depth		0.41	0.71	0.64			0.19	0.09		0.36			0.38	0.31	1.25	0.54			0.39
Origin	well	well	well	well	well	well	well	well	well	well	well	well	well	well	well	well	well	well	piezometer
Sampling month	Mar-17	Oct-17	Feb-18	Jun-18	Nov-16	Mar-17	Jun-17	Oct-17	Feb-18	Jun-18	Nov-16	Mar-17	Jun-17	Oct-17	Feb-18	Jun-18	Feb-18	Jun-18	Mar-18
D	L2				L6						L7						L11		L18

in Ladji, the seasonal fluctuations in the water table between the wet and dry seasons vary by 1.3 m (Fig. 5). In comparison, in Agla, the seasonal amplitude of the water table variation is lower (0.5 m). Since Ladji is located on the shore of Lake Nokoué, the lake fluctuations likely influence the variation of the water table depths. An increase of about 90 cm in the water table is observed at Ladji between the wet and dry seasons is consistent with the observed elevation of the lake level during the same period (IRD/IRHOB, unpublished data). The rise in water level starts in January and increases gradually to April/May, which is a 2-month time lag from Agla and St Jean.

With seasonal variations in the water table depth, the groundwater EC also shows varying trends (Fig. 3). In St Jean, the groundwater EC values (297-1.285 uS/cm) are relatively weak compared to Agla and Ladji, and show variable seasonal changes (Fig. 3). Wells J1 and J2 recorded higher EC values in the dry season (511-1,285 µS/cm) when groundwater levels were low compared with the wet season (297-799  $\mu$ S/cm). In comparison, the well J5 and the piezometer had higher EC values (444-1,169 µS/cm) in the rainy season (rise of the water table) compared with the dry season (497–685  $\mu$ S/cm). In Agla, the groundwater EC ranges between 353 and 1,468 µS/cm, which is low compared with local surface waters (including temporary ponds and swamps; 2,800–2,900  $\mu$ S/cm). EC values are similar during the rainy season and the dry season (Fig. 3). In Ladji, the groundwater EC values are higher than in the other neighbourhoods (633–5,340  $\mu$ S/cm) and similar to the local temporary ponds (480–4,090  $\mu$ S/cm), but strongly lower than the Lake Nokoué (up to 49,700 µS/cm during dry season). With the exception of well L2, no difference was observed in the variation of the EC during the dry season and the rainy season (1,478–5,340  $\mu$ S/cm).

#### 4.2. Seasonal variations of major ions

The composition of major ion concentrations of waters is different in each of the neighbourhoods as highlighted in Fig. 4. In St Jean, the groundwater is of Na-Ca-HCO<sub>3</sub>-Cl type. In comparison, the groundwater in Ladji is of Na-HCO<sub>3</sub>-Cl type, and Agla groundwater is more of the Na-Ca-HCO<sub>3</sub>-SO<sub>4</sub>-HCO<sub>3</sub> type (Fig. 4). The ponds sampled in both Ladji and Agla show some similarities in major ion compositions with local groundwater, thus indicating a potential connection between surface and subsurface waters. However, this connection is spatially and/or temporally variable, in particular in Agla, since some of the pond samples have a greater concentration of HCO<sub>3</sub> relative to Cl and SO<sub>4</sub>, and increased Ca relative to Na and Mg compared with groundwater. In addition, in Ladji, the Lake Nokoué has a greater ratio of Cl to SO<sub>4</sub> and HCO<sub>3</sub> compared with groundwater.

The temporal evolution of these major ions at each site is presented in Fig. 5 and, similar to EC values, the seasonal changes vary both between sites and between neighbourhoods. Greatest seasonal variations are observed in Ladji, and particularly for groundwater sampled at L6 and L7 where dry season conditions results in increases in Na (by 244–374 mg/L), Ca (by 76–81 mg/L), Cl (by 301–447 mg/L) and HCO<sub>3</sub> (by 530–622 mg/L). These sites are located between 100 and 150 m from the Lake shore. The increase of the ions' concentration during dry season is associated with a deepening of the water table and may therefore indicate influences of lake infiltration. The Lake Nokoué has greater concentrations of Na (3,168–12,219 mg/L), Ca (146–419 mg/ L), Cl (6,166–1,9310 mg/L), and HCO<sub>3</sub> (73–110 mg/L) compared with all groundwater samples (Tables 1 and 2).

In Agla, the seasonal variations in major ions remain relatively stable except for  $HCO_3$  and  $SO_4$  concentrations (Fig. 5). However, between sites, there are large differences in seasonal trends. Four of the eight groundwater wells (A4, A5, A9 and A10) show an increase in Na (by 47–128 mg/L), Ca (by 20–88 mg/L),  $HCO_3$  (by 155–750 mg/L) and Cl (by 61–155 mg/L) during the month of February (dry season). During the wet seasons (June and October/November), the remaining four wells (A2, A6, A8 and A7) show an increase in Na (by 93–158 mg/L), Ca (by 34–78 mg/L),  $HCO_3$  (by 99–436 mg/L), Cl (by 110–176 mg/L), Ca (by 34–78 mg/L),  $HCO_3$  (by 99–436 mg/L), Cl (by 110–176 mg/L).

Table 2	
Chemical parameters and isotopic values in surface water.	

	r				mate																	
ID	Sampling month	Origin	T°C	pН	DO	EC	NO <sub>x</sub>	$HCO_3^-$	$Cl^{-}$	$NO_2^-$	$Br^{-}$	$NO_3^-$	PO4 <sup></sup>	SO4 <sup></sup>	Na <sup>+</sup>	$\mathrm{NH_4}^+$	K <sup>+</sup>	Mg <sup>++</sup>	Ca <sup>++</sup>	DOC	d <sup>18</sup> O	d <sup>2</sup> H
					a	<i>C</i> /	a	a													0/	
Units	No. 16		00.40	7.00	mg/L	µS/cm	meq/L	mg/L	00.10	DDI	0.00	DDI	1.00	10.41		0.00	14.01	7.04	(0.50		%0 1 = 1	0.10
AZ	NOV-16	pond	32.40	7.02	7 77	818		287.54	82.19	BDL	0.26	BDL	1.98	19.41	/6.54	0.26	14.21	7.84	62.59 1.70	10.01	- 1.51	-3.18
	Juli-17	pond	25.20	8.72	1.//	113		41.02	5.19	BDL	BDL 0.57	BDL	0.00	4.2/	0.27	0.41 BDI	3.47	0.74	1.79	10.91	-2.13	-12.69
	Uct-17	pond	20.20	7.25	4.05	970	0.02	338.08	108.79	6DL	0.57	BDL	4.49	10.05	90.12	5DL 1.00	22.87	1.32	69.99	25.43	- 2.05	-12.20
4.2	Juli-18 New 16	pond	26.80	0.90	1.51	740	0.02	360.03	95.54	0.74	6DL	BDL 0.00	2.20	11.58	/9.5/	1.22	52.02	14.01	03.34 F0.61		- 2.89	-12.18
AS	INDV-10	pond	20.10	0.01	0 52	1610	0.02	409.10	240.00	0.39 PDI	0.43	0.99 PDI	1 5 2	154.29	120.17	1.70 PDI	50.54 67 E4	16.90	29.01	26.00	- 0.90	-0.03
	Juli-17 Oct 17	pond	29.10	0.24	9.55	246		400.70	249.90	BDL	0.39	BDL	2 41	0.60	20.27	BDL	14.99	10.00	09.00	10 /E	- 2.20	- 0.04
	Uct-17	pond	29.80	8.43 7.05	11.01	340 009		137.80	24.35	BDL	0.18 PDI	BDL	3.41 0.57	0.09	29.30 64 E1	6DL 0.19	14.22	4.19	20.47	18.45	- 2.41	-14.50
16	Jun 17	pond	28.40	7.05 8.67	7.45	990 75		250.20	4 50	0.20	BDL	BDL	0.57 RDI	4/./4	6.83	0.10 RDI	4 94	7.55	5 20	9 1 2	- 1 20	- 17.80
A7	Nov 16	pond	20.40	6.20	7.45	504		408.68	127 55	0.20 RDI	0.27	BDL	4 5 2	17.86	174.65	BDL	9.22	0.39	27.26	0.12	2.99	24 74
A/	Iup 17	pond	20.70	0.20 8.04	6 66	250		102.05	127.33	0.27	BDI	BDL	4.55 RDI	15.70	17 78	BDL	6.42	1.00	27.50	7.86	-2.74	24.74 
	Oct 17	pond	25.00	0.04	15 70	1070		103.83	02.01	0.37 RDI	0.24	BDL	4 50	13.79	117.64	BDL	0.42 82.62	1752	41.35	20.75	- 2.74	-6.12
	Uct-17	pond	20.70	9.05	13.70	E24		470.31 172.0E	20.60	DDL	0.24 PDI	BDL	F 77	12.00	40.25	PDI	25.02	7.02	21.07	39.75	4.07	-0.12
10	Juli-16	pond	20.20	6.40	4.55	700		1/0.00	30.00 74.4E	BDL	0.22	0.12	0.// 1 10	10.90	49.23	6 4 2	10.10	1.92	40 10		-4.27	-23.07
Ao	INDV-10	pond	25.00	0.40	10.00	1070	0.05	210.23	74.43 116 7E	0.67	0.32	1.90	1.10 PDI	20E 60	112 71	0.43	20.75	4.90	40.10	26.26	- 2.97	- 11.99
	Juli-17 Oct 17	pond	35.90	9.04	12.38	12/8	0.05	97.00	110.75	0.07 PDI	0.10	1.89 PDI	BDL	393.09 0 15	113./1	0.30 PDI	39.75 12.72	9.80	20 16	20.20	-0.33	-1.29
	Uct-17	pond	29.70	6.00	2.34	60E	0.09	147.02 052.15	42.90	2 46	0.45 PDI	BDL		0.40 E2 E2	73.47	0.20	24.74	0.70	30.40 17.66	15.41	- 2.03	-11.30
4.0	Juli-16	pond	20.90	6.00	0.08	764	0.08	205.10	43.30	3.40 DDI	0.24	BDL	1.00	32.33	30.39	0.39	24.74	0.4/	47.00		- 3.45	- 10.37
A9	NOV-10	pond	31.10	0.88	0.01	704		305.53	74.35	BDL	0.24	BDL	2.13 PDI	4.88	71.43	BDL 2.22	19.02	12.90	44.00	25.20	0.35	3.38
	Juli-17	pond	29.40	7.07	0.01	1690		/ 38.10	102.81	BDL	0.17	BDL	E DE	11 54	/5.29	3.32 BDI	145.23	33.92	147.00	23.28	- 2.09	-11.65
	Uct-17	pond	28.00	0.41	2.40	1020		361.33	240.73	BDL	0.50	BDL	5.25	11.54	100.41	BDL	102.78	20.22	38.85	51./5	-0.09	-6.20
110	Jun-18	pond	31.20	8.58	10.72	/11	0.00	260.47	69.58	BDL	BDL	BDL	0.40	16.46	/.35	BDL	35.68	12.37	34.24		- 3.29	-16.12
A10	NOV-16	pond	30.10	0.69	0.07	1310	0.02	544.58	127.78	BDL	0.44	1.51	0.50	2.21	12.24	0.18	4.32	14.42	/3.28	40.00	-0.93	-1.43
	Juli-17	pond	31.40	7.48	2.07	1114		314.15	124.09	BDL	0.18	0.42	5.99	130.02	92.28	BDL	5.3Z	13.50	95.01	43.28	- 2.55	-11.18
	Oct-17	pond	28.70	6.96	1.82	621		250.71	69.50	BDL	0.19	BDL	0.85	0.44	62.84	BDL	21.67	7.69	41.23	27.14	- 3.30	- 16.92
410	Jun-18	pond	28.70	/.1/	3.29	/58		2/1.45	/5.4/	BDL	BDL	0.25	BDL	15.37	/2.84	0.3/	23.60	8.9/	54.34	10.00	- 2.90	-11.70
A12	Feb-18	pond	28.80	6.03	5.72	768		42.70	153.98	BDL	0.99	BDL	BDL	254.62	162.77	3.64	12.15	13.69	27.38	18.06	-2.71	-11.35
	Jun-18	pond	28.10	6.69	0.17	605	18.55	176.90	53.70	BDL	BDL	BDL	1.47	19.75	63.75	0.25	15.75	6.33	35.76	14	- 3.98	- 19.48
A13	Feb-18	pond	31.90	4.12	6.77	2900	17.55	0.00	256.87	0.59	BDL	188.27	12.89	24.59	219.67	28.23	95.68	68.63	221.89	55.14	-2.44	-9.10
	Jun-18	pond	29.30	6.72	0.03	537		247.05	43.53	BDL	BDL	BDL	BDL	6.82	46.36	BDL	12.94	9.75	39.46	01.00	- 3.27	-14.28
A14	Feb-18	pond	29.60	7.28	0.01	1671	0.10	456.28	189.73	BDL	0.43	BDL	5.98	291.84	146.16	3.67	92.25	25.54	139.40	31.90	-1.51	-0.95
	Jun-18	pond	27.90	6.90	0.30	685	0.19	271.45	55.38	0.32	BDL	11.15	4.33	7.70	54.90	0.42	5.35	9.86	42.82		- 3.08	-13.31
A15	Jun-18	pond	27.80	6.93	0.12	829	0.14	298.90	65.99	BDL	BDL	8.95	4.87	18.13	69.75	0.25	3.56	1.98	66.66		- 3.35	-14.37
A16	Jun-18	pond	28.40	7.32	0.86	712		282.43	57.09	BDL	0.14	BDL	2.75	6.71	55.33	BDL	37.75	1.63	53.39		-2.87	-11.27
A17	Jun-18	pond	26.10	7.29	1.06	846		378.20	64.69	BDL	0.29	BDL	4.74	12.48	83.16	BDL	21.60	9.51	67.19		-2.31	-8.49
A18	Jun-18	pond	31.90	7.44	7.33	563		250.10	40.80	BDL	0.92	BDL	1.45	16.12	45.58	BDL	26.43	6.77	42.95		-3.60	-16.65
A19	Jun-18	pond	27.30	7.15	0.19	796		284.26	74.60	BDL	0.38	BDL	1.83	14.76	78.96	0.28	17.67	6.92	62.75		-2.05	-7.31
L2	Oct-17	pond	24.80	7.63	2.10	732		211.06	84.00	BDL	0.49	BDL	7.38	37.13	83.44	3.52	36.63	5.85	28.74	15.26	-1.98	-11.15
L3	Nov-16	lake	32.90	7.18	/	18,630	0.01	73.43	6166.30	BDL	18.98	0.82	BDL	811.84	3168.64	BDL	144.99	375.64	146.79	27.19	-3.29	-14.44
	Jun-17	pond	29.10	8.05	6.15	480		128.41	53.24	BDL	0.12	BDL	3.45	34.35	53.23	0.77	22.55	4.37	23.57		-1.86	- 4.57
	Jun-17	pond	30.40	8.73	7.19	4090		401.08	946.12	BDL	2.21	BDL	BDL	339.79	655.14	BDL	158.33	48.63	85.54	27.60	-1.85	-12.12
	Oct-17	pond	28.80	7.43	2.60	1371		235.46	314.06	BDL	1.34	BDL	3.41	31.84	180.00	BDL	27.68	22.49	35.97	22.60	-1.93	-9.03
L6	Jun-17	pond	29.90	8.17	0.02	2050		555.56	255.63	BDL	0.55	BDL	14.98	98.67	275.76	27.97	137.28	12.36	28.24	28.67	-0.98	0.71
L8	Nov-16	lake	29.60	7.01	/	23,200		93.10	8273.96	BDL	25.72	0.34	BDL	134.38	4495.24	BDL	158.90	54.36	171.41		-0.71	-0.85
	Mar-17	lake	31.00	7.65	5.10	47,100	0.01	109.80	15655.71	0.33	49.97	BDL	BDL	1867.28	8478.39	8.64	251.58	153.00	268.12		1.13	7.60
	Jun-17	lake	28.90	7.55	3.75	38,800		103.70	14687.98	BDL	5.48	BDL	BDL	2232.55	848.96	1.94	286.80	966.60	298.53	2.30	1.09	8.46
	Oct-17	lake	25.70	7.29	2.00	886		102.48	229.88	BDL	0.94	BDL	BDL	19.31	123.87	BDL	11.32	14.36	16.98	13.01	-2.20	-10.87
	Feb-18	lake	28.70	7.86	3.32	47,300		109.80	18119.73	BDL	62.32	BDL	BDL	2473.38	9449.17	0.71	36.92	928.45	34.41	0.98	0.97	7.46
	Jun-18	lake	29.70	7.41	2.54	39,100		128.71	16114.88	BDL	62.43	BDL	BDL	2293.29	8415.52	0.47	36.52	151.93	276.92		- 4.99	-27.32
L9	May-17	pond	31.10	9.85	11.40	1204		156.62	254.45	BDL	0.23	BDL	2.25	89.27	195.47	0.10	64.58	2.42	11.95	45.49	-0.57	3.33
L10	Jun-17	pond	31.80	8.16	0.02	2000		491.97	408.44	BDL	0.66	BDL	8.95	8.72	295.17	3.12	96.45	15.95	64.45	27.02	-1.77	-16.01
	Oct-17	pond	28.70	9.61	3.28	2440		500.20	509.98	BDL	0.82	BDL	8.83	158.87	384.79	2.74	12.76	12.65	4.88	39.97	-1.13	-5.51
L14	Feb-18	lake	29.60	7.63	4.40	47,600		109.80	19310.02	BDL	74.44	BDL	BDL	2667.71	12219.87	21.24	381.49	1256.91	419.23	0.71	0.75	6.26
	Jun-18	lake	30.10	7.60	3.82	39,400		131.15	16379.80	BDL	57.53	BDL	BDL	236.46	8487.56	0.51	323.79	158.49	263.38		-3.36	-8.85

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Ð	Sampling month	Origin	T°C	Ηd	DO	EC I	NOx	$HCO_3^{-}$	Cl -	$NO_2^-$	$\mathrm{Br}^{-}$	$NO_3^-$	$PO_4^{}$	$\mathrm{SO_4}^{-}$ -	Na +	$\mathrm{NH_4}^+$	$\mathbf{K}^+$	Mg <sup>+ +</sup>	Ca <sup>+ +</sup>	DOC	d <sup>18</sup> O	d²H
L15	Feb-18	lake	30.20	7.90	7.29	49,700		113.46	18031.65	BDL	69.73	BDL	BDL	249.49	9579.63	25.33	32.24	955.30	323.52	1.02	0.93	7.34
	Jun-18	lake	37.00	7.67	5.85	38,100		57.95	15173.94	BDL	52.19	BDL	BDL	2165.47	7884.58	0.34	296.81	994.89	254.81		-1.34	-14.50
L16	Mar-18	puod	29.20	8.65	1.16	3830		1115.08	824.56	BDL	3.41	BDL	7.25	2.78	698.15	BDL	34.17	73.68	11.74	40.37	0.56	5.05
	Jun-18	puod	29.10	7.37	0.01	1024		289.75	120.41	BDL	0.17	BDL	6.98	41.34	121.28	0.83	39.94	12.58	39.45		-1.34	-4.37
L17	Mar-18	puod	28.90	7.20	0.01	2000		274.50	356.31	BDL	5.95	BDL	BDL	224.64	314.95	1.96	12.20	19.63	43.30	33.59	-2.02	-6.86
	Jun-18	puod	30.90	6.86	0.35	831		259.25	46.16	BDL	BDL	BDL	1.66	1.43	47.63	2.57	1.98	8.63	44.32		0.50	5.33
L19	Jun-18	puod	30.30	6.88	0.04	633		251.32	53.73	BDL	BDL	BDL	3.55	8.52	62.68	0.24	26.85	5.28	32.71		0.63	6.15
AO1	Jan-19	Ocean				51,000		132.00	19091.00	BDL	7.00	BDL	BDL	2317.00	126.00	0.70	384.00	1219.00	391.00		0.52	4.68
A02	Jan-19	Ocean				51,300		135.00	19172.00	BDL	64.00	BDL	BDL	2329.00	1178.00	0.40	382.00	1216.00	386.00		0.54	4.69
*BDL: E	selow Detection L	imit.																				

Table 2 (continued)

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L), and SO<sub>4</sub> (by 75–276 mg/L) concentrations. These trends correspond to changes in the water table depths, where the increasing wet season major ion concentrations occurs in wells where the groundwater is (sub)artesian during the wet season (A2 and A8), compared to wells where groundwater remains well below the surface and has increased dry season major ion concentrations (A4). These trends may highlight the influence of groundwater mixing with pond water in the discharge areas, compared with infiltrating rainfall diluting groundwater in other areas during the wet season. The temporary and permanent pond waters have similar concentrations of Na (56–166 mg/L), Ca (34–147 mg/ L), Cl (43–240 mg/L), and HCO<sub>3</sub> (97–738 mg/L) compared with Agla groundwater samples (Tables 1 and 2).

In St Jean, groundwater at seven of the ten wells sampled (J1, J2, J10, J3, J4, J6 and J9) show an increase in Na (by 36–115 mg/L), Ca (by 49–114 mg/L),  $HCO_3$  (by 119–439 mg/L) and Cl (by 42–111 mg/L) concentrations during the dry season (February and March, Fig. 5). This occurs at the time when the water table is at its deepest level. In addition, small wet season peaks in major ion concentrations are also observed in groundwater from wells J5, J7 and J8. These increases include Na (by 63–65 mg/L), Ca (by 63–80 mg/L),  $HCO_3$  (by 128–272 mg/L) and Cl (by 61–79 mg/L) concentrations. Since St Jean does not have any surface water bodies, the wet and dry season major ion variations are probably due to variations in the composition of infiltrating water, mixing between subsurface waters, reactions during water table fluctuations, and evaporation.

#### 4.3. Stables isotopes

The  $\delta^{18}$ O and  $\delta^{2}$ H values of rainfall in Cotonou (obtained from 95 samples from the IITA and IAEA Global Network of Isotopes in Precipitation (GNIP) from 2005 to 2016) has a weighted average of  $-2.8 \pm 1.6\%$  for  $\delta^{18}$ O and  $-11.2 \pm 11.7\%$  for  $\delta^{2}$ H. The local meteorological water line (LMWL) of rainfall obtained from these data has a slope of  $\sim$ 7 and is presented together with the global meteoric water line (GMWL; slope of ~8, Craig, 1961) in Fig. 6. These data were also used to calculate the mean weighted average values of  $\delta^{18}O$  and  $\delta^2$ H for different ranges in monthly rainfall. The results show that when monthly rainfall is < 50 mm, the isotope values are greater compared with monthly rainfall ranging from 50 mm to 500 mm (Fig. 6a). The isotope values of rainfall collected in Cotonou show an overall mass effect; a depletion in heavy isotopes correlates with an increase in rainfall amounts. In Cotonou, the local rainfall has stable isotope values covering a large range; from -5.4 to 0.8‰ for  $\delta^{18}$ O, and from -30.9to 14.5‰ for  $\delta^2$ H (Fig. 6a, black crosses). The groundwater values from the three sites are also presented in Fig. 6a, which also highlights a large range in values (from -4.17 to 0.35% for  $\delta^{18}$ O, and from -21.9to 4.3% for  $\delta^2$ H).

In St Jean, the stable isotope values for groundwater vary between -4.17 and -2.37% for  $\delta^{18}O$  and between -21.87 and -8.98% for  $\delta^{2}H$ . These values show that the groundwater in St Jean is depleted at  $\delta^{18}O$  and  $\delta^{2}H$ . The linear regression between  $\delta^{18}O$  and  $\delta^{2}H$  values (slope of 7.5) lie close to the LMWL (Fig. 6a), suggesting that groundwater likely originates from local rainfall.

In Ladji, groundwater has enriched in  $\delta^{18}$ O and  $\delta^{2}$ H compared with St Jean (-3.36 to 0.35 and -12.96 to 4.17‰, respectively; Fig. 6b). This discrepancy may be due to three distinct phenomena: (i) the recharge of relatively low volume rainfall events (< 50 mm/month) in Ladji compared with St Jean; (ii) evaporation effects (some values lie to the right of the LMWL indicating a slope of 5.2), or (iii) groundwater mixing with lake water. During the dry season, groundwater isotopic values are close to the lake Nakoue values (Fig. 6b), which suggests a strong mixing between the lake and groundwater during this season. There are also a number of groundwater samples whose  $\delta^{18}$ O and  $\delta^{2}$ H mixing ratios indicate ranges from 72 to 74% of shallow groundwater mixing with lake water during the dry season in Ladji.

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Fig. 3. Relationship between water-table variation and electrical conductivity at each site.

Like in Ladji, groundwater in Agla is also enriched in  $\delta^{18}O$  and  $\delta^{2}H$  (-3.57 to -0.03, and -18.22 to 2.78‰, respectively) compared with St Jean. Some samples from Agla (mostly during the dry season) lie to the right of the MWL with a slope of 5.4 (Fig. 6c). Therefore, these groundwaters may also be subject to evaporation and mixing with enriched surface waters such as the lake water (via the lowlands) and the pond water. In addition, groundwaters (e.g., wells A9 and A10) that are close to lake water are located in discharge areas.  $\delta^{18}O$  and  $\delta^{2}H$  mixing ratios indicate ranges from 54% to 62% of shallow groundwater mixing with lake water in Agla.

#### 4.4. Nitrogen

The Beninese quality standard for drinking water for nitrogen is 45 mg/L (0.70 mmol/L) for NO<sub>3</sub>, 3.2 mg/L (0.07 mmol/L) for NO<sub>2</sub>, and 250 mg/L (7.0 mmol/L) for Cl (Decree N  $^{\circ}$  2001–094 of February 20th). In this study, 26% (values of 0.73–5.06 mmol/L) of the groundwater sampled exceeds these limits in terms of NO<sub>3</sub>, and 7.0% (values of 0.12–2.04 mmol/L) for NO<sub>2</sub> (Table 1).

Highest values of NOx ( $\Sigma$ NO<sub>2</sub> + NO<sub>3</sub>) are observed in groundwater of St Jean, where the concentrations reach values of 5.06 mmol/L (average of 0.90 mmol/L). For NH<sub>4</sub>, the groundwater concentrations remain low (up to 0.80 mmol/L, average of 0.04 mmol/L). The highest concentrations of NOx are observed during the dry season (> 3 mmol/L) compared with wet season samples (NOx < 3.37 mmol/L). Two sites (J6 and J8) have elevated NOx concentrations in both the wet and dry seasons (1.15–5.06 and 0.99–2.58 mmol/L, respectively). NOx in St Jean groundwater may originate from natural fixation of nitrogen in the soil and human pollution via leaky latrines and human defacation in the streets.

In Ladji, the groundwater NOx concentrations ranges up to 2.51 mmol/L (average of 0.54 mmol/L), and NH<sub>4</sub> is up to 0.43 mmol/L (average of 0.11 mmo/L). During the wet season, the groundwater NOx concentrations are highest (1.06–2.51 mmol/L) compared to dry season concentrations (< 0.13 mmol/L). The wet season increase in NOx of groundwater in Ladji may either be due to infiltration of anthropogenic pollution, or nitrogen fixed by vegetation in the soils. The NOx concentrations are low in the permanent ponds and the lake (< 0.01 mmol/L). However, the wet season temporary ponds have high concentrations of NH<sub>4</sub> (up to 1.55 mmol/L), which may undergo nitrification during infiltration and may also result in the NOx contaminantion of groundwater.

The highest value of  $NH_4$  in groundwater is observed in Agla, but only at one site (A10: 4.91 mmol/L). All other sites have  $NH_4$  concentrations in groundwater ranging from 0.00 to 0.52 mmol/L (average of 0.17 mmol/L). Compared with Ladji and St Jean, groundwater NOx



Fig. 4. Piper diagram for groundwaters, ponds and lake at each site.



Fig. 5. Temporal variation between, depth water table, electrical conductivity, major ion and isotopic stable at each site.

concentrations in Agla are low, with a maximum of 0.86 mmol/L (average of 0.15 mmol/L). In contrast, permanent ponds in the Agla lowlands have high NOx levels only at one site (A13: 17.50 mmol/L), particularly in the dry season.

As described by Katz et al. (2011), the Cl/Br ratio can be a valuable first assessment of septic tank contamination of shallow groundwater. This is based on the assumption that sewerage waters and septic tank effluent exhibit distinct ranges and higher values of Cl/Br molar ratios



Fig. 6. Relationship between  $^{18}$ O and  $^{2}$ H with Global Meteoric Water Line (GMWL) and Local Meteoric Water Line (GMWL).

and Cl concentrations compared with rainwater (Davis et al., 1998), as observed in many samples form this study (Fig. 7a). Elevated Cl/Br ratios may also result from the dissolution of halite. However, halites have not been reported in the local Quaternary aquifer. In addition, many of the groundwater values correspond to ranges reported for sewerage and septic tank effluent (Davis et al., 1998; Vengosh and Pankratov, 1998; Katz et al., 2011) compared with waters with halite dissolution (Davis et al., 1998; Pastén-Zapata et al., 2014; Panno et al., 2006), rainwater from coastal areas, and seawater (Alcalá and Custodio



**Fig. 7.** (a) Groundwater, pond and lake water Cl/Br molar ratios with changes in Cl concentrations highlights different sources of Cl in the waters. (b) Relationship between Cl/Br ratios and Cl concentrations showing higher concentrations in NOx (circle sizes are relative to NOx concentrations).

2008; Davis et al., 1998). According to Fig. 7b, groundwater values with similar Cl/Br ratios and Cl concentrations than sewerage wastewater also show higher concentrations of NOx. Three samples (wells J2, L9 and A6) plot above the Cl/Br ratios for sewage or septic tank effluent (Cl/Br molar ratios 676–1350; Cl < 1000 ml/L), likely indicating animal manure. As evidenced in Fig. 7b, values that fall into the animal manure/animal urine (Cl/Br molar ratios, 2810–3730; Cl < 1000 mg/L) also have high NOx concentrations (0.76–0.81 mmol/L). Other evidence for a septic tank and animal manure influence are the corresponding elevated dissolved organic carbon (DOC) concentrations (Table 1). However, not all the groundwater samples have high DOC concentrations. According to Kortelainen and Karhu (2006), rapid oxidation of DOC into carbon dioxyde may account for the low DOC concentration in groundwater.

Alternatively, samples with low  $NO_x$  concentrations and relatively low Cl/Br ratios, such as groundwater from Agla (Fig. 7b), can indicate areas of organic biodegradation (e.g. McArthur et al., 2012). The relationship between HCO<sub>3</sub>,  $NO_x$  and DO for all groundwater sampled at Agla is shown in Fig. 8. The samples where dissolved oxygen (DO) and NOx are low compared with HCO<sub>3</sub> correspond to discharge areas. This is especially the case for groundwater sampled during the dry season at wells A10, A9 and piezometers A11, J11 where HCO<sub>3</sub> values increases (4.80–12.31 mmol/L) and NOx decreases (0.0–0.40 mmol/L) with low DO values (0.06–4.22 mg/L). However, this may also indicate areas of recent rainfall infiltration resulting in a dilution of NOx concentrations (e.g. for well A4).Fig. 9.



Fig. 8. Relationship between HCO3, NOx and DO for all groundwater sampled at Agla.

#### 5. Discussion

#### 5.1. Variations in the drivers of groundwater degradation

The vulnerability of groundwater degradation from either salinization or sewage leakage in Cotonou is temporally and spatially variable. The Quarternary aquifer is exposed to large fluctuations in the water table in response to rainfall changes; all sites show seasonal water table fluctuations higher than 0.5 m. However, both the timing of the water table fluctuations and the seasonal changes in groundwater quality varies between urban areas.

In Agla, a strong driver of groundwater quality is the proximity to lowlands. Agla is scattered by these low elevation zones, which have either temporary or permanent ponds where groundwater seasonally discharges. As highlighted by the major ion results, wet season increases in the water table can either result in increased groundwater salinity (EC up to 1468 µS/cm) in these lowland discharge areas due to mixing with pond water, or can result in lower groundwater salinity (EC <  $842 \mu$ S/cm) due to the dilution effect from infiltrating rainfall. During the dry season, the groundwater stable isotope values also highlight increased mixing with pond water and saltier Lake Nokoué water. Elevated NOx concentrations in groundwater at Agla is due to sewerage contamination and was recordered during both dry and wet seasons (NOx concentrations up to 0.86 mmol/L). Groundwater contamination from sewerage may infiltrate directly from leaky latrines or from mixing with the permanent ponds, which have accumulated NOx concentrations up to 17.50 mmol/L. According to Starr and Gillham (1993), denitrification tends to occur in aquifers with very shallow groundwater compared to aquifers with deeper groundwater (more than 2 m). Low dissolved oxygen levels combined with low NOx levels in shallow wells (even less than 1.0 m) in the study area (Fig. 8) where higher levels can be expected, may be related to biodegradation or denitrification processes (Postma et al., 1991, Jorgensen et al., 2004, Hassane et al., 2016; Kadjangaba et al., 2018). Either by reducing NO<sub>3</sub> to HCO<sub>3</sub> (1) or N<sub>2</sub> (2). According to Postma et al. (1991) and Anornu et al. (2017), the absence or low concentrations of NO2 and NH4 in groundwater is probably related to the reduction of NO3 to N2. The overall denitrification process can be described as (Berner, 1980):





Fig. 9. Schematic of conceptual model for contaminant sources and transfers at (a) Agla, and (b) Ladji and Saint-Jean.

$$5CH_2O + 4NO_3^- \rightarrow 2N_2 + 4HCO_3^- + CO_2 + 3H_2O$$
 (1)

$$2NO_3^- + 12H^+ + 10e^- \rightarrow N_2 + 6H_2O$$
 (2)

However, denitrification may not be the only process that contributes to the increase of  $HCO_3$  in shallow groundwater, which probably also results from the decomposition of organic matter and mineral dissolution reactions (e.g. Alassane et al., 2015).

In Ladji, the main influence on groundwater salinization (EC up to 5,340  $\mu$ S/cm) is the mixing with the Lake Nokoué waters. This was obvious during the dry season where both stable isotopes values and major ion concentrations clearly highlight mixing, and the water level also shows an early dry season rise in values due to lake infiltration. In comparison, the sewerage contamination of groundwater is mostly evidenced during the wet season (groundwater NOx up to 2.5 mmol/L). During the wet season, increases in NOx from sewerage sources may result from both (i) rises in the water table mobilising NOx in the unsaturated zone, and/or (ii) infiltrating rainfall and temporary ponds recharging the shallow groundwater (as suggested by stable isotope results) and therefore transporting NOx to the saturated zone.

St Jean is the only studied neighbourhood that does not have any surface water bodies and no groundwater discharge sites. Therefore, the wet and dry season variations in groundwater degradation are potentially due to variations in the composition of infiltrating water, mixing between subsurface waters, and reactions during water table fluctuations. Although St Jean has low groundwater salinity levels (EC up to 1,285 uS/cm), this area has the highest NOx concentrations recorded in this study (NOx up to 5.06 mmol/L). Since the local rainfall is the only origin of the shallow groundwater (as seen from stable isotope values), it is expected that either recharging rainfall or rising water tables transfers NOx from the sewerage sources to the groundwater system during the wet season. However, at St Jean, groundwater NOx concentrations are greater during the dry season (up to 5.06 mmol/L) compared with the wet season (up to 2.58 mmol/L). So, instead, the contamination may be constant leakage from latrines throughout the year and wet season rainfall may act to dilute this contamination. Almost all the pit latrines and septic tanks in Cotonou have depths between 1.50 and 2.50 m (Hounkpe et al., 2014; Yadouléton, 2015). In St Jean, the maximum depth to water table is observed at 1.53 m in the dry season, which means that dry season saturated zones remain close to leaking sewerage sources.

#### 5.2. Periods of increased risk due to groundwater degradation

Generally speaking, groundwater samples with nitrate levels that exceed the Beninese quality standards for drinking water originated from wells J1, J3, J6, J7, J8, J9 and J11 (piezometer) during the dry season (February and March) in St Jean, from wells L7 and L11 during the rainy season (June and October) in Ladji, and from wells A2, A6 in the dry season (February and November) in Agla. Thus, risk for nitrate pollution in shallow groundwater shows high seasonal variation between sites. Similar results were obtained by Boukari (1998) and Totin et al. (2013) at different sites in Cotonou with higher NO<sub>3</sub> levels (up to 1,61 mmol/L) observed during recharge of the wet season.

The measured EC values show a large amplitude of spatial and temporal variation. The EC values in Agla and St Jean are in the same order of variations (200–1,800  $\mu$ S/cm) whereas the groundwater in Ladji records EC values in the order of 750–5,340  $\mu$ S/cm. In St Jean, the waters are more saline during the dry season. Unlike St Jean, the wells in Agla and Ladji are saline during both the dry and the rainy seasons due to mixing processes with lake water.

## 5.3. Shallow groundwater, an unregulated water resource in expanding urban environments

In urban areas of sub-saharan Africa, groundwater from shallow

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wells is commonly used to partially or fully supply drinking water resources (Okotto et al., 2015). In major cities, such official groundwater resources are monitored and treated. For example, in the coastal city of Douala (Cameroon), shallow groundwater is the main source for domestic and drinking purposes (Takem et al., 2015). Likewise, in Bamako (Mali), around 55% of the population uses water from aquifer resources (British Geological Survey, 2002). Where shallow groundwater is not the official resource, the shallow groundwater usually free and therefore commonly used in the impoverished areas of urban sprawls, including for domestic uses. Normally this resource is meant for washing only, but it commonly ends up for drinking, dish washing, shower and cooking water supplies. For example, the interviews conducted during this study showed that 10 on 10 of the households in St Jean. 9 on 10 in Agla, and 4 on 10 in Ladji reported to use well water only for dish washing. In St Jean, 1 on 10 households declared to use well water for drinking water supply compared with Agla and Ladji where no households have reported such use. The results of the interviews have therefore showed that there are differences in groundwater use between St Jean, Ladji and Agla. The underprivileged areas of the city are subject to greater groundwater quality issues, and this also correspond to the parts of the city where the residents are more dependent on the groundwater as a domestic water resource. This is the case in Kinshasa (Democratic Republic of Congo) where in peri-urban and rural inhabitants widely use the unregulated and untreated shallow groundwater resources for drinking water supply (Ndembo, 2009).

Unfortunately, as seen in this case study of Cotonou, the shallow groundwater in urban environments is often contaminated. This was also observed in Blantyre (Malawi) where drinking water from shallow groundwater was heavily polluted by a lack of sanitation facilities and indiscriminate waste disposal (Mkandawire, 2008). Likewise, high NO<sub>3</sub> concentrations in urban shallow groundwater resources have been observed in Dakar, Senegal (NO<sub>3</sub> > 100 mg/L; Ndeye et al., 2017); Douala, Cameroon (NO<sub>3</sub> up to 241 mg/L; Ketchemen-Tandia et al., 2017) and in different regions of Ghana (NO<sub>3</sub> up to 507 mg/L; Rossiter et al., 2010).

In many urban cases, the nitrate contamination of shallow groundwatear is from anthropogenic sources (Martínez-Santos et al., 2017). This was also observed in this study and previous work in Cotonou (Boukari, 1998; Maliki, 1993; Boukari et al., 1996; Totin et al., 2013), and as also noted in other major sub-saharan cities such as N'djaména, Tchad, (Kadjangaba et al., 2018) and Djibouti (Ahamed et al., 2017). As highlighted in the study of Ouagadougou, Burkina Faso (Yameogo, 2008), such nitrate contamination is linked to the high population density that relies on archaic or non-existent sanitation systems. This is a particularly significant challenge in informal settlements, like in the cities of Douala (Takem et al., 2010; Ketchemen-Tandia et al., 2017) and Kampala, Ouganda (Nyenje et al., 2013) where the increase in nitrates and chlorides in shallow groundwater are related to faeces from pit latrines, sewages, landfills, surface discharges and droppings from domestic animals.

#### 6. Conclusion

Although pollution sources are identical for each of the three neighbourhoods studied, the resultant transfers and reactions controlling concentrations are distinct. In the neighbourhood where there is no surface inundation and acts as a local recharge area, the groundwater salinity values remain low (EC < 1285  $\mu$ S/cm), however NOx concentrations are the highest recorded in this study (up to 5.1 mmol/L). In the neighbourhood bordering a lake where there is seasonal inundation and groundwater discharge, the dry season interaction with lake water results in groundwater with highest observed EC values (up to 5340  $\mu$ S/cm). Stable isotope ( $\delta^2$ H and  $\delta^{18}$ O) mixing ratios indicate mixing of up to 74% with lake water; In the neighbourhood located in a swamp that is subject to inundation during both the small and large wet seasons also shows mixing with lake water (up to 62% using  $\delta^2$ H and  $\delta^{18}$ O)

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values) resulting in groundwater EC values up to 1468  $\mu$ S/cm. This discharge site notably has lower NOx concentrations (up to 0.86 mmol/L), however this is not indicative of less sewerage contamination only greater degradation processes. Understanding the seasonal changes in processes controlling groundwater quality between each site is key to identifying risks to the residents who use this unregulated shallow groundwater resource for domestic purposes, including drinking water supplies. Seasonal variations highlight heightened risks from sewerage and septic tank leakage during the wet season in neighbourhoods located in discharge areas compared with increased risk during the dry season in the recharge area. In addition, there are increased risks of shallow groundwater salinization during both the small wet and dry seasons in discharge areas.

Stable water isotopes showed a direct relationship between local rainfall water and groundwater at St Jean compared with Ladji and Agla where groundwaters are also be subjected to mixing with enriched surface waters such as the lake water (via the lowlands) and the pond water, particularly during the dry and small wet seasons.

Groundwaters chemistry in each of the neighbourhoods are different. In St Jean, the composition of major ion concentrations of waters is dominated by Na-Ca-HCO<sub>3</sub>-Cl groundwater type, while Ladji is of the Na-HCO<sub>3</sub>-Cl type and Agla of the Na-Ca-HCO<sub>3</sub>-SO<sub>4</sub>-HCO<sub>3</sub> one. The ponds sampled in both Ladji and Agla showed some similarities in major ion compositions with local groundwater, thus indicating a potential connection. The Lake Nokoué has greater concentrations of Na and Cl compared with all groundwater samples.

Groundwater samples indicated that 26% for NO<sub>3</sub> and 7.0% for NO<sub>2</sub> do not comply with the Beninese quality standard for drinking water. Based on Cl/Br molar ratios, sources of NOx in groundwater appear to be dominated by infiltration of sewerage and septic tanks in dry season in St Jean, while in Agla and Ladji, contamination was obvious in wet season following infiltrating rainfall and ponds recharging. Low NOx for some of the groundwater samples may indicate effects from biodegradation in discharge area or dilution from rainfall.

#### CRediT authorship contribution statement

Honoré Houéménou: Conceptualization, Writing - original draft, Writing - review & editing, Methodology, Investigation, Formal analysis, Visualization. Sarah Tweed: Conceptualization, Methodology, Writing - original draft, Visualization, Supervision. Gauthier Dobigny: Conceptualization, Supervision, Writing - original draft. Daouda Mama: Supervision. Abdoukarim Alassane: Supervision. Roland Silmer: Resources. Milanka Babic: Resources. Stéphane Ruy: Writing - original draft. Alexis Chaigneau: Writing - original draft. Philippe Gauthier: Investigation. Akilou Socohou: Investigation. Henri-Joël Dossou: Investigation. Sylvestre Badou: Investigation. Marc Leblanc: Supervision, Resources, Project administration.

#### **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.

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#### Appendix A. Supplementary data

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