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Hydrochemical study of bottled water in Rwanda and relationship with their origin

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ABSTRACT

In Rwanda, commercialized bottled water comes mainly from aquifers located in Rwanda and in neighboring countries. In this study, we compare geochemical parameters of bottled water from the great lakes region and seek to correlate mineralization, hydrochemistry and geological facies of the water reservoir. Based on the results obtained from major ions, trace elements and stable isotopes, 15 bottled waters were analyzed. Bottled waters show low mineralization and various chemical facies, with mainly the dominance of sodium due to the occurrence of alkaline granites in the East Africa region. Bottled water quality was evaluated using various classification systems. All bottled waters show low mineralization attributed to low rock-water interaction with granite, and to unconfined aquifer close to the surface fed by direct infiltration and possibly with high transit fractured circulation. In this region, the very high population density with high agricultural activities lead to strong anthropogenic conditions, but most of the bottled waters do not show any evidence of influence. However, this does not prevent regular control of water quality. Stable water isotopes (δ^2H and δ^{18}O) highlighted the altitude of aquifers and waters origin of the bottled waters, which is linked to direct rainfall infiltration.

Key words: bottled waters, great lakes region, hydrochemistry, Rwanda, trace elements, δ^2H and δ^{18}O isotopes

HIGHLIGHTS

- Adequacy between water quality and its origin.
- Water bottles are good samples for studying aquifers.
- Heavy isotope tools to confirm aquifer recharge.
- Good quality of bottled water in the neighboring countries of the great African lake.

INTRODUCTION

On a global scale, bottled water is the most important and dynamic market of the beverage and food industry. The growing consumption reflects a certain way of life and a healthy alternative to other beverages (Ferrier 2001), with an average economic growth of 5.5% per annum in Rwanda (NBR 2017). Life standards have noticeably improved for some Rwandans, whose financial means and mindset have increased, allowing them to consume bottled water. Rwandans consume bottled water locally produced and from other East African countries.

Numerous studies dealing with physicochemical characterization of bottled water have been carried out across the world (Qian 2018; Willis *et al.* 2019). However, few studies have focused on analysing and improving the quality of groundwater in the study area (Biryabarema 2001). This is an important issue because, in Africa, the region of great lakes is known for high human population density, mainly in Rwanda, which has a population density of more than 600 people/km^2. In these

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countries, the main income-generating activity is agriculture, which uses chemical fertilizers and pesticides. These fertilizers are susceptible to infiltrate and pollute groundwaters that feed water sources used by people and for bottling, for one use. Since the harmful effects of fertilizers on human health and biodiversity have been clearly documented (Berndtsson *et al.* 2006; Scanlon *et al.* 2007), an efficient and sound strategy to protect water resources from pollution should be implemented.

Drinking mineral or spring water, as an essential source of minerals, can play an important role in human nutrition (WHO 2017). Mineral bottled waters are not subject to treatment before bottling and conserve their chemical quality, which is not the case for bottled table water, some of whose mineral nutrients can be removed during water treatment processes such as membrane filtration, reverse osmosis and softening, leading to nutrients deficit (Ahmed *et al.* 2016). However, the mineral and spring bottled waters from the region of Great Lake marketed in Rwanda generally have a low mineralization. Long-term consumption of waters poor in minerals may increase deficiencies in minerals (Mihayo & Mkoma 2012). Unfortunately, during the last two decades, globally, little research has been carried out into the beneficial or harmful effects of some minerals from drinking water in Rwanda.

The present study aims to determine physico-chemical properties of 15 bottles of water marketed in Rwanda to evaluate their chemical quality and their origin according to the geology of the water reservoir. In addition, the water isotope signature brings information on aquifer recharge conditions.

SAMPLING AND ANALYTICAL METHODS

In this study, we randomly collected fifteen brands of bottled water brands. A total of 45 samples of 3–5 50 mL bottled waters were purchased from supermarkets in Kigali City in 2017–2018 (Dege 2011). Bottle caps, protective seals, label information and expiration dates were checked. The bottled water brands purchased were protected from heat and odors. Afterward, the samples were transported to the laboratory of water quality of ICRP Kigali for physico-chemical analysis.

These bottled waters come from five countries from the region of great lakes: Rwanda, Tanzania, Uganda, Kenya and Democratic Republic of the Congo. The origins of the different water brands are given in Figure 1. The selected bottled waters were classified into three categories: three purified drinking waters qualified as table water (T), ten mineral waters (M) and two spring waters (S). Mineral and spring water have not undergone any prior chemical treatment. Mineral water has therapeutic properties. The spring water is bottled without any chemical treatment but meets quality standards. Table water, taken from the drinking water network, which distributes chlorinated water, was filtered by reverse osmosis or ultrafiltration before being bottled.

The standards of quality for bottled water in the East African countries are similar. They require that bottled water be safe to drink. They define the types of bottled water and set the conditions for packaging, storage and labeling.

The physicochemical parameters were measured immediately after opening. Once opened, the bottles were refrigerated at 4 °C. The dates mentioned on the labels of the same brand were different but without any variability of chemical composition. The physicochemical parameters were measured three times and average values are given.

The ions were stabilized by adding 2 mL of nitric acid in 50 mL of water before the analysis of trace elements. For isotopic sampling, 20 ml airtight glass bottles were completely filled with each water sample. The water samples used for isotopic analyses were placed in airtight glass bottles of 20 ml capacity.

The pH and electrical conductivity (EC) were measured using an MP512-03 pH meter and a DDS-307 conductivity meter respectively. Major elements were determined, by various methods, in the water quality laboratory of the Integrated Polytechnic Regional Center (IPRC-KIGALI). Chloride and bicarbonate determinations were carried out by titration. Calcium, magnesium, sulfate and nitrate were determined using a DR3900 spectrophotometer. Ionic balances show a good agreement for all analysis ($<3\%$). Trace elements were measured by ICP-MS (i CAP Q Thermofisher) and stable isotopes ($\delta^{18}O$ and $\delta^{2}H$) by IRMS (Isoprime, standard errors are 0.06‰ and 0.8‰ respectively for ^{18}O and deuterium) at Hydrosciences Montpellier University (Tables 1, 3 and 4).

GEOLOGICAL SETTING

The geology of Rwanda consists of granite, migmatites, gneisses and micaschists, which are of Paleoproterozoic age (Ruzizian, 1.8-1.6 Ga) as basement overlain by the Mesoproterozoic Kibaran belt (1.6-0.98 Ga). Most of the country is covered by the Kibaran, composed of folded and metamorphosed sediments with dominance of schist and quartzite intruded by granite (Rutagarama & Uhorakeye 2010).

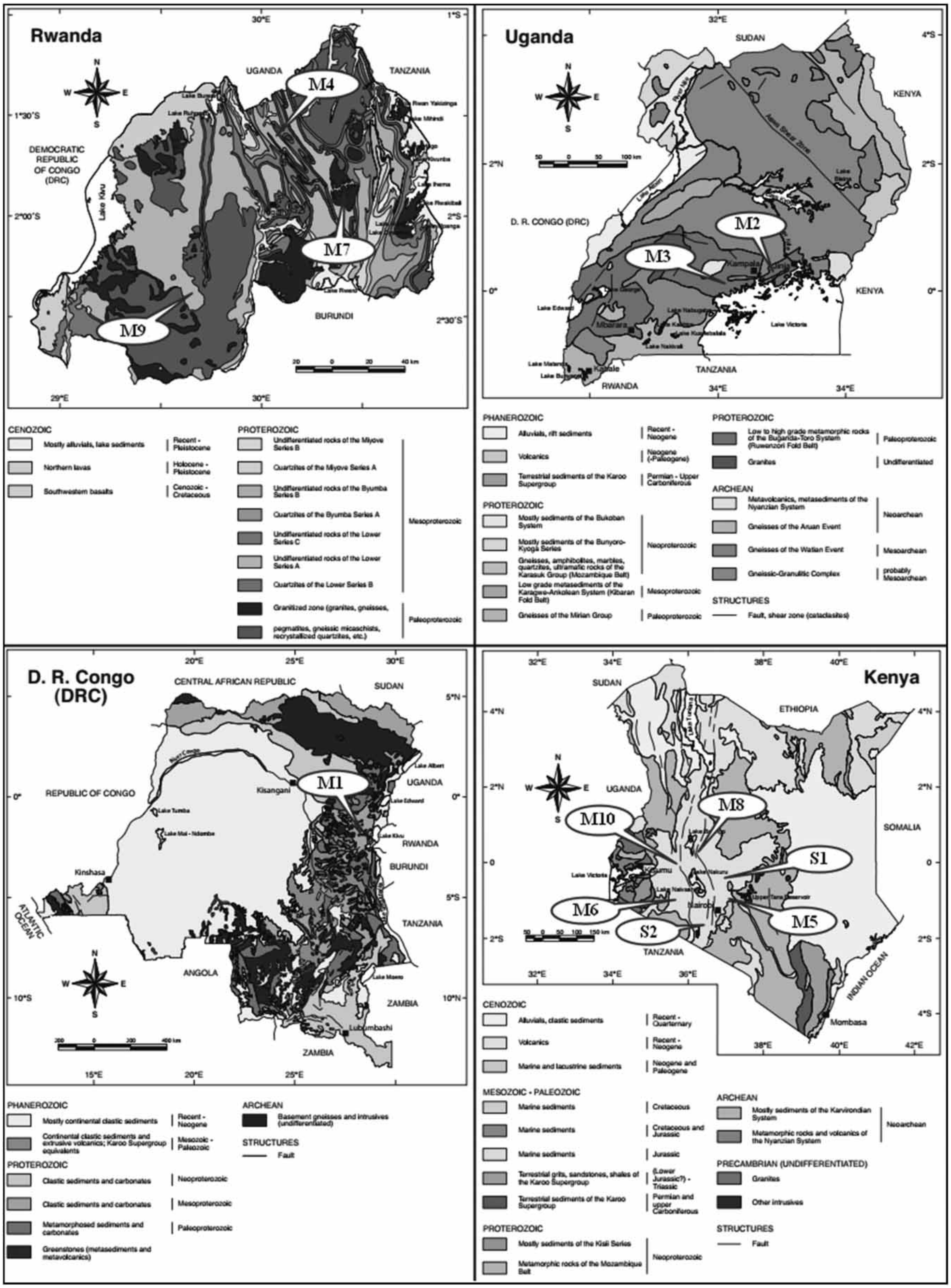

Figure 1 | Geological context of the Great Lakes Region (Schlüter 2008) and location of emergence zones for springs and mineral waters marketed in Rwanda.

Table 1 | Physico-chemical parameters for the 15 bottled water brands

Label	Water brand	Type	Country/location	pH	Cond µS/cm	TDS mg/L	Na mg/L	K mg/L	Mg mg/L	Ca mg/L	Cl mg/L	SO_4 mg/L	HCO_3 mg/L	NO_3 mg/L	SiO_2 mg/L	TH mg/L $CaCO_3$
M1	La vie	Mineral water	Rutshuru/RD Congo	7.41	31.6	20.2	2.64	3.2	0.41	0.46	4.99	1.61	3.45	1.45	1.85	2.83
M2	Rwenzori	Mineral water	Namamve/Ouganda	6.95	129.5	82.8	8.12	3.31	2.26	8.5	13.2	25.9	5.92	2.41	65.7	4.75
M3	Riham	Mineral water	Kampala/Ouganda	6.91	245.1	157	22.9	2.85	3.49	13.5	35.2	32.5	18.6	3.41	22.7	15.2
M4	Akandi	Mineral water	Rulindo/Rwanda	6.75	154.7	99	23.9	0.78	1.25	2.46	30.8	9.25	10.9	2.67	11.8	11.3
M5	Waba	Mineral water	Thika/Kenya	7.61	249.4	160	48.6	2.51	0.45	3.25	22.3	9.15	92.2	2.9	33.9	9.96
M6	Highlands	Mineral water	Nyeri/Kenya	7.31	102.6	65.7	6.25	1.19	1.61	8.05	11.2	20.6	5.14	0.54	14.4	26.7
M7	Inyange	Mineral water	Gasabo/Rwanda	6.62	236.2	151	16.7	2.5	7.32	10.6	27.9	44.9	5.9	6.5	16.5	56.4
M8	Aquamist	Mineral water	Great Rift valley/Kenya	6.61	183.6	117	18.6	6.03	1.88	7.15	30.1	22.4	6.23	3.24	48.0	25.6
M9	Huye	Mineral water	Huye/Rwanda	6.84	126.8	81.2	20.7	0.27	1.6	3.09	4.96	12.3	39.6	8.91	7.65	14.3
M10	Keringet	Mineral water	Molo/Kenya	7.29	91.4	58.5	14.7	2.27	0.03	0.13	24.2	0.39	1.17	1.9	37.7	0.45
S1	Kenyan	Spring water	Nairobi/kenya	7.23	130.8	83.7	18.1	2.61	0.1	3.61	28.9	8.84	2.84	0.05	58.5	9.72
S2	Grange park	Spring water	Nairobi/ Kenya	7.21	146.3	93.6	22.5	4.17	0.73	4.38	5.99	13.4	49.2	6.75	34.8	13.9
T1	Safina	Packaged drinking water	Tanzania	7.76	131.3	84	14.2	7.4	1.1	4.2	9.4	20.2	19.7	6.1	1.41	15.3
T2	Hema	Packaged drinking water	Ouganda	6.98	115.9	74.2	8.2	1.6	2.3	8.4	5.4	21.4	15.2	8.3	1.32	30.4
T3	Nil	Packaged drinking water	Rwanda	7.01	210	134	9	5.6	4.7	14.9	44.2	9.9	6.1	8.4	1.56	56.5
RS				6.5–8.5	1,500	700	200	–	100	150	250	300	–	10	–	–
WHO				6.5–8.0	–	–	–	–	–	–	250	250	–	50	–	–

In Uganda, two-thirds of the country is mostly represented by Precambrian rocks including Archaean rocks (>2.5 Ga), Lower and Middle Proterozoic Groups of varied lithology (2.5-1.0 Ga). The younger rocks are Cretaceous and Tertiary age (carbonatite), and Quaternary (syenite and alkaline volcanic rocks) from Pleistocene to Holocene located in the Rift Valley (Gabert 1984). In the area of Kampala, the basement is of Precambrian age (granite gneisses, quartizites, schists, phylites, and amphibolites), overlain by Pleistocene to recent alluvium and lacustrine deposits (Biryabarema 2001).

In Kenya, the west part of the country along Lake Victoria outcrops Neoarchean rocks (2.8-2.5 Ga), in the northern part metamorphic rocks of the Neoproterozoic Mozambique Belt (1.0-054 Ga), whereas the central part of the country outcrops sediments ranging from Late Paleozoic to recent age along the coast and predominantly younger volcanism associated with the rift formation (Schlüter 2008).

The east of the Democratic Republic of the Congo is underlain by the PanAfrican Lufilian Arc (2.2-2.1 Ga). Along the eastern part, at the border to the Western Rift of the East African Rift System, occur numerous Tertiary to recent volcanoes and some carbonatites (Schlüter 2008).

RESULTS AND DISCUSSION

Water mineralization

Average values of physico-chemical parameters of the bottled water brands are presented in Table 1 together with Rwanda standards (RS) and maximum acceptable concentration recommended by WHO (2017).

EC values of the bottled water brands show large variability. Likewise, EC values depend on total dissolved solids (TDS) content, the origin of the water, and the treatment or purification method applied during the bottling process could explain this high variability.

An exception is observed for water brand M1, which has the lowest TDS value of 20.2 mg/L, while most of the waters show low mineralization with TDS values varying from 58.5 to 160 mg/L. According to Chapelle (2005), bottled mineral water is obtained from a protected underground formation with TDS content greater than 250 mg/L with any added minerals. Van der Aa (2003) suggested that the high concentration of bicarbonate (HCO_3^-) for brand M5 (highest EC) could be attributed to the reaction of hydrolysis of alkaline feldspars contained in granite and the solvation of organic materials.

The high values of silica in mineral waters from Uganda (M2) could be attributed to the geology of the region, which is underlain by granite and schist basement (Pallister 1959). The high values of silica in Kenyan bottled waters (M8 and S1) could be attributed to the granitic rocks present in the Proterozoic Mozambique Belt, which is most extensive in Central Kenya.

pH value is lowest in brand M8 (6.61) and the highest in brand T1 (7.76). Most bottled waters have a total hardness (TH) between 0 and 50 mg/L, and are considered as soft water. Hardness value varies between 0.45 and 46.5 with an average of 14.3 mg/L ($CaCO_3$). In exception are two samples, M7 and T3, which have hardness between 50 and 100 mg/L (56.4 and 56.5 respectively), and are considered as moderately hard. The significant mineralization of M7 can be attributed to the aquifer being located in the granite-rich zone (Figure 1). For table waters, the origin of mineralization is complex and multiple due to possible mixing of water and treatment.

All bottled water brands have NO_3^- values less than 50 mg/L, indicating no influence of human activity (Madison 1984).

The water samples studied are qualified as being Soft-Fresh according to the TH-TDS diagram shown in Figure 2 (Du *et al.* 2017). The bottled waters marketed in Rwanda are classified as Fresh-Soft, and are good quality.

Hydrochemical facies

Bottled water brands marketed in Rwanda were classified using a bar chart and Piper's diagram (Figures 3 and 4). Both types of classification are based on dominant ions (Oyebog *et al.* 2012). Referring to the water classification scheme by Davis & De Wiest (1966), the fifteen analyzed samples have been classified into ten water types as presented in Table 2. For most of the brands (60%), sodium is the dominant cation, while for others (40%), calcium is dominant. For the anions, chloride is dominant (47%), followed by sulfate (33%) and finally bicarbonate (27%).

The presence of sodium, in most of the samples of bottled water, could be attributed to the presence of many alkaline granites in the East African region (Pohl *et al.* 2013). The sodium in water could be also linked to salts and evaporite deposits (e.g. halite and gypsum) which may be in the granite faults.

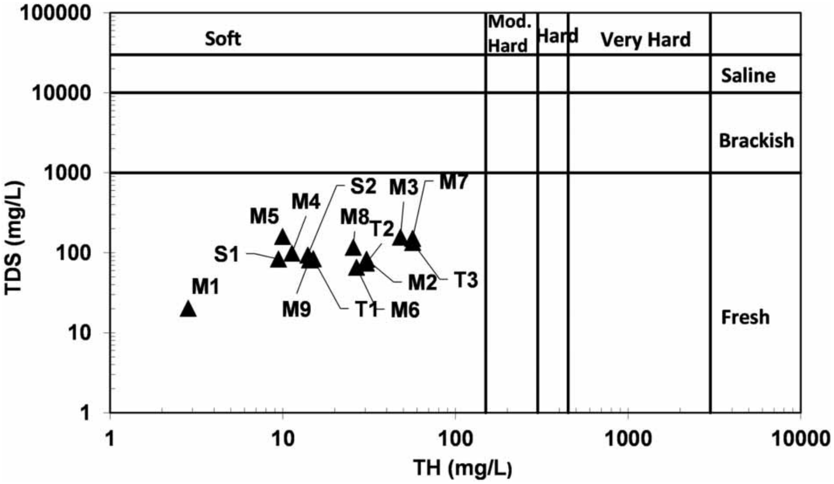

Figure 2 | Classification of bottled waters based on TH-TDS diagram.

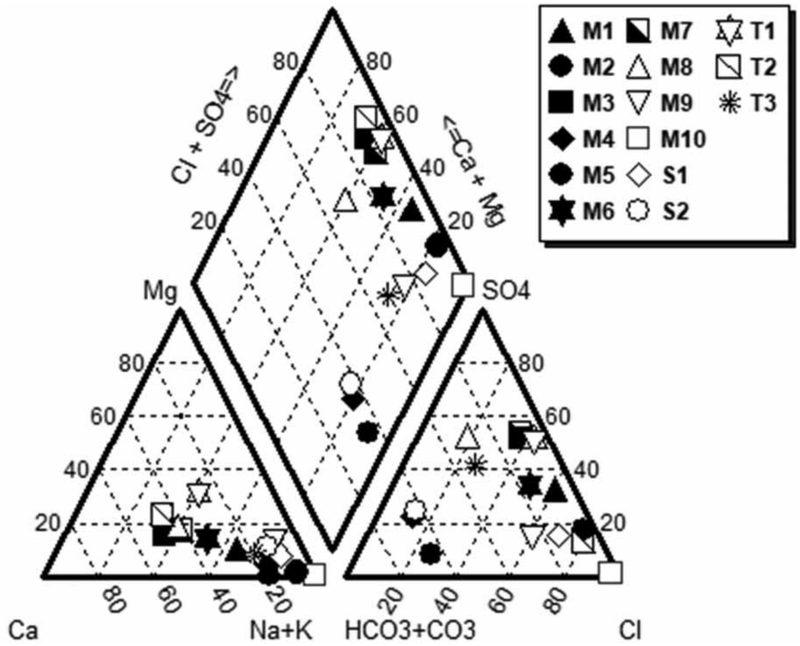

Figure 3 | Piper Diagram.

The excess of concentration of Na compared to chloride (M1, M3, M4, M5, M7, M8, M9, M10, S1, S2 and T1) is attributed to a Na source, such as albite (plagioclase), or natural softening in addition to Na in rainfall:

$$Ca^{2+} + 2Na\text{-albite} \rightarrow 2Na^{+} + Ca\text{-feldspar}$$

Only some samples show a concentration of chloride greater with respect to sodium (M2, M6, T2 and T3). This could be attributed probably to the reverse softening:

$$2Na^{+} + Ca\text{-Clay} \rightarrow Ca^{2+} + Na\text{-Clay}$$

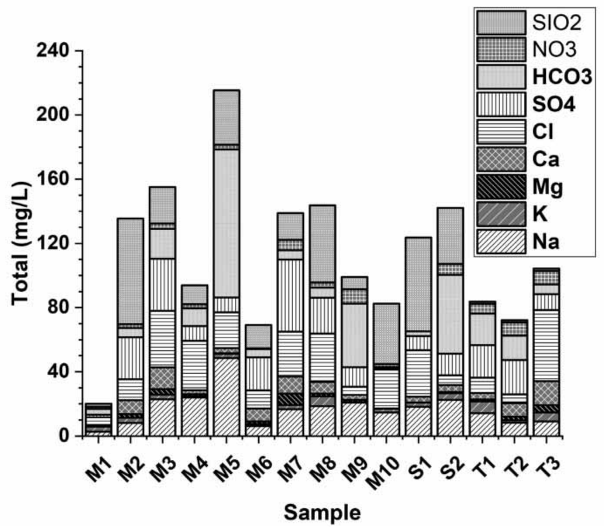

Figure 4 | Weight distribution.

Table 2 | Hydrochemical facies

Labels	Water brand	Water type
M1	La vie	Na-Cl
M2	Rwenzori	Ca-SO_4
M3	Riham	Na-Cl
M4	Akandi	Na-Cl
M5	Waba	Na-HCO_3
M6	Highlands	Ca-SO_4
M7	Inyange	Na-SO_4
M8	Aquamist	Na-Cl
M9	Huye	Na-HCO_3
M10	Keringet	Na-Cl
S1	Kenyan	Na-Cl
S2	Grange park	Na-HCO_3
T1	Safina	Na-SO_4
T2	Hema	Ca-SO_4
T3	Nil	Ca-Cl

The highest values of sulfates were observed in M7 and M3, (44.9 and 32.5 mg/L), which may be due to evaporite deposits (e.g. gypsm) which may be in the granite substrate but more probably from pyrite because Ca does not associated to sulfates.

Figure 4 shows the weight and relative distribution of major ions, highlighting the high *Ca* variability of ion contents linked to origins of water. M1 is the least mineralized water.

Trace elements

Spring and mineral waters have natural trace elements which are in relation with the water-rock contact (Table 3). However, for purified drinking waters the concentration of trace elements is affected by physico-chemical treatments; then, no traces measurement was done, this study emphasizing the relationship between mineral water and its geologic origin.

The bottled waters marketed in Rwanda do not contain significant amounts of trace elements. The average values obtained are negligible compared to the guide values tolerated by the standards provided by WHO and Rwanda Standard Board (RSB).

Table 3 | Trace elements in spring and mineral waters

	μg/L																		
	Li	**B**	**Al**	**P**	**V**	**Cr**	**Mn**	**Fe**	**Ni**	**Cu**	**Zn**	**As**	**Se**	**Rb**	**Sr**	**Mo**	**Ba**	**Pb**	**U**
M1	0.41	7.15	0.59	1.06	0.33	< **DL**	< **DL**	< **DL**	< **DL**	0.3	0.32	0.01	< **DL**	1.21	6.7	0.04	0.25	0.01	0.02
M2	4.07	1.51	1.05	148	1.93	0.67	0.86	< **DL**	0.45	2.27	1.68	0.01	0.08	0.72	108.2	0.47	58.4	0.01	0.43
M3	2.47	26.6	0.83	31.9	0.84	0.62	0.52	< **DL**	1.22	0.07	0.96	0.01	0.15	1.29	169	0.73	78	< **DL**	0.29
M4	2.82	5.43	49.4	< **DL**	< **DL**	< **DL**	106.9	< **DL**	4.85	0.88	15.7	0.03	0.08	1.03	40	< **DL**	117.4	0.14	0.03
M5	7.03	43.3	3.81	136	2.22	< **DL**	< **DL**	0.56	< **DL**	< **DL**	0.52	3.51	0.7	3.7	15	5.83	0.26	0.01	1.57
M6	3.29	19.3	0.81	3.27	4.27	0.48	< **DL**	0.54	< **DL**	< **DL**	1.95	0.66	0.02	9.63	44	2.75	0.24	0.01	0.66
M7	7.5	2.31	3.2	< **DL**	< **DL**	0.18	0.1	1.03	0.74	< **DL**	2.38	0.02	0.38	11.3	98.5	0.01	56.9	< **DL**	< **DL**
M8	7.09	6.68	2.12	11.8	< **DL**	0.05	0.06	0.44	0.05	0.08	34.2	0.03	0.09	29	29.5	0.21	1.71	0	0.22
M9	0.48	4.1	6.95	0.94	0.16	0.16	1.42	< **DL**	0.85	2.96	2.74	0.08	0.01	0.57	25.3	0.01	20.1	0.04	0.02
M10	3.69	6.88	5.78	123	< **DL**	< **DL**	3.34	6.25	< **DL**	1.6	0.29	0.1	< **DL**	5.03	0.51	1.08	0.04	0.09	0.02
S1	3.73	27.9	0.41	12.7	1.11	< **DL**	< **DL**	< **DL**	< **DL**	0.13	1.5	0.36	< **DL**	6.13	1.55	1.03	0.04	0.03	0.07
S2	0.28	2.57	28.6	0.97	0.4	0.06	< **DL**	< **DL**	0.05	0.15	8.57	0.03	< **DL**	3.36	51.5	0.12	13.5	< **DL**	0.01
RS	–	300	100	–	–	50	100	300	–	1,000	5,000	10	10	–	–	–	700	10	–
WHO	–	2,400	200	–	–	50	400	–	70	2,000	5,000	10	40	–	–	70	1,300	10	30

* < **DL**, below detection limit.

Brand (M4) has the highest concentrations in Al, Ba, Ni and Zn. The highest levels of Ba present in drinking water are likely due to low pH of igneous rocks (granite), alkaline igneous and manganese-rich sedimentary rocks. The high values of Zn in brand (M4) is most likely due to the use of fertilizers and wood preservatives that contain zinc (Oyebog *et al.* 2012). Tin was detectable only in one brand (M10). Silver was not detectable in any of the bottled water samples.

The high values of phosphorus in some brands (M2, M10 and M5) could be related to the decomposition of organic matter. This was also reported by Chapman (1996).

Li and Sr concentrations are the highest values in M7 and M3 respectively. The high concentration of Li could be attributed to the weathering of silicate rocks.

Stable isotopes δ^{18}O and δ^{2}H

The results of the isotopic measurement of stable isotopes of oxygen (δ^{18}O) and hydrogen (δ^{2}H) for fifteen samples are presented in the Table 4.

All samples are close to the Global Water Meteoric Line (GWML) in Figure 5. GWML describes the water isotopic behavior of rainfall, an issue only of condensation process at the world scale (Craig 1961; Dansgaard 1964). The Dansgaard's equation gives a close relationship between δ^{2}H and δ^{18}O in precipitation: $\delta^{2}H = 8 * \delta^{18}O + 10$ ‰

All the water brands studied are located on the GMWL, with the exception of M3, which is located below the line, representing an evaporation process that affects water before infiltration, also marked with a low d-excess values (Levin *et al.* 2009). For M3, the shallow groundwater is close to Victoria Lake, which constitutes a recharge zone for the aquifer, submitted to a high evaporation rate. The same result has been obtained by Odada & Olago (2006) based on stable isotopic composition of east African lake waters.

The deuterium excess (Table 4) is deducted from Dansgaard's equation: d-excess = 8 * δ^{2}H - δ^{18}O. The continental precipitation issue from oceanic moisture gives a d-excess close to +10‰ (Dansgaard 1964). The continental moisture issue from surface water evaporation and/or evapotranspiration (recycling continental process) gives rainfall with d-excess values higher to +10‰ (Njitchoua *et al.* 1999). Half of waters present a higher d-excess, showing rainfall then recharge of aquifer issue from a recycling continental process conforming to the results of rainfall in central eastern Africa (Balagizi & Liotta 2019), explained by an area located in the middle of Africa with large forest zone and surface water under tropical climate.

Determining the altitudes and recharge areas of the mineral and spring waters is essential for the estimation of groundwater resources. Recharge altitudes can be estimated from stable isotope contents. The isotopic signal of the infiltration water is generally a function of temperature (annual temperature above the surface) where the infiltration took place (Gat 1980).

Table 4 | Stable isotope compositions of the 15 bottled waters

Code	Water brand	δ^{18}O ± 0.07 ‰	δ^{2}H ± 0.7 ‰	d-excess (‰)	Altitude
M1	La vie	− 2.82	− 12.8	11.8	1130
M2	Rwenzori	− 1.53	− 1.6	10.6	1180
M3	Riham	2.80	21.9	− 0.5	1200
M4	Akandi	− 3.41	− 11.1	16.2	1445
M5	Waba	− 2.89	− 14.2	8.9	1631
M6	Highlands	− 3.59	− 13.0	15.7	1753
M7	Inyange	− 3.27	− 12.6	13.6	1800
M8	Aquamist	− 4.04	− 19.2	13.1	1830
M9	Huye	− 2.59	− 9.6	11.1	1900
M10	Keringet	− 3.24	− 12.0	13.9	2457
S1	Kenyan	− 3.65	− 17.5	11.7	1795
S2	Grange park	− 3.96	− 20.1	11.6	1795
T1	Safina	− 2.26	− 1.8	16.3	55
T2	Hema	− 1.88	− 3.7	11.3	1200
T3	Nil	− 2.61	− 7.6	13.3	1800

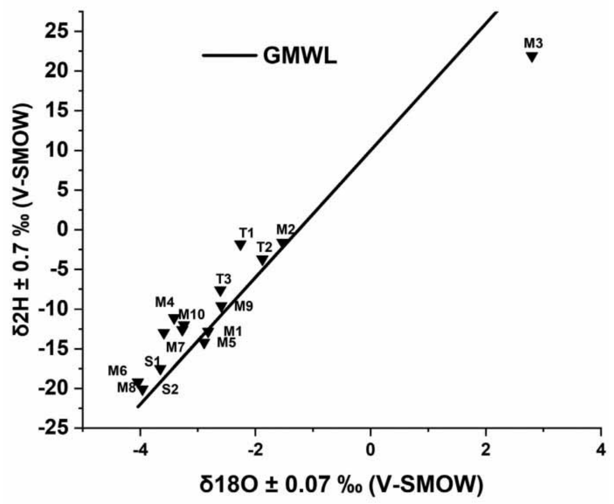

Figure 5 | A plot of δ^2H (‰) against δ^{18}O (‰) together with the GMWL.

Negative relationships were found between both brand-weighted δ^{18}O and δ^2H and altitude (Figure 6). The vertical oxygen gradient is −0.12 ‰/100 m and that of δ^2H is −0.9 ‰ /100 m. This value is similar to that found for precipitation waters in Central-Eastern Africa (−0.15 ‰ /100 m for δ^{18}O and −0.86 ‰/100 m for δ^2H) (Balagizi & Liotta 2019). The altitude gradient is −0.16‰ /100 m for δ^{18}O in the Mount Cameroon region (Fontes & Olivry 1977).

Statistical analysis

Pearson correlation coefficients (r) were calculated (Table 5). The results show a very high positive correlation (>0.8) between Na^+ and HCO_3^-, highlighting the dominant process hydrolysis of alkaline granites, a high positive correlation (>0.9) between Ca^{2+} and SO_4^{2}; Mg^{2+} and SO_4^{2}; and a moderate positive correlation (>0.7) between Ca^{2+} and Mg^{2+}.

In order to examine the overall variation in the data set, factor analysis was used. Principal component analysis was used to reduce the number of variables in the data set, select the most discriminating parameters and investigate the variance. Eigenvalues, the percentage of variance, the cumulative eigenvalue and the cumulative percentage of variance associated with each other are registered in Table 6.

Principal components analysis (PCA) shows four trends (F1-F4) based on the Kaiser Criterion (Kaiser 1960). F1 (HCO_3^-, B, V, As, Ce, Mo, U and Zn) represents the largest part (24.5%) of the total variance (Figure 7). F1 is characterized by high amounts of HCO_3^- and trace elements, reflecting the influence of carbonate-rich lithologies. F2 (Mg^{2+}, Ca^{2+}, SO_4^{2-}, Cr, Sr and Pb) represents 19.2% of the total variance and indicates the control of evaporite deposits (e.g. gypsum). F3 (Na^+, Al, Mn, Ni and Ba) represents 15.2% of the total variance and is characterized by the occurrence of heavy metals (Al, Mn, Ni and Ba), indicating the contributions of weathering of rocks and anthropogenic supplies to aquifers. F4 (K^+, Cl^-, Li, Zn and Rb) represents 12% of the total variance. F3 and F4, together, reflect the predominance of chloride and sodium.

CONCLUSION

This study gives an insight into the diversity of bottled water marketed in Rwanda in terms of physicochemical quality linked to their geologic origin as well as the mode of water circulation and recharge from their stable water isotopic content. The results show all bottled waters marketed in Rwanda have a low mineralization. For the mineral and spring bottled water this low mineralization could be attributed to different parameters: geology (low rock-water interaction inside granite with high transit fractured circulation), groundwater (unconfined aquifer) close to the surface, and direct infiltration of rainwater.

There are different chemical facies in bottled water with mainly the dominance of sodium linked to alkaline geologic facies present in East African region. The occurrence of many trace elements can be directly related to aquifer lithology.

The water quality is inside that of different standards for major ions and trace elements and does not present any risk for human health; however, the low mineralization requires a nutritional supplement in the diary diet in particular for trace

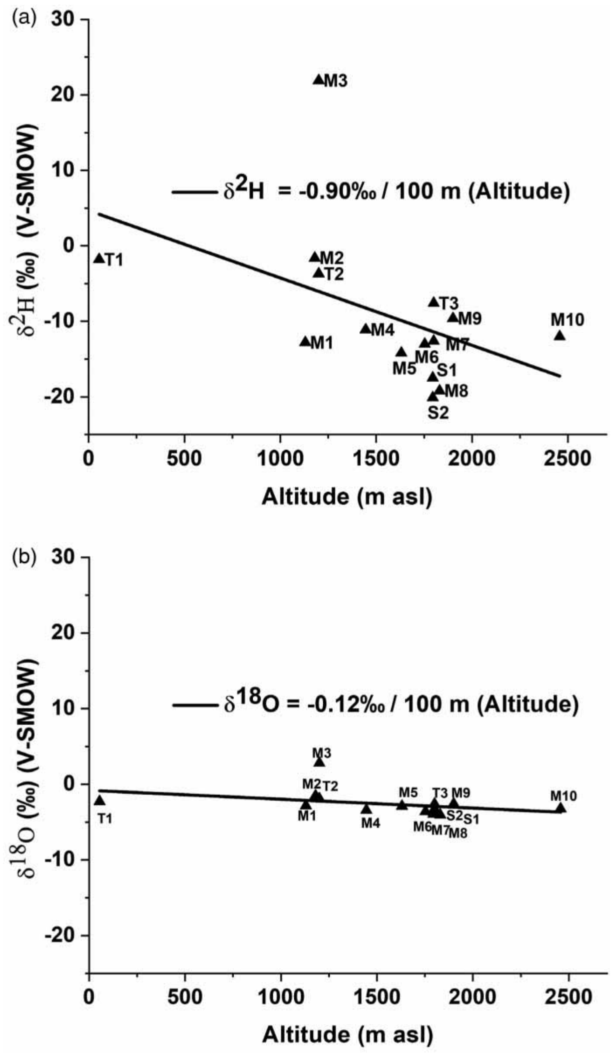

Figure 6 | δ^2H (a) and δ^{18}O (b) values plotted vs. altitude.

Table 5 | Correlation matrixes of chemical composition

Variables	Na^+	K^+	Mg^{2+}	Ca^{2+}	Cl^-	SO_4^{2-}	HCO_3^-	NO_3^-	SiO_2
Na^+	**1**								
K^+	– 0.041	**1**							
Mg^{2+}	– 0.092	0.012	**1**						
Ca^{2+}	– 0.054	0.201	**0.751**	**1**					
Cl^-	0.339	0.148	0.305	0.365	**1**				
SO_4^{2-}	– 0.066	0.169	**0.925**	**0.927**	0.326	**1**			
HCO_3^-	**0.843**	– 0.062	– 0.209	– 0.148	– 0.196	– 0.166	**1**		
NO_3^-	0.245	– 0.098	0.416	0.131	– 0.275	0.314	0.384	**1**	
SiO_2	0.080	**0.526**	– 0.187	0.120	0.257	0.047	– 0.055	– 0.305	**1**

Values of correlation coefficient higher than 0.5 are set in bold.

Table 6 | Eigenvalues, percentage of variance, and cumulative percentage values obtained from correlation matrixes of chemical composition

	F1	F2	F3	F4
Eigenvalues	6.867	5.383	4.267	3.367
Percentage of variance (%)	24.524	19.224	15.240	12.024
Cumulative percentage %	24.524	43.747	58.987	71.011

Four trends (F1-F4) are observed.

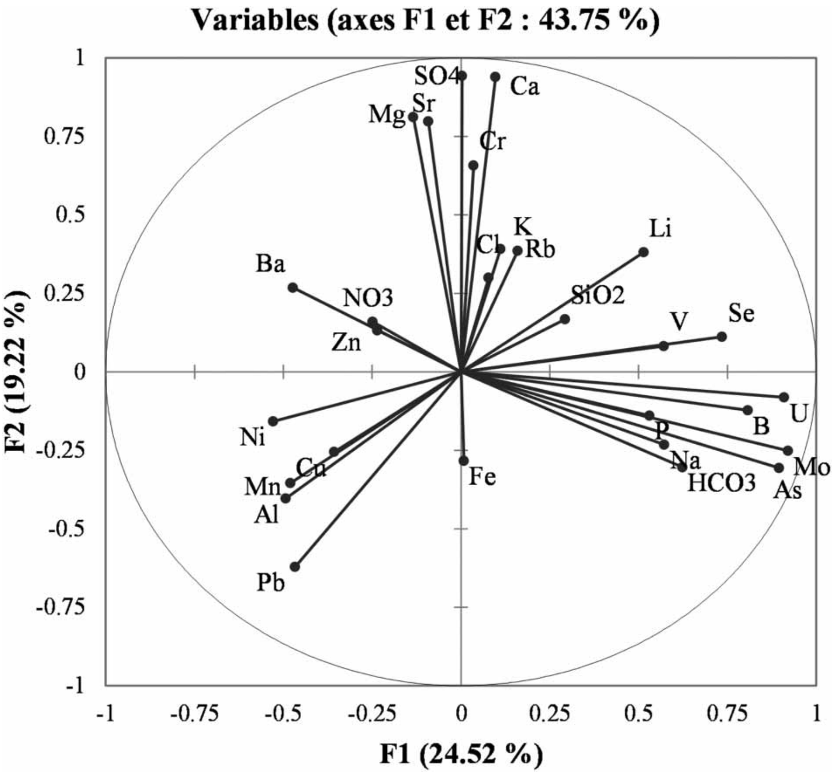

Figure 7 | Graphic representation in the factorial plane.

elements. Despite a high level of cultivation and the fast recharge conditions, few nitrates are found in the mineral and spring bottled water, but it would be necessary to regularly control the chemical quality to ensure to a standard sufficient for drinking water with the increase of agricultural activities.

Stable water isotopes (δ^2H and δ^{18}O) have showed that the recharge in these aquifers is fast and by direct recharge from rainfall without evaporation except one (M3), whose recharge is indirect through the bed of Victoria lake (evaporated water). In addition, the isotope technique allows characterization of the recharge altitude for each mineral and spring bottled water. Finally, in term of climate circulation, isotopes show that the aquifers are mainly recharged by rainfall issued from continental recycling.

We have studied brands of bottled waters purchased in supermarkets of Kigali city in 2017-2018. Despite the representativeness of the samples, other brands could have appeared after this date and deserve to be studied.

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DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

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