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

3

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

7

8 **Affiliations**

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

10 ²UMR G-eau, IRD, Montpellier, France

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

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

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

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

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

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

19

20 *Corresponding author:

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

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

23

24 **Abstract**

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

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

44

45 **1. Introduction**

46 With cities in developing countries expanding at unprecedented rates, ensuring clean water
47 supplies for all inhabitants is becoming increasingly more challenging. This is particularly the case in
48 more impoverished urban areas, where infrastructure development often lags behind population
49 growth (Lapworth et al., 2017). In addition, there often exist two types of water supplies: firstly, there
50 is the official water supply that is monitored, treated and comes at a cost for consumers; and then there
51 is the second unregulated water supply that is often sourced from wells accessing shallow
52 groundwater that is unmonitored, untreated but free.

53 The health risks in using the unregulated groundwater resource for domestic purposes,
54 including drinking water supplies, is high due to water quality issues in many urban areas (e.g.
55 Ouedraogo et al., 2016; Hassane, 2010). The same factors driving the demand for water supplies,
56 accelerated urban growth and the expansion of informal settlements, are also significant drivers of
57 groundwater pollution (UNESCO, 2017). One of the many threats to shallow groundwater quality in
58 cities is from sanitary wastewater and solid waste disposal (Lu, 2015). Uncontrolled seepage of
59 wastewater from septic tanks and human activities as well as infiltration of urban stormwater lead to
60 groundwater contamination (Dhanasekarapandian et al., 2016). Many parts of the world are now
61 reporting groundwater and surface water nitrate pollution issues (Roy et al. 2007; Stuart et al. 2007;
62 Zhang et al., 2014; Ogrinc et al., 2019). For example, in the Coimbatore city, India, population
63 growth, pit latrines and septic tanks, industrial effluents, and irrigation water return flows are the main
64 sources of groundwater contamination (Selvakumar et al. 2017). In Florida, the proximity of wells to
65 septic tanks contributes to increasing fecal coliform, nitrate and phosphate concentrations during wet
66 season compared with the dry season (Arnade, 1999). Many studies have identified sewerage and
67 latrine contamination as a significant public health issue due to the resultant presence of faecal
68 bacteria in waters such as *Escherichia coli*, faecal *Streptococci*, *Salmonella* and *Shigella* (Boukari,
69 1998; Odoulami et al. 2013; Yadouléton, 2015). During this study, for the first time, the seasonal
70 variation in the contamination of waters by sewerage and latrine contamination is investigated using
71 environmental tracers.

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

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

92

93 **2. Study Area**

94

95 **2.1 Location and climate**

96 The city of Cotonou is bordered by Lake Nokoué to the north, and the Atlantic Ocean to the
97 south. The location of Cotonou and the average monthly interannual rainfall at Cotonou station from
98 1971 to 2016 are shown in Figures 1 and 2. The topography of the city is relatively flat with an
99 altitude varying between 0 m and 6 m (Boukari, 1998). The average annual rainfall for Cotonou is

100 1,300 mm (Yabi et al. 2006). The city's climate is characterised by 4 seasons; main dry season
101 (average rainfall is 25 mm from mid-November to mid-March), main wet season (average rainfall is
102 152 mm from mid-March to mid-July), small dry season (average rainfall is 55 mm in mid-July to
103 mid-September), and small wet season (average rainfall is 75 mm from mid-September to mid-
104 Novembre).

105

106 **2.2 Aquifer geology**

107 Cotonou is located in the coastal sedimentary basin comprised of Quaternary (Holocene)
108 sediments, which include facies of the littoral plain (sands) and alluvial deposits, underlain by
109 sediments from the Mio-Pliocene (Continental Tenuinal), Paleocene and Upper Cretaceous (Maliki,
110 1993). Previous studies by Oyédé (1991), Maliki (1993), Alidou (1994) and Boukari (1998) have
111 described in detail the different sedimentary units of this coastal sedimentary basin and the
112 distribution are presented in Figure 1. The shallow Quaternary sediments consist predominantly of
113 fine to medium sands (85.5%), silts (5.5%) and clays (9%) (Oyédé, 1991). The low clay content in
114 these sediments results in the high permeability of the shallow sandy soils (Maliki, 1993), and
115 therefore a high vulnerability of the aquifers to the transfer of surficial pollutants. The clays in
116 Quaternary sediments are constituted by smectite and a kaolinite content, which increases with depth.
117 The traditional domestic wells were sampled in this study so that we could analyse the water
118 consumed and used by local residents. However, due to the intense pumping of these wells, the
119 fluctuations in hydraulic head are heavily influenced by pumping, therefore we were unable to use the
120 hydraulic head data to infer subsurface flow directions. There has however been previous work that
121 has addressed this question, the work of Maliki (1993) and Boukari (1998) focused on the piezometry
122 of the Cotonou water table during the flood and low water periods, and the interactions with the Lake.
123 It appears from these studies that during the flood period, there is a flow direction of groundwater
124 from the center of Cotonou (piezometric dome) to outlets such as Lake Nokoué, the lagoon, the
125 Atlantic Ocean and swamps. In comparison, during the low-water period it was observed that water
126 from the Lake Nokoué flows into the groundwater aquifer

127

128 **2.3 Sewerage and waste**

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

145 **2.4 Groundwater quality and use**

146 Several studies have evaluated the impact of human activities on the quality of the shallow
147 groundwater in the Cotonou region and have reported relatively high levels of nitrates (e.g. up to 100
148 mg/L; Maliki, 1993; Boukari, 1998; Odoulami et al. 2013; Totin et al. 2013). In addition, the
149 vulnerability of this groundwater resource is also related to its proximity to the Atlantic Ocean and the
150 saltwater or brackish lakes (Boukari, 1998; Totin et al. 2013). Data from previous studies have shown
151 an increase in salinity levels in groundwater near Lake Nokoué. The salinity of the Lake Nokoué also
152 exhibits a marked seasonal cycle (0 psu during the small wet season in October and 25-30 psu at the
153 end of the dry season) (Stephen et al. 2010; McInnis et al. 2013; Totin et al. 2013). The coastal
154 groundwater system in Cotonou has relatively high chloride (23.6 to 160 mg/L) and sulphate (6.4 to
155 25.7 mg/L) concentrations (Alassane et al. 2015; Nlend et al. 2018). Lake Nokoué and the Atlantic

156 Ocean are therefore a major concern in terms of the source of salinization of groundwater resources in
157 the coastal zone (Alassane et al. 2015).

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

164 Groundwater from the shallow Quaternary aquifer, is accessed by the residents of Cotonou via
165 large-diameter wells installed on private household property. These large-diameter wells are shallow
166 and are designed to pump groundwater from the water table surface so as not to pump deeper saline
167 groundwater (Maliki, 1993). The Quaternary aquifer sands have a high permeability (in the order of
168 10^{-2} to 10^{-4} m/s) and locally contains fresh to brackish groundwater whose exploitation is related to the
169 position of the lake water and seawater intrusions, and to the replenishment of freshwater lenses.

170 In Cotonou, 81% of the neighbourhoods have wells, and whilst 9% have not any water supply
171 (no wells nor stand pipe for drinking water supply) for domestic purposes, they sometimes buy water
172 for drinking purposes from neighbours (Houngpè et al., 2014). Furthermore, the groundwater from the
173 large-diameter private wells is used in households for drinking, cooking, laundry, bathing and washing
174 dishes (Yadouléton, 2015; EAA, 2018).

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

178

179 **3. Methods**

180 Three neighbourhoods in Cotonou were selected for sampling based on differences in their
181 hydrogeological environments: (i) St Jean, where there is no long-term surface inundation and the
182 heavy rainfall infiltrates the soils or leaves the site via overland flow; (ii) Ladji, which is located at the

183 shore of Lake Nokoué and is inundated towards the small wet season; and (iii) Agla, which is located
184 in a flood plain and is inundated early during both the small and large wet seasons. The locations of
185 these three neighbourhoods and sampling sites are presented in Figure 1. In each neighbourhood,
186 groundwater wells were selected for sampling and, where/when possible, surface water samples
187 (permanent and temporary pools, as well as Lake Nokoué) were also included. The depths to the water
188 table and the electrical conductivity (EC) of groundwater in the Quaternary aquifer were monitored
189 monthly at 9 wells and 3 piezometers from each of the 3 neighbourhoods from June 2017 to June
190 2018. The wells are separated by an average distance of ~200 m within the same neighbourhood
191 probe.

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

200 Water samples were collected for the analyses of cations (filtered at 0.45 μm and acidified
201 with HNO_3), anions and stable isotopes, that were conducted at the Laboratory of Hydrogeology of the
202 University of Avignon, France. Major ions were analysed using ion chromatography (Dionex;
203 ICS1100 and autosampler AS-AP). Ion analysis uncertainty is in the order of 3 %, and all ionic
204 balances were ≤ 5 %. The alkalinity was measured using a HACH digital titrator, and stable isotopes
205 were analysed using a Picarro Analyser L 2130-I. For the stable isotopes of waters, the error is $\pm 0.1\%$
206 for $\delta^{18}\text{O}$ and $\pm 1\%$ $\delta^2\text{H}$. The results are presented in Tables 1 and 2. The mixing ratio between lake and
207 shallow groundwater is calculated by using stable isotopes values according to Paran et al. (2015):

$$208 \quad \delta^{18}O_m = f \cdot \delta^{18}O_L + (1 - f) \cdot \delta^{18}O_{GW}.$$

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

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

217

218 **4. Results**

219 **4.1 Seasonal variations of the water table and groundwater EC**

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

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

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

243 With seasonal variations in the water table depth, the groundwater EC also shows varying
244 trends (Fig. 3). In St Jean, the groundwater EC values (297-1,285 $\mu\text{S}/\text{cm}$) are relatively weak
245 compared to Agla and Ladji, and show variable seasonal changes (Fig. 3). Wells J1 and J2 recorded
246 higher EC values in the dry season (511-1,285 $\mu\text{S}/\text{cm}$) when groundwater levels were low compared
247 with the wet season (297-799 $\mu\text{S}/\text{cm}$). In comparison, the well J5 and the piezometer had higher EC
248 values (444-1,169 $\mu\text{S}/\text{cm}$) in the rainy season (rise of the water table) compared with the dry season
249 (497-685 $\mu\text{S}/\text{cm}$). In Agla, the groundwater EC ranges between 353-1,468 $\mu\text{S}/\text{cm}$, which is low
250 compared with local surface waters (including temporary ponds and swamps; 2,800-2,900 $\mu\text{S}/\text{cm}$). EC
251 values are similar during the rainy season and the dry season (Fig. 3). In Ladji, the groundwater EC
252 values are higher than in the other neighbourhoods (633-5,340 $\mu\text{S}/\text{cm}$) and similar to the local
253 temporary ponds (480-4,090 $\mu\text{S}/\text{cm}$), but strongly lower than the Lake Nokoué (up to 49,700 $\mu\text{S}/\text{cm}$
254 during dry season). With the exception of well L2, no difference was observed in the variation of the
255 EC during the dry season and the rainy season (1,478-5,340 $\mu\text{S}/\text{cm}$).

256

257 **4.2 Seasonal variations of major ions**

258 The composition of major ion concentrations of waters is different in each of the
259 neighbourhoods as highlighted in Figure 4. In St Jean, the groundwater is of Na-Ca-HCO₃-Cl type. In
260 comparison, the groundwater in Ladji is of Na-HCO₃-Cl type, and Agla groundwater is more of the
261 Na-Ca-HCO₃-SO₄-HCO₃ type (Fig. 4). The ponds sampled in both Ladji and Agla show some
262 similarities in major ion compositions with local groundwater, thus indicating a potential connection
263 between surface and subsurface waters. However, this connection is spatially and/or temporally

264 variable, in particular in Agla, since some of the pond samples have a greater concentration of HCO_3
265 relative to Cl and SO_4 , and increased Ca relative to Na and Mg compared with groundwater. In
266 addition, in Ladji, the Lake Nokoué has a greater ratio of Cl to SO_4 and HCO_3 compared with
267 groundwater.

268 The temporal evolution of these major ions at each site is presented in Figure 5 and, similar to
269 EC values, the seasonal changes vary both between sites and between neighbourhoods. Greatest
270 seasonal variations are observed in Ladji, and particularly for groundwater sampled at L6 and L7
271 where dry season conditions results in increases in Na (by 244-374 mg/L), Ca (by 76-81 mg/L), Cl (by
272 301-447 mg/L) and HCO_3 (by 530-622 mg/L). These sites are located between 100 to 150 m from the
273 Lake shore. The increase of the ions' concentration during dry season is associated with a deepening
274 of the water table and may therefore indicate influences of lake infiltration. The Lake Nokoué has
275 greater concentrations of Na (3,168-12,219 mg/L), Ca (146-419 mg/L), Cl (6,166-1,9310mg/L), and
276 HCO_3 (73-110 mg/L) compared with all groundwater samples (Tables 1 and 2).

277 In Agla, the seasonal variations in major ions remain relatively stable except for HCO_3 and
278 SO_4 concentrations (Fig. 5). However, between sites, there are large differences in seasonal trends.
279 Four of the eight groundwater wells (A4, A5, A9 and A10) show an increase in Na (by 47-128 mg/L),
280 Ca (by 20-88 mg/L), HCO_3 (by 155-750 mg/L) and Cl (by 61-155 mg/L) during the month of
281 February (dry season). During the wet seasons (June and October/November), the remaining four
282 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
283 99-436 mg/L), Cl (by 110-176 mg/L), and SO_4 (by 75-276 mg/L) concentrations. These trends
284 correspond to changes in the water table depths, where the increasing wet season major ion
285 concentrations occurs in wells where the groundwater is (sub)artesian during the wet season (A2 and
286 A8), compared to wells where groundwater remains well below the surface and has increased dry
287 season major ion concentrations (A4). These trends may highlight the influence of groundwater
288 mixing with pond water in the discharge areas, compared with infiltrating rainfall diluting
289 groundwater in other areas during the wet season. The temporary and permanent pond waters have
290 similar concentrations of Na (56-166 mg/L), Ca (34-147 mg/L), Cl (43-240 mg/L), and HCO_3 (97-738
291 mg/L) compared with Agla groundwater samples (Tables 1 and 2).

292 In St Jean, groundwater at seven of the ten wells sampled (J1, J2, J10, J3, J4, J6 and J9) show
293 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-
294 111 mg/L) concentrations during the dry season (February and March, Fig. 5). This occurs at the time
295 when the water table is at its deepest level. In addition, small wet season peaks in major ion
296 concentrations are also observed in groundwater from wells J5, J7 and J8. These increases include Na
297 (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)
298 concentrations. Since St Jean does not have any surface water bodies, the wet and dry season major
299 ion variations are probably due to variations in the composition of infiltrating water, mixing between
300 subsurface waters, reactions during water table fluctuations, and evaporation.

301

302 4.3 Stables isotopes

303 The $\delta^{18}\text{O}$ and $\delta^2\text{H}$ values of rainfall in Cotonou (obtained from 95 samples from the IITA and
304 IAEA Global Network of Isotopes in Precipitation (GNIP) from 2005-2016) has a weighted average
305 of $-2.8 \pm 1.6 \text{ ‰}$ for $\delta^{18}\text{O}$ and $-11.2 \pm 11.7 \text{ ‰}$ for $\delta^2\text{H}$. The local meteorological water line (LMWL) of
306 rainfall obtained from these data has a slope of ~ 7 and is presented together with the global meteoric
307 water line (GMWL; slope of ~ 8 , Craig, 1961) in Figure 6. These data were also used to calculate the
308 mean weighted average values of $\delta^{18}\text{O}$ and $\delta^2\text{H}$ for different ranges in monthly rainfall. The results
309 show that when monthly rainfall is < 50 mm, the isotope values are greater compared with monthly
310 rainfall ranging from 50 mm to 500 mm (Fig. 6a). The isotope values of rainfall collected in Cotonou
311 show an overall mass effect; a depletion in heavy isotopes correlates with an increase in rainfall
312 amounts. In Cotonou, the local rainfall has stable isotope values covering a large range; from -5.4 to
313 0.8 ‰ for $\delta^{18}\text{O}$, and from -30.9 to 14.5 ‰ for $\delta^2\text{H}$ (Fig. 6a, black crosses). The groundwater values
314 from the three sites are also presented in Figure 6a, which also highlights a large range in values (from
315 -4.17 to 0.35 ‰ for $\delta^{18}\text{O}$, and from -21.9 to 4.3 ‰ for $\delta^2\text{H}$).

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

320 In Ladji, groundwater has enriched in $\delta^{18}\text{O}$ and $\delta^2\text{H}$ compared with St Jean (-3.36 to 0.35 and
321 -12.96 to 4.17 ‰, respectively; Fig. 6b). This discrepancy may be due to three distinct phenomena: (i)
322 the recharge of relatively low volume rainfall events (< 50 mm/month) in Ladji compared with St
323 Jean; (ii) evaporation effects (some values lie to the right of the LMWL indicating a slope of 5.2), or
324 (iii) groundwater mixing with lake water. During the dry season, groundwater isotopic values are
325 close to the lake Nakoue values (Fig. 6b), which suggests a strong mixing between the lake and
326 groundwater during this season. There are also a number of groundwater samples whose $\delta^{18}\text{O}$ and $\delta^2\text{H}$
327 values remain close to those of the temporary and permanent ponds. $\delta^{18}\text{O}$ and $\delta^2\text{H}$ mixing ratios
328 indicate ranges from 72 to 74 % of shallow groundwater mixing with lake water during the dry season
329 in Ladji.

330 Like in Ladji, groundwater in Agla is also enriched in $\delta^{18}\text{O}$ and $\delta^2\text{H}$ (-3.57 to -0.03, and -18.22
331 to 2.78 ‰, respectively) compared with St Jean. Some samples from Agla (mostly during the dry
332 season) lie to the right of the MWL with a slope of 5.4 (Fig. 6c). Therefore, these groundwaters may
333 also be subject to evaporation and mixing with enriched surface waters such as the lake water (via the
334 lowlands) and the pond water. In addition, groundwaters (e.g., wells A9 and A10) that are close to
335 lake water are located in discharge areas. $\delta^{18}\text{O}$ and $\delta^2\text{H}$ mixing ratios indicate ranges from 54% to
336 62% of shallow groundwater mixing with lake water in Agla.

337

338 **4.4 Nitrogen**

339 The Beninese quality standard for drinking water for nitrogen is 45 mg/L (0.70 mmol/L) for
340 NO_3 , 3.2 mg/L (0.07 mmol/L) for NO_2 , and 250 mg/L (7.0 mmol/L) for Cl (Decree N ° 2001-094 of
341 February 20th). In this study, 26 % (values of 0.73-5.06 mmol/L) of the groundwater sampled exceeds
342 these limits in terms of NO_3 , and 7.0 % (values of 0.12-2.04 mmol/L) for NO_2 (Table 1).

343 Highest values of NO_x ($\sum\text{NO}_2+\text{NO}_3$) are observed in groundwater of St Jean, where the
344 concentrations reach values of 5.06 mmol/L (average of 0.90 mmol/L). For NH_4 , the groundwater
345 concentrations remain low (up to 0.80 mmol/L, average of 0.04 mmol/L). The highest concentrations
346 of NO_x are observed during the dry season (> 3 mmol/L) compared with wet season samples (NO_x <

347 3.37 mmol/L). Two sites (J6 and J8) have elevated NO_x concentrations in both the wet and dry
348 seasons (1.15-5.06 and 0.99-2.58 mmol/L, respectively). NO_x in St Jean groundwater may originate
349 from natural fixation of nitrogen in the soil and human pollution via leaky latrines and human
350 defecation in the streets.

351 In Ladji, the groundwater NO_x concentrations ranges up to 2.51 mmol/L (average of 0.54
352 mmol/L), and NH₄ is up to 0.43 mmol/L (average of 0.11 mmol/L). During the wet season, the
353 groundwater NO_x concentrations are highest (1.06-2.51 mmol/L) compared to dry season
354 concentrations (<0.13 mmol/L). The wet season increase in NO_x of groundwater in Ladji may either
355 be due to infiltration of anthropogenic pollution, or nitrogen fixed by vegetation in the soils. The NO_x
356 concentrations are low in the permanent ponds and the lake (<0.01 mmol/L). However, the wet season
357 temporary ponds have high concentrations of NH₄ (up to 1.55 mmol/L), which may undergo
358 nitrification during infiltration and may also result in the NO_x contamination of groundwater.

359 The highest value of NH₄ in groundwater is observed in Agla, but only at one site (A10: 4.91
360 mmol/L). All other sites have NH₄ concentrations in groundwater ranging from 0.00 to 0.52 mmol/L
361 (average of 0.17 mmol/L). Compared with Ladji and St Jean, groundwater NO_x concentrations in
362 Agla are low, with a maximum of 0.86 mmol/L (average of 0.15 mmol/L). In contrast, permanent
363 ponds in the Agla lowlands have high NO_x levels only at one site (A13: 17.50 mmol/L), particularly
364 in the dry season.

365 As described by Katz et al. (2011), the Cl/Br ratio can be a valuable first assessment of septic
366 tank contamination of shallow groundwater. This is based on the assumption that sewerage waters and
367 septic tank effluent exhibit distinct ranges and higher values of Cl/Br molar ratios and Cl
368 concentrations compared with rainwater (Davis et al. 1998), as observed in many samples from this
369 study (Fig. 7a). Elevated Cl/Br ratios may also result from the dissolution of halite. However, halites
370 have not been reported in the local Quaternary aquifer. In addition, many of the groundwater values
371 correspond to ranges reported for sewerage and septic tank effluent (Davis et al. 1998; Vengosh and
372 Pankrativ, 1998; Katz et al. 2011) compared with waters with halite dissolution (Davis et al. 1998;
373 Pastén-Zapata et al. 2014; Panno et al. 2006), rainwater from coastal areas, and seawater (Alcalá and
374 Custodio 2008; Davis et al. 1998). According to Figure 7b, groundwater values with similar Cl/Br

375 ratios and Cl concentrations than sewerage wastewater also show higher concentrations of NO_x. Three
376 samples (wells J2, L9 and A6) plot above the Cl/Br ratios for sewage or septic tank effluent (Cl/Br
377 molar ratios 676-1350; Cl <1000 ml/L), likely indicating animal manure. As evidenced in Figure 7b,
378 values that fall into the animal manure/animal urine (Cl/Br molar ratios, 2,810-3,730; Cl <1000 mg/L)
379 also have high NO_x concentrations (0.76-0.81 mmol/L). Other evidence for a septic tank and animal
380 manure influence are the corresponding elevated dissolved organic carbon (DOC) concentrations
381 (Table 1). However, not all the groundwater samples have high DOC concentrations. According to
382 Broun et al. (2009), rapid oxidation of DOC into carbon dioxide may account for the low DOC
383 concentration in groundwater.

384 Alternatively, samples with low NO_x concentrations and relatively low Cl/Br ratios, such as
385 groundwater from Agla (Fig.7b), can indicate areas of organic biodegradation (e.g. McArthur et al.,
386 2012). The relationship between HCO₃, NO_x and DO for all groundwater sampled at Agla is
387 shown in Figure 8. The samples where dissolved oxygen (DO) and NO_x are low compared with HCO₃
388 correspond to discharge areas. This is especially the case for groundwater sampled during the dry
389 season at wells A10, A9 and piezometers A11, J11 where HCO₃ values increases (4.80-12.31
390 mmol/L) and NO_x decreases (0.0-0.40 mmol/L) with low DO values (0.06-4.22 mg/L). However, this
391 may also indicate areas of recent rainfall infiltration resulting in a dilution of NO_x concentrations (e.g.
392 for well A4).

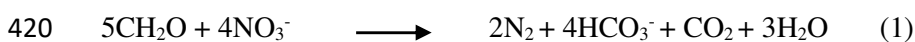
393

394 **5. Discussion**

395 **5.1 Variations in the drivers of groundwater degradation**

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

401 In Agla, a strong driver of groundwater quality is the proximity to lowlands. Agla is scattered
 402 by these low elevation zones, which have either temporary or permanent ponds where groundwater
 403 seasonally discharges. As highlighted by the major ion results, wet season increases in the water table
 404 can either result in increased groundwater salinity (EC up to 1,468 $\mu\text{S}/\text{cm}$) in these lowland discharge
 405 areas due to mixing with pond water, or can result in lower groundwater salinity (EC < 842 $\mu\text{S}/\text{cm}$)
 406 due to the dilution effect from infiltrating rainfall. During the dry season, the groundwater stable
 407 isotope values also highlight increased mixing with pond water and saltier Lake Nokoué water.
 408 Elevated NO_x concentrations in groundwater at Agla is due to sewerage contamination and was
 409 recorded during both dry and wet seasons (NO_x concentrations up to 0.86 mmol/L). Groundwater
 410 contamination from sewerage may infiltrate directly from leaky latrines or from mixing with the
 411 permanent ponds, which have accumulated NO_x concentrations up to 17.50 mmol/L. According to
 412 Starr et al. (1993), denitrification tends to occur in aquifers with very shallow groundwater compared
 413 to aquifers with deeper groundwater (more than 2 m). Low dissolved oxygen levels combined with
 414 low NO_x levels in shallow wells (even less than 1.0 m) in the study area (Fig. 8) where higher levels
 415 can be expected, may be related to biodegradation or denitrification processes (Postma et al. 1991,
 416 Jorgensen et al. 2004, Hassane et al. 2016 and Kadjangaba et al. 2018). Either by reducing NO₃ to
 417 HCO₃⁻ (1) or N₂ (2). According to Postma et al. (1991) and Anornu et al. (2017), the absence or low
 418 concentrations of NO₂ and NH₄ in groundwater is probably related to the reduction of NO₃ to N₂. The
 419 overall denitrification process can be described as (Berner, 1980):



422 However, denitrification may not be the only process that contributes to the increase of HCO₃⁻
 423 in shallow groundwater, which probably also results from the decomposition of organic matter and
 424 mineral dissolution reactions (e.g. Alassane et al. 2015).

425 In Ladji, the main influence on groundwater salinization (EC up to 5,340 $\mu\text{S}/\text{cm}$) is the
 426 mixing with the Lake Nokoué waters. This was obvious during the dry season where both stable
 427 isotopes values and major ion concentrations clearly highlight mixing, and the water level also shows
 428 an early dry season rise in values due to lake infiltration. In comparison, the sewerage contamination

429 of groundwater is mostly evidenced during the wet season (groundwater NO_x up to 2.5 mmol/L).
430 During the wet season, increases in NO_x from sewerage sources may result from both (i) rises in the
431 water table mobilising NO_x in the unsaturated zone, and/or (ii) infiltrating rainfall and temporary
432 ponds recharging the shallow groundwater (as suggested by stable isotope results) and therefore
433 transporting NO_x to the saturated zone.

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

448

449 **5.2 Periods of increased risk due to groundwater degradation**

450 Generally speaking, groundwater samples with nitrate levels that exceed the Beninese quality
451 standards for drinking water originated from wells J1, J3, J6, J7, J8, J9 and J11 (piezometer) during
452 the dry season (February and March) in St Jean, from wells L7 and L11 during the rainy season (June
453 and October) in Ladji, and from wells A2, A6 in the dry season (February and November) in Agla.
454 Thus, risk for nitrate pollution in shallow groundwater shows high seasonal variation between sites.
455 Similar results were obtained by Boukari (1998) and Totin et al. (2013) at different sites in Cotonou
456 with higher NO₃ levels (up to 1,61 mmol/L) observed during recharge of the wet season.

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

462

463

464 **5.3 Shallow groundwater, an unregulated water resource in expanding urban environments**

465 In urban areas of sub-saharan Africa, groundwater from shallow wells is commonly used to
466 partially or fully supply drinking water resources (Okotto et al. 2015). In major cities, such official
467 groundwater resources are monitored and treated. For example, in the coastal city of Douala
468 (Cameroon), shallow groundwater is the main source for domestic and drinking purposes (Takem et
469 al. 2015). Likewise, in Bamako (Mali), around 55 % of the population uses water from aquifer
470 resources (British Geological Survey, 2002). Where shallow groundwater is not the official resource,
471 the shallow groundwater usually free and therefore commonly used in the impoverished areas of urban
472 sprawls, including for domestic uses. Normally this resource is meant for washing only, but it
473 commonly ends up for drinking, dish washing, shower and cooking water supplies. For example, the
474 interviews conducted during this study showed that 10 on 10 of the households in St Jean, 9 on 10 in
475 Agla, and 4 on 10 in Ladji reported to use well water only for dish washing. In St Jean, 1 on 10
476 households declared to use well water for drinking water supply compared with Agla and Ladji where
477 no households have reported such use. The results of the interviews have therefore showed that there
478 are differences in groundwater use between St Jean, Ladji and Agla. The underprivileged areas of the
479 city are subject to greater groundwater quality issues, and this also correspond to the parts of the city
480 where the residents are more dependent on the groundwater as a domestic water resource. This is the
481 case in Kinshasa (Democratic Republic of Congo) where in peri-urban and rural inhabitants widely
482 use the unregulated and untreated shallow groundwater resources for drinking water supply (Ndembo,
483 2009).

484 Unfortunately, as seen in this case study of Cotonou, the shallow groundwater in urban
485 environments is often contaminated. This was also observed in Blantyre (Malawi) where drinking
486 water from shallow groundwater was heavily polluted by a lack of sanitation facilities and
487 indiscriminate waste disposal (Mkandawire, 2008). Likewise, high NO₃ concentrations in urban
488 shallow groundwater resources have been observed in Dakar, Senegal (NO₃ > 100 mg/L; Ndeye et al.
489 2017); Douala, Cameroon (NO₃ up to 241 mg/l; Ketchemen-Tandia et al. 2017) and in different
490 regions of Ghana (NO₃ up to 507 mg/L; Rossiter et al. 2010).

491 In many urban cases, the nitrate contamination of shallow groundwater is from
492 anthropogenic sources (Martínez-Santos et al. 2017). This was also observed in this study and previous
493 work in Cotonou (Boukari, 1998; Maliki, 1993; Boukari et al., 1996; Totin et al. 2013), and as also
494 noted in other major sub-saharan cities such as N'djaména, Tchad, (Kadjangaba et al. 2018) and
495 Djibouti (Ahmed et al. 2017). As highlighted in the study of Ouagadougou, Burkina Faso (Yaméogo,
496 2008), such nitrate contamination is linked to the high population density that relies on archaic or non-
497 existent sanitation systems. This is a particularly significant challenge in informal settlements, like in
498 the cities of Douala (Takem et al. 2010; Ketchemen-Tandia et al. 2017) and Kampala, Ouganda
499 (Nyenje et al. 2013) where the increase in nitrates and chlorides in shallow groundwater are related to
500 faeces from pit latrines, sewages, landfills, surface discharges and droppings from domestic animals.

501

502

503 **6. Conclusion**

504 Although pollution sources are identical for each of the three neighbourhoods studied, the
505 resultant transfers and reactions controlling concentrations are distinct. In the neighbourhood where
506 there is no surface inundation and acts as a local recharge area, the groundwater salinity values remain
507 low ($EC < 1285 \mu S/cm$), however NO_x concentrations are the highest recorded in this study (up to 5.1
508 $mmol/L$). In the neighbourhood bordering a lake where there is seasonal inundation and groundwater
509 discharge, the dry season interaction with lake water results in groundwater with highest observed EC
510 values (up to $5340 \mu S/cm$). Stable isotope (δ^2H and $\delta^{18}O$) mixing ratios indicate mixing of up to 74%
511 with lake water; In the neighbourhood located in a swamp that is subject to inundation during both the
512 small and large wet seasons also shows mixing with lake water (up to 62% using δ^2H and $\delta^{18}O$
513 values) resulting in groundwater EC values up to $1468 \mu S/cm$. This discharge site notably has lower
514 NO_x concentrations (up to $0.86 mmol/L$), however this is not indicative of less sewerage
515 contamination only greater degradation processes. Understanding the seasonal changes in processes
516 controlling groundwater quality between each site is key to identifying risks to the residents who use
517 this unregulated shallow groundwater resource for domestic purposes, including drinking water
518 supplies. Seasonal variations highlight heightened risks from sewerage and septic tank leakage during

519 the wet season in neighbourhoods located in discharge areas compared with increased risk during the
520 dry season in the recharge area. In addition, there are increased risks of shallow groundwater
521 salinization during both the small wet and dry seasons in discharge areas.

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

526 Groundwaters chemistry in each of the neighbourhoods are different. In St Jean, the
527 composition of major ion concentrations of waters is dominated by Na-Ca-HCO₃-Cl groundwater
528 type, while Ladji is of the Na-HCO₃-Cl type and Agla of the Na-Ca-HCO₃-SO₄-HCO₃ one. The ponds
529 sampled in both Ladji and Agla showed some similarities in major ion compositions with local
530 groundwater, thus indicating a potential connection. The Lake Nokoué has greater concentrations of
531 Na and Cl compared with all groundwater samples.

532 Groundwater samples indicated that 26 % for NO₃ and 7.0 % for NO₂ do not comply with the
533 Beninese quality standard for drinking water. Based on Cl/Br molar ratios, sources of NO_x in
534 groundwater appear to be dominated by infiltration of sewerage and septic tanks in dry season in St
535 Jean, while in Agla and Ladji, contamination was obvious in wet season following infiltrating rainfall
536 and ponds recharging. Low NO_x for some of the groundwater samples may indicate effects from
537 biodegradation in discharge area or dilution from rainfall.

538

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544

545

546 **References**

- 547 Ahamed H A, Rayaleh WE, Zghibi A, Ouddane B (2017) Assessment of chemical quality of
548 groundwater in coastal volcano-sedimentary aquifer of Djibouti, Horn of Africa. *Journal of*
549 *African Earth Sciences* 131 (july): 284-300. <https://doi.org/10.1016/j.jafrearsci.2017.04.010>.
- 550 Alassane A, Trabelsi R, Dovonon LF, Odeloui DJ, Boukari M, Zouari K, Mama D (2015) Chemical
551 evolution of the continental terminal shallow aquifer in the south of the coastal sedimentary
552 basin of Benin (West Africa) using multivariate factor analysis. *J. Water Resour. Protection* 7,
553 496–515.
- 554 Alcalá FJ, Custodio E (2008) Using the Cl/Br ratio as a tracer to identify the origin of salinity in
555 aquifers in Spain and Portugal *J. Hydrol.* 359 189–207
- 556 Alidou S, Boukari M, Oyedé LM, Gaye CB, Faye A, Gelinat P, Isabel D, Locat J (1994) Rapport
557 technique final du projet "hydrogéologie du quaternaire du sud-bénin", phase 2. CRDI 3-p-89-
558 1017. Tomes 1, ii, iii et iv. Univ. nat. Bénin, univ. CA diop, dakar, univ. Laval québec.
- 559 Anornu G, Abass G, Dickson A (2017) Tracking nitrate sources in groundwater and associated health
560 risk for rural communities in the White Volta River basin of Ghana using isotopic approach
561 ($\delta^{15}\text{N}$, $\delta^{18}\text{ONO}_3$ and 3H). *Science of The Total Environment* 603-604 (december): 687-98.
562 <https://doi.org/10.1016/j.scitotenv.2017.01.219>.
- 563 Arnade LJ (1999) Seasonal Correlation of Well Contamination and Septic Tank Distance.
564 *Groundwater* 37 (6): 920-23. <https://doi.org/10.1111/j.1745-6584.1999.tb01191.x>.
- 565 Barker, AP, Newton RJ, Bottrell SH, Tellam JH (1998) Processes affecting groundwater chemistry in
566 a zone of saline intrusion into an urban sandstone aquifer. *Applied Geochemistry* 13 (6):
567 735-49. [https://doi.org/10.1016/S0883-2927\(98\)00006-7](https://doi.org/10.1016/S0883-2927(98)00006-7).
- 568 Berner RA (1980). *Early Diagenesis: A Theoretical Approach*. Princeton University Press.
- 569 Boukari M (1998) Fonctionnement du système aquifère exploité pour l’approvisionnement en eau de
570 la ville de Cotonou sur le littoral béninois. Impact du développement urbain sur la qualité des
571 ressources. Thèse Doctorat ès- Science, Université C. A. Diop de Dakar. Sénégal.
- 572 Boukari M, Gaye CB, Faye A, Faye S1 (1996) The Impact of Urban Development on Coastal
573 Aquifers near Cotonou, Benin. *Journal of African Earth Sciences* 22 (4): 403-8.

574 [https://doi.org/10.1016/0899-5362\(96\)00027-9](https://doi.org/10.1016/0899-5362(96)00027-9).

575 British Geological Survey (2002) Groundwater Quality: Mali.

576 <http://www.bgs.ac.uk/downloads/start.cfm?id=1284>

577 Cary L, Petelet-Giraud E, Bertrand G, Kloppmann W, Aquilina L, Martins V, Hirata R (2015) Origins
578 and processes of groundwater salinization in the urban coastal aquifers of Recife
579 (Pernambuco, Brazil): A multi-isotope approach. *Science of The Total Environment* 530-531
580 (october): 411-29. <https://doi.org/10.1016/j.scitotenv.2015.05.015>.

581 Craig H (1961) Standards for reporting concentrations of deuterium and oxygen-18 in natural waters.
582 *Science*, 133, 1833-1834.

583 Davis S N, Whittemorw DO, Martin JF (1998) Uses of Chloride/Bromide Ratios in Studies of Potable
584 Water. *Groundwater* 36 (2): 338-50. <https://doi.org/10.1111/j.1745-6584.1998.tb01099.x>.

585 Decret N° 20001-094 du 200/02/2001 fixant les normes de qualité de l'eau potable en République du
586 Bénin.

587 Dhanasekarapandian M, Chandran S, Saranya Devi D, Kumar V (2016) Spatial and temporal variation
588 of groundwater quality and its suitability for irrigation and drinking purpose using GIS and
589 WQI in an urban fringe. *Journal of African Earth Sciences* 124 (december): 270-88.
590 <https://doi.org/10.1016/j.jafrearsci.2016.08.015>.

591 EAA (2018) Etude de reference sur les comportements, attitudes et pratiques des populations de
592 Cotonou sur la chaîne de l'eau dans la ville de cotonou. Rapport technique du projet SAC-
593 TIC. 72.

594 Hassane AB (2010) Aquifères superficiels et profonds et pollution urbaine en Afrique : Cas de la
595 communauté urbaine de Niamey (NIGER). 250.

596 Hassane AB, Leduc C, Favreau G, Bekins BA, Margueron T (2016) Impacts of a Large Sahelian City
597 on Groundwater Hydrodynamics and Quality: Example of Niamey (Niger). *Hydrogeology*
598 *Journal* 24 (2): 407-23. <https://doi.org/10.1007/s10040-015-1345-z>.

599 Hounkpe SP, Adjovi EC, Crapper M, Awuah E (2014) Wastewater Management in Third World
600 Cities: Case Study of Cotonou, Benin. *Journal of Environmental Protection* 05 (april) : 387.
601 <https://doi.org/10.4236/jep.2014.55042>.

602 INSAE (2016) Principaux indicateurs socio-démographiques et économiques, mai 2013, synthèse des
603 résultats d'analyse. Rapport, Direction des études démographiques, Cotonou, Bénin.

604 INSAE (2015) Quatrième Recensement Général de la Population et de l'Habitat, mai 2013, synthèse
605 des résultats d'analyse. Rapport, Direction des études démographiques, Cotonou, Bénin.

606 Jorgensen PR, Urup J, Helstrup T, Jensen MB, Eiland F, Vinther FP (2004) Transport and Reduction
607 of Nitrate in Clayey till underneath Forest and Arable Land. *Journal of Contaminant*
608 *Hydrology* 73 (1) : 207–26. <https://doi.org/10.1016/j.jconhyd.2004.01.005>.

609 Kadjangaba E, Djoret D, Doumnang JC, Ndoutamia GA, Mahmoud Y (2018) Impact des Processus
610 Hydrochimique sur la Qualité des Eaux souterraines de la Ville de N'Djaména-Tchad. Vol.
611 14. <https://doi.org/10.19044/esj.2018.v14n18p162>.

612 Katz BG, Eberts SM, Kauffman JL (2011) Using Cl/Br ratios and other indicators to assess potential
613 impacts on groundwater quality from septic systems: A review and examples from principal
614 aquifers in the United States. *Journal of Hydrology* 397 (3): 151-66.
615 <https://doi.org/10.1016/j.jhydrol.2010.11.017>.

616 Ketchemen-Tandia B, Boum-Nkot SN, Ebondji SR, Nlend BY, Emvoutou H, Nzegue O (2017)
617 Factors Influencing the Shallow Groundwater Quality in Four Districts with Different
618 Characteristics in Urban Area (Douala, Cameroon). *Journal of Geoscience and Environment*
619 *Protection* 05 (August): 99. <https://doi.org/10.4236/gep.2017.58010>.

620 Lapworth DJ, Nkhuwa DCW, Okotto-Okotto J, Pedley S, Stuart ME, Tijani MN, Wright J
621 (2017) Urban Groundwater Quality in Sub-Saharan Africa: Current Status and Implications
622 for Water Security and Public Health. *Hydrogeology Journal* 25 (4): 1093-1116.
623 <https://doi.org/10.1007/s10040-016-1516-6>.

624 Liu Y, Jiu JJ, Wenzhao L, Xingxing K (2017) Hydrogeochemical Characteristics in Coastal
625 Groundwater Mixing Zone. *Applied Geochemistry* 85 (october): 49-60.
626 <https://doi.org/10.1016/j.apgeochem.2017.09.002>.

627 Lu Y, Shuai S, Ruoshi, Liu Z, Meng J, Sweetman AJ, Jenkins A (2015) Impacts of Soil and Water
628 Pollution on Food Safety and Health Risks in China. *Environment International* 77 (april):
629 5-15. <https://doi.org/10.1016/j.envint.2014.12.010>.

630 Maliki R (1993) Etude hydrogéologique du littoral béninois dans la région de Cotonou (A.O). Thèse
631 de Doctorat de 3ème cycle. Université C. A. Diop de Dakar. Sénégal.

632 Martínez-Santos P, Martín-Loeches M, García-Castro N, Solera D, Díaz-Alcaide S, Montero E,
633 García-Rincón J (2017) A survey of domestic wells and pit latrines in rural settlements of
634 Mali: Implications of on-site sanitation on the quality of water supplies. *International Journal
635 of Hygiene and Environmental Health* 220 (7): 1179-89.
636 <https://doi.org/10.1016/j.ijheh.2017.08.001>.

637 McArthur J M, Sikdar P K, Hoque MA, Ghosal U (2012) Waste-water impacts on groundwater: Cl/Br
638 ratios and implications for arsenic pollution of groundwater in the Bengal Basin and Red
639 River Basin. *Vietnam Sci. Total Environ.* 437 390–402.

640 McInnis D, Silliman S, Boukari M, Yalo N, Orou-Pete S, Fertenbaugh C, Sarre K, Fayomi H
641 (2013) Combined Application of Electrical Resistivity and Shallow Groundwater Sampling to
642 Assess Salinity in a Shallow Coastal Aquifer in Benin, West Africa. *Journal of Hydrology* 505
643 (november): 335-45. <https://doi.org/10.1016/j.jhydrol.2013.10.014>.

644 Mkandawire T (2008) Quality of groundwater from shallow wells of selected villages in Blantyre
645 District, Malawi. *Physics and Chemistry of the Earth, Parts A/B/C, Integrated Water
646 Resources Management - From Concept to Practice*, 33 (8): 807-11.
647 <https://doi.org/10.1016/j.pce.2008.06.023>.

648 Najib S, Fadili A, Mehdi K, Riss J, Makan A (2017) Contribution of hydrochemical and geoelectrical
649 approaches to investigate salinization process and seawater intrusion in the coastal aquifers of
650 Chaouia, Morocco. *Journal of Contaminant Hydrology* 198 (march): 24-36.
651 <https://doi.org/10.1016/j.jconhyd.2017.01.003>.

652 Ndembo LJ (2009) Apport des outils hydrogéologiques et isotopiques à la gestion de l'aquifère du
653 Mont Amba. Thèse de doctorat. Kinshasa / République Démocratique du Congo. 203.

654 Ndeye DM, Orban P, Otten J, Stumpp C, Faye S, Dassargues A (2017) Temporal changes in
655 groundwater quality of the Saloum coastal aquifer. *Journal of Hydrology: Regional Studies* 9
656 (february): 163-82. <https://doi.org/10.1016/j.ejrh.2016.12.082>.

657 Ngo et al., 2015. The sustainability risk of Ho Chi Minh City, Vietnam, due to saltwater intrusion,
658 Geosciences Journal, 19 (3), 547-560.

659 Nlend B, Celle-Jeanton H, Huneau F, Ketchemen-Tandia B, Fantong WY, Ngo Boum-Nkot S, Etame
660 J (2018) The Impact of Urban Development on Aquifers in Large Coastal Cities of West
661 Africa: Present Status and Future Challenges. Land Use Policy 75 (june): 352-63.
662 <https://doi.org/10.1016/j.landusepol.2018.03.007>

663 Nyenje PM, Foppen JW, Kulabako R, Muwanga A, Uhlenbrook S (2013) Nutrient pollution in
664 shallow aquifers underlying pit latrines and domestic solid waste dumps in urban slums.
665 Journal of Environmental Management 122 (june): 15-24.
666 <https://doi.org/10.1016/j.jenvman.2013.02.040>.

667 Odoulami L, Gbesso F, Hounguevou S (2013) Qualité de l'eau de consommation et maladies
668 hydriques dans la commune de Ze (Benin).10.

669 Ogrinc N, Tamše S, Zavadlav S, Vrzel J, Jin L (2019) Evaluation of geochemical processes and nitrate
670 pollution sources at the Ljubljansko polje aquifer (Slovenia): A stable isotope perspective.
671 Science of The Total Environment 646 (january): 1588-1600.
672 <https://doi.org/10.1016/j.scitotenv.2018.07.245>.

673 Okotto L, Okotto-Okotto J, Price H, Pedley S, Wright J (2015) Socio-economic aspects of domestic
674 groundwater consumption, vending and use in Kisumu, Kenya. Applied Geography 58
675 (march): 189-97. <https://doi.org/10.1016/j.apgeog.2015.02.009>.

676 Ouedraogo I, Defourny P, Vanclooster M (2016) Mapping the groundwater vulnerability for pollution
677 at the pan African scale. Science of The Total Environment 544 (february): 939-53.
678 <https://doi.org/10.1016/j.scitotenv.2015.11.135>.

679 Oyédé LM (1991) Dynamique sédimentaire actuelle et messages enregistrés dans les séquences
680 quaternaires et néogènes du domaine margino littoral du Bénin (l'Afrique de l'Ouest). Thèse
681 présentée pour l'obtention du doctorat en géologie sédimentaire. Nouveau régime. Université
682 de Bourgogne, Paris; 302 p.

683 Panno SV, Hackley KC, Hwang HH, Greenberg SE, Krapac IG, Landsberger S, O'Kelly DJ (2006)
684 Characterization and Identification of Na-Cl Sources in Ground Water. Groundwater 44 (2):

685 176-87. <https://doi.org/10.1111/j.1745-6584.2005.00127.x>.

686 Paran F, Arthaud F, Novel M, Graillot D, Bornette G, Piscart C, Marmonier P, Lavastre V, Travi Y,
687 Cadilhac L (2015) *Caracterisation Des Échanges Nappes/Rivieres En Milieu Alluvionnaire*
688 *Guide Méthodologique. Bassin Rhône-Méditerranée et Corse. Septembre. 180.*

689 Pastén-Zapata E, Ledesma-Ruiz R, Harter T, Ramírez AI, Mahlknecht J (2014) Assessment of sources
690 and fate of nitrate in shallow groundwater of an agricultural area by using a multi-tracer
691 approach. *Science of The Total Environment* 470-471 (february) : 855-64.

692 Petelet-Giraud E, Négrel P, Aunay B, Ladouche B, Bailly-Comte V, Guerrot C, Flehoc C, Pezard P,
693 Lofi J, Dörfliger N (2016) Coastal Groundwater Salinization: Focus on the Vertical
694 Variability in a Multi-Layered Aquifer through a Multi-Isotope Fingerprinting (Roussillon
695 Basin, France). *Science of The Total Environment* 566–567 (October): 398–415.
696 <https://doi.org/10.1016/j.scitotenv.2016.05.016>.

697 Postma D, Boesen C, Kristiansen H, Larsen F (1991) Nitrate Reduction in an Unconfined Sandy
698 Aquifer: Water Chemistry, Reduction Processes, and Geochemical Modeling. *Water*
699 *Resources Research*. <https://agupubs.onlinelibrary.wiley.com/doi/abs/10.1029/91WR00989>.

700 Rossiter HMA, Peter AO, Awuah E, MacDonald AM, Schäfer AI (2010) Chemical drinking water
701 quality in Ghana: Water costs and scope for advanced treatment. *Science of The Total*
702 *Environment* 408 (11): 2378-86. <https://doi.org/10.1016/j.scitotenv.2010.01.053>.

703 Roy S, Speed C, Bennie J, Swift R, Wallace P (2007) Identifying the significant factors that influence
704 temporal and spatial trends in nitrate concentrations in the Dorset and Hampshire Basin Chalk
705 aquifer of Southern England. *Quarterly Journal of Engineering Geology and Hydrogeology*,
706 40(4), 377–392. doi:10.1144/1470-9236/07-025

707 Selvakumar S, Chandrasekar N, Kumar G (2017) Hydrogeochemical characteristics and groundwater
708 contamination in the rapid urban development areas of Coimbatore, India. *Water Resources*
709 *and Industry* 17 (june): 26-33. <https://doi.org/10.1016/j.wri.2017.02.002>.

710 Starr RC, Gillham RW (1993) Denitrification and Organic Carbon Availability in Two Aquifers.
711 *Groundwater* 31 (6): 934–47. <https://doi.org/10.1111/j.17456584.1993.tb00867.x>.

712 Stephen SE, Borum BI, Boukari M, Yalo N, Orou-Pete S, McInnis D, Fertenbaugh C, Mullen

713 AD (2010) Issues of Sustainability of Coastal Groundwater Resources: Benin, West
714 Africa. *Sustainability* 2 (8): 2652-75. <https://doi.org/10.3390/su2082652>.

715 Stuart ME, Chilton PJ, Kinniburgh DG, Cooper DM (2007) Screening for long-term trends in
716 groundwater nitrate monitoring data. *Quarterly Journal of Engineering Geology and*
717 *Hydrogeology*, 40(4), 361–376. doi:10.1144/1470-9236/07-040

718 Takem GE, Dornadula C, Ayonghe SN, Thambidurai P (2010) Pollution Characteristics of Alluvial
719 Groundwater from Springs and Bore Wells in Semi-Urban Informal Settlements of Douala,
720 Cameroon, Western Africa. *Environmental Earth Sciences* 61 (2): 287-98.
721 <https://doi.org/10.1007/s12665-009-0342-8>.

722 Takem GE, Kuitcha D, Ako AA, Mafany GT, Takounjou-Fouepe A, Ndjama J, Ntchancho R, Ateba
723 BH, Chandrasekharam D, Ayonghe SN (2015) Acidification of Shallow Groundwater in the
724 Unconfined Sandy Aquifer of the City of Douala, Cameroon, Western Africa: Implications for
725 Groundwater Quality and Use. *Environmental Earth Sciences* 74 (9): 6831-46.
726 <https://doi.org/10.1007/s12665-015-4681-3>.

727 Totin HSV, Amoussou E, Odoulami L, Edoh P A, Boukari M, Boko M (2013) Groundwater
728 Pollution and the Safe Water Supply Challenge in Cotonou Town, Benin (West Africa).191-
729 196.

730 UNESCO (2017) *L'industrialisation et l'urbanisation au service de la transformation de l'Afrique*.
731 Addis-Abéba. Commission économique pour l'Afrique.

732 Vengosh A, Pankratov I (1998) Chloride/Bromide and Chloride/Fluoride Ratios of Domestic Sewage
733 Effluents and Associated Contaminated Ground Water. *Groundwater* 36 (5): 815-24.
734 <https://doi.org/10.1111/j.1745-6584.1998.tb02200.x>.

735 Yabi I, Afouda F (2012) Extreme rainfall years in Benin (West Africa). *Quaternary International*,
736 Volume 262, ISSN 1040-6182, 39-43. <https://doi.org/10.1016/j.quaint.2010.12.010>.

737 Yadouléton M J (2015) *Assainissement environnemental à Cotonou et lutte contre le choléra* ». Thèse
738 de Doctorat Unique. Université d'Abomey-Calavi. 313.

739 Yameogo S (2008) *Ressources en eau souterraine du centre urbain de Ouagadougou au Burkina Faso,*
740 *qualité et vulnérabilité*. Univ. Avignon. 254.

741 Zhang Y, Li F, Zhang Q, Li J, Liu Q (2014) Tracing Nitrate Pollution Sources and Transformation in
742 Surface- and Ground-Waters Using Environmental Isotopes. Science of The Total
743 Environment 490 (august): 213-22. <https://doi.org/10.1016/j.scitotenv.2014.05.004>.

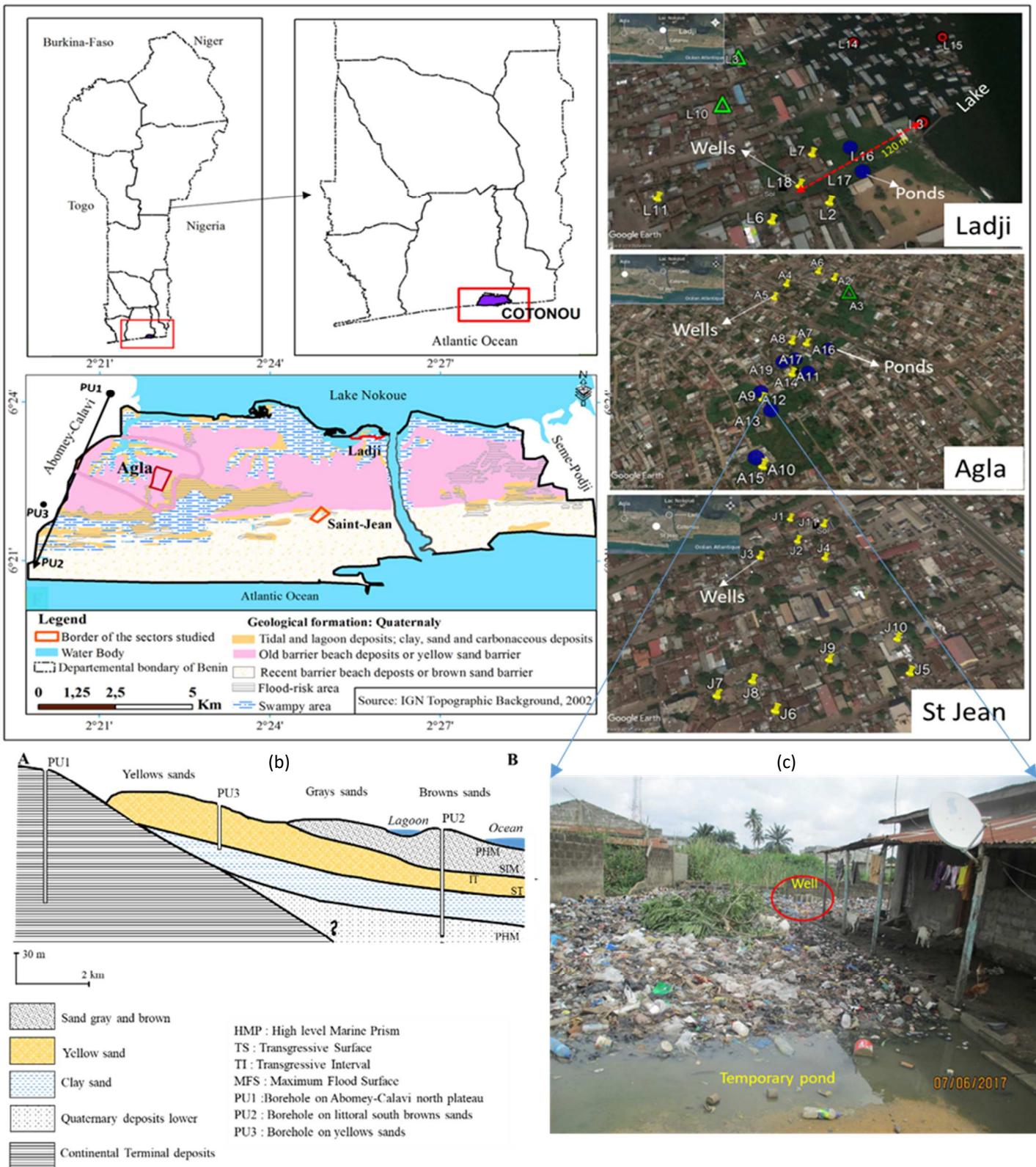


Figure 1. (a) Location of Cotonou and sampling sites in the three neighbourhoods; (b) Sedimentary unit cross section (A-B) of marginal-littoral area of Benin (modified from Boukari, 1998); and (c) Environmental situation at Agla (A9 site)

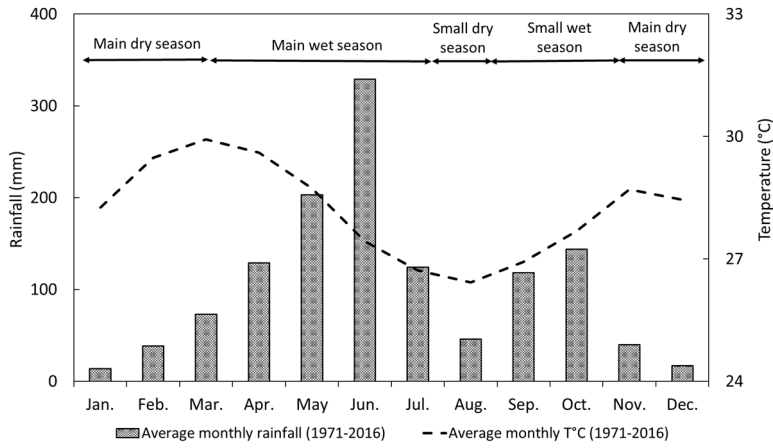


Figure 2: Average monthly interannual rainfall at Cotonou station from 1971 to 2016 (ASECNA station)

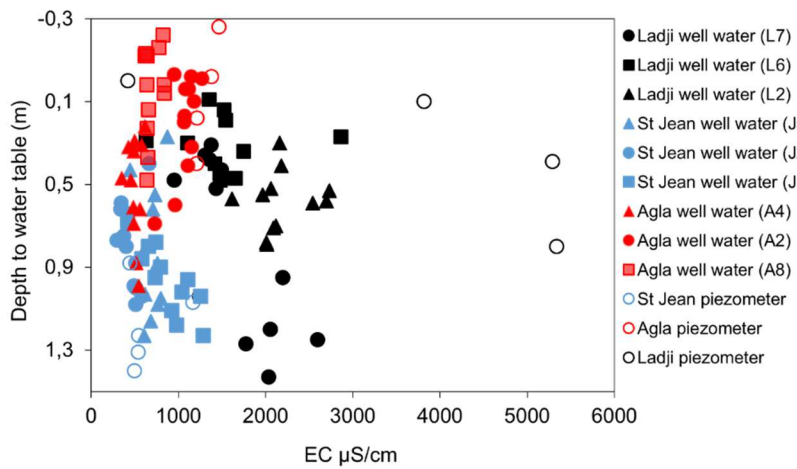


Figure 3. Relationship between water-table variation and electrical conductivity at each site

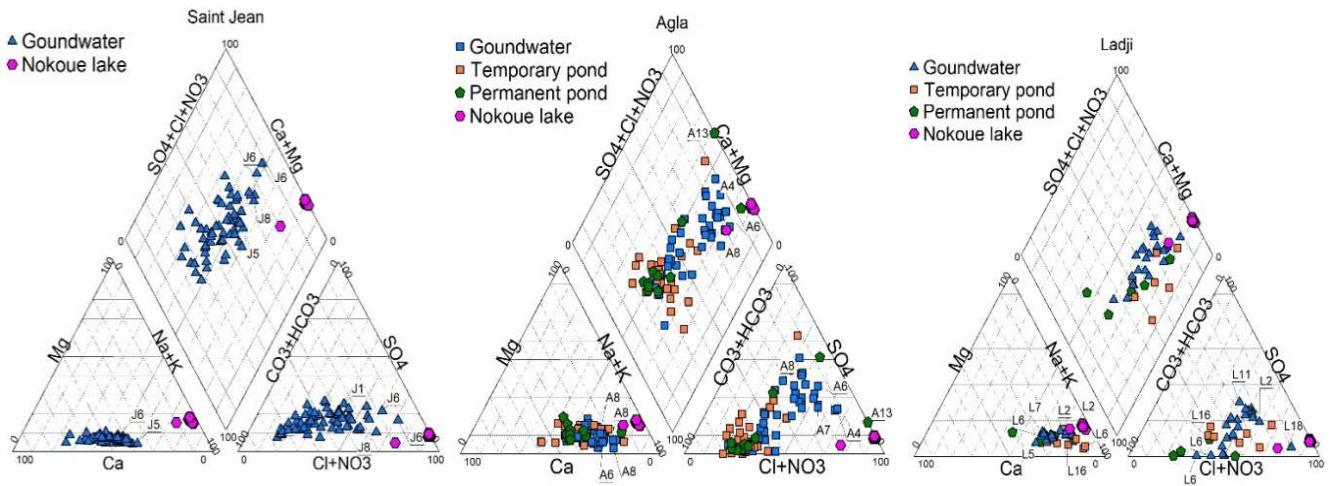


Figure 4. Piper diagram for groundwaters, ponds and lake at each site

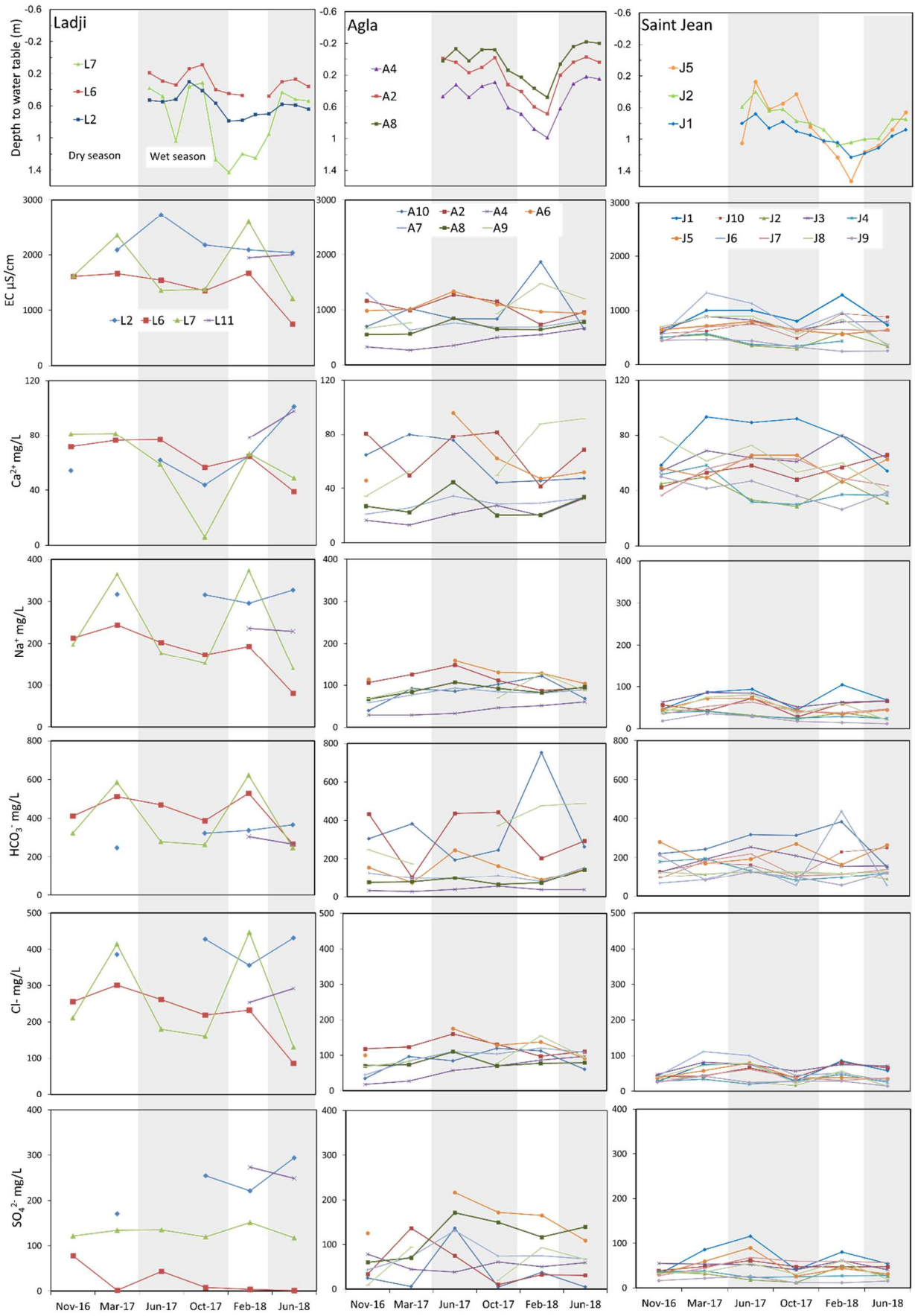


Figure 5. Temporal variation between, depth water table, electrical conductivity, major ion and isotopic stable at each site.

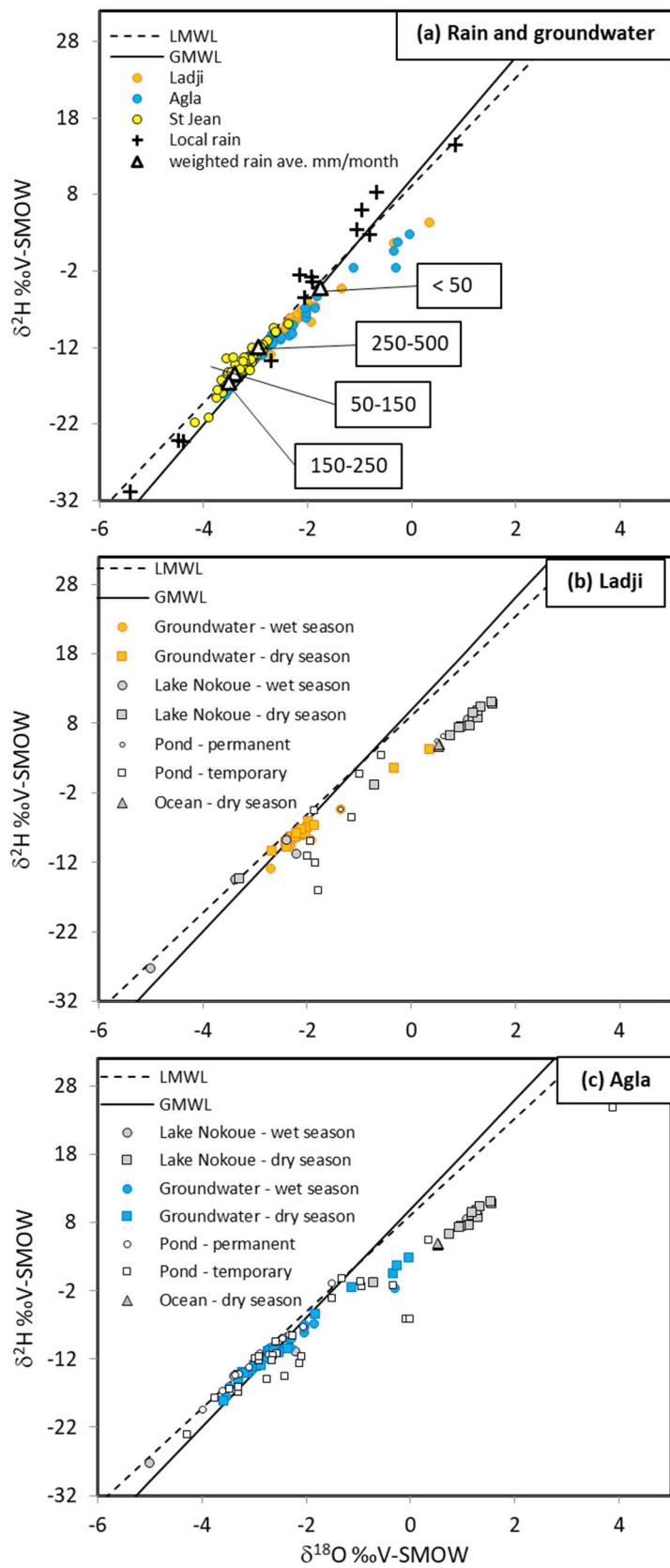


Figure 6. Relationship between ^{18}O and ^2H with Global Meteoric Water Line (GMWL) and Local Meteoric Water Line (GMWL).

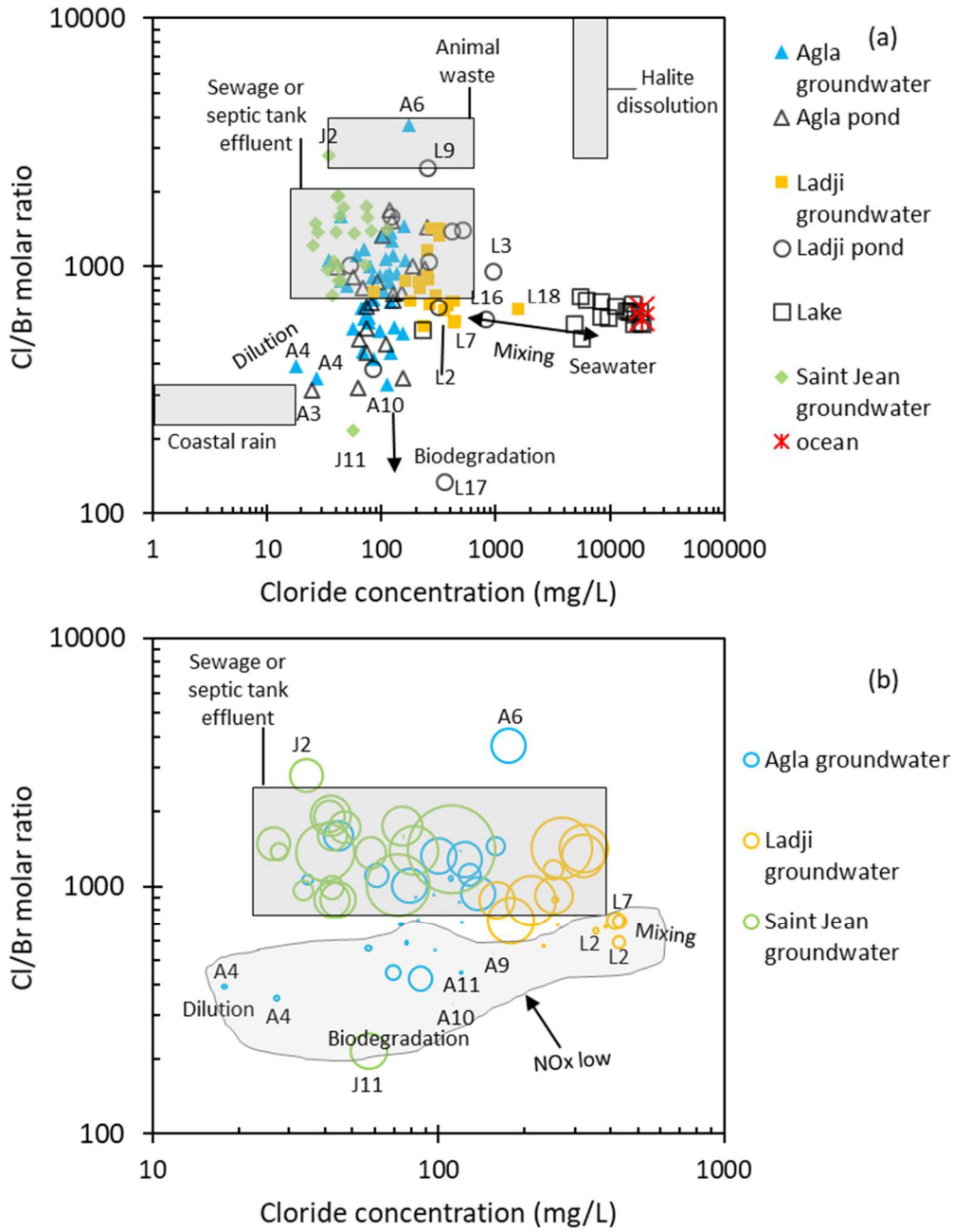


Figure 7. (a) Groundwater, pond and lake water Cl/Br molar ratios with changes in Cl concentrations highlights different sources of Cl in the waters. (b) Relationship between Cl/Br ratios and Cl concentrations showing higher concentrations in NO_x (circle sizes are relative to NO_x concentrations).

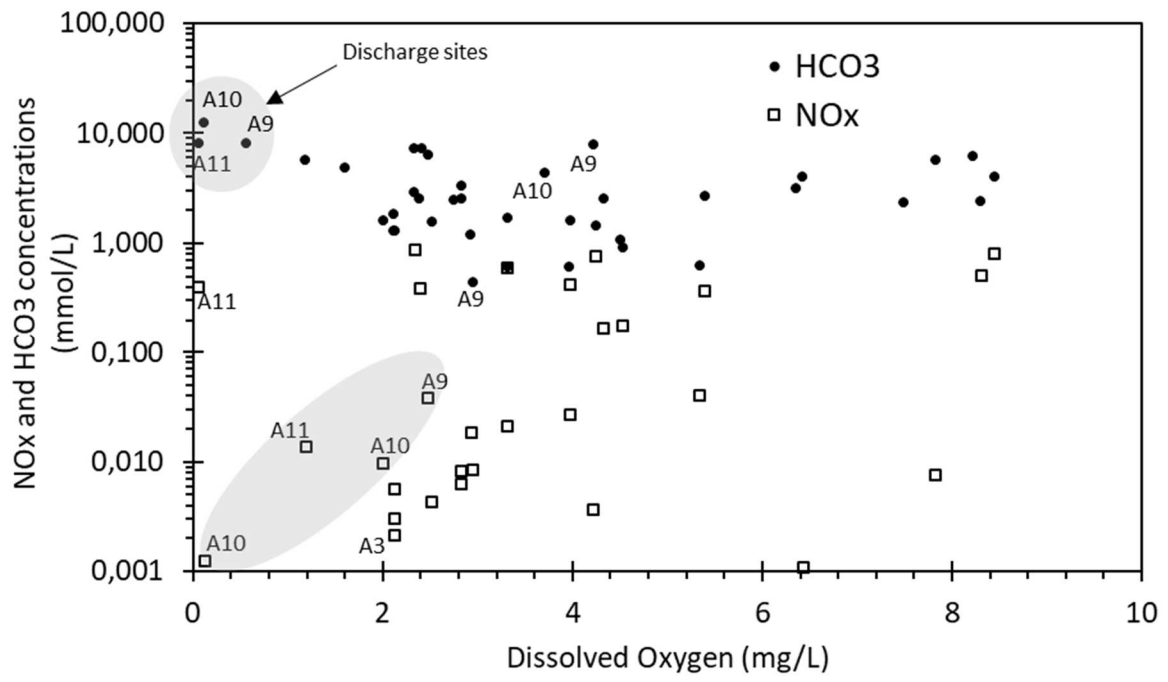
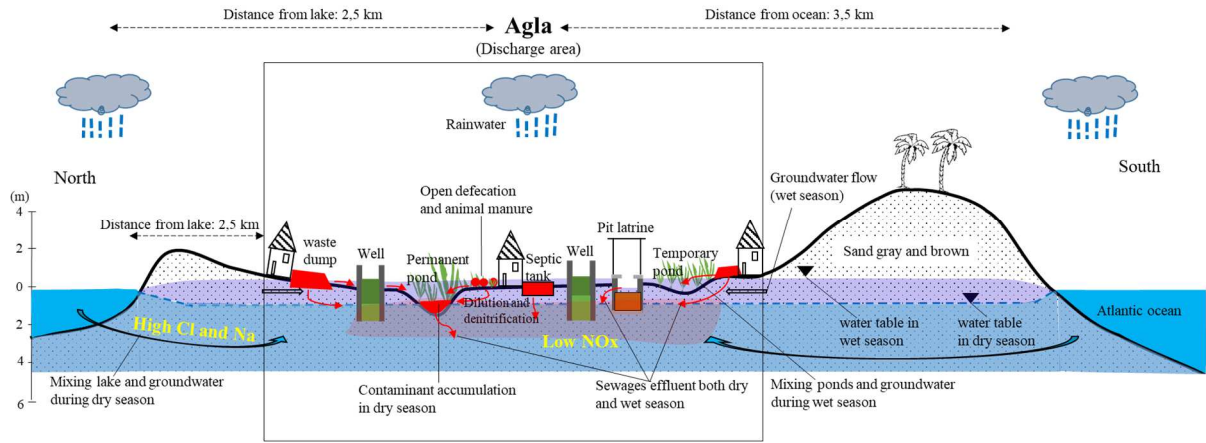


Figure 8. Relationship between HCO₃, NOx and DO for all groundwater sampled at Agla

(a)



(b)

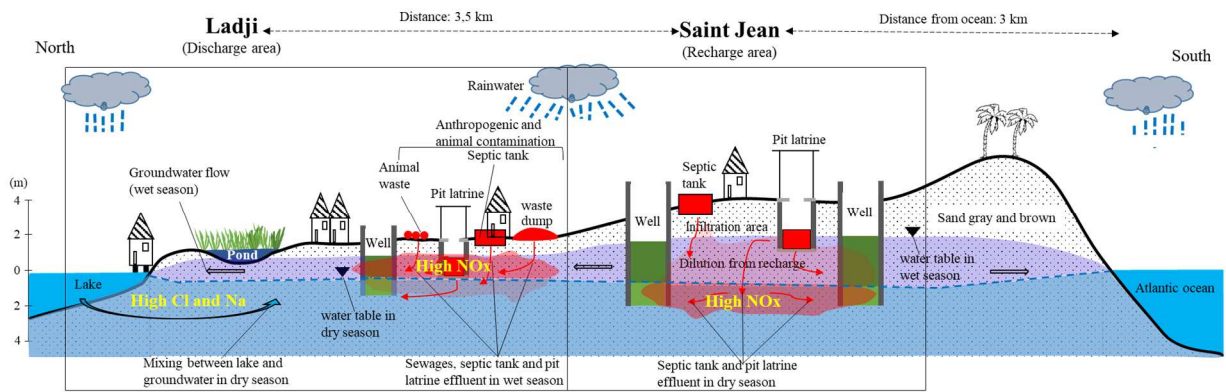
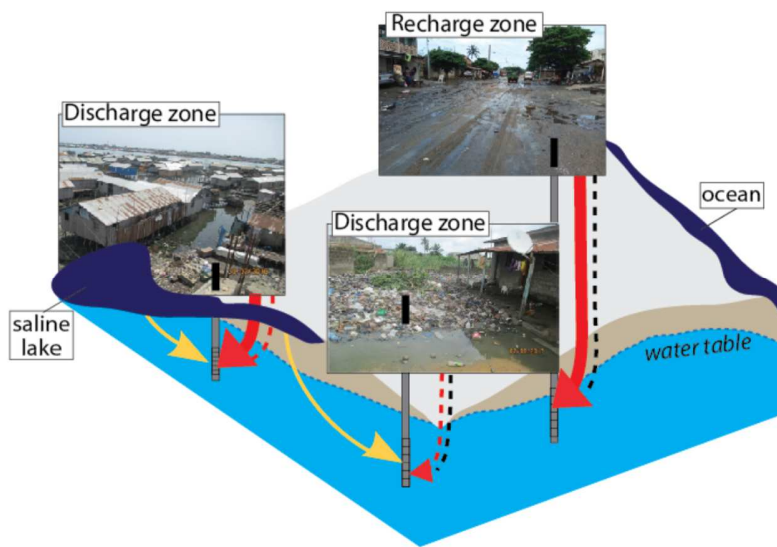


Figure 9. Schematic of conceptual model for contaminant sources and transfers at (a) Agla, and (b) Ladji and Saint-Jean.



■ High NOx contaminantion

- - - DRY SEASON denitrification

— DRY SEASON salinisation

- - - WET SEASON dilution