Research papers

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

Honoré Houéménou, Sarah Tweed, Gauthier Dobigny, Daouda Mama, Abdoukarim Alassane, Roland Silmer, Milanka Babic, Stéphane Ruy, Alexis Chaigneau, Philippe Gauthier, Akilou Socohou, Henri-Joël Dossou, Sylvestre Badou, Marc Leblanc

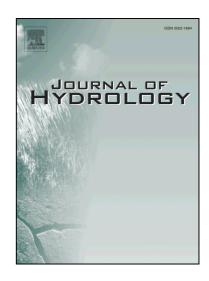
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case study of the unregulated shallow aquifer in Cotonou.

Authors: Honoré Houéménou^{1,5*}, Sarah Tweed², Gauthier Dobigny^{3,4}, Daouda Mama⁵, Abdoukarim Alassane⁵, Roland Silmer¹, Milanka Babic¹, Stéphane Ruy¹, Alexis Chaigneau^{6,7,8}, Philippe Gauthier^{3,4}, Akilou Socohou⁵, Henri-Joël Dossou⁴, Sylvestre Badou⁴, Marc Leblanc¹.

Affiliations

¹University of Avignon, Hydrogeology Laboratory, UMR EMMAH, Avignon, France

²UMR G-eau, IRD, Montpellier, France

³ UMR CBGP, IRD, INRA, Cirad, Montpelllier SupAgro, MUSE, Montpellier France

⁴ Laboratory of Research in Applied Biology, University of Abomey-Calavi, EPAC, Cotonou, Benin

⁵Laboratory of Applied Hydrology, University of Abomey-Calavi, 01 B.P. 4521 Cotonou, Benin

⁶Laboratoire d'Études en Géophysique et Océanographie Spatiale (LEGOS), Université de Toulouse, CNES, CNRD, IRD, UPS, Toulouse, France.

⁷Institut de Recherches Halieutiques et Océanologiques du Bénin (IRHOB), Cotonou, Benin,

⁸International Chair in Mathematical Physics and Applications (ICMPA–UNESCO Chair), University of Abomey-Calavi, Cotonou, Benin

*Corresponding author:

E-mail address: houemenou.honore@univ-avignon.fr

Postal address: 301 rue Baruch de Spinoza, BP 21239, 84911 Avignon Cedex 9, France

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 nonvolontary (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 $\delta^2H-\delta^{18}O$ 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.

Key words: Groundwater, sewerage and septic tank contamination, salinization, urbanization, recharge and discharge.

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, 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 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 et al. 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.

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 (δ^2 H and δ^{18} 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 (Ladji) 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 Figures 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

(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 (1994) and Boukari (1998) have described in detail the different sedimentary units of this coastal sedimentary basin and the distribution are presented in Figure 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

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 Figure 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 still 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 et al. 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). Lake Nokoué and the Atlantic

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 (Hounkpè et al., 2014). Furthermore, the groundwater from the large-diameter private wells is used in 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

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 Figure 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 ~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 (δ^2 H and δ^{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 μ m and acidified with HNO₃), 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 ≤ 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 δ^{18} O and $\pm 1\%$ δ^{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):

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

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 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 Figures 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, in Ladji, the seasonal fluctuations in the water table

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 μS/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 μS/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 μS/cm). In Agla, the groundwater EC ranges between 353-1,468 μS/cm, which is low compared with local surface waters (including temporary ponds and swamps; 2,800-2,900 μS/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 μS/cm) and similar to the local temporary ponds (480-4,090 μS/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 μS/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 Figure 4. In St Jean, the groundwater is of Na-Ca-HCO₃-Cl type. In comparison, the groundwater in Ladji is of Na-HCO₃-Cl type, and Agla groundwater is more of the Na-Ca-HCO₃-SO₄-HCO₃ 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

relative to Cl and SO₄, 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 SO₄ and HCO₃ compared with groundwater.

The temporal evolution of these major ions at each site is presented in Figure 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 HCO₃ (by 530-622 mg/L). These sites are located between 100 to 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 (6,166-1,9310mg/L), and HCO₃ (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 HCO₃ and SO₄ 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), HCO₃ (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), HCO₃ (by 99-436 mg/L), Cl (by 110-176 mg/L), and SO₄ (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 HCO₃ (97-738 mg/L) compared with Agla groundwater samples (Tables 1 and 2).

an increase in Na (by 36-115 mg/L), Ca (by 49-114 mg/L), HCO₃ (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), HCO₃ (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 δ^2H values of rainfall in Cotonou (obtained from 95 samples from the IITA and IAEA Global Network of Isotopes in Precipitation (GNIP) from 2005-2016) has a weighted average of -2.8 ± 1.6 % for $\delta^{18}O$ and -11.2 ± 11.7 % for δ^2H . The local meteorological water line (LMWL) of rainfall obtained from these data has a slope of ~7 and is presented together with the global meteoric water line (GMWL; slope of ~8, Craig, 1961) in Figure 6. These data were also used to calculate the mean weighted average values of $\delta^{18}O$ and δ^2H 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 -30.9 to 14.5 % for $\delta^{2}H$ (Fig. 6a, black crosses). The groundwater values from the three sites are also presented in Figure 6a, which also highlights a large range in values (from -4.17 to 0.35 % for $\delta^{18}O$, and from -21.9 to 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.

-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 δ^{18} O and δ^{2} H values remain close to those of the temporary and permanent ponds. δ^{18} O and δ^{2} H mixing ratios indicate ranges from 72 to 74 % of shallow groundwater mixing with lake water during the dry season in Ladji.

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 NO₃, 3.2 mg/L (0.07 mmol/L) for NO₂, 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 NO₃, and 7.0 % (values of 0.12-2.04 mmol/L) for NO₂ (Table 1).

Highest values of NOx (\sum NO₂+NO₃) are observed in groundwater of St Jean, where the concentrations reach values of 5.06 mmol/L (average of 0.90 mmol/L). For NH₄, 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 <

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 NH₄ 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 NH₄ (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 NH₄ in groundwater is observed in Agla, but only at one site (A10: 4.91 mmol/L). All other sites have NH₄ 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 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 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 Pankrativ, 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 2008; Davis et al. 1998). According to Figure 7b, groundwater values with similar Cl/Br

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 Figure 7b, values that fall into the animal manure/animal urine (Cl/Br molar ratios, 2,810-3,730; 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 Broun et al. (2009), 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 HCO₃, NO_x and DO for all groundwater sampled at Agla is shown in Figure 8. The samples where dissolved oxygen (DO) and NOx are low compared with HCO₃ 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 HCO₃ 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).

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.

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 1,468 µS/cm) in these lowland discharge areas due to mixing with pond water, or can result in lower groundwater salinity (EC < 842 µS/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 et al. (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 and Kadjangaba et al. 2018). Either by reducing NO₃ to HCO₃ (1) or N₂ (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):

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

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

However, denitrification may not be the only process that contributes to the increase of HCO₃ 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 μ 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

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 (Hounkpè 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 NO₃ levels (up to 1,61 mmol/L) observed during recharge of the wet season.

in Agla and St Jean are in the same order of variations (200-1,800 μ S/cm) whereas the groundwater in Ladji records EC values in the order of 750-5,340 μ 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.

In urban areas of sub-saharan Africa, groundwater from shallow wells is commonly used to partially or fully supply drinking water resources (Okotto at 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 NO₃ concentrations in urban shallow groundwater resources have been observed in Dakar, Senegal (NO₃ > 100 mg/L; Ndeye et al. 2017); Douala, Cameroon (NO₃ up to 241 mg/l; Ketchemen-Tandia et al. 2017) and in different regions of Ghana (NO₃ up to 507 mg/L; Rossiter et al. 2010).

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 (Ahmed et al. 2017). As highlighted in the study of Ouagadougou, Burkina Faso (Yaméogo, 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 μ 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 μ S/cm). Stable isotope (δ^2 H and δ^{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 δ^2 H and δ^{18} O values) resulting in groundwater EC values up to 1468 μ 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

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-HCO₃-Cl groundwater type, while Ladji is of the Na-HCO₃-Cl type and Agla of the Na-Ca-HCO₃-SO₄-HCO₃ 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 NO₃ and 7.0 % for NO₂ 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.

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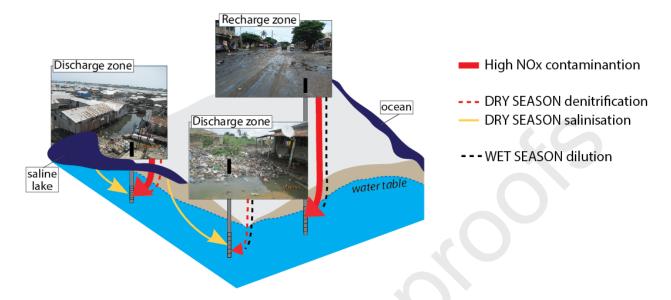
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Abstract

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, CI/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

contamination.

Key words: Groundwater, sewerage and septic tank contamination, salinization, urbanization, recharge and discharge.

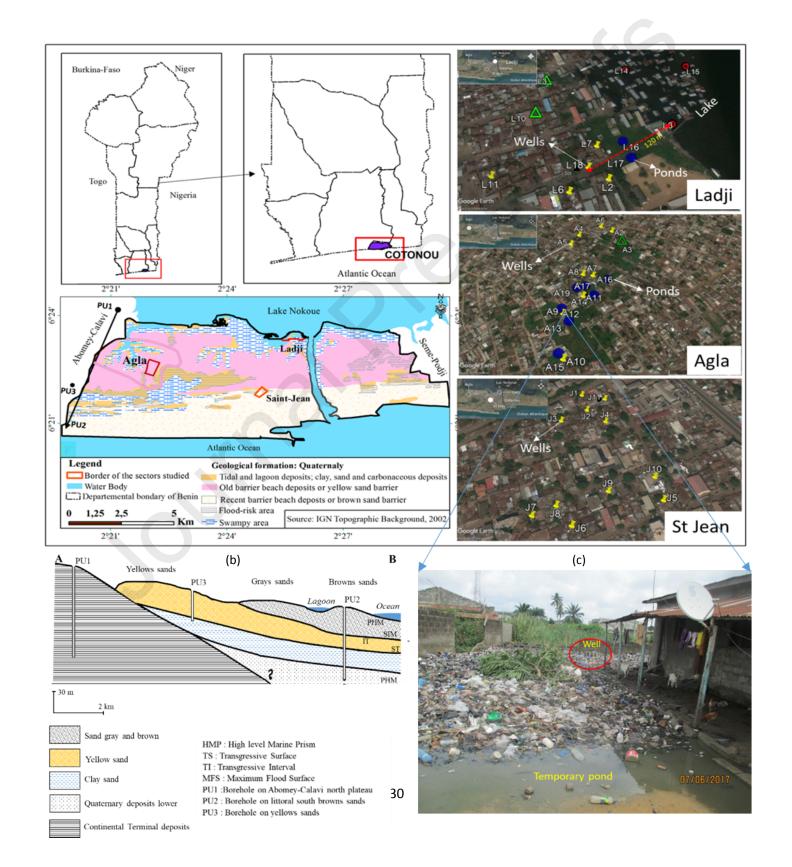


Figure 1. (a) Location of Cotonou and sampling sites in the three neighbourhoods; (b) Sedimentary unit cross section (A-B) of marginal-littoral area of Benin (modified from Boukari, 1998); and (c) Environmental situation at Agla (A9 site)

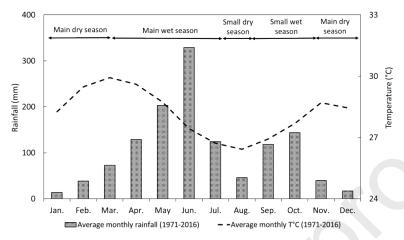


Figure 2: Average monthly interannual rainfall at Cotonou station from 1971 to 2016 (ASECNA station)

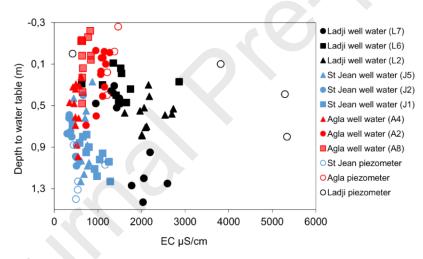
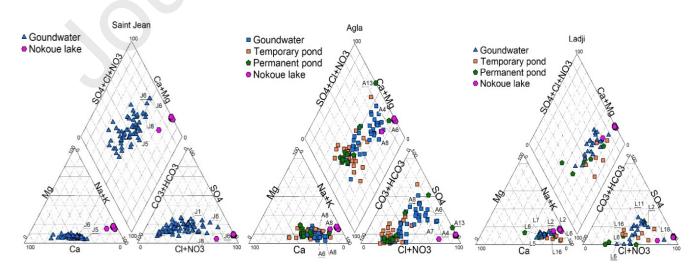


Figure 3. Relationship between water-table variation and electrical conductivity at each site





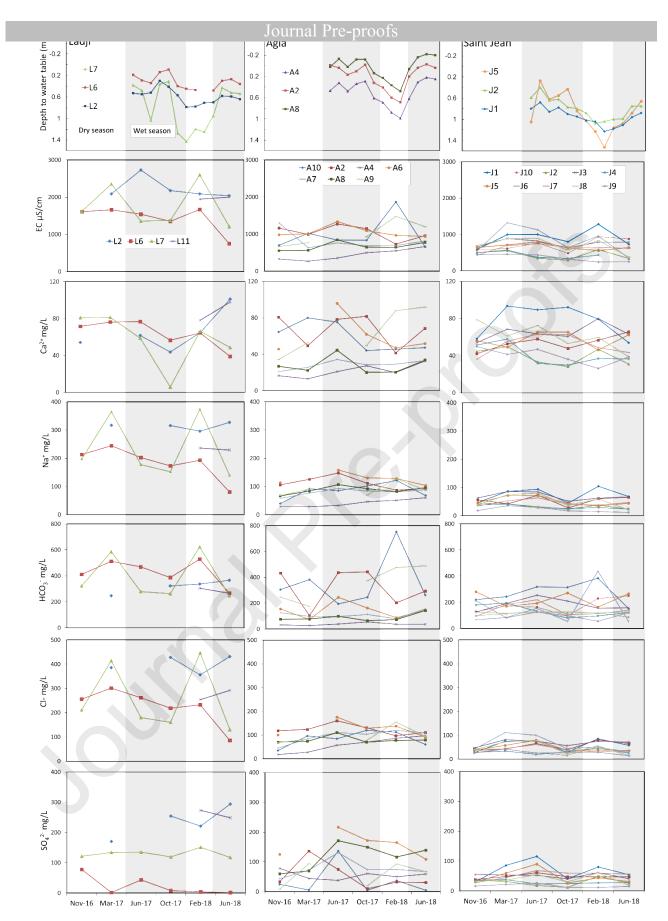


Figure 5. Temporal variation between, depth water table, electrical conductivity, major ion and isotopic stable at each site.

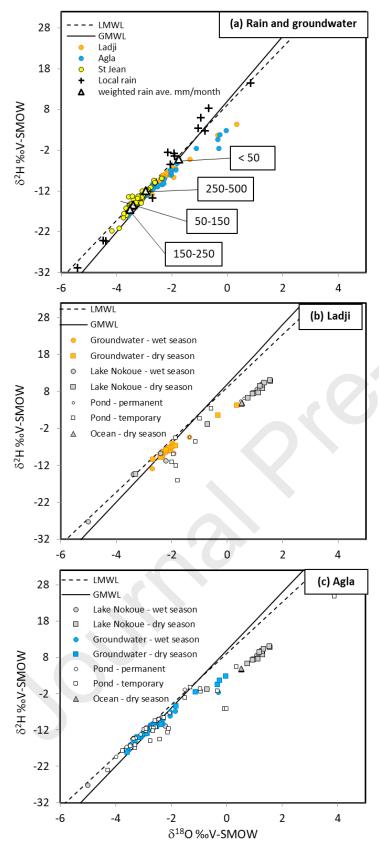


Figure 6. Relationship between ¹⁸O and ²H with Global Meteoric Water Line (GMWL) and Local Meteoric Water Line (GMWL).

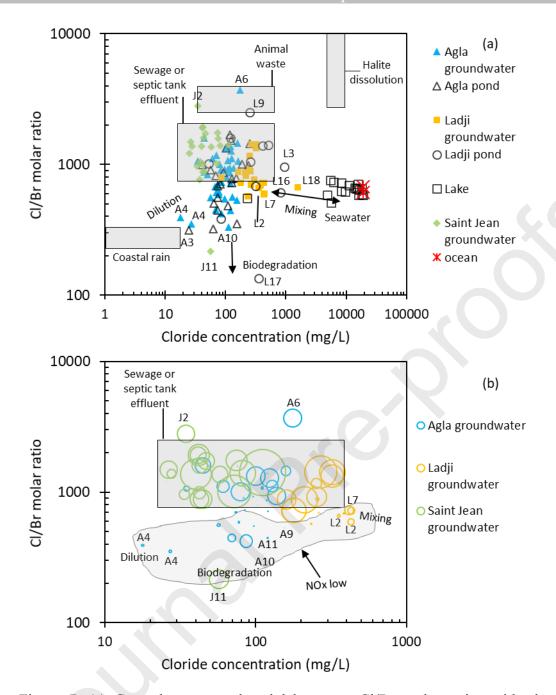


Figure 7. (a) Groundwater, pond and lake water Cl/Br molar ratios with changes in Cl concentrations highlights different sources of Cl in the waters. (b) Relationship between Cl/Br ratios and Cl concentrations showing higher concentrations in NOx (circle sizes are relative to NOx concentrations).

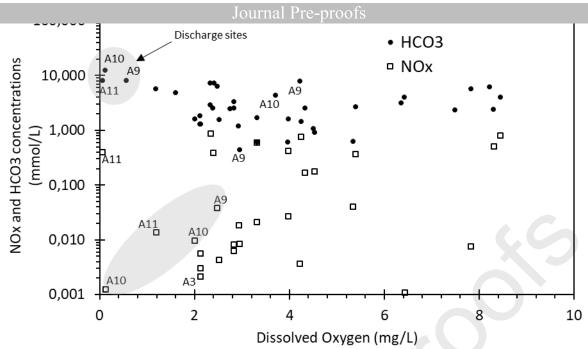


Figure 8. Relationship between HCO3, NOx and DO for all groundwater sampled at Agla

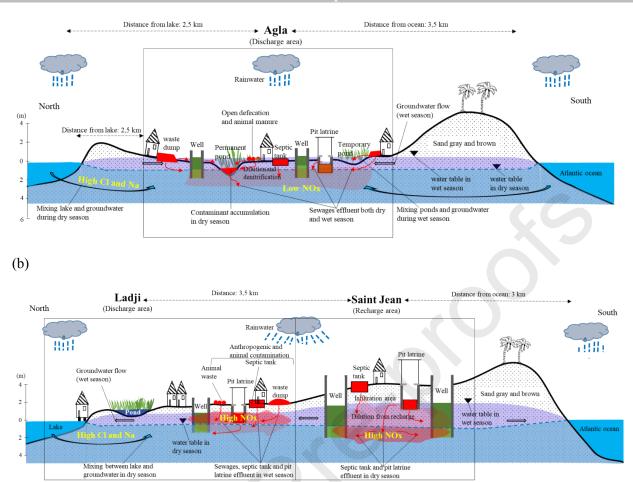


Figure 9. Schematic of conceptual model for contaminant sources and transfers at (a) Agla, and (b) Ladji and Saint-Jean.

Table 1: Chemical parameters and isotopic values in shallow groundwater

								,	Jou	rnal	Pr	e-pi	coof	Ŝ									
Units			m			mg/l	μS/cm		05:						mg/l							9	
J1	Nov-16 Mar-17 Jun-17 Oct-17 Feb-18 Jun-18	well well well well well	0.80 0.90 1.23 0.88	29.50 29.70 28.20 27.90 29.10 29.20	6.97 7.24 7.45 7.57 7.16 7.40	2.59 3.60 3.93 2.35 3.24	590 998 1002 799 1285 731	0.67 1.84 0.44 1.18 1.16	221.49 244.00 318.12 314.76 384.30 152.50	27.59 74.90 77.18 28.52 85.78 57.74	DBL 0.80 DBL 7.13 DBL 1.19	DBL 0.97 DBL DBL DBL DBL	37.68 66.12 DBL 17.67 73.45 7.37	8.69 8.26 6.99 5.68 8.34 4.97	3.90 85.49 115.78 4.87 8.62 54.63	47.20 86.41 94.37 44.68 14.51 68.32	DBL 0.25 DBL DBL 13.65 0.58	19.57 29.43 3.86 22.27 35.65 19.86	5.42 9.19 9.99 9.69 8.76 5.16	58.13 93.52 89.42 92.45 79.28 53.95	10.85 5.89 6.80	-3.07 -2.85 -2.94 -3.89 -3.36 -3.22	-14.04 -12.48 -12.61 -21.22 -16.10 -13.37
J2	Nov-16 Mar-17 Jun-17 Oct-17 Feb-18	well well well well well	0.59 0.77 1.04	29.20 29.00 29.90 28.60 28.60 29.70	6.92 6.93 7.04 7.36 6.88	2.42 3.13 5.13 2.56	502 552 347 297 586	0.76 0.70	99.56 113.46 128.10 121.60 119.56	34.52 42.75 25.67 15.94 52.64	0.12 0.17 DBL DBL 0.43	0.28 0.65 DBL DBL DBL	46.95 37.70 DBL DBL 3.49	6.93 7.13 5.27 6.52 6.40	36.23 31.75 17.90 13.00 49.37	45.47 41.98 31.66 22.97 37.97	0.36 0.30 DBL DBL 0.62	12.67 18.60 7.32 7.66 13.28	2.80 3.24 1.94 1.56 3.14	44.77 49.58 33.24 28.22 46.95	8.30 6.31 5.55	-3.22 -2.81 -3.35 -3.65 -4.17 -3.39	-13.37 -11.64 -15.40 -16.31 -21.87 -15.90
J3	Jun-18 Nov-16 Mar-17 Jun-17 Oct-17 Feb-18	well well well well well	0.75	29.03 29.70 29.20 27.60 29.00 29.20	7.06 7.02 7.15 7.35 5.91 7.00	3.39 4.61 7.85 3.55 3.61	341 658 891 822 630 791	0.24 0.74 1.59 0.63	86.93 125.58 191.54 254.68 29.84 156.16	2.19 46.97 81.73 75.52 56.65 74.76	DBL 0.53 0.31 DBL DBL 0.29	0.61 0.13 0.18 DBL DBI	12.43 45.39 98.13 0.39 DBL 73.98	3.77 9.82 6.28 5.25 5.19 4.26	26.87 54.88 53.17 52.61 4.84 61.19	22.87 62.97 86.30 84.25 51.28 62.85	0.22 1.64 2.25 DBL DBL 0.33	8.45 17.99 21.71 22.82 13.35 15.11	1.79 3.46 4.96 4.27 4.54 6.18	31.72 53.96 68.47 63.45 6.62 79.89	1.17 4.53 5.37	-3.54 -3.01 -3.02 -3.19 -3.56 -3.19	-15.48 -13.16 -12.99 -13.76 -16.94 -13.90
J4	Jun-18 Nov-16 Mar-17 Jun-17 Oct-17	well well well well well		29.40 29.80 29.90 27.90 28.70	6.90 7.11 7.15 7.63 8.06	3.90 4.95 3.98 5.35	792 500 572 368 342	1.12 0.22 0.31 0.33	158.60 18.45 195.20 132.37 81.74	7.88 27.63 33.68 2.34 29.18	DBL 0.77 0.11 0.15 DBL	DBL 0.45 0.79 DBL DBL	69.65 13.48 18.69 DBL DBL	3.83 2.47 2.36 1.99 2.45	41.29 36.46 37.86 23.22 24.79	66.47 37.84 41.44 29.65 25.31	0.38 0.31 0.12 DBL DBL	14.90 17.42 13.45 15.53 7.95	5.79 3.77 6.22 2.62 2.97	63.11 51.35 57.91 31.66 29.79	14.96 4.24	-3.38 -2.37 -2.58 -3.23	-15.19 -8.98 -9.93 -14.08
J5	Feb-18 Jun-18 Nov-16 Mar-17 Jun-17 Oct-17	well well well well well	1.05 0.43	29.20 28.80 29.70 29.70 28.50 27.10	7.15 7.41 6.83 6.89 7.03 7.03	3.33 4.08 3.25 1.96 1.31	434 379 642 715 798 630	0.22 0.33 0.45 0.69 0.48	96.38 122.00 28.68 17.80 193.22 272.60	47.54 25.24 38.17 57.97 79.80 37.12	DBL DBL 0.43 0.22 DBL	DBL 0.82 0.96 DBL 0.19	13.85 18.82 0.28 42.66 DBL DBL	2.27 2.37 0.82 0.21 DBL DBL	27.26 28.23 3.38 59.64 89.97 26.37	28.87 24.75 46.38 72.32 72.19 43.66	0.81 0.48 DBL 0.34 0.58 DBL	3.52 16.77 43.42 37.83 35.18 19.78	3.64 3.00 6.31 3.69 6.18 5.82	36.94 36.36 56.38 49.65 65.25 65.35	3.79 11.57 4.93	-3.07 -3.52 -3.22 -3.06 -3.17 -3.61	-13.61 -15.23 -14.19 -13.09 -13.43 -17.56
J6	Feb-18 Jun-18 Nov-16 Mar-17 Jun-17 Oct-17	well well well well well well	1.53 0.66	28.60 27.80 29.10 29.40 29.20 28.50	6.96 6.97 6.76 6.99 7.06 6.99	2.34 2.03 3.60 5.12 4.40	558 639 548 1326 1129 637	0.46 0.25 1.15 5.67 3.80 1.94	164.70 265.35 66.49 85.40 154.79 54.90	38.37 35.12 41.99 111.13 99.84 44.28	DBL DBL 1.95 54.79 2.28	DBL DBL 0.49 0.18 DBL DBL	2.89 1.53 71.59 311.33 157.26 117.19	DBL DBL 5.27 2.68 2.57 2.33	44.12 31.40 51.16 91.61 87.74 53.18	34.22 44.85 47.46 115.00 99.79 46.00	0.50 0.62 0.65 0.73 0.39 DBL	16.46 19.72 21.98 37.66 4.98 21.69	3.98 5.49 3.87 13.46 1.37 4.32	46.12 62.37 4.27 17.85 87.46 48.44	8.59 16.65 8.31	-2.89 -3.46 -3.24 -2.85 -2.60 -3.28	-12.01 -15.23 -15.12 -11.68 -10.19 -15.11
J7	Feb-18 Jun-18 Nov-16 Mar-17 Jun-17	well well well well well		29.00 29.00 30.10 29.90 30.20	6.97 7.17 6.87 7.05 7.00	4.35 5.96 5.12 3.78	961 346 431 695 742	3.37 0.43 0.74 0.87 0.56	439.20 54.90 93.25 184.22 22.36	51.14 29.35 26.42 44.50 61.44	0.17 DBL 0.19 0.22 0.26	DBL DBL 0.40 0.11 DBL	28.87 24.95 45.76 53.78 DBL	2.62 4.54 3.19 0.79 1.14	47.94 2.49 27.42 44.46 68.21	93.79 26.22 34.79 53.47 63.46	0.16 0.27 0.39 0.18 DBL	29.53 11.87 13.95 2.82 22.43	15.13 2.27 2.89 4.77 5.45	114.74 24.73 36.26 55.50 63.31	8.63 6.80	-3.32 -3.23 -2.85 -3.08 -2.61	-15.88 -13.82 -12.03 -12.98 -9.98
Ј8	Oct-17 Feb-18 Jun-18 Nov-16 Mar-17 Jun-17	well well well well well		28.90 29.00 29.40 29.40 29.70 29.20	7.14 6.90 7.08 6.99 6.78 7.27	4.20 4.19 4.19 3.62 4.67	652 647 616 701 885 894	1.29 1.55 0.66 2.21 2.58 2.56	16.14 113.46 14.30 126.41 115.90 128.86	38.98 3.99 37.00 4.12 72.18 79.23	13.15 0.49 DBL 0.58 0.48 93.70	DBL DBL DBL 0.66 0.16 DBL	57.27 95.59 4.89 136.47 159.26 32.54	0.73 1.18 1.87 4.61 5.87 4.43	59.96 59.66 56.59 32.89 36.16 54.90	4.94 38.17 47.58 38.52 74.78 8.34	DBL 0.23 0.24 0.32 2.78 0.71	15.58 24.95 31.95 18.17 29.87 25.57	5.28 4.69 4.20 5.39 4.94 5.65	62.60 48.66 43.35 78.67 61.28 72.48	4.59 7.08	-3.13 -3.04 -3.38 -3.35 -2.77 -2.92	-14.66 -13.22 -14.29 -15.92 -11.16 -11.84
J9	Oct-17 Feb-18 Jun-18 Nov-16 Mar-17 Jun-17	well well well well well		27.80 28.50 28.40 29.50 30.10 29.10	7.49 6.91 7.65 7.19 6.98 7.20	4.75 3.30 4.56 4.82 4.06	564 840 377 448 461 437	0.99 2.60 0.62 0.34 0.88 0.49	128.10 118.34 128.10 213.58 82.96 126.58	22.92 56.71 14.14 25.43 42.48 24.64	45.57 0.46 DBL 0.23 0.69 8.35	DBL DBL DBL 0.47 0.19 DBL	DBL 16.63 38.59 0.18 54.42 19.20	5.67 6.20 4.18 DBL DBL DBL	3.47 6.15 18.97 16.48 21.57 26.35	36.54 59.92 19.89 18.58 36.85 29.73	DBL 0.32 0.46 4.60 0.33 0.26	19.17 25.37 1.39 2.88 11.14 1.53	3.18 4.63 2.17 3.11 2.94 3.20	53.58 59.75 38.72 49.79 41.27 46.72	5.54 4.09 8.89	-3.64 -3.12 -3.55 -2.96 -2.64 -3.06	-17.99 -13.91 -13.45 -12.98 -9.55 -12.08
J10	Oct-17 Feb-18 Jun-18 Nov-16 Mar-17	well well well well		28.10 28.60 28.60 29.20 28.50	7.21 7.00 7.04 6.95 6.89	4.04 4.63 3.67 2.37	323 242 251 572 615	0.46 0.13 0.15 0.75 0.39	97.60 56.12 122.00 13.63 173.24	27.93 28.38 14.44 41.52 42.31	1.56 DBL DBL 0.32 0.86	DBL DBL DBL 0.49 0.96	0.42 8.23 9.45 43.32 23.27	DBL DBL DBL 2.64 DBL	11.57 11.53 15.59 38.84 48.34	17.19 14.34 11.91 56.63 42.65	0.58 0.60 0.43 0.45 1.74	5.12 7.64 5.34 26.72 37.39	2.22 1.82 1.62 2.86 4.64	36.84 26.23 38.59 42.15 52.68	0.41 4.58	-3.44 -3.74 -3.43 -3.44 -3.17	-15.80 -18.62 -13.35 -16.28 -14.39
J11	Jun-17 Oct-17 Feb-18 Jun-18 Feb-18 Jun-18	well well well well piezometer piezometer	1.40 0.88	29.50 29.30 29.60 30.50 31.30 29.40	7.13 8.03 6.90 6.75 7.21 7.33	3.43 4.94 2.77 0.73	770 481 943 878 497 444	0.99 0.90 0.89 0.65	163.18 98.82 229.36 25.10 92.72 141.52	65.67 39.22 8.94 65.43 57.13 13.61	DBL 0.56 DBL DBL DBL	DBL DBL DBL DBL 0.60	69.32 DBL 6.44 55.54 55.30 4.26	DBL 2.51 0.52 1.52 4.48 6.47	61.25 47.65 47.17 47.69 27.92 22.82	72.53 28.13 6.59 66.77 42.63 13.44	1.46 DBL 14.36 5.41 2.29 0.15	33.84 13.14 28.62 27.62 11.97 12.46	4.70 3.82 5.34 6.57 4.67 4.81	57.83 47.88 56.68 65.57 46.88 49.92	11.67 3.81 7.19 13.50	-3.17 -3.17 -3.14 -3.28 -3.09 -3.73	-13.29 -14.61 -13.91 -14.91 -15.03 -17.66
A2	Nov-16 Mar-17 Jun-17 Oct-17 Feb-18	well well well well well	-0.01 0.04 0.69	28.60 29.90 27.30 26.50 28.30	6.60 6.28 7.21 7.02 6.96	3.32 2.42 2.33 2.83	1164 990 1273 1150 733	0.72 0.87 0.83	432.95 13.70 435.85 441.64 22.52	118.77 123.64 16.53 14.00 97.21	DBL 0.39 DBL DBL DBL	0.38 0.22 0.35 0.53 0.40	0.44 53.39 DBL DBL 0.51	DBL DBL DBL DBL DBL	33.32 136.56 74.85 1.45 32.23	15.85 125.35 147.93 111.34 86.88	0.25 0.23 DBL DBL 0.32	35.77 28.66 35.17 29.87 15.55	1.85 7.45 17.46 15.27 14.75	8.73 49.37 78.28 81.80 41.54	22.27 25.62 14.65	-1.81 -2.72 -2.32 -2.02 -2.90	-5.39 -10.94 -9.17 -7.51 -12.08
A4	Jun-18 Nov-16 Mar-17 Jun-17 Oct-17 Feb-18	well well well well well well	0.04 0.47 0.29 0.99	28.90 29.60 30.90 30.70 28.00 29.50	7.02 6.02 5.85 6.01 5.95 5.84	1.61 2.95 5.34 4.53 3.97	959 326 269 353 497 548	0.27 0.39 0.43 0.18 0.42	292.80 31.95 27.00 37.82 54.90 36.60	111.26 17.78 27.70 56.98 69.62 86.61	DBL 0.35 0.57 DBL DBL DBL	0.12 0.17 0.23 0.35 0.46	DBL 1.63 2.32 2.52 1.95 25.86	DBL DBL DBL DBL DBL DBL	3.56 78.71 44.29 38.15 61.86 5.46	93.69 29.22 29.25 32.98 46.93 51.32	DBL 0.16 0.38 0.54 DBL 0.76	24.14 6.91 6.53 8.24 1.46 1.49	1.24 3.73 3.67 5.30 6.77 7.16	68.46 16.65 13.12 2.99 27.46 2.14	9.86 8.79 10.38	-2.75 -3.22 -3.12 -2.96 -2.68 -2.89	-11.56 -14.01 -14.05 -12.84 -10.44 -12.28
A5	Jun-18 Nov-16 Oct-17 Feb-18 Jun-18	well well well well well	0.25	29.00 30.70 27.20 29.00 29.20	5.76 6.00 7.01 6.55 6.74	3.32 2.75 2.39 4.33	666 1037 500 652 652	0.66 0.27 0.39 0.17	36.60 13.62 15.60 154.94 155.55	97.38 158.50 5.92 6.87 48.84	DBL DBL DBL 0.20 DBL	DBL 0.25 0.13 0.12 DBL	37.26 16.97 DBL 23.85 1.53	DBL DBL DBL DBL DBL	59.26 135.17 19.18 58.67 52.19	6.43 131.15 51.31 67.19 52.72	0.10 0.90 DBL 2.58 0.12	12.96 17.93 9.32 13.73 11.19	1.98 6.68 6.26 9.77 8.15	32.71 37.45 33.27 47.38 4.83	10.06 8.85	-2.81 -2.77 -3.52 -3.18 -3.46	-11.54 -11.89 -17.61 -14.14 -16.07
A6 A7	Nov-16 Jun-17 Oct-17 Feb-18 Jun-18 Nov-16	well well well well well		30.30 29.00 27.60 30.30 29.60 34.00	6.16 7.12 6.50 6.22 6.72 7.51	8.45 5.40 4.25 8.31	983 1338 1095 967 935 1302	0.86 0.90 0.37 0.77 0.59 0.36	155.55 245.53 162.26 87.84 144.88 125.58	1.56 175.75 128.13 137.65 91.65 44.84	1.67 8.61 16.70 DBL DBL DBL	0.17 0.17 0.26 0.33 DBL 0.12	51.84 38.63 0.14 47.68 31.57 0.23	1.38 DBL DBL DBL DBL DBL	125.27 216.32 172.62 165.24 19.87 43.42	113.75 158.45 13.59 128.49 13.93 58.32	DBL DBL 2.42 0.24 1.63	22.17 38.76 28.97 26.57 25.35 13.37	5.98 1.17 6.95 7.92 6.24 3.69	45.76 96.61 62.43 47.19 51.88 21.27	14.60 7.43 7.08	-2.99 -2.02 -2.49 -2.62 -2.66 -3.57	-12.98 -8.14 -10.75 -10.75 -11.51 -18.22
Ai	Mar-17 Jun-17 Oct-17 Feb-18 Jun-18	well well well well well		30.20 30.70 29.10 29.30 28.30	6.21 6.31 6.28 6.20 6.38	2.01 2.52 2.12 2.12 2.83	631 763 687 689 816	0.75 0.42 0.35 0.56 0.63	97.60 95.75 112.24 79.30 155.55	84.84 11.86 13.59 12.42 18.42	DBL 0.52 DBL DBL DBL	0.26 0.28 0.29 0.38 DBL	0.46 0.19 0.19 0.35 0.39	DBL DBL DBL DBL DBL	72.31 131.16 74.26 74.59 67.64	76.93 93.31 84.18 8.83 89.20	0.91 0.68 0.36 1.77 0.89	19.39 16.73 21.11 27.33	8.34 1.26 6.42 8.15 9.20	25.66 34.31 28.45 29.82 32.96	6.80 6.19 7.79	-2.96 -2.92 -2.89 -2.99 -3.02	-13.00 -12.16 -13.15 -13.11 -13.62
A8	Nov-16 Mar-17 Jun-17 Oct-17 Feb-18	well well well well	0.02 -0.12 0.48 -0.20	29.50 29.10 29.70 27.80 28.90	6.20 6.03 6.37 6.03 6.29	2.13 3.98 4.51 2.93	551 560 842 645 642 778	0.25 0.28 0.27 0.18	75.41 78.80 98.67 64.23 73.20 143.35	7.68 73.69 11.17 70.00 77.37 78.94	DBL 0.19 0.19 DBL DBL DBL	0.13 0.24 0.24 0.26 0.30 DBL	0.16 1.26 1.39 DBL 1.14 DBL	DBL DBL DBL DBL DBL DBL	59.96 69.90 171.18 149.72 116.63 139.61	66.77 84.16 16.87 92.14 82.51	0.12 1.28 2.49 0.22 0.88	6.86 7.44 13.32 3.37 5.91 7.69	1.90 2.28 6.34 4.96 4.88 9.16	26.92 22.49 44.49 2.22 2.54 33.59	9.95 7.31 8.89	-2.58 -2.90 -2.87 -2.67 -2.69 -2.64	-10.57 -12.29 -12.41 -10.51 -10.74 -10.29
A9	Jun-18 Nov-16 Mar-17 Oct-17 Feb-18 Jun-18	well well well well well	-0.20	29.10 29.00 29.70 28.90 28.40 27.40	6.15 7.04 6.85 7.78 7.84 7.29	7.49 2.34 8.22 4.22 0.56	765 931 1475 1200	0.84	246.67 174.46 374.83 475.80 488.00	67.77 82.83 77.28 155.00 94.55	DBL 0.53 DBL DBL DBL	0.23 0.26 0.29 0.66 DBL	DBL 0.45 DBL 0.23 DBL	1.46 0.39 4.26 5.15 3.19	8.92 93.83 2.17 93.18 66.50	95.79 67.94 91.12 7.46 127.81 86.96	0.91 DBL 0.34 2.98 5.78 1.48	18.82 1.21 71.94 55.96 53.93	9.32 9.13 15.24 19.57 2.64	34.30 52.77 49.57 87.88 92.00	20.79 26.91	-2.64 -0.33 -2.52 -2.29 -0.03 -2.27	-10.29 0.51 -11.02 -10.29 2.78 -9.04
A10	Nov-16 Mar-17 Jun-17 Oct-17 Feb-18	well well well well		28.50 31.10 30.20 29.20 28.70	6.56 7.55 7.60 7.84 7.77	2.48 6.36 6.43 0.12	700 1023 837 835 1870	0.96 0.96 0.17 0.12	35.84 381.86 193.68 245.22 751.52	34.77 96.39 84.73 119.79 112.19	0.62 0.14 DBL DBL DBL	0.74 0.24 DBL 0.20 0.76	5.14 0.50 DBL 0.66 0.77	3.69 DBL DBL DBL DBL	25.72 5.69 136.69 4.98 37.48	39.59 92.29 85.33 11.90 121.80	9.44 2.52 DBL DBL 88.40	16.33 31.14 22.51 26.97 44.42	5.00 1.53 7.26 8.75 9.35	64.39 8.56 75.40 44.20 45.65	14.76 22.53 26.47	-3.29 -1.11 -1.85 -0.29 -0.26	-14.98 -1.57 -6.87 -1.64 1.72
A11	Jun-18 Feb-18 Jun-18 Mar-17 Oct-17 Feb-18	well piezometer piezometer well well well	0.40 -0.26 0.41 0.71	28.80 32.20 28.20 29.30 26.20 28.00	7.33 6.61 7.23 6.80 6.73 6.93	3.71 1.19 0.06 1.69 3.10 3.75	654 1212 1468 2090 2180 2090	0.14 0.42 0.17 1.67	262.30 346.48 491.66 247.66 323.30 337.94	6.83 12.41 148.36 385.88 428.57 355.74	DBL 0.56 DBL 1.26 1.63 1.22	DBL 0.69 DBL 0.14 0.11 0.39	DBL 0.94 24.94 0.62 5.57 2.40	DBL DBL DBL DBL DBL DBL	4.68 84.86 54.75 17.50 254.40 221.44	67.89 85.96 114.51 317.38 315.83 295.92	DBL 7.45 5.11 2.52 0.48 2.56	14.47 43.27 64.13 53.61 55.56 46.93	6.75 9.63 13.96 32.94 29.42 26.65	47.42 8.93 91.52 54.18 61.73 43.73	23.4 18.69 15.16	-2.03 -2.86 -2.46 -2.21 -2.25 -2.20	-6.96 -13.02 -10.21 -8.43 -8.75 -7.84
L6	Jun-18 Nov-16 Mar-17 Jun-17 Oct-17	well well well well well	0.71 0.64 0.19 0.09	28.00 28.50 29.50 28.70 26.30	6.92 6.87 7.12 7.73 7.69	2.52 2.37 2.29 1.61	2040 1608 1660 1543 1352	0.63	366.61 41.99 512.40 47.50 386.74	431.66 255.80 31.17 261.68 219.13	1.35 0.65 0.89 0.85 0.63	0.15 0.41 0.35 0.96 DBL	8.59 2.57 0.19 0.59 DBL	DBL DBL 2.17 DBL 2.54	294.16 78.11 1.91 43.27 7.87	327.83 213.45 244.35 22.77 172.98	0.23 2.75 1.97 4.32 DBL	62.13 43.28 45.50 38.96 38.28	31.20 23.14 23.73 21.14 17.52	64.83 71.64 76.22 76.80 56.54	24.63 15.12	-2.38 -1.98 -0.33 -2.30 -1.92	-8.85 -6.20 1.60 -9.74 -8.77
L7	Feb-18 Jun-18 Nov-16 Mar-17 Jun-17 Oct-17	well well well well well	0.36 0.38 0.31	28.50 28.60 31.90 30.70 29.40 27.00	7.84 7.39 7.13 7.13 7.69 7.39	1.63 0.56 1.70 7.32 4.32	1669 750 1623 2350 1357 1378	0.13 0.17 0.39 5.46	529.48 266.57 323.22 585.60 279.69 263.52	232.88 85.77 21.94 414.29 18.85 16.51	0.91 0.24 0.55 1.29 0.56 0.41	0.18 DBL 1.66 0.23 1.44 0.90	1.59 DBL 12.69 14.25 89.49 48.44	1.84 1.83 4.45 2.38 4.15 3.49	4.19 1.46 121.78 134.38 135.36 119.67	192.75 8.56 197.25 365.33 177.48 152.12	5.76 0.15 0.65 1.26 0.19 DBL	48.46 26.70 48.74 39.34 44.73 47.78	17.66 9.42 24.86 42.15 18.83 19.40	64.46 39.00 81.14 81.31 58.85 66.91	7.03 5.93	0.35 -1.34 -2.66 -1.99 -2.32 -2.70	4.27 -4.37 -10.42 -6.96 -8.25 -12.96
L11 	Feb-18 Jun-18 Feb-18 Jun-18 Mar-18	well well well well piezometer	1.25 0.54 0.39	29.10 29.70 30.00 29.70 33.40	7.17 7.68 7.00 6.89 6.84	1.98 4.87 3.60 3.32 0.10	2600 1210 1945 2004 5340	0.15 2.51	622.20 248.27 35.00 266.57 4.16	446.98 13.48 253.78 292.14 1561.35	1.68 DBL 0.62 DBL 5.28	DBL 1.21 0.94 1.72 0.87	DBL 75.72 57.98 13.25 0.54	0.83 4.45 2.16 4.33 DBL	151.54 117.46 273.55 248.81 136.77	374.26 14.17 236.26 229.91 832.90	2.51 0.33 2.69 0.76 7.33	31.75 44.64 59.67 7.81 13.91	39.95 15.60 17.98 19.18 63.48	66.38 48.68 78.17 97.83 13.00	13.96 11.85 8.45	-2.11 -3.36 -2.38 -2.41 -1.86	-7.32 -14.50 -9.83 -9.59 -6.72

Table 2: Chemical parameters and isotopic values in surface water

Journal Pre-proofs																						
Units					mg/L	μS/cm	meg/L							mg/L							%	00
A2	Nov-16	pond	32.40	7.02		818		287.54	82.19	BDL	0.26	BDL	1.98	19.41	76.54	0.26	14.21	7.84	62.59		-1.51	-3.18
	Jun-17	pond	25.20	8.72	7.77	113		41.02	5.19	BDL	BDL	BDL	0.60	4.27	6.27	0.41	3.47	0.74	1.79	10.91	-2.13	-12.69
	Oct-17	pond	26.20	7.25	4.65	970	0.00	358.68	108.79	BDL	0.57	BDL	4.49	16.65	96.12	BDL	22.87	1.32	84.99	25.43	-2.65	-12.20
A3	Jun-18	pond	28.80 34.10	6.96 7.51	1.51	740 1242	0.02	380.03 459.10	95.54 149.59	0.74	BDL 0.43	BDL 0.99	2.26	11.58 24.29	79.57 128.17	1.22	52.62 58.34	14.61 13.90	63.34 59.61		-2.89 -0.96	-12.18 -0.63
AS	Nov-16 Jun-17	pond	29.10	8.24	9.53	1619	0.02	408.70	249.90	BDL	0.43	BDL	1.53	154.25	26.27	BDL	67.54	16.88	89.66	36.99	-2.26	-0.63
	Oct-17	pond	29.80	8.43	11.61	346		137.86	24.35	BDL	0.18	BDL	3.41	0.69	29.56	BDL	14.22	4.19	26.47	18.45	-2.41	-14.50
	Jun-18	pond	30.10	7.05	1.07	998		256.20	62.27	BDL	BDL	BDL	0.57	47.74	64.51	0.18	22.14	7.55	57.43		-3.74	-17.80
A6	Jun-17	pond	28.40	8.67	7.45	75		26.00	4.50	0.20	BDL	BDL	BDL	7.13	6.83	BDL	4.84	0.39	5.39	8.12	-1.30	-0.28
A7	Nov-16	pond	28.70	6.20		504		498.68	127.55	BDL	0.37	BDL	4.53	17.86	174.65	BDL	8.33	9.77	37.36		3.88	24.74
	Jun-17	pond	28.00	8.04	6.66	250		103.85	13.47	0.37	BDL	BDL	BDL	15.79	17.78	BDL	6.42	1.99	27.58	7.86	-2.74	-15.01
	Oct-17 Jun-18	pond pond	35.70 30.70	9.05 8.46	15.70 4.33	1070 534		470.31 173.85	92.01 38.68	BDL BDL	0.24 BDL	BDL BDL	4.50 5.77	42.50 18.98	117.64 49.25	BDL BDL	83.62 35.93	17.53 7.92	41.35 21.97	39.75	-0.01 -4.27	-6.12 -23.07
A8	Nov-16	pond	30.20	6.48	4.55	700		218.23	74.45	BDL	0.32	0.12	1.18	77.43	77.99	6.43	19.18	4.98	48.18		-2.97	-11.99
710	Jun-17	pond	35.90	9.64	12.38	1278	0.05	97.60	116.75	0.67	0.16	1.89	BDL	395.69	113.71	0.30	39.75	9.80	131.91	26.26	-0.33	-1.29
	Oct-17	pond	29.70	6.38	2.34	659		147.62	62.96	BDL	0.45	BDL	BDL	8.25	73.47	BDL	13.73	6.78	38.46	15.41	-2.63	-11.50
	Jun-18	pond	26.90	6.80	0.08	695	0.08	253.15	43.38	3.46	BDL	BDL	1.85	52.53	56.39	0.39	24.74	8.47	47.66		-3.45	-16.37
A9	Nov-16	pond	31.10	6.88		764		305.53	74.35	BDL	0.24	BDL	2.13	4.88	71.43	BDL	19.62	12.90	44.65		0.35	5.38
	Jun-17	pond	29.40	7.07	0.01	1643		738.10	102.81	BDL	0.17	BDL BDL	BDL	0.73	75.29	3.32	145.23	33.92	147.66	25.28	-2.09	-11.65
	Oct-17 Jun-18	pond pond	28.00 31.20	8.41 8.58	2.40 10.72	1620 711		581.33 260.47	240.73 69.58	BDL BDL	0.56 BDL	BDL	5.25 0.40	11.54 16.46	166.41 7.35	BDL BDL	162.78 35.68	26.22 12.37	38.85 34.24	51.75	-0.09 -3.29	-6.20 -16.12
A10	Nov-16	pond	30.10	6.69	10.72	1310	0.02	544.58	127.78	BDL	0.44	1.51	0.50	2.21	12.24	0.18	4.32	14.42	73.28		-0.93	-1.43
	Jun-17	pond	31.40	7.48	2.07	1114		314.15	124.09	BDL	0.18	0.42	5.99	136.62	92.28	BDL	5.32	13.56	95.61	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	6.44	62.84	BDL	21.67	7.69	41.23	27.14	-3.30	-16.92
	Jun-18	pond	28.70	7.17	3.29	758		271.45	75.47	BDL	BDL	0.25	BDL	15.37	72.84	0.37	23.60	8.97	54.34		-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
A13	Jun-18 Feb-18	pond pond	28.10 31.90	6.69 4.12	0.17 6.77	605 2900	17.55	176.90 0.00	53.70 256.87	BDL 0.59	BDL BDL	BDL 188.27	1.47 12.89	19.75 24.59	63.75 219.67	0.25 28.23	15.75 95.68	6.33 68.63	35.76 221.89	55.14	-3.98 -2.44	-19.48 -9.10
AIS	Jun-18	pond	29.30	6.72	0.03	537	17.55	247.05	43.53	BDL	BDL	BDL	BDL	6.82	46.36	BDL	12.94	9.75	39.46	33.14	-3.27	-14.28
A14	Feb-18	pond	29.60	7.28	0.01	1671		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 A18	Jun-18 Jun-18	pond	26.10 31.90	7.29 7.44	1.06 7.33	846 563		378.20 250.10	64.69 40.80	BDL BDL	0.29	BDL BDL	4.74 1.45	12.48 16.12	83.16 45.58	BDL BDL	21.60 26.43	9.51 6.77	67.19 42.95		-2.31 -3.60	-8.49 -16.65
A19	Jun-18	pond pond	27.30	7.44	0.19	796		284.26	74.60	BDL	0.92	BDL	1.45	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	1	18630	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 BDL	1.34 0.55	BDL BDL	3.41	31.84 98.67	180.00	BDL	27.68	22.49	35.97	22.60	-1.93	-9.03
L6 L8	Jun-17 Nov-16	pond lake	29.90 29.60	8.17 7.01	0.02	2050 23200		555.56 93.10	255.63 8273.96	BDL	25.72	0.34	14.98 BDL	134.38	275.76 4495.24	27.97 BDL	137.28 158.90	12.36 54.36	28.24 171.41	28.67	-0.98 -0.71	0.71 -0.85
LU	Mar-17	lake	31.00	7.65	5.10	47100	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	38800		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	47300		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
L9	Jun-18	lake	29.70 31.10	7.41 9.85	2.54 11.40	39100		128.71	16114.88	BDL BDL	62.43 0.23	BDL BDL	BDL 2.25	2293.29 89.27	8415.52 195.47	0.47	36.52 64.58	151.93 2.42	276.92 11.95	45.40	-4.99 -0.57	-27.32
L10	May-17 Jun-17	pond pond	31.80	8.16	0.02	1204 2000		156.62 491.97	254.45 408.44	BDL	0.23	BDL	8.95	8.72	295.17	3.12	96.45	15.95	64.45	45.49 27.02	-0.57	3.33 -16.01
210	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	47600		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	39400		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
L15	Feb-18	lake	30.20	7.90	7.29	49700			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	38100		57.95	15173.94	BDL	52.19	BDL	BDL	2165.47	7884.58	0.34	296.81	994.89	254.81	40.07	-1.34	-14.50
L16	Mar-18 Jun-18	pond pond	29.20 29.10	8.65 7.37	1.16 0.01	3830 1024		1115.08 289.75	824.56 120.41	BDL BDL	3.41 0.17	BDL BDL	7.25 6.98	2.78 41.34	698.15 121.28	BDL 0.83	34.17 39.94	73.68 12.58	11.74 39.45	40.37	0.56 -1.34	5.05 -4.37
L17	Mar-18	pond	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	-1.34	-6.86
	Jun-18	pond	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	00.00	0.50	5.33
L19	Jun-18	pond	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				51000		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
AO2	Jan-19	Ocean				51300		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: Below Detection Limit

Highlights

- Predominant drivers of the unregulated shallow aquifer degradation in Cotonou were analysed using major ions, Cl/Br molar ratios and stables isotopes as environmental tracers.
- Vulnerability of this unofficial groundwater resource is due to either salinization or sewage leakage, which is temporally and spatially variable.
- Increasing risks from sewage and septic tank leakage during the wet season in neighbourhoods located in discharge areas compared with increasing risk during the dry season in the recharge area.
- Mixing with pond water and saline Lake Nokoué water highlight increased risks of shallow groundwater salinization during the dry and small wet seasons in discharge areas.

Author Contributions

Honoré Houéménou: Conceptualization, Writing – original draft, Writing – review and 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

Honore HOUEMENOU Avignon University No. 301 Baruch de Spinoza Road, Avignon 84911, France

E-mail: honore.houemenou@univ-avignon.fr

May 28, 2019

To the Editor of Journal of Hydrology Elsevier

Dear Editor,

We would like to submit our manuscript entitled "Degradation of groundwater quality in expanding cities in West Africa. A case study of the unregulated shallow aquifer in Cotonou" for publication in Journal of Hydrology as a research article.

This manuscript fits in the aims and scope of the journal by undertaking original environmental science research in the themes of contaminants, global change, and groundwater hydrogeochemistry.

This study is our own work and no parts have been copied from other manuscripts, including those of the authors; the manuscript is unpublished and is not currently under review with another journal; all data measurements are genuine results and have not been manipulated. There are no conflicts of interest associated with this work. The manuscript has been prepared in accordance with the journal of Hydrology guide for authors.

The authors look forward to receiving the reviews of this manuscript and thank you in advance for your time during this process.

Sincerely yours,

Corresponding author

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