ORIGINAL ARTICLE

Hydrochemistry of nutrients in groundwater under farmland in the Benue River Basin, North-Cameroon

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Abstract

In this study, we investigated the distribution and the principal sources of macronutrients (N, P, K, S) in groundwater in agro-pastorally dominated North Cameroon. Graphical methods and multivariate statistics were used to understand the impact of land use and regional geology on nutrient concentrations (K^+, NO_3^-, SO_4^{2-}, PO_4^{3-}). Groundwater was acidic (median pH 6.8) and more mineralized (EC range = 35–2500 μS/cm, median = 376 μS/cm) than surface water (EC range = 50–222 μS/cm, median = 81 μS/cm). The median concentration of nutrients was (in mg/L) in the order NO_3^- (41.3) > SO_4^{2-} (7.7) > K^+ (5.2) > PO_4^{3-} (0.1) in groundwater and rather K^+ (3.3) > NO_3^- (0.3) > SO_4^{2-} (0.2) > PO_4^{3-} (0.2) in surface water. Using hierarchical cluster analysis and Schoeller diagram, we identified three distinct water quality groups. Group I, which contained cluster 1 and 2 were generally less mineralized and characterised as Na + K − HCO_3 water type found in the urban area such as Garoua and Lagdo. Group II, containing cluster 3 was much higher mineralized and characterised as the Na + K − HCO_3 water type with elevated nitrate. The occurrence of these groups is linked to the outcrops of cretaceous terrestrial sediments of the Garoua basin. Group III, which contained cluster 4 were heavily mineralized and characterised as the Ca + Mg − NO_3 water type. The occurrence was controlled by the outcrop of the crystalline Proterozoic basement. The nitrate median (41 mg/L) found in groundwater demonstrates the influence exerted by anthropogenic activities. Management strategies are required for the whole study area to limit further deterioration of groundwater resources in the wake of the planned expansion of agricultural productivity in the North Region of Cameroon.

Introduction

The North Region of Cameroon which belongs to the Sudano-Sahelian agro-ecological zone (Sy et al. 2008; Achancho 2013) is one of the main agro-pastoral zones

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of Cameroon. Drained by the upper Benue River Basin (BRB), an important fraction of Niger River Basin (NRB), the region has been a pole of attraction for farmers from Cameroon and beyond (Boutrais 1973; Brabant and Gavaud 1985; Koulandi 2006). Farmers from this region, their majority working only 1–2 ha, have developed extensive production systems including the use of fertilizers and pesticides. They developed on to previously considered unfavorable soils like hydromorphic soils of the large plains and to river banks to achieve and sustain higher yields of cotton, maize, millet, sorghum, onion and peanuts (Barbier et al. 2002). Moreover, Cameroon has a national strategy for the development of agriculture to accommodate the projected population growth (Document de Stratégie de Développement du Secteur Rural, DSDSR (Republique du Cameroun 2005; MINADER 2006; Abia et al. 2016) seeking to increase productivity by mitigating the constraints to production

(Achancho 2013; ZEF et al. 2017). The areas of intervention include intensification and diversification of agricultural production with a focus on the North Region. This puts water resource at risk through a growing demand for water for agro-pastoral, domestic and industrial purposes and at the same time negatively impacts water quality. In the hot semi-arid climate type (Cheo et al. 2013; Ankidawa et al. 2015) surface water is scarce, essentially limited from October to April during the dry season to the perennial Benue. Groundwater is the only reliable source in both seasons. Livestock breeding, the use of pesticides, fertilization and unsustainable land use lead to water pollution. Production enhancing agricultural programs in the North region have started. The increased application of inorganic fertilizer, animal droppings and composted plant remains are the most direct way to increase productivity (Abia et al. 2016) which will have an additional bearing on groundwater and surface water chemistry (Njitchoua et al. 1997).

The quality of groundwater or surface water is a function of a number of natural processes and of human activity. Without human influence, water quality would evolve from infiltrating rainwater through biogeochemical weathering with minerals and organic matter in the root zone, the rest of the unsatured zone or vadose zone and in the aquifer, modified before recharge by atmospheric processes like evapotranspiration or the deposition of reactive minerals and salts by dust (Khatri and Tyagi 2015). Agricultural activities, untreated waste water discharge from industries and domestic activities often result in the run-off and infiltration of untreated or partially treated wastewater into the environment, hence contaminating groundwater and surface water. A major consequence of this contamination is the flux of wastewater-derived nutrients to groundwater and surface water. The dissolved chemical species of macronutrients (N, P, K and S) behave differently in groundwater and surface water. Nutrients stem from a variety of sources such as the application of mineral and farm fertilizer in agriculture, onsite sanitation, dispersion of animal droppings from migrating livestock, wastewater discharge into ditches and creeks and a slow contribution from rock weathering (Molua and Lambi 2006; Cheo et al. 2013; Nyenje 2014; Yu et al. 2018). A non-negligible nutrient background is coming from both wet and dry atmospheric deposition with the special mention of dust during seasonal Harmattan (Abdus-Salam et al. 2011; Galy-Lacaux and Delon 2014; Vet et al. 2014; Akpo et al. 2015; Fantong et al. 2020).

Nutrients are essential to the growth of (aquatic) organisms, yet excess nutrient loading by N and P causes eutrophication of water bodies and can severely impact designated uses of water and impairing ecosystem services through excessive growth of cyanobacteria, algae and macrophytes. This can be seen in the nitrogen river load of major river basins in the world (Caraco and Cole, 1999). Fish kills due to the depletion of oxygen and/or free ammonia can result (Nyenje et al. 2010; Ndjama et al. 2017). From the river catchment perspective, the dynamics of nutrients in groundwater and linked surface water is an important issue in water quality management, in particular in areas with intensive agriculture (Smith et al. 2008; Van Geer et al. 2016). Shallow, unprotected phreatic aquifers under agro-pastoral land use are highly vulnerable to nitrate contamination due to excess application of mineral and farm fertilizer (Carpenter et al. 1998; Cherry et al. 2008; Ako et al. 2014; Khatri and Tyagi 2015). It is well known that nitrate-rich drinking water causes public health problems like infant methemoglobinemia, risk of specific cancers and birth defects and is therefore rightly regulated ($NO_3^- < 50$ mg/L) (Thornton et al. 1999; WHO 2004; Ward et al. 2018).

However, the BRB as most catchments in sub-Saharan Africa is vulnerable to water quality deterioration by increasing levels of nutrients (N, P, K, S) which can be further concentrated by evapotranspiration (Njitchoua et al. 1997; Molua and Lambi 2006; Yong 2013; Jokam Nenkam et al. 2019 and Fantong et al. 2020). Consequently, water quality and baseline studies in the BRB are needed to understand and manage the NRB (Chippaux et al. 2002; Edet et al. 2011). Thus, this study provides a background status of nutrient concentrations in ground and surface water in the upper BRB, before a large-scale increase in agricultural productivity.

In sub-Saharan Africa, various authors have studied the issue of eutrophication of surface water (Carpenter et al. 1998; Nyenje et al. 2010) and the contamination of groundwater resources in various agricultural and climatic settings (Roose and Lelong 1981; Cronin et al. 2006; Ohou et al. 2008; Kelderman et al. 2009; Douagui et al. 2012; Nyenje 2014; Kringel et al. 2016; Lapworth et al. 2017; Bello et al. 2020). Njitchoua et al. (1997) conducted a study to identify recharge mechanisms and the geochemistry of groundwater in Garoua in July and December 1991 based on a smaller dataset. Key results, more than 25 years ago, already showed the vulnerability of groundwater to nitrate pollution in boreholes tapping the upper 40 m of the aquifer. Therefore, studies are urgent to assess the current status and identify the main factors affecting the water quality in the BRB with an emphasis on groundwater. To this end principal component analysis (PCA) and hierarchical cluster analysis (HCA) techniques were employed as key methods in this study, evaluated against land cover and geology.

This study provides the first basin-scale assessment of the spatial distribution of dissolved nutrients in groundwater and surface water in the upper Benue River Basin (BRB). The upper BRB has been earmarked for a leap in agricultural productivity as part of the national development strategy by government actors (Achancho, 2013; Abia et al. 2016; ZEF et al. 2017) supported by international donors. This study examines the concentrations of nitrate (NO_3^-),

sulphate (SO_4^{2-}), potassium (K^+) and phosphorus (PO_4^{3-}) along with chloride (Cl^-) as a conservative tracer in natural water with an emphasis on groundwater. This is done in relation to surface geology and a publicly available land cover classification. Furthermore, we try to derive the geogenic background concentration of these nutrients from a critical investigation of the univariate distribution of concentrations and the prevalent lithologies.

Materials and methods

Description of the study area

The study area (Fig. 1) is administratively located in the North Region of Cameroon bordered to the east by the Republic of Chad and Central Africa, to the west by the

Federal Republic of Nigeria, to the north by the Far North Region and to the south by the Adamaoua Region. It has a surface area of 65,576 km^2 with an estimated population of 3,993,007 in 2015 and a birth rate estimated at 45.6 births per thousand people (INS and UNFPA Cameroun, 2016). The land cover is characterized by a dry grassy savanna, a shrubby and wooded savanna, a wooded savanna, forest galleries and mountain vegetation (Djoufack et al. 2012). Geographically, the study area is located along the Benue catchment upstream and downstream of Garoua between 8° 30.0′ and 10° 0.0′ N and 12° 8.0′–14° 3.0′ E. There are 4 administrative divisions (Departments) in the North Region of Cameroon, while this research focused on 3 divisions: Benue, Mayo-Louti and Mayo-Rey, the two latter being synonymous to tributaries to the Benue. The development of croplands and the relative fertility of the soil in these divisions have created dense human settlements. The
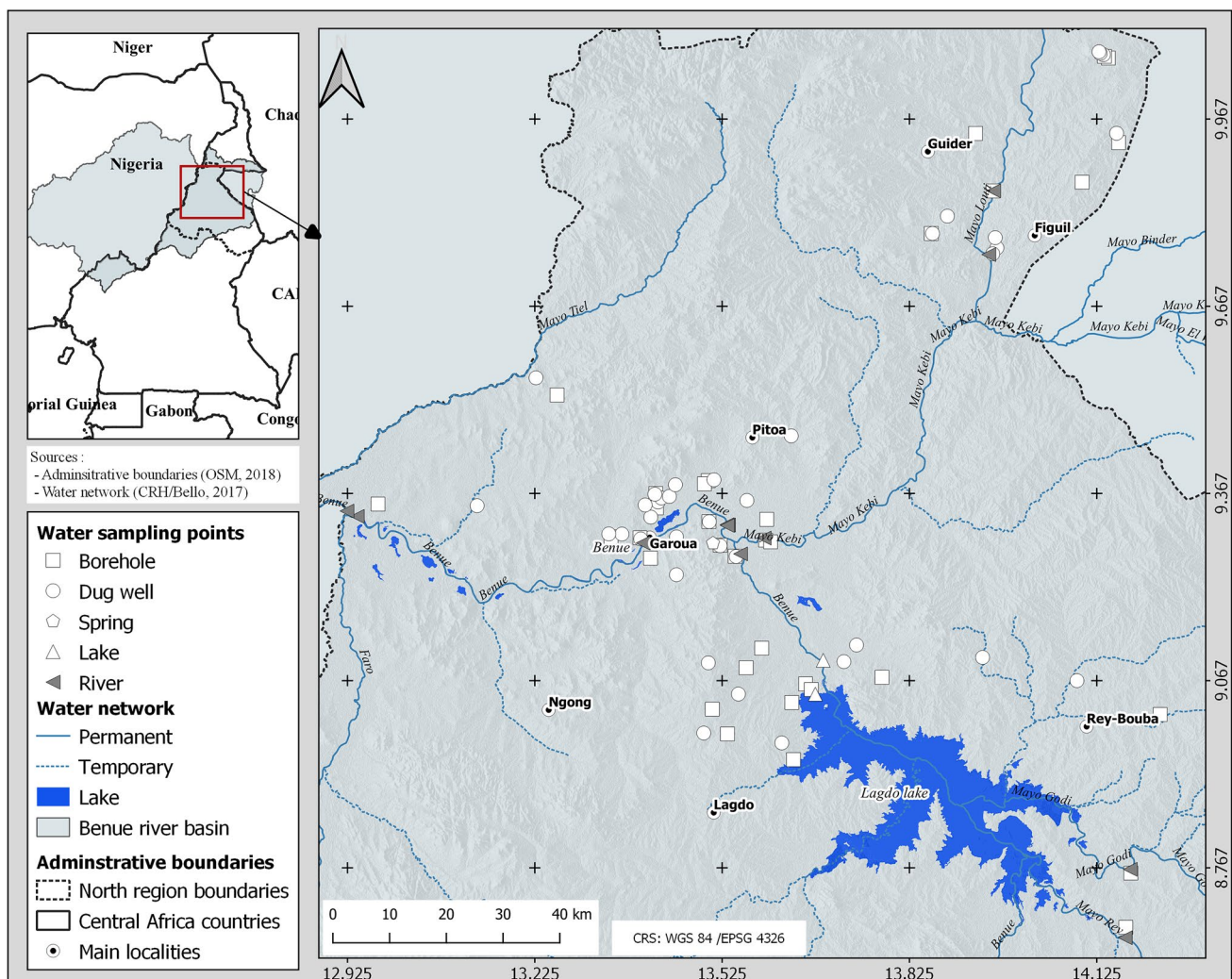

Fig. 1 Location of water sampling sites in the Benue River Basin (BRB) an important fraction of Niger River Basin (NRB); data source: Open Streetmap

subdivisions Benue (about 312 inhabitants/km^2) followed by Figuil (163 inhabitants/km^2) and Guider (145 inhabitants/km^2) (MINEPAT 2016) have the highest population densities. These subdivisions are largely characterised by low-income urban catchment with poor access to safe drinking water and poor on-site sanitation.

The semi-arid climate type (Ndenecho and Lambi 1998) of the study area which lies within the Sudano-Sahelian zone of Africa is characterized by two seasons: a long dry season from October to April and a rainy season that lasts from May to September. Rainfall is generally below 1000 mm with major variability from year to year and from month to month (Molua and Lambi 2006). The annual rainfall pattern is variable with about 70% of rain falling between July and September (Molua and Lambi 2006). Precipitation occurs either as low altitude monsoon rain or as occasional high altitude squally showers. Between 1951 and 1989, the annual precipitation was 1018 mm on the average (Molua and Lambi 2006; Cheo et al. 2013). Despite the high value of the annual rainfall, only a small fraction contributes to groundwater recharge because of the high annual potential evapotranspiration (about 1800 mm), which is about double the mean annual rainfall (Olivry 1986; Njitchoua et al. 1995; Fantong et al. 2020).

The BRB has been modified from its original savannah environment by widespread agriculture, overgrazing and urbanisation. It has been estimated in 2016 that 782,320.40 ha of cultivated land yielded a production of 1,578,034 tons of food crops, such as maize, millet, peanut, vegetable, onion and cotton (MINEPAT 2016). Rainfed agricultural production is vulnerable to climatic variability. Consequently, problems with crop cultivation, livestock, wildlife and fuelwood are very pronounced in this area (Djoufack et al 2012; Abia et al. 2016).

Geology and rock-forming lithologies

The E–W trending Yola-Garoua arm of the NE-SW intra-plate tectonic Benue trough formed on pan-african proterozoic crust beginning latest in the Aptian of the Cretaceous (Benkhelil 1989), accumulating terrestrial sediments of up to 5000 m in Nigeria (Ogunmola et al. 2016; Finthan and Mamman 2020) and its Garoua basin in Cameroon (Kamguia et al. 2005; Abate Essi et al. 2019). Six smaller satellite basins trend with the Garoua basin which are known for their pelitic/psammitic terrestrial/lacustrine sediments with some intercalated basalts (Maurin and Giraud 1990; Ntsama 2013). The basement into which these basins subsided is proterozoic crust made up of migmatitic gneisses (ortho and para), granitoids, as well as subordinate mica-schists and meta-dolerites (Schwoerer 1965) (Fig. 2).

The 1:500,000 hydrogeological ORSTOM map (Dauteloup et al. 1971) shows the central Garoua basin, specifying more pelitic middle cretaceous sediments, equated to the Aptian lower Bima formation (and older, eastern Benoue grits), towards its southern margin and the east in contrast to the butte-forming coarse arkosic sandstones and conglomerates, equated to the upper Bima formation (western Garoua grits), in the center and towards the north. Finthan and Mamman (2020) describe the more distal sedimentology and facies relationships of the Bima sandstone in Nigeria. Bessong (2012) investigated geological sections, distributed over the Garoua basin sedimentologically and mineralogically (Bessong 2012; Bessong et al. 2011, 2015) and revealed arkosic and wacke sandstones containing primarily quartz, followed by K-feldspar with hematitic and goethitic cements as the main mineral assemblage of the terrestrial sediments. This arkosic lithology reflects the dominant granito-gneissic pan-african basement as source rock. Significant indicators of compaction were observed, further reducing primary porosity. Calcitic cements, often as traces, were discovered in less than half of the samples. The ubiquitous clay mineral group found was kaolinite/dickite (Bessong 2012; Bessong et al. 2011, 2015). Close to the Benue and its tributaries, the sandstones in the Garoua basin are eroded and overlain by quarternary siliciclastic alluvial deposits made of sand, gravel and clay (Njitchoua et al. 1995) with description in Tillement (1972).

Numerous (31) volcanic and sub-volcanic bodies of calc-alkaline composition with ages around 37 Ma, ranging from small volumes of basalt over dominant trachyte to rare rhyolite are cropping out in the center of the Garoua basin. The basalt has apatite contents exceeding 1 wt% P_2O_5 (Ngounouno et al. 2003).

Hydrogeology

Tillement (1972) distinguishes aquifers of the altered and fractured basement, aquifers of the cretaceous deposits and quarternary aquifers (in Olivry 1986). Granular disintegration has formed shallow (5–15 m), unconfined aquifers on granitoid-migmatitic pan-african basement with a yield generally below 2 m^3/h. In the Garoua grits, secondary porosity and fractures may enhance the hydraulic conductivity (K) to 10^{-4} to 10^{-5} m/s with a porosity estimate of 25%, making yields of 50 m^3/h possible. However, K is much lower in the finer-grained Benue grits as well as in the satellite basins which contain more silt, clay and beds of marls, with a reference to higher mineralization (EC) due to lithology being made here. The quaternary Benue sediments attain a thickness of up to 40 m, making it an important regional aquifer. In the 1:500,000 hydrogeological ORSTOM map (Dauteloup et al. 1971) the distance to the water table was indicated to be between 5 and 10 m below surface with a deeper water table on hill crests and shallower water table in the temporary rivers locally called
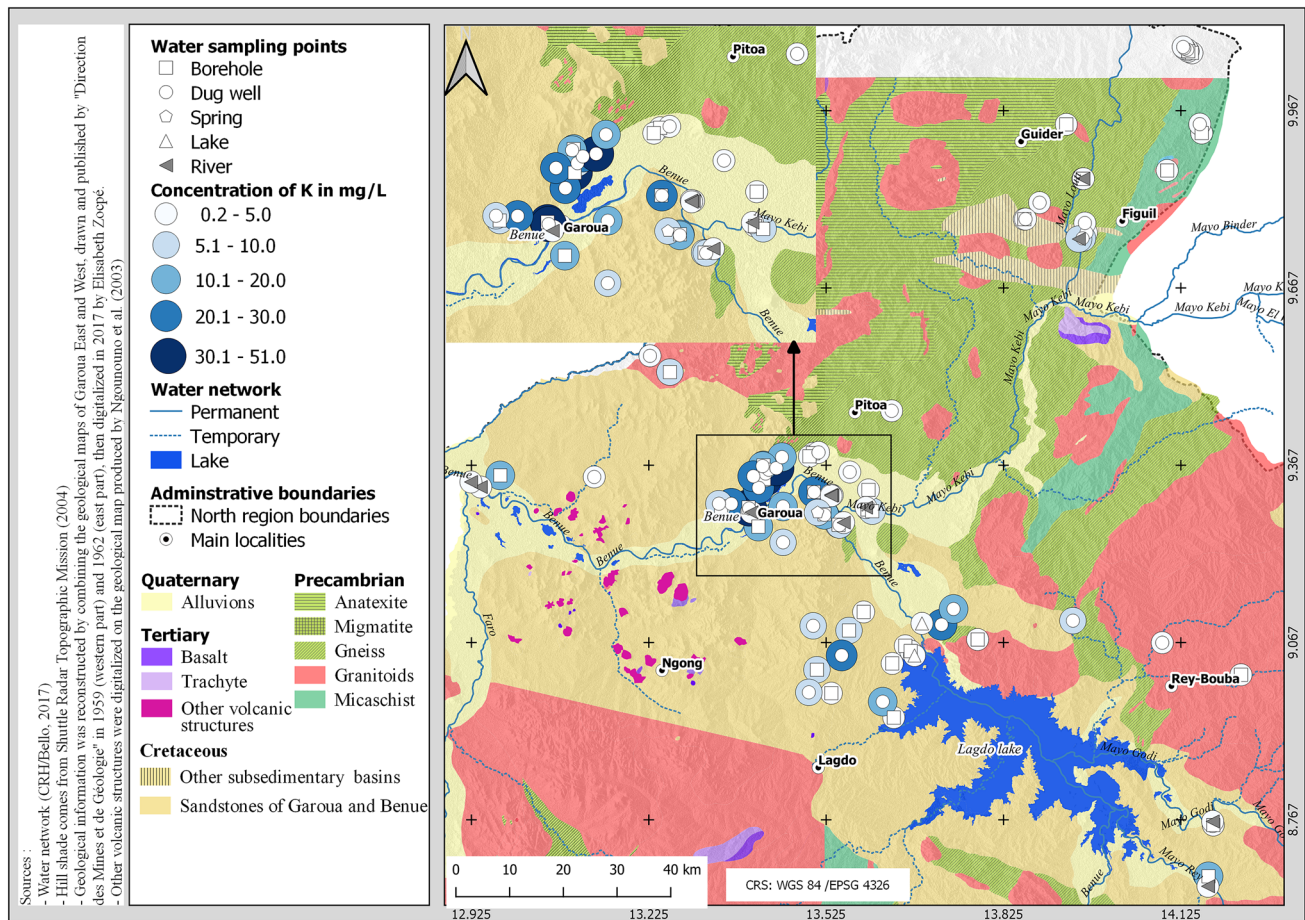
Fig. 2 Spatial distribution of potassium in ground- and surface water of the Upper Benue catchment in relation to surface geology of the area—dominated by siliclastic cretaceous sediments on proterozoic crystalline basement, tertiary volcanics digitized from Ngounouno et al. (2003), Lower Left: close up of urban Garoua; data Schwoerer (1962) source: 1:500,000 sheets Garoua East and West from Koch (1959) (https://doi.org/10.13140/RG.2.2.15769.36964) and, in https://doi.org/10.13140/RG.2.2.32546.58562), SRTM, OpenStreetmap

"mayos" and depressions. During dry season depths to underflow in the mayos is from 0.5 to 1 m. Nbendah et al. (2022) in preparation assessed the degree of maintenance and salubrity of 139 water points, including all in this study (Fig. 2). The median static water level of 7 m below surface in the dry season 2016/2017 appears not significantly different from the range indicated by Dauteloup et al. (1971). It has to be stressed, that virtually all water points in the tropics are used for water production and are usually very frequented by the local population, dedicated groundwater observation wells are practically unknown.

Hydrology

The River Benue with a total length of 1400 km flows only 350 km within the Cameroon territory (Molua and Lambi 2006). It descends from the central northern slopes of the Adamawa plateau. The tributaries which originate from the eastern sector are the Mayo Rey and the Mayo Godi. From the central part, it is the Mayo Farda from the Poli mountains. The Faro, Mayo Njal and Mayo Deo, which form the western sector of the tributaries all converge to join the Benue at the exit point into Nigeria (Olivry 1986). The mean climatic values for Garoua are 28.2 °C mean air temperature, precipitation of 1034 mm/a (32 years since hydrological year 1949–1950) and a potential evapotranspiration of 1922 mm/a. The discharge at Garoua descends to a minimum in April of 2.8 m^3/s and reaches its maximum in September with 1699 m^3/s (range 516–2803 m^3/s) with 96% of the annual discharge between June and end of November. The mean annual discharge at Garoua has been 360 m^3/s for a catchment area of 60 500 km^2, before completion of the Lagdo reservoir in 1982 (Olivry 1986). Midstream of the upper BRB the Lagdo reservoir has a surface area of between about 697 km^2 and 840 km^2 with a (nominal) corresponding volume between 6.0×10^9 m^3 and

7.7×10^9 m^3 of water for irrigation and hydroelectricity (Blanken and Pecher 2013; Grijsen and Worldbank 2014). Sedimentation has since reduced the dead volume[1] (below 206 m.a.s.l) of the lake. Net evaporation from the lake and irrigation water uptake reduce discharge by 17 m^3/s and on the order of 23 m^3/s, respectively (Grijsen and Worldbank 2014). In the dry season irrigation, navigation and fishing activities are much restricted according to Toro (1997). The river banks are being used as transhumance corridors for cattle herds at the beginning of the dry season, but they are also used by farmers who practice market gardening (e.g. tomatoes) or flood recession crops (e.g. cereals).

The quality of wet deposition in the savanna

In their worldwide rainwater quality assessment, Vet et al. (2014) came up with atmospheric transport model-based spatial distributions of precipitation chemistry parameters like sea salt, sulphate, nitrogen, base cations, acetate and formate, acidity as well as phosphorus, thereby surpassing the explicatory value of time series at individual stations. For the Sahel, the two savanna IDAF-sites in Banizoumbou (Western Niger) and in Djougou (Benin) can be considered more representative than the Cameroonian IDAF-site at Nsimi-Zoetele (Mfoupo Mewouo et al. 2009) in the humid forest zone. This is due to the important control of annual Harmattan dust in the dry season. Dust profoundly alters the chemistry of rain water by offering fine-grained mineral surfaces for reaction (Fantong et al. 2020). The transport of sea salt and with it, the background of chloride and sodium in rainwater are low in the savanna, due to distance from the coast and generally low wind velocity prevailing in the intertropical zone (Vet et al. 2014). The following ranges can be expected for the precipitation chemistry as an annual mean: the total deposition of sea salt is much lower than 10 kg/ha a. The sum of sulfur deposition is up to 2 kg S/ha a (6 kg SO_4/ha a) in the Sahel, the bulk being brought with rain. The sum of nitrogen deposition is in the range of 5–7 kg N/ha a (24–34 kg NO_3/ha a) in the Sahel with less than 50% occurring as dry deposition. The proportion of reduced and acid neutralizing NH_3 as N-species is rising in the savanna with emission from animals. Base cations (Ca^{2+}, Mg^{2+}, Na^+ and K^+) counter the natural acidity of rain water and are highly dependent of geology and dust provenance. Carbonic weak acid deposition (acetate, formate) depends on productive plant cover and in particular on biomass burning. It is higher in the wet savanna (Lamto) than in the dry savanna (Banizoumbou) with between 4 µeq/L (formate)

and 16 µeq/L (acetate). Acidity of rain in the wet savanna is reduced to pH 5.16 by neutralization from bases and ammonia, further increasing to pH 6.06 in the dry savanna of Niger. The Sahara and its adjacent Sahel zone stand out as a world hotspot for total phosphorous (TP) in aerosols. TP deposition can be up to 500 g P/ha a in the Sahel (Vet et al. 2014).

Sampling and chemical analysis

A sampling campaign was undertaken at the peak of the dry season in January 2017 during which 86 water samples were randomly[2] collected to obtain a snapshot of surface water and groundwater quality in the Benue catchment. These samples included 72 functional and frequented groundwater water points (37 dug wells, 34 boreholes and 1 spring) and 14 surface water sites (12 rivers and 2 lakes) above and below confluences. Most of the stream and springs were already dried up in dry season. Each densely populated sub-area of the catchment was represented. Surface water samples were collected under base-flow conditions (no run-off influence). Dug wells were of two types: open wells with depths-to-water level measured and hand pump wells sealed with unknown depths-to-water. While water points were in near constant use by the local population hand pump wells and boreholes were pumped for 5–15 min before sampling. Water was collected into a collector after thorough rinsing with the sample. The geographic coordinates and barometric altitudes of the selected sampling points were identified in the field using a Garmin 64S GPS receiver. In addition, water point management, land use, human activities and rock type were noted at each sampling site using a mobile Open Data Kit (ODK) smartphone application (Nbendah et al. 2018).

Hydronium activity pH (± 0.05 pH), electrical conductivity (EC ($\pm 0.5\%$)) and water temperature were determined on-site with portable Hanna kits. Alkalinity measurements were carried out within 10 h of sample collection by acid titration from the volume of 0.02 N HCl added to the sample to reach the titration end-point, which was marked by a pH of 4.5. Groundwater and surface water samples were collected using a bottle set consisting of: (1) cations, silica and trace elements (100 mL PE bottle, prepared with 1 wt% (1 mL) HNO_3cc_{sp}, sample filtrated through 0.45 µm cellulose acetate membrane disposable filter after conditioning of syringe and filter) and (2) anions and isotopes (50 mL PE bottle, pre-rinsed with sample).

Hydrochemical analysis was done after DHL delivery to the laboratory of the Federal Institute for Geosciences

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[1] The dead volume storage in a reservoir is determined as the storage volume between the streambed and the lowest elevation from which water can be withdrawn by gravity (Mahmood 1987).

[2] River samples taken upstream and downstream of major confluences.

and Natural Resources (BGR), Hanover/Germany within 4 weeks. Cooled storage was assured. Cations were analysed via inductively coupled plasma optical emission spectrometry using a Ciros by Spectro (DIN EN ISO 11885 (1998)). Nitrate, sulphate, chloride and bromide were analysed by ion chromatography (IC-Dionex ICS-3000) (DIN EN ISO 10304–1_(1997). Ammonium was determined spectro-photometrically with a Unicam UV300 (Thermo Electron Corporation) with the blue indophenole reaction (DIN38406E5:(1983)). Stable environmental isotopes ratios $\delta^{18}O$ and $\delta^{2}D$ of water were determined on a PICARRO cavity ring-down spectrometer (CRDS model L2120-i) following the procedures described by Brand et al. (2009) and Lis et al. (2008). Parameters analyzed included K^+, Na^+, Mg^{2+}, Ca^{2+}, NH_4^+, Fe^{2+}, Mn^{2+}, Cl^-, F^-, Br^-, NO_3^-, NO_2^-, SO_4^2 PO_4^{3-}, SiO_2. For the major elements, reliabilities of the chemical measurements were verified by charge balance (Appelo and Postma 2005) and were within the limit of less than $\pm 5\%$ for all investigated samples.

Data analysis

Data analysis started with descriptive statistics and boxplots for the most pertinent parameters to gain insight into the state of water quality of the samples using the R program (ver. 3.6.1; http://www.r-project.org) (R Core Team 2019). The Schoeller diagram (Schoeller, 1960) was used to present the average chemical composition of groundwater using the "DIAGRAMMES" program package (version 6.59, http://www.lha.univ-avignon.fr) (Roland, 2017). QGIS software (ver. 2.18; QGIS Development Team, 2018), was used to symbolize the spatial distribution of water quality parameters. To attribute surface geology the 1: 500,000 map sheets from Koch (1959) Garoua West and Schwoerer (1965) Garoua East digitalized by Elisabeth et al. (2018a, b) has been used.

Multivariate statistical analyses

Multivariate statistics especially principal component analysis (PCA) and hierarchical cluster analysis (HCA) are key methods to evaluate the multivariate relationship between the water quality parameters and land cover to identify the key drivers affecting water quality and influences on the aquatic ecosystem (Rodríguez-Romero et al. 2018). They have been extensively used to characterize and evaluate groundwater and surface water quality, revealing temporal and spatial variations caused by natural and anthropogenic factors (Güler et al. 2002; Ouyang 2005; Ako et al. 2014; Kamtchueng et al. 2016; Nyenje et al. 2010). They are designed to reduce the number of variables to a small number of indices (i.e., principal components or dimensions) while attempting to preserve the relationships present in the

original data. The problems of identification of hydrogeochemical processes, impact of anthropogenic activities, classification of water types, data reduction and interpretation and characteristic change in water quality parameters can be approached through the use of the PCA and HCA (Nyenje et al. 2010; Thin et al. 2018).

We applied PCA and HCA via R program to better understand the variability in both groundwater and surface water composition and processes likely controlling the fate of nutrients in the BRB. Principal component analysis (PCA) has been chosen to aggregate the effects of many variables into a small subset of factors called principal components (PCs). The PCs may reveal the effects of water–rock interaction, climate, and human activity on surface and groundwater quality. Hierarchical cluster analysis (HCA), as a branch of cluster analysis was used to subdivide water samples so that samples resulting subgroups or clusters are similar to each other, but distinct from other groups. HCA which successively joins the most similar observations, is widely used in hydrochemical analyses to identify different chemical characteristics (Li et al., 2018) with the advantage of not demanding any prior knowledge of the number of the cluster that the non-hierarchical method does (Yang et al. 2015; Reghunath et al. 2002).

The PCA and HCA methods were applied to the hydrochemical dataset of 86 water samples including 14 surface waters. Data consisted of 17 hydrochemical parameters (variables) including EC, pH, Na^+, Ca^{2+}, K^+, Mg^{2+}, NH_4^+, Cl^- SO_4^{2-}, NO_3^-, NO_2^-, HCO_3^-, F^-, PO_4^{3-}, Mn^{2+}, Fe^{2+} and SiO_2. The parameters NH_4^+, NO_2^-, PO_4^{3-}, Fe^{2+} and Mn^{2+} were excluded from the analyses because they were below detection limits for a large number of samples. Güler et al. (2002) reported that such data is not appropriate for multivariate analysis. Hence multivariate statistics were applied only on 12 variables. Before running multivariate analyses, data were first screened for homoscedasticity (homogeneity of variance) and normality via histograms and Q–Q plots. The distributions of most parameters were highly skewed to the right. Hence, all data were log-transformed so as to correspond to normally distributed data and to remove heteroscedasticity. Secondly, data were first standardized using median and the mean absolute deviation to ensure that each variable was weighted equally. Furthermore, using median values minimizes errors created by outliers (Güler et al. 2002; Nyenje et al. 2014). For the initial factor extraction Kaiser's (1958) eigenvalue criterion of retaining only those components/factors whose eigenvalues are greater than 1 was adopted. Other criteria used to determine the appropriate number of components to retain included scree plot, variance and residuals. Kaiser's varimax rotation, an orthogonal rotation procedure which produces a set of component loadings having the maximum variance of the squares of the loadings is used in conducting the PCA to make the factor

solutions more interpretable without altering the underlying mathematical structure (Mertler and Vannatta 2005).

PCA reduced the remaining 12 variables to 3 uncorrelated PC's explaining 77% of the total variance present in the dataset. PCA results were presented in terms of component scores (the individual transformed observations) and loadings (the weight by which each standardized original variable should be multiplied to get the component score in the PCs). A varimax rotation, a form of orthogonal rotation, was applied to the PCs to minimise the contribution of variables that had minimal significance (i.e. low loadings) and maximize the contribution of variables with high loadings (Mertler and Vannatta 2005; Owoseni et al. 2013).

HCA results were presented as a dendrogram using the euclidean distance for similarity measures and Ward's method for linkage (Güler et al. 2002; Nyenje et al. 2014). To further evaluate the geochemistry of groundwater, major constituents where polled in Shoeller diagram. In this case, major cations ($Na^+ + K^+$, Ca^{2+} and Mg^{2+}) where plotted on the left side of the diagram with the major anions (SO_4^{2-}, $HCO_3^- + CO_3^{2-}$ and Cl^-) plotted on the right side.

Land cover attribution

Global Land Cover data from Africa land cover characteristics database version 2.0 (USGS, 2018) was used to attribute sampling sites with an additional variable to expose the spatial relationship with multivariate statistical analysis. This is because the complexity of information in water quality data needs new statistical approaches which uncovers spatio-temporal variability (Afed Ullah et al. 2018).

Results and discussion

Descriptive statistics and concentration ranges of nutrients

Table 1 shows the descriptive statistics of the hydro-chemical data for the surface water and groundwater samples collected from the upper Benue River Basin (BRB) (Fig. 1) compared to concentrations from the groundwater of Garoua (Njitchoua et al., 1997) and rainfall from IDAF savannah site Djougou (Benin) (Akpo et al. 2015). Values are given as minimum, percentiles p10, p25, p50 (median value), p75, p90 and maximum. Generally, values of most parameters are not normally distributed and are right skewed, median has been used as the center of the distribution of data instead of arithmetic mean, to avoid a strong influence of outliers. The complete list of all major ions and stable isotope ratios of groundwater ($n = 72$) and surface water ($n = 14$) in the BRB is presented in Table 1 in the ESM appendix.

The median water temperature of surface water (SW) was lower than that of groundwater (GW). In SW, values ranged from 24.1 to 32.2 °C (median = 27.4 °C) while GW ranged from 23.9 to 32.3 °C (median = 30.0 °C). For comparison, Njitchoua et al. (1997), who carried out a similar study in the area, obtained values ranged from 30.1 to 31.9 °C (median = 30.7 °C), similar to our result (Table 1). While surface water undergoes a diurnal temperature cycle, groundwater generally does not.

In surface water (SW) pH varied from 6.4 into the alkaline range at 8.9 (median = 7.7) and from 5.8 to 7.8 (median = 6.8) in GW. Meanwhile, the pH of rain water of savannah site Djougou was acidic with a mean of 5.1 (range = 5.0–5.3) (Table 1). All water samples were, therefore, circum-neutral to mildly acidic within the HCO_3 buffer range of natural waters. The concentration of dissolved salts varied over a wide range with electrical conductivity (EC) ranging from 50 to 222 μS/cm (median = 81 μS/cm) in SW and from 35 to 2500 μS/cm (median = 376 μS/cm) in GW (Table 1). The considerably higher median EC obtained in groundwater indicates the amount of dissolved salts (ions) added from natural sources and human activities combined.

A boxplot of the univariate distribution of the macronutrients (NO_3^-, SO_4^{2-}, K^+ and PO_4^{3-}) including chloride (Cl^-) as a conservative parameter is given in Fig. 3. The concentration of these parameters-related nutrients is in the abundance order of $K^+ > NO_3^- > Cl^- > SO_4^{2-} > PO_4^{3-}$ in surface water compared to $NO_3^- > Cl^- > SO_4^{2-} > K^+ > PO_4^{3-}$ in groundwater.

The median values of nitrate (NO_3^-) were 0.32 mg/L (range = 0.01–2.08 mg/L) and 41.25 mg/L (range = 0.01–823 mg/L), respectively, in SW and GW (Fig. 3 and Table 1). The rain water shows much lower concentrations with a mean of 0.05 mg/L (range = 0.004–0.006 mg/L) (Table 1). As a limiting nutrient for plant growth, NO_3^- is readily assimilated by macrophytes and algae in SW, especially when the flow rate and turbulence is low. NO_3^- in GW shows an enrichment factor of 129 compared to SW and 825 compared to rain water. Rain water clearly is a source of nitrogen at low concentrations (Vet et al. 2014; Akpo et al. 2015). Nitrate above 10 mg/L reflects anthropogenic contamination (Kloppmann 2013; Ollagnier and Vittecoq 2007). The common sources of nitrate in an agriculturally dominated catchment like BRB are the application of fertilizer, animal excreta and domestic wastewater discharge (Khatri & Tyagi 2015). With the majority of NH_4^+ concentration close to or below the detection limit (8 from 86 ≥ 0.3 mg/L), dissolved nitrogen is significant only in its fully oxidized form nitrate. Thus, in a general way, the prevailing geochemical milieu of the upper unconfined aquifer can be constrained as overwhelmingly « oxic». This falls in line with generally very low concentrations of dissolved iron (median = 0.01 mg/L, 5 outliers)

Table 1 Descriptive statistics of the hydrochemistry of the surface water and groundwater samples collected from Benue catchment (around Garoua) compared to concentrations from the groundwater of Garoua (Njitchoua et al. 1997) and rainfall from IDAF savannah site Djougou (Benin) (Akpo et al. 2015)

Parameter	Temp	pH	EC	Na+	K+	Mg2+	Ca2+	NH4+	HCO3-	Cl-	NO3-	NO2-	SO4²⁻	PO4³⁻	Fe2+	Mn2+	Al3+
Unit	°C	–	µS/cm	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L	µg/L
Rain water, IDAF savannah site Djougou (Benin) (Akpo et al. 2015) form 2005 to 2009																	
Minimum	–	5.0	–	0.0004	0.0006	0.0001	0.0020	0.0015	–	0.0008	0.0038	–	0.0027	–	–	–	–
Maximum	–	5.3	–	0.0012	0.0010	0.0003	0.0028	0.0031	–	0.0015	0.0056	–	0.0035	–	–	–	–
Mean	–	5.1	–	0.0009	0.0007	0.0002	0.0025	0.0077	–	0.0012	0.0048	–	0.0031	–	–	–	–
Std. dev.	–	0.1	–	0.0003	0.0001	0.0000	0.0003	0.0005	–	0.0002	0.0006	–	0.0003	–	–	–	–
Surface water (SW)																	
n (number of samples) in each cluster	14	14	14	14	14	14	14	12	14	14	14	2	14	8	14	14	14
Minimum	24.1	6.4	50	3.70	1.90	2.03	6.16	0.01	0.21	0.26	0.01	< 0.003	0.09	< 0.003	0.004	0.001	0.003
p10	24.5	6.7	63	3.73	2.30	2.03	6.18	0.01	6.39	0.29	0.06	–	0.09	0.03	0.006	0.001	0.003
p25	25.0	7.2	65	3.82	2.40	2.05	6.31	0.01	22.25	0.32	0.14	–	0.11	–	0.009	0.001	0.006
p50	27.4	7.7	81	4.85	3.30	2.69	7.74	0.01	30.62	0.41	0.32	0.520	0.17	–	0.057	0.026	0.008
p75	29.2	8.2	137	6.67	3.80	3.99	13.15	0.02	45.96	0.79	1.02	–	0.62	–	0.182	0.151	0.094
p90	29.9	8.6	181	13.70	4.00	4.39	18.11	0.06	54.39	3.07	1.76	–	1.58	–	0.339	0.492	0.124
Maximum	32.2	8.9	222	16.50	5.50	4.55	24.80	0.07	69.24	6.27	2.08	0.722	2.65	0.60	0.691	0.837	0.381
Mean	27.4	7.6	106	6.75	2.93	3.03	10.77	0.02	33.05	1.15	0.66	0.515	0.56	0.15	0.137	0.149	0.060
Std Dev	3.0	0.9	61	4.35	1.01	1.19	6.24	0.02	23.30	1.44	0.78	0.413	0.73	0.17	0.191	0.237	0.090
Groundwater, Garoua 1991 (Njitchoua et al. 1997)																	
n	25	25	–	23	23	23	23	–	–	25	25	–	25	–	–	–	–
Minimum	30.1	5.0	–	2.10	1.80	0.10	0.90	–	–	0.2	0.20	–	0.60	–	–	–	–
p10	30.4	5.1	–	2.52	3.90	0.34	3.26	–	–	0.20	0.70	–	0.74	–	–	–	–
p25	30.5	5.4	–	3.80	4.70	1.80	7.05	–	–	0.30	0.90	–	0.90	–	–	–	–
p50	30.7	6.2	–	7.00	5.90	2.60	13.40	–	–	0.80	2.00	–	1.30	–	–	–	–
p75	31.1	7.4	–	27.45	7.55	6.35	17.70	–	–	3.30	16.90	–	3.10	–	–	–	–
p90	31.5	7.6	–	130.78	15.04	12.36	27.82	–	–	5.46	36.20	–	7.06	–	–	–	–
Maximum	31.9	8.2	–	159.50	21.10	27.90	39.7	–	–	21.4	50.70	–	11.50	–	–	–	–
Mean	30.8	6.3	–	34.06	7.56	5.86	14.33	–	–	2.75	10.67	–	2.74	–	–	–	–
Std. dev.	0.5	1.3	–	48.62	5.10	7.24	11.07	–	–	3.44	14.90	–	2.95	–	–	–	–
Groundwater (GW)																	
n	72	72	72	72	72	72	72	59	72	72	72	54	72	50	72	72	34
Minimum	23.9	5.8	35	1.10	0.20	0.18	0.98	< 0.001	0.24	0.24	0.01	< 0.003	0.01	< 0.003	< 0.003	0.000	< 0.003
p10	27.1	6.1	83	3.91	1.21	1.23	5.21	0.01	3.24	0.80	0.80	0.030	0.20	0.04	0.003	0.001	0.003
p25	28.8	6.5	227	12.45	2.00	3.68	16.85	0.01	8.80	2.37	9.16	0.004	1.50	0.05	0.004	0.002	0.003
p50	30.0	6.8	376	23.90	5.20	8.46	33.90	0.01	49.65	17.20	41.25	0.010	7.70	0.11	0.012	0.011	0.005
p75	30.9	7.1	662	48.43	12.05	23.85	64.27	0.04	144.55	27.07	92.37	0.023	16.37	0.33	0.080	0.040	0.008

Table 1 (continued)

Parameter	Temp	pH	EC	Na^+	K^+	Mg^{2+}	Ca^{2+}	NH_4^+	HCO_3^-	Cl^-	NO_3^-	NO_2^-	SO_4^{2-}	PO_4^{3-}	Fe^{2+}	Mn^{2+}	Al^{3+}
Unit	°C	–	µS/cm	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L	mg/L	µg/L
p90	31.5	7.4	1011	70.70	25.97	40.50	90.53	0.05	314.58	74.06	208.60	0.071	39.10	1.10	0.663	0.171	0.120
Maximum	32.3	7.8	2500	244.00	55.50	124.00	254.00	0.09	608.29	345.00	823.00	0.388	177.00	2.14	3.130	0.607	2.040
Mean	29.7	6.8	518	37.60	9.31	17.55	48.24	0.02	106.80	29.03	83.35	0.030	16.00	0.35	0.204	0.060	0.087
Std. dev.	1.7	0.5	463	40.27	10.55	20.53	46.87	0.01	138.90	38.91	110.00	0.042	22.00	0.46	0.035	0.094	0.161
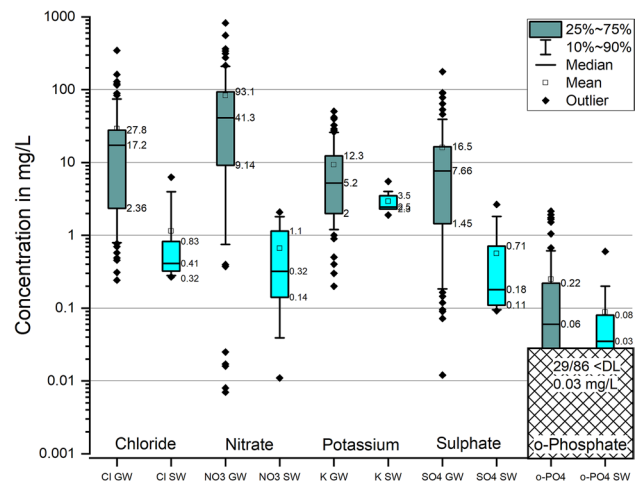
Fig. 3 Boxplot of macronutrients and chloride of groundwater and surface water

and manganese (median = 0.01 mg/L, 6 outliers) (Table 1), indicating precipitation of oxyhydroxides during weathering.

As seen in Fig. 4 nitrate is strongly correlated with chloride ($r = 0.8$) in both SW and GW, thus reflecting pollution of anthropogenic origin. Furthermore, nitrate positively correlates with SO_4^{2-}, Ca^{2+}, Mg^{2+} and EC ($r = 0.6$) in groundwater further implying a linkage with anthropogenic input. A weak correlation between nitrate and Na^+, K^+ and HCO_3^- ($r = 0.4, 0.3, 0.3$) (Fig. 4) in GW on the other hand indicates that the latter stem largely from water–rock interaction. Our median concentration already exceeds three times the concentrations observed 25 years ago by Njitchoua et al. (1997). It is very concerning that the median concentration of nitrate is already approaching the WHO recommendation value of 50 mg/L and that already 44.4% of all groundwater samples in the BRB exceed this value considerably.

The median potassium (K^+) concentration of 3.30 mg/L (range: 1.9–4.0 mg/L) in SW was only half that of GW with 5.2 mg/L (range: 0.2–55.5 mg/L) (Fig. 3 and Table 1). A mean K^+ concentration in rainwater of 0.0007 mg/L (Table 1) (Akpo et al 2015) shows a negligible influence of wet deposition. In groundwater, K^+ was 2 times higher concentrated than in SW and correlates negatively with fluoride ($r = -0.6$) (Table 1 and Fig. 3). In SW, K^+ was positively correlated with Cl^-, Na^+, Ca^{2+}, Mg^{2+}, SO_4^{2-} and NO_3^- (r = 0.9, 0.8, 0.8, 0.7, 0.7 and 0.6) (Fig. 4). Mineral sources of K^+ are present abundantly in the BRB (Fig. 2) primarily as K-feldspars (here largely microcline) but also in muscovite/biotite (micas) giving rise to subordinate illite (Bessong 2012). A significant geogenic background concentration is thus to be expected from their continuing weathering (Eq. 1) where K-feldspars dissolve incongruently, leaving kaolinite as a solid residue. According to the weathering rates typical for arkosic and wacke sandstones of

the Cretaceous (Bessong 2012; Ntsama 2013; Bessong et al. 2015) and of the proterozoic crystalline basement (Bessong 2012) underlying the region, there is continuous mobilization of K^+ by weathering. This geogenic background of K^+ is relevant in the light of plant nutrition as outlined by Manning (2010). K^+ in surface water might be linked to anthropogenic inputs, as follows from a high concentration of KCl in urine or use of agricultural fertilizers (Putnam, 1971; Skowron et al. 2018). It is to remind here that surface water sites are preferred places for cattle and their migration:

$$2KAlSi_3O_8 + 2H_2CO_3 + H_2O \rightarrow Al_2(OH)_4Si_2O_5$$
$$+ 2HCO_3^- + 2K^+ + 4SiO_{2(aq)}. \tag{1}$$
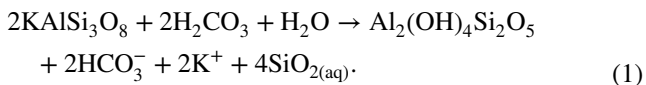
In the context of K-feldspar weathering, we briefly return to N here, as K-feldspar is a well-known, slow-release source for geogenic nitrogen (Deer et al. 1962; Orberger et al. 2005; Ciceri and Allanore 2019), because NH_4^+ can take the position of K^+ in the mineral lattice of K-Feldspar and micas. Locally even N-feldspar is possible in geological settings influenced by N-rich late magmatic and/or volcanic fluids giving rise to buddingtonite (Svensen et al. 2008). While localized volcanism was widespread 37 Ma ago (e.g. Ngounounou et al. 2003), in all cases a slow release rate, yet steady supply of geogenic mineral NH_4^+ from weathering of rock forming K-feldspar and micas (Bessong 2012) is to be expected.

The principal sources of phosphorus to surface water and groundwater systems include overlying soils, fertilizer application, animal waste, biocide application, leaking septic systems or infiltration of wastewater and dissolution of minerals that contain phosphate in aquifer sediments (Fuhrer et al. 1999; Welch et al 2010). The phosphate (o-PO_4^{3-}) in SW had a low median concentration of 0.08 mg/L ranging from below the detection limit of <0.003 mg/L to a cattle droppings contaminated site in Mayo Kebi with 0.60 mg/L as an extreme outlier (Fig. 3 and Table 1). Concentrations of o-PO_4^{3-} in SW showed a correlation with NO_3^- ($r = 0.5$) suggesting an anthropogenic source (Welch et al 2010). In contrast, GW with a high median concentration of 0.11 mg/L, ranged from 23 water points below the detection limit (<0.003 mg/L) to 13 sites above 0.5 mg/L with an extreme outlier of 2.1 mg/L. The water points above 0.5 mg/L were situated in (sub)urban Garoua (I, II, III) and Lagdo. In spite of these anthropogenically high concentrations, the lower median concentration of 0.11 mg/L is a strong indication, that o-PO_4^{3-} is not readily sorbed in the geochemical aquifer environment of the BRB as compared to e.g. the ferralitic subsurface of Yaounde, where P is consistently below the detection limit (Kringel et al. 2016). Feldspars are well known for impurities of phosphorus (P) in varying proportions with their weathering linked to P-content (Rogers 1995). So, a significant geogenic background

for this growth limiting key nutrient is to be expected in the regional geological setting of arkosic and wacke sandstones and the proterozoic granito-gneissic basement rich in K-feldspar, undergoing incongruent dissolution of silicates as outlined in Njitchoua et al. (1997) and Fantong et al. (2020). In addition to feldspar, accessory apatite is to be expected in granitoid terrain and concentrated local sources exist in tertiary basalts (Ngounounou et al. 2003). Moreover, the evaluation of vivianite ($FePO_4 \cdot 4H_2O$) deposits for fertilizer use has been reported from upstream (Fodoue et al. 2015). This points to the several sources of geogenic contributions this key nutrient appears to have in the upper BRB.

The sulphate (SO_4^{2-}) concentration for its part follows the pattern rain water $\approx$ SW < GW with median values of 0.0031 mg/L (range = 0.0027–0.0035 mg/L), 0.17 mg/L (range = 0.09–2.65 mg/L) and 7.7 mg/L (range: 0.0–177.0 mg/L), respectively (Fig. 3 and Table 1). In groundwater, SO_4^{2-} was 45 times higher concentrated than in SW and was positively correlated with Ca^{2+}, Mg^{2+}, Cl^- and Na^+ ($r = 0.8, 0.8, 0.8$ and 0.7) (Fig. 3). Higher concentrations than 30 mg/L were found only outside the Cretaceous sandstones at Figuil, Pitoa and Barndaké. At Figuil a geogenic contribution from the local Cretaceous mudstones and marls (Fig. 2) may be expected. While sulphate contribution from the weathering of sulphides in local basement rock is likely very low or nil, anthropogenic pollution (fertilization, dissolution of organic matter) is assumed to be dominant with the addition of a small atmospheric contribution on the order of 6 kg SO_4/ha an according to Vet et al. (2014)

Principal component analysis

Principal component analysis (PCA) has been carried out to complement correlation analysis which just reveals similarities or differences in the concentration of pairs of ions to identify groups of ions that behave similarly. PCA was applied to 12 variables (EC, pH, Na^+, Ca^{2+}, K^+, Mg^{2+}, Cl^- SO_4^{2-}, NO_3^-, HCO_3^-, F^- and SiO_2) which produced three uncorrelated ($p < 0.001$) components (dimensions) with eigen values greater than 1 (ESM Table 1). This criterion was proposed by Kaiser (1958) and has been widely used (Ako et al. 2011; Nyenje et al. 2014; Kamtchueng et al. 2016). The three dimensions selected in this study all together account for 77% of the total data variability which adequately explain and reflect the major variation of the data and water chemistry found in ESM Table 1.

The contribution of variables to the selected dimensions is shown in bold and underlined type in Table 2, with loadings above 0.5 considered significant. The first dimension (Dim-1) has the highest variance of 6.58 representing 55% of the total variation and exhibits positive weightings of 8 variables in the order EC > Mg^{2+} > Ca^{2+} > Cl^- > NO_3^- > SO_4^{2-} > Na^+ (Table 2). The dominance of Mg^{2+} and Ca^{2+}

Table 2 Principal component loadings (–) and explained variance for three components with varimax normalized rotation

Variables	Principal components		
	Dim-1	Dim-2	Dim-3
pH	− 0.03	**0.64**	**-0.5**7
EC	**0.98**	− 0.01	0.00
HCO$_3$	0.47	0.15	0.39
K$^+$	− 0.11	**-0.73**	0.13
Na$^+$	**0.83**	0.20	0.00
Cl$^-$	**0.91**	− 0.28	− 0.20
Mg^{2+}	**0.96**	0.07	− 0.07
Ca^{2+}	**0.93**	− 0.07	− 0.07
SO$_4^{2-}$	**0.88**	− 0.18	− 0.24
NO$_3^-$	**0.90**	− 0.28	− 0.13
F$^-$	0.48	**0.63**	0.26
SiO$_2$	**0.52**	0.16	**0.65**
Eigen value	6.58	1.62	1.10
Explained variance (%)	54.87	13.45	9.21
Cumulative % of variance	54.87	68.34	77.56

The bold and underlined figures are loadings that are significant (> 0.5)

which strongly correlate with EC is indicative of an increase in mineralization, whereas the participation of the anions Cl$^-$ and NO$_3^-$ indicates their key role in this dimension, reflecting the anthropogenic source. Pollution could be attributed to the use of fertilizers manure, livestock droppings and are generally related to land use (Ako et al. 2011; Nyenje et al. 2014; Egbi et al. 2019). The second dimension (Dim-2), accounting for 13% of the total variance is characterized by negative loadings of K$^+$ and positive loadings of F$^-$ > pH (ESM Table 1). A negative loading of K$^+$ goes with

reduced concentrations while a positive loading of F$^-$ and pH is associated with increased concentration and pH. The potassium map in Fig. 2 shows that low concentrations are mostly found in aquifers on outcrops of migmatitic-granitoid proterozoic basement, in particular to the N of the Benue. Referring to the fluoride map of Fantong et al. (2020) shows these water points are of concern because of their high fluoride concentrations exceeding the WHO recommendation value. Therefore Dim-2 is essentially reflecting the lithological contrast between fluoride-rich proterozoic basement aquifers and cretaceous and quaternary sediment aquifers. The variables SiO$_2$ and pH contribute most to the third dimension Dim-3, which explains 9% of the total variance. The strong positive correlation with SiO$_2$ and negative correlation with pH is likely due to H$_2$CO$_3$ induced weathering of the silicate minerals in the study area (Table 2). It can be attributed to the incongruent dissolution of silicates as suggested by Fantong et al. (2020). The PCA results suggest that anthropogenic input use of fertilizers, waste of livestock and water–rock interaction are the most important processes affecting groundwater chemistry in the catchment.

Hierarchical cluster analysis, land cover and water quality

Hierarchical cluster analysis (HCA) was applied to combine the water samples in the study area into homogeneous groups due to their chemical composition. In this study, 12 transformed variables (EC, pH, Na$^+$, Ca^{2+}, K$^+$, Mg^{2+}, Cl$^-$ SO$_4^{2-}$, NO^{3-}, HCO$_3^-$, F$^-$ and SiO$_2$) were selected and used to group the 86 water samples using Ward's method. The dendrogram obtained (Fig. 5) has three distinct hydrochemical groups: I, II and III with a total of five subgroups (clusters). These are cluster 1 (blue) and cluster 2 (magenta)

Fig. 4 Visualisation of the correlation matrix of major ions with Spearman correlation coefficient
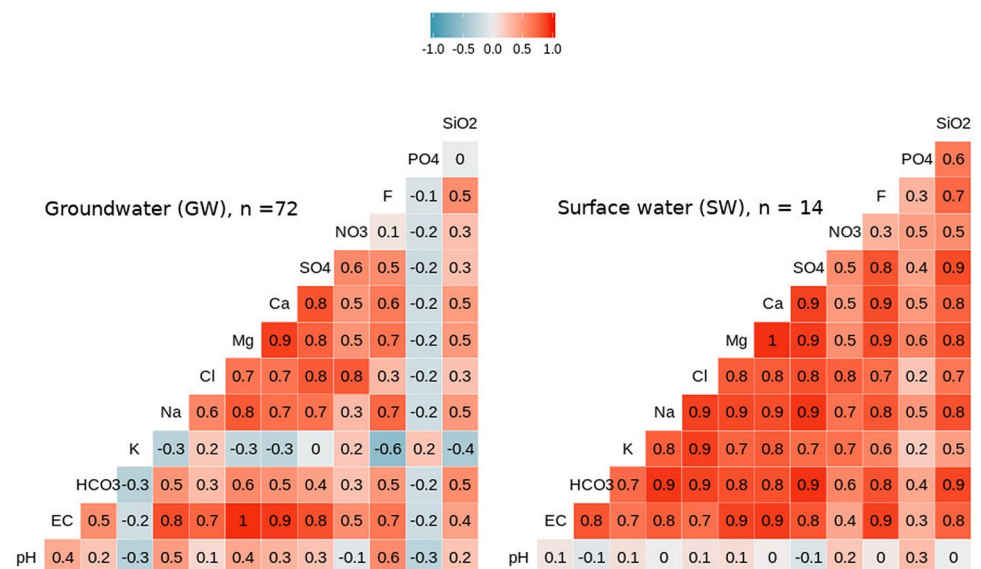
under group I (to the right), a single cluster 3 (green) under group II and clusters 4 and 5 under group III (to the left). A much higher linkage distance of 4.3 separates group III (Fig. 5) from the other two groups, indicating a wider compositional difference to these groups. Group II was linked to group I at a linkage distance of about 1.3 indicating a much smaller compositional difference.

The first three clusters 1, 2 and cluster 3 share a similar anionic composition dominated by HCO_3 with the equivalent concentrations in decreasing order $HCO_3^- > NO_3^- > Cl^- \geq SO_4^{2-}$ in Fig. 6. These clusters also share a similar cationic composition, dominated by $Na + K$ with the equivalent concentration in decreasing order $Na^+ + K^+ > Ca^{2+} > Mg^{2+}$ in (Fig. 6). The chemical composition of these clusters is characterized by the $Na + K - HCO_3$ hydrological facies or water type (Schoeller, 1960; Thin et al. 2018). Cluster 1 (blue) is composed of all surface water samples including two wells, two boreholes and the spring which share a low mineralization (median EC = 70 µS/cm, median $NO_3^- = 0.32$ mg/L). Cluster 3 (green) samples are considerably higher concentrated tending towards the brackish range with a median EC = 638 µS/cm (median $NO_3^- = 37.25$ mg/L) (Table 3). The samples of the last clusters 4 (dark yellow) and the single sample (outlier W081) cluster 5 are characterized by high EC values (cluster 4 = 421 µS/cm and cluster 5 = 2500 µS/cm). They show a different ionic composition dominated by NO_3^-, with the equivalent concentration decreasing order of $NO_3^- > HCO_3^- > Cl^- > SO_4^{2-}$ for anions and $Ca^{2+} > Mg^{2+} > Na^+ + K^+$ for cations (Fig. 6). Thus, they are characterized as the $Ca + Mg - NO_3$ water type. From this above and the Schoeller plot (Fig. 6) it follows that there

is a transition from low mineralization to high mineralization with only minor compositional change from cluster 1 to cluster 3, while cluster 4 (and outlier 5) are compositionally different and strongly mineralized.

A relevant question is whether these 5 clusters and their composition can be (partly) explained by the land cover pattern in the upper BRB upstream and downstream of Garoua. To test the relationship between the statistical cluster groups obtained with the land cover attributes (Global Land Cover 2018) samples for each cluster were plotted on the land cover (Fig. 7). Clusters 1 and 2 are located either in the urban area/cultivated land around Garoua and Lagdo or in grassland along with BRB. Clusters 3 and 4 (and outlier 5) are located N and E of the course of the Benue in Grassland, cultivated area and urban area. The compositionally different cluster 4 with samples at Lagdo, Ngong, Guider ($n = 3$) and Figuil (e.g. emissions from cement mill) are all either from aquifers on proterozoic basement or from the northern satellite cretaceous basins, which are different in lithology from the Garoua basin (e.g. calcareous mudstones of the Figuil basin) in the geological map in Fig. 2. The spatial distribution of the cluster groups superimposed on the land cover map shows a significant heterogeneity (Fig. 7) and suggests an underlying spatial geological influence. This result shows an increase of groundwater mineralization by anthropogenic factors linked to agriculture, use of fertilizer and manure, animal husbandry, pollution due to industrial effluents and domestic sewage and mining. While biocide use is visibly prevalent, it's possible influence on groundwater quality has not yet been assessed. Olina Bassala (2010) reports that Glyphosate (Phosphonomethyl aminoacetic acid, $C_3H_8NO_5P$) is a biocide used frequently at least once a year,
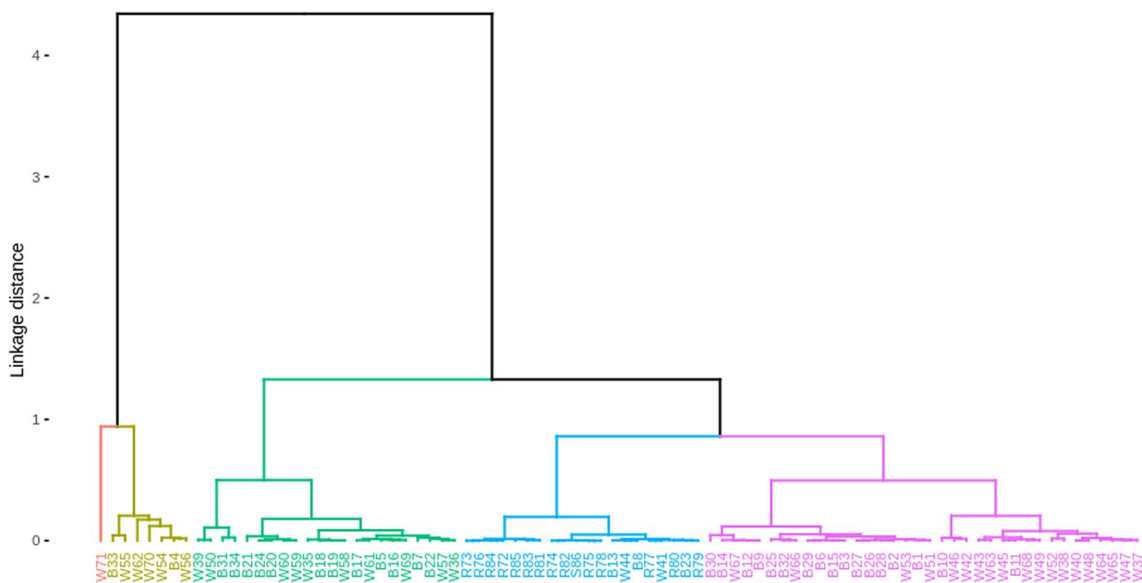

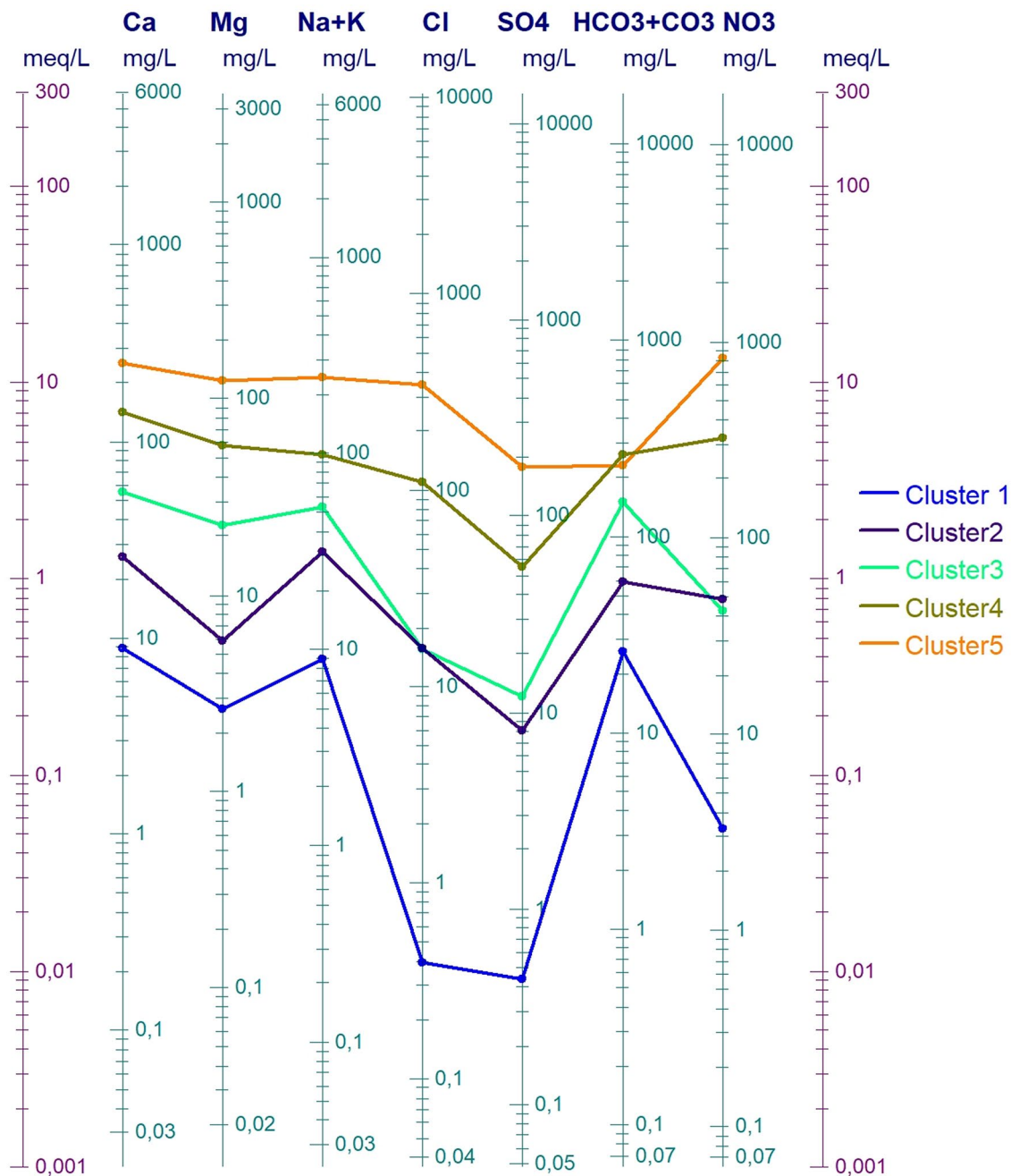**Fig. 5** Dendrogram of the hierarchical cluster analysis

Fig. 6 Schoeller diagram of each cluster in mg/L concentration scales linked to the meq/L concentration scale (on both sides)

at an average application rate of 1440 g/ha on soils used for cotton farming, but also for the other crops in the three-year crop rotation schema with maize and groundnuts.

Compared to European fertilizer use, total fertilizer imports to Cameroon (amounting to 22,400 tons Total Nitrogen in 2012, http://www.fertilizer.org, IFA 2015 in Kringel et al. (2016)) shows that presently the use is still low, due to the high cost to small holders which typically farm an area of 1–2 ha in the North Region. Yet the parastatal

SODECOTON company is the largest user and distributor of fertilizer in the region. Between 1994 and 1995, 95% of the cotton area received 150 kg/ha of fertiliser (SODECON 2003). For the 1999–2000 growing season, cotton crops consumed 8684 tons of fertilizer formulated 22–10–15, 15,150 tonnes of fertiliser formulated 15-20-15, 63 tons of fertiliser formulated 5-19-19 and 4911 tons of urea, e.g., 28,858 tons of fertiliser in total (SODECOTON 2004, 2005, 2006). The recommended application rates for the area are 200 kg/ha

Table 3 Median values of parameters of surface water and groundwater clusters. Values of variables are given in mg/L except for Temperature (Temp, °C), pH (–) and electrical conductivity (EC, µS/cm)

Tableau 1

Group	Cluster	n (number of samples in each cluster)	pH	EC	Na$^+$	K$^+$	Mg^{2+}	Ca^{2+}	HCO$_3^-$	Cl$^-$	NO$_3^-$	SO$_4^{2-}$	F$^-$
I	1	16	7.6	70	4.20	2.45	2.10	6.45	24.16	0.48	0.32	0.15	0.13
	2	40	6.6	256	15.25	9.50	5.45	22.00	17.69	9.25	31.30	2.25	0.18
II	3	22	7.2	638	46.10	2.15	22.70	56.95	121.63	16.30	37.25	10.60	1.20
III	4	7	6.7	1421	65.50	3.00	58.50	129.00	237.23	116.00	313.00	53.10	1.10
	5	1	7.1	2500	244.00	1.00	120.00	252.00	230.80	345.00	823.00	177.00	0.82

n number of samples in each cluster

of $15 - 20 - 15 + 50$ kg urea. This increase in chemical fertiliser consumption reached 41,000 tonnes of fertiliser in 2004 (Olina Bassala 2010).

Evidence of anthropogenic source of pollution

The combination of hydrological stable isotope ratios (δ^{18}O and δ^2D) with hydrochemical data such as chloride
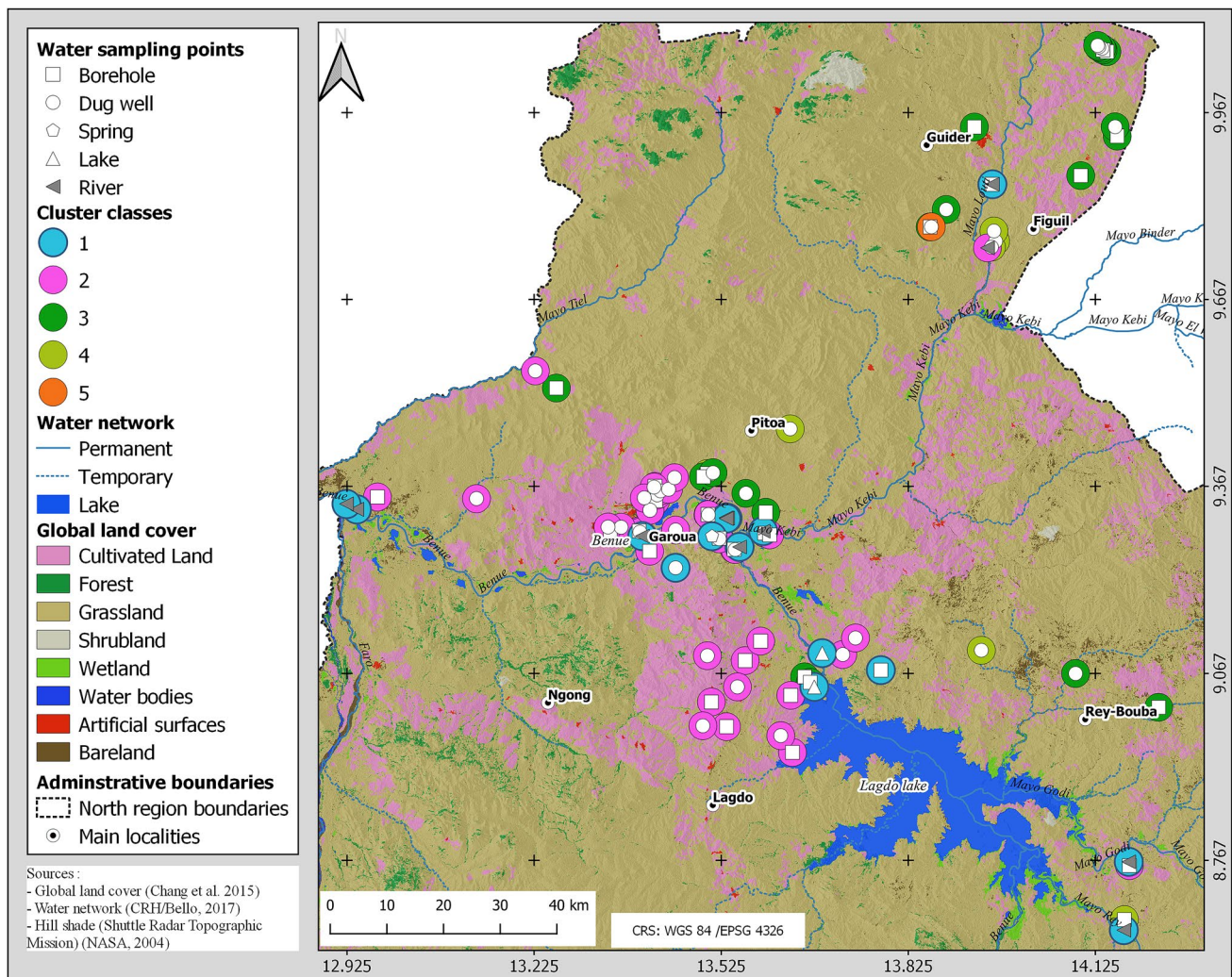
Fig. 7 Spatial distribution of HCA classified ground- and surface water samples—5 classes—with a background of land cover for the Upper Benoue catchment, Lower Left: closeup of urban Garoua; data source: SRTM, Global Land Cover (Chang et al. 2015), OpenStreetmap

Fig. 8 Evidence of pollution of groundwater in the BRB: **a** relationship between δ^2H and $\delta^{18}O$, **b** relationship between Cl^- and $\delta^{18}O$, **c** correlation plot of SO_4^{2-} and Cl^-, **c** correlation plot of NO_3^- and Cl^-, both with Spearman coefficient and *p* value
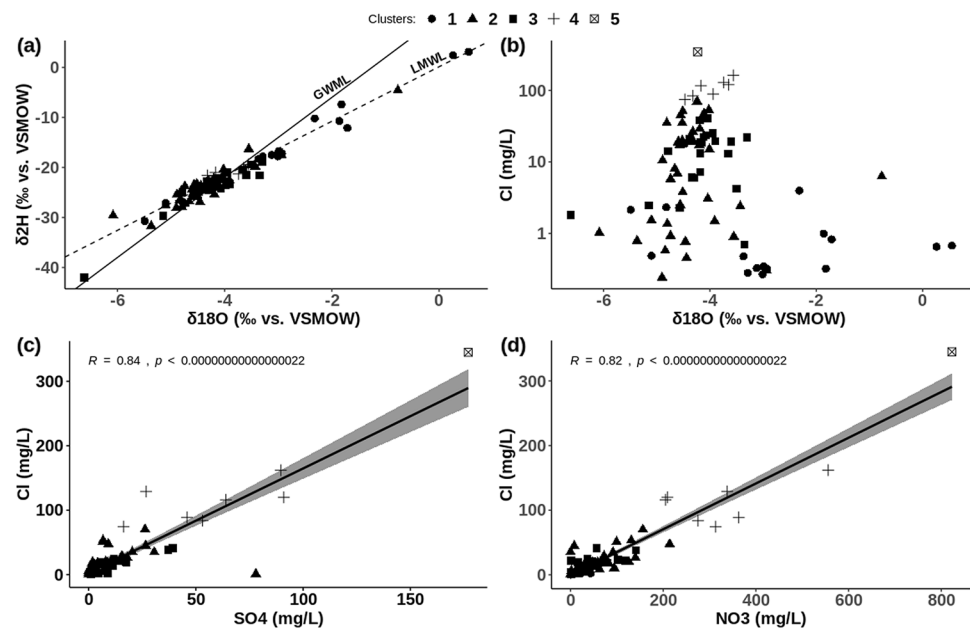
concentration gives a better understanding of the groundwater recharge process (Njitchoua et al. 1997 and Fantong et al. 2020). These parameters are widely used as tracers in hydrological studies to identify the cause of an increase in salinization of surface and groundwater (Albek 1999; Wenninger 2020). A low background concentration of Cl^- with a mean value of 0.0012 mg/L in rainwater (Akpo et al. 2015) compared to a median of 0.41 mg/L in surface water and a median of 17.2 mg/L in groundwater (Table 1) may be linked to contamination that makes it an anthropogenic (strongly positively correlated to NO_3^-) rather than a hydrological tracer in our study area (Albek 1999).

The $\delta^{18}O$ vs. δ^2D relationship of water clusters along the Global Meteoric Water Line (GMWL) of Craig (1961) and the Local Meteoric Water Line (LMWL) of Fantong et al. (2020) presented in the Fig. 8a. This indicates that groundwater has been recharged by meteoric water without significantly being affected by evapotranspiration during the recharge period. In Fig. 8b, Cl^- and $\delta^{18}O$ show no correlation, indicating that leaching of salts (minerals and/or anthropogenic) plays an important role in the mineralisation of groundwater. Furthermore, Cl^- has been plotted against SO_4^{2-} and NO_3^- to visualise their relationships (Fig. 8c, d). The result shows strong positive correlations between Cl^- with SO_4^{2-} and NO_3^- ($r = 0.84$, 0.82, *p* value < 0.001) which emphasizes that groundwater in the BRB is strongly affected by anthropogenic. In the study area, pollution of surface and groundwater is linked to agricultural activities such as the use of inorganic and organic fertilizers, excretions of migrating livestock as well as direct domestic wastewater discharge into the environment. A complete absence of water protection perimeters around water points, combined
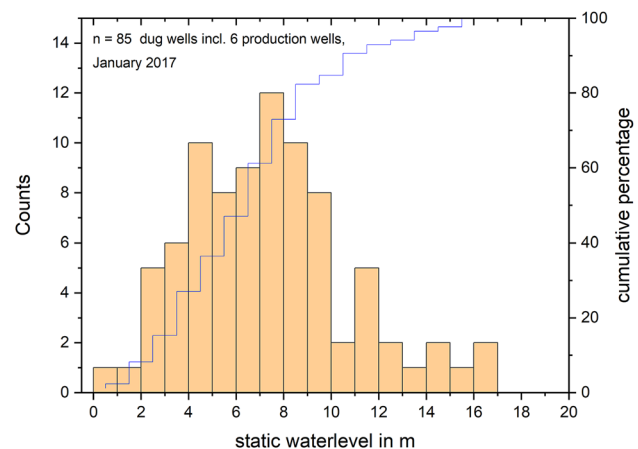

Fig. 9 Depth distribution of measured water levels in 86 water points of the North Region in dry season 2017

with widespread mismanagement of water points (absence of concrete cover slab, great concentration of livestock confinement close to the water point) allows the rapid pollution of the groundwater resource in the area. Nbendah et al. (in prep.) measured the depth to groundwater in 85 wells and dug wells (Fig. 9). They found the groundwater table at a depth between min. 1.9 m and max. 16.9 m below surface with an outlier of 0.3 m in river bed. The median depth to the phreatic groundwater in the BRB was 7.1 m below surface (mean 7.4 m ± 3.4 m) in dry season 2017 (January). Further investigation towards the identification of the actual (mineral vs. excretions) sources of NO_3^- of the BRB with its associated health risks is crucial for pollution control strategies and the avoidance of water-related health issues for humans

and cattle in the light of the planned intensification of agriculture and intended leap of productivity.

Constraining the geogenic background concentration of nutrients

Nutrient cycling is a key process of any ecosystem, including those left with only negligible human influence. Savanna and gallery forest would be the predominant natural ecosystems of the North Region without the strong human influence as witnessed today (Brabant and Gavaud 1985; Barbier et al. 2002). Potassium, nitrogen (as nitrate), phosphorus and sulphate as macronutrients would have distinct concentration ranges[3] in such remote ecosystems (Njitchoua et al. 1995). These extensively used parts of the North Region—far from road access—were outside the focus of our sampling strategy and are thus underrepresented in the data (Fig. 3). An univariate statistical analysis of the nutrients may, however, yield important clues to the non-anthropogenic concentration ranges caused by the geochemical baseline, atmospheric and biological processes, summarized for simplicity as "geogenic" (Matschullat et al. 2000; Appelo and Postma 2005). These will mainly be atmospheric input (wet and dry deposition), N-assimilation and in particular rock weathering (Manning 2010) in the root zone and in the phreatic aquifer. As our perspective here is rather the geogenic supply of nutrients, added an unknown contribution by wild fauna, the lower percentiles of our observed distribution are of interest. This perspective is different from natural baseline approaches adapted to regulatory issues and/or detection of contamination, choosing much higher percentiles (e.g. 50–p90, Cave et al. (2012), Lucon et al. (2018)). Assuming slow release by incongruent dissolution of silicate minerals, the contrast between more depleted, but K-feldspar-rich arkosic and wacke sandstone aquifers of the Garoua basin and presumably less depleted residual aquifers developed on migmatitic-granitoid proterozoic basement is of interest.

The best estimate of geogenic background concentrations of macronutrients in groundwater of the upper BRB to date is to be derived from Fig. 1 (ESM appendix). Remembering these log-normal distributions, in Fig. 3, geogenic background will be chosen as the 25% percentiles for nitrate with 9.14 mg/L, sulphate with 1.45 mg/L and potassium with 2 mg/L. Chloride, being an anthropogenic tracer in our setting, has 2.3 mg/L in groundwater. It has to be stressed, that lower concentrations do occur in the outliers in Fig. 3 in the direction of rainwater as the starting point. Strictly speaking,

these values hold only for siliclastic sediment aquifers of the Garoua basin and not the mudstones/marls of the Figuil basin and the proterozoic basement as indicated with the HCA and PCA analysis above.

To put dissolved nutrients in the Benue during dry season and base flow (visible in Fig. 3) into perspective, the dissolved median concentrations of nitrate (0.32 mg/L), potassium (2.5 mg/L), sulphate (0.18 mg/L) and o-phosphate (0.03 mg/L) together with a low chloride concentration (0.41 mg/L) are given (Table 1, and Fig. 3). An EC of 65 μS/cm was measured both upstream of Lagdo Dam (Blue Lagoon, R010) as well as downstream of Garoua (near Barndaké, R006a). Thus, the Benue water appears remarkably unaffected by discharge and/or GW-exfiltration from Garoua in dry season 2017. These values can be compared to the EC ranging from 64 μS/cm to 139 μS/cm from 2012 to 2013 at Markurdi in Nigeria (Akahaan et al. 2015), 561 km (beeline) downstream of Garoua. There is no indication of extreme EC values as given by Hong et al. (2014) for Jimeta/Yola. Yet, the lower limits of the concentration ranges for Makurdi, nitrate (2.2–3.8 mg/L), phosphate (0.9–1.47 mg/L) and sulphate (10.4–17.2 mg/L) (Akahaan et al. 2015) seem to be high compared to our data (Fig. 3). They need to be taken with caution especially when compared to chloride[4] in the light of our median values given above.

The current non-availability of post-Lagdo construction discharge data for the Benue in Cameroon, dissolved nutrient exports of the upper BRB during the dry season (and wet season) yet waits to be constrained. New key hydrological investigations and groundwater level monitoring are highly necessary in order to properly advise on the intended agricultural productivity increase (adapted plant nutrition, higher abstraction for irrigation). This planned increase should neither impact negatively on ecosystem services nor on the regional water budget, both on the national scale and on the international Benue River as the largest Niger tributary.

Conclusion

Our results show that nutrient migration (N, P, K, S) from anthropogenic sources to groundwater is pronounced in the upper Benue River basin (BRB). Nitrate concentration in the surface water is low as a result of assimilation by plants, a significant increase of nitrate since 1991 is observed for groundwater for Garoua. Groundwater generally flows in unprotected siliclastic arkosic phreatic aquifers of cretaceous and quarternary age or in the weathering

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[3] It should be noted that even in the completing absence of migrating livestock, the pre-existing migrating savanna fauna would have had the dominant control on nitrogen and sulphur dynamics as well as on chloride concentration in groundwater.

[4] Chloride 117 – 173 mg/L at Makurdi being in disagreement with EC.

zone and the fractures of the migmatitic-granitoid proterozoic basement aquifers with a low distance (median = 7.5 m below surface) to the water table. The setting allows rapid downward movement of nutrients with apparently rapid groundwater recharge, occurring around the peak of rainy season with little evaporation as shown by an apparently low fractionation of hydrological isotopes. Groundwater is slightly acidic (median pH 6.8) and more mineralized (EC range = 35–2500 μS/cm, median = 376 μS/cm) than surface water (EC range = 50–222 μS/cm, median = 81 μS/cm). The concentration of nutrient-related parameters is in equivalent concentration decreasing order of $K^+ > NO_3^- > Cl^- > SO_4^{2-} > PO_4^{3-}$ in surface water compared to $NO_3^- > Cl^- > SO_4^{2-} > K^+ > PO_4^{3-}$ in groundwater. PCA results, in addition to correlation analysis, show that these macronutrients and chloride have mostly an anthropogenic origin and are associated with the influence of agricultural land use in the study area. Any released ammonium is readily oxidized. This is in line with a noticeable P-mobility as compared to the ferralitic soils of the forested south. Nitrate is the key pollutant of concern in the groundwater with high concentrations attributed to the proximity of water sources to point sources such as areas of concentrated livestock confinement, leaky septic tanks or sewers and refuse dumps increased by the mismanagement of water supplies. Already 44.4% of the groundwater sampling points—all of which were used for human consumption—in the study area exceed the WHO guideline concentration for nitrate of 50 mg/L, a situation aggravated since 1991. Using HCA, we identified three distinct water quality groups. Group I, which contained cluster 1 and 2 were generally less mineralized and characterised by the $Na + K - HCO_3$ water type found in the urban area such as Garoua and Lagdo in the cretaceous Garoua basin; Group II, which contained cluster 3 were higher mineralized and also characterised by the $Na + K - HCO_3$ water type. Group III, which contained cluster 4 (and cluster 5) were heavily mineralized and were characterised by the $Ca + Mg - NO_3$ water type, associated with the outcrop of cretaceous mudstones/marls or the migmatitic-granitoid proterozoic basement. Based on the observed groundwater quality deterioration in the upper BRB and in the wake of needed agricultural expansion and intensification, the urgent need for groundwater protection measures is evident. Among these is the introduction of a locally viable concept for groundwater protection perimeters including awareness building. For a better management, there is firstly an urgent need to measure and monitor the necessary hydrological data required for a regional water budget (discharge, groundwater levels, soil water balance, evapotranspiration and climate data) for the upper BRB. Water allocation in the wake of agricultural expansion and global climate change may become critical. Secondly, there is a need to better understand the local processes/mechanisms leading to nutrient pollution, migration and retardation and their fate in both groundwater and surface water.

Declarations

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