

Article

Geochemical Characterization of Nyamyumba Hot Springs, Northwest Rwanda

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Abstract: A hot spring is a hot water source that naturally occurs on the surface of the Earth from underground; it is typically heated by subterranean volcanic activity and the local underground geothermal gradient. There are four main hot springs in Rwanda, such as: Kalisimbi, Bugarama, Kinigi, and Nyamyumba, formerly named Gisenyi hot springs. It is often believed that soaking in a hot spring is a great way to naturally detox human skin. This research focuses on the geochemical analysis of Nyamyumba hot springs, located near the fresh water supply of Lake Kivu, with the purpose of understanding its healing capacity and safety. The Nyamyumba hot springs are located in the western branch of the East African Rift System, near the Virunga volcanic complex, which explains the rising and heating mechanism of the water. The concentrations of sulfate, iron, ammonia, silica, and phosphate, and the conductivity, alkalinity, and salinity of the water were measured using standard procedures. The results showed that the hot spring water has higher concentrations of chemicals compared to the Lake Kivu water, and the geochemistry of these hot springs may be associated with rock dissolution by hot water. The measured parameters were compared with World Health Organization (WHO) standards for recreational waters, and it has been identified that the Nyamyumba hot springs are safe to use for swimming and therapeutic activities.

Keywords: geochemistry; hot springs; Lake Kivu; rift system; safety; Nyamyumba



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1. Introduction

A hot spring, known as a geothermal spring, is a spring produced by the emergence of groundwater that has been strongly heated geothermally below the Earth's surface. Hot spring water often contains large amounts of dissolved minerals. According to [1], the chemistry of hot springs ranges from acid sulfate springs with a pH as low as 0.8 to alkaline chloride springs saturated with silica and bicarbonate springs saturated with carbon dioxide and carbonate minerals. Some springs also contain abundant dissolved iron. The minerals brought to the surface in hot springs often feed communities of extremophiles, microorganisms adapted to extreme conditions, and it is possible that life on Earth had its origin in a hot springs [2].

Generally, the chemical composition of a hot spring varies from alkaline to acid sulfate to bicarbonate to iron-rich, and each of them defines an end member of a range of possible chemical compositions of that hot spring [3]. According to [4], alkaline chloride hot springs are derived from the hydrothermal fluids that form when groundwater containing dissolved chloride salts reacts with silicate rocks at high temperatures. These springs are saturated with silica (SiO₂) but their pH is nearly neutral. Since silica solubility depends on temperature, upon cooling, the silica is deposited as geyserrite, a form of opal (opal-A: SiO₂·nH₂O) [4]. On the other hand, acid sulfate hot springs are made when the

hydrothermal fluids rich in hydrogen sulfide (H_2S) are oxidized to form sulfuric acid, H_2SO_4 [3].

The resulting pH in this formed fluid is lowered to an acidic value less than 0.8 [1]. The so-formed acid reacts with the rock to change it to clay minerals and a silica residue. In the case where carbon dioxide (CO_2) and groundwater react with carbonate rocks, this would lead to the formation of the hydrothermal fluid of bicarbonate hot springs [3]. When these hydrothermal fluids reach the surface, CO_2 is rapidly evaporated and carbonate minerals precipitate to form travertine, thus, bicarbonate hot springs tend to form high-relief structures around their openings [5]. Iron-rich hot springs are characterized by the presence of groups of microorganisms that produce small groups of oxidized iron from the iron found in the hydrothermal fluids, which then feed the springs [6].

Recreational waters may contain chemicals of organic or inorganic sources. Generally, chemicals get into hot springs' water through rock dissolution where it passes, and the soil in which the water percolates [7]. This is due to the fact that dissolution increases with temperature. Other than temperature, the low pH of the water also causes greater leaching of inorganic materials from rocks and soil, increasing the chance that naturally occurring inorganic substances will accumulate at higher concentrations than expected [8].

The main sources of sulfate ion in underground waters are: (i) fumarolic gas of sulfur dioxide, (ii) rocks forming minerals of gypsum ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$), and (iii) rocks forming minerals of pyrite [9]. However, pyrite needs a lot of oxygen to form sulfate ion ($\text{FeS}_2 + 4\text{O}_2 + 3\text{H}_2\text{O} \rightarrow \text{Fe}(\text{OH})_3 + 2\text{SO}_4^{2-} + 6\text{H}^+$) [10]. Geothermal water tends to reduce the resistivity and increase the conductivity of its host rock which is caused by high concentrations of dissolved particles in the fluid and secondary alteration minerals that are formed when the thermal waters interact with the host rock. The type of alteration minerals being formed depends on the type of the host rock and the temperature and salinity of the fluid [10]. In complex circulation processes, water–rock interactions play an important part in the hydro-geochemical indicators of the waters in the geothermal systems which can be used to examine the properties of geothermal reservoirs and the mixing behavior of groundwater [11–14]. Mixing processes between thermal and non-thermal water can be enhanced by faults in a geothermal area [13].

The minerals in hot springs have a detoxifying capacity and remedy skin ailments including acne, eczema, and psoriasis because of their dissolved chemicals including sodium bicarbonate, chloride, and sulfur [15,16]. Additionally, the Nyamyumba hot springs attract many people for swimming purposes.

2. Geological and Geochemical Background of the Area

Kivu Rift is the middle part of the Western branch of the East African Rift System. Different geodynamic processes including faulting and magmatism controlled and contributed to the formation of the lake and different structures along the margin of Lake Kivu (Figure 1).

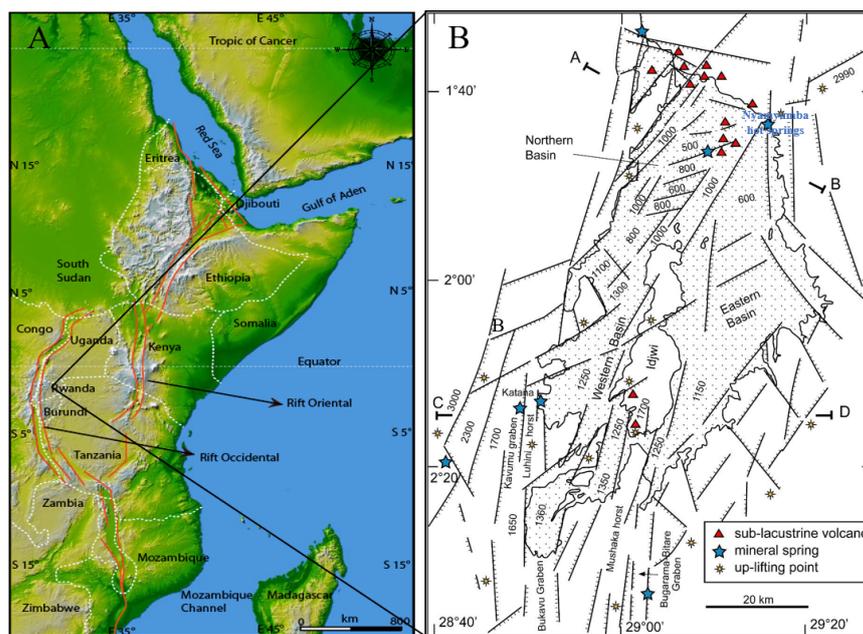


Figure 1. (A) Digital elevation model showing the location of the East African Rift System and Rwanda. (B) Structural map of the Kivu basin. A–B and C–D indicate the cross-sections. Adapted with permission from [17], 2016, Elsevier.

The East African Rift System resulted from two mantle plumes beneath the Afar and Kenyan Plateau [18–20]. The tectonic uplift and an extension led to the creation of the East African Rift (EAR) [21], the best example of an active rift system. The plateaus are dynamically supported by the convective activity under the asthenosphere [22], providing heat transfer for partial melting of the lithospheric mantle. All of these parameters make the East Africa Rift System (EARS) a very good potential area for geothermal resources [23].

The western branch of the East African Rift System to which Nyamyumba belongs has a limited and localized volcanic product with a more diverse chemistry than the eastern branch. The Kivu rift valley is composed of deep lacustrine basins and structural heights which are overlain by volcanic rocks [17] indicating the presence of a mantle plume beneath the lithosphere.

The northern basin of Lake Kivu (the closest to the Nyamyumba hot springs) contains about 0.5 km of sediments which overlie a basement believed to be of crystalline rocks of Precambrian age [24].

The faults around the mineral springs in the western part of Rwanda suggest that the rising temperature of Nyamyumba hot springs' water is structurally controlled.

Lake Kivu is known to possess high concentrations of carbon dioxide and methane in its deep waters. A minor amount of nitrogen is also present in all depths and the dissolved oxygen decreases with depth. The pH of the oxygenated waters lies around 9 and drops below 7 in the anoxic waters. Moreover, the distribution of principal cations shows the increase in salt content with water depth, and they are at a relatively uniform concentration level at a given depth in an independent geographical position [24].

The chemical composition of water can be produced by (1) a long circulating fluid system; (2) spatial variations in the biomass distribution; and (3) water–rock interactions [25].

3. Methods

3.1. Sampling Procedure

Fifteen water samples were collected at seven locations: 0 (hot spring emerging point), 1, 2, 3, 4, 5, and 6 using clean plastic narrow-mouthed bottles (Figure 2). Collections were made from the hottest regions of the pools, near the vents down to the lake, in order to compare the chemistry at different locations and analyze the impact of hot spring water

chemistry as compared to the chemistry of Lake Kivu's water. There exist two emerging points at the male site. Therefore, two samples were collected at each point for comparison.

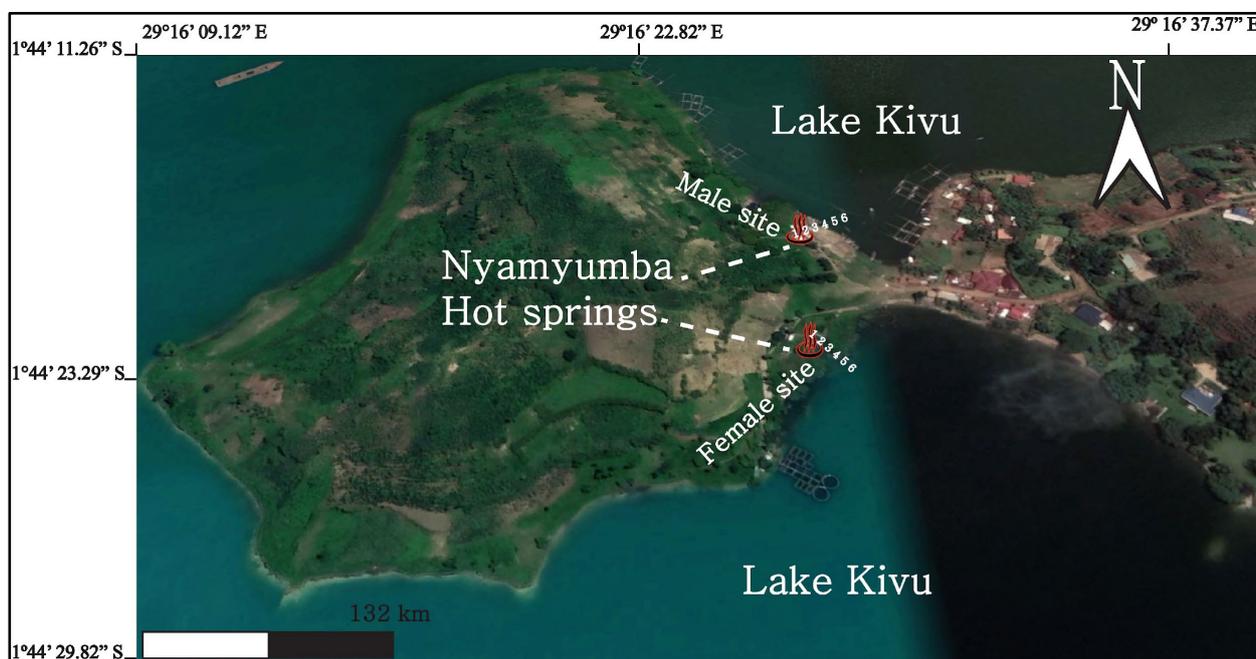


Figure 2. The location of Nyamyumba hot springs shows the location of the two sites (female and male) relative to Lake Kivu with corresponding longitude (x -axis) and latitude (y -axis). Names: the male and female names were named by local people based on the location reserved for males and females, respectively. Numbers from 1 to 6 represent the sampled locations. N: North, S: South, E: East.

3.2. Analytical Procedure

Conductivity as the measure of the ability of water to conduct electricity increases with increasing salinity [26]. The conductivity of water samples in seven locations was measured using CTD Sonde. Measuring the conductivity at each location helps to compare the dissolved ions and factors favoring the dissolution at different locations.

The concentrations of sulfate, iron, ammonia, silica, and phosphate and the alkalinity of the water were measured in water samples using Hach test kits and procedures.

The silica content was measured using the silico-molybdate method (silica, high range (0 to 75.0 mg/L)). The measurement of the silica concentration was preceded by warming the samples at room temperature before analyzing them. A silica standard solution, 50-mg/L as SiO_2 , was used. The silica and phosphate in the sample reacted with molybdate ion under acidic conditions to form yellow silicomolybdic acid complexes and phosphomolybdic acid complexes. Citric acid was added to destroy the phosphate complexes. Silica was then determined by measuring the remaining yellow-colored complex.

Moreover, the alkalinity which is the capacity of water to resist acidification, was measured by using a digital titrator. Unlike other chemicals, the sample bottles for phosphate analysis were first cleaned with 1:1 hydrochloric acid solution and rinsed with deionized water. The sample was poured into the mixing bottle. One phenolphthalein indicator powder pillow was added and mixed. A 0.035 N sulfuric acid standard solution was added in drops. The solution was mixed after each drop until the color changed from pink to colorless. The number of drops until the color changed was multiplied by 20 to obtain the alkalinity of phenolphthalein as CaCO_3 . One bromocresol green-methyl red powder pillow was added and swirled to mix the solution. A 0.035 N sulfuric acid standard solution was added in drops. The solution was again mixed after each drop and the number of drops was counted until the color changed from green to pink. The total number of drops for the

whole procedure was calculated and multiplied by 20 to obtain the total alkalinity (methyl orange) as CaCO_3 . The standard solution used was 500 mg/L as CaCO_3 using the above stated procedure.

The measurement of the iron concentration was preceded by collecting water samples in acid-cleaned plastic containers. Since the water samples were analyzed immediately, no acid was added. The Ferro Ver method for iron (0 to 3.00 mg/L) was used for the analysis. Iron standard solution of 1.0 mg/L was prepared by diluting 1.00 mL of iron standard solution and 100 mg/L Fe to 100 mL with deionized water. Then, the test was run using the AccuVac Ampuls procedure.

The concentration of phosphate was measured by filling 25 mL of sample within a 25 mL sample cell. Using a 1 mL calibrated dropper, 1 mL of molybdate reagent was added. This was followed by the addition of 1 mL of amino acid reagent solution. The sample was well mixed, and the reaction took 10 min to complete. 25 mL of the sample was poured into the sample cell. When the timer beeped, mg/L PO_4 was displayed. The blank was placed into the cell holder. The sample cell was covered with the instrument cap and after pressing 'zero', the cursor moved to the right and 0.0 mg/L PO_4 was displayed. The prepared sample was placed into the cell holder and covered with the instrument cap. After pressing 'read', the cursor moved to the right and the final concentration of the sample was displayed. This was completed for each sample to measure its concentration. For phosphorus, a reactive (0 to 30.0 mg/L PO_4^{3-}) method was used. A 10.0-mg/L phosphate standard by pipetting 10.0 mL of a phosphate standard solution, 50 mg/L as PO_4^{3-} into a 50-mL volumetric flask was prepared. The sample was diluted to the volume with deionized water. The procedure was followed as stated above and the concentration was 10 mg/L.

The concentration of ammonia was also measured by filling two tubes with 5 mL of the sample. One tube was put in the left opening of the color comparator box, whereas with the second tube, ammonia salicylate reagent powder pillow was added. The tube was shaken until the powder was fully dissolved. After a few minutes, an ammonia cyanurate reagent powder pillow was added and shaken, and after 15 min, a green color developed. The second tube was put in the color comparator box which was held in front of the light source. Color matching was found by turning the color disc and the result was read in the scale window.

The ammonia present in the water is in the form of ammonium ion (NH_4^+) and unionized ammonia (NH_3). So, this method measures both NH_4^+ and NH_3 as ammonia nitrogen ($\text{NH}_3\text{-N}$). One mg/L $\text{NH}_3\text{-N}$ of nitrogen ammonia standard solution was used. The mg/L NH_3 in the sample was calculated as follows:

- $\text{mg/L NH}_3 = ((\text{mg/L NH}_3\text{-N} \times \text{percent NH}_3 \text{ of water sample at a given temperature and pH}) \div 100) \times 1.2$ [27].

A CDC401 conductivity probe was used in the measurement of the salinity of the water samples. The probe was rinsed with deionized water and dried afterward with a lint-free cloth. The shroud was installed. The probe was put in the sample with the sensor fully in the sample. Air bubbles were removed by shaking the probe. After shaking, it was stirred, and the salinity was read.

Furthermore, sulfate analysis was completed using the Sulfa-Ver 4 method for sulfate (0 to 70 mg/L). The method was first calibrated with the standard solution of 50 mg/L of sulfate. The standard solution was prepared by pipetting 1 mL of a PourRite ampule standard for sulfate (2500 mg/L) into a 50 mL volumetric flask. The sample was diluted with deionized water. In this method, sulfate ions react with barium in Sulfa-Ver 4, a sulfate reagent forming barium sulfate precipitate. The turbidity formed is proportional to the sulfate concentration, and the stabilizing agent in Sulfa-Ver 4 holds the suspended precipitates.

4. Results

Chemical analysis of the water samples at all of the locations indicated that the male site has higher concentrations of chemicals than the female site (Tables 1 and 2). Compared with the other locations, the water samples from the hot spring sources have higher concentrations.

Table 1. The concentrations of chemicals at seven locations taken at the female site.

Parameter/ Location	Ammonia (mg/L)	Silica (mg/L)	Conductivity ($\mu\text{s}/\text{cm}$)	Salinity (ppt)	Alkalinity (eq/L)
0 (Hot springs)	0.145	47	2400	1.2	19
1	0.16	53.7	2550	1.4	20.7
2	0.15	49.6	2100	1.14	18.7
3	0.135	16	1450	0.8	17.0
4	0.087	7.5	375	0.6	4.6
5	0.08	5	210	0.56	4.0
6 (Lake)	0.05	1.7	185	0.49	3.5

Table 2. The concentrations of chemicals at seven locations taken at the male site.

Parameter/ Location	Ammonia (mg/L)	Silica (mg/L)	Conductivity ($\mu\text{s}/\text{cm}$)	Salinity (ppt)	Alkalinity (eq/L)
0 (Hot springs)	0.15	53.7	2470	1.3	22.0
	0.16	57.0	2450	-	23.8
1	0.41	55.3	2670	1.26	22.0
2	0.58	54	2600	1.3	21.4
3	0.28	36	2050	1.0	19.0
4	0.13	10	540	0.74	16.0
5	0.08	5	210	0.6	10.0
6 (Lake)	0	5	185	0.6	3.5

The large variation in ammonia content was observed at location 2 at both sites. The male site has a very high ammonia concentration of around 0.58 mg/L compared to 0.15 mg/L at the female site.

Another significant variation was also observed in conductivity and salinity contents which are lower at the sources compared to location 1 at all sites (Figure 3B,C). Furthermore, the conductivity of the water at the source is higher than the conductivity in Lake Kivu and the other locations (Figure 3B).

Like other chemicals, the concentration of silica decreases from the hot spring sources to the lake. An abrupt change was marked between locations 2 and 3 (Figure 3D).

The alkalinity at location 4 shows a greater decrease in concentration at the female site than at the male site (Figure 3E). The significant change in alkalinity was observed between locations 3 and 4.

Generally, the concentrations in the lakes are lower than the concentrations at the source of the hot springs as shown by the linear and curved relationships in Figure 3.

The average concentrations calculated from Tables 1 and 2 at the male and female sources in Table 3 show that ammonia is in a very low concentration compared to the other chemicals. Sulfate and silica have larger concentrations than other chemicals, with their concentrations being 75.7 mg/L and 52.57 mg/L, respectively (Table 3).

Table 3. Average concentrations of the chemicals at the source of the hot springs.

Parameter/ Location	Ammonia (mg/L)	Silica (mg/L)	Conductivity ($\mu\text{s}/\text{cm}$)	Salinity (ppt)	Alkalinity (eq/L)	Iron (mg/L)	Phosphate (mg/L)	Sulfate (mg/L)
Source	0.152	52.57	2440	1.25	21.6	0.2	0.31	75.7

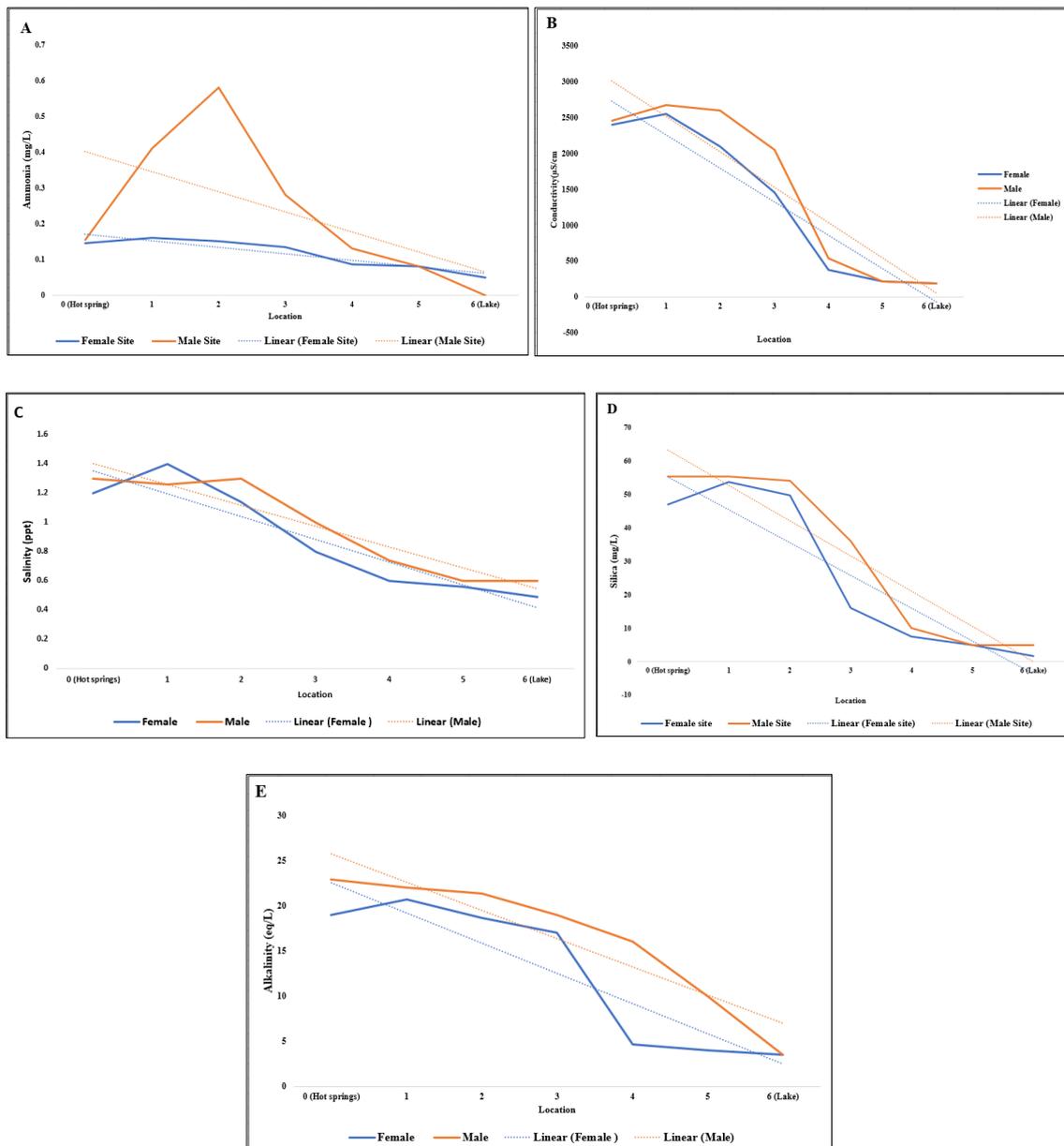


Figure 3. Comparison of chemical composition of water samples taken at seven locations at two sites (female and male). (A) ammonia, (B) conductivity, (C) salinity, (D) silica, and (E) alkalinity.

5. Discussion

The results of this study show that all of the analyzed parameters have higher concentrations at the hot spring sources than the other locations as shown in Figure 3 above. The concentration of these parameters tends to decrease from the source of the hot spring to Lake Kivu; this variation has been explained by this study to be caused by the dissolution of the rocks encountered by hot water due to its high temperature before it emerges on the surface. This explains why the concentration of chemicals in the hot spring is very high compared to the lake water.

Table 4 below shows the comparison of the chemical parameters' concentration measured from the Nyamyumba Hot Springs and the World Health Organization standards for swimming water.

Alkalinity refers to the water's capacity of withstanding changes in pH while allowing the water quality to be maintained. It also refers to the measure of the buffering ability of the water. The alkalinity level recommended by the World Health Organization standards

in swimming pools is limited to 2.7–1668 eq/L, below which it can turn the pool water green and cause eye and skin irritation, and above which it turns the pool water cloudy.

Table 4. The comparison of chemicals from the hot springs and the WHO standards.

No	Chemicals	WHO (Min)	WHO (Max)	Hot Spring Data	Reference
1	Ammonia (mg/L)	0.2	3	0.152	[28,29]
2	Silica (mg/L)	-	100	52.57	[30]
3	Conductivity ($\mu\text{s}/\text{cm}$)	-	-	2440	
4	Salinity (ppt)	0.3	1.7	1.25	[28]
5	Alkalinity (eq/L)	2.7	1668	21.6	[28,31]
6	Iron (mg/L)	0.5	50	0.2	[29]
7	Phosphate (mg/L)	-	5	0.31	[29]
8	Sulfate (mg/L)	-	500	75.7	[29]

The alkalinity measurement of 21.6 eq/L obtained from the hot springs' data falls within the acceptable World Health Organization range. The alkalinity at the location 4 female site immediately decreases and continues to be constant at locations 5 and 6. This is caused by the mixing of lake water and hot spring water. The dilution of this water leads to the decrease in alkalinity [32].

Ammonia is considered as one of the most unwanted pollutants in waters used for swimming. The results show that the average concentration of ammonia is low in the hot springs' water compared to the acceptable limits of WHO standards. The increase in the concentration of ammonia at location 2 of the male site (see Figure 3A) may have been caused by the decay of organic matter and chemical fertilizers as the location is in the swamp area with water and bushes.

The silica contents in groundwater are found in two forms such as dissolved particulate matter. Dissolved silica is an indicator of weathering and water circulation [33]. According to [34], the silica contents in groundwater are mostly from rock–water interactions. The weathering process which releases silica into underground water is controlled by the water saturation deficit of the aeration zone, precipitation and temperature fluctuations, mineral stability, and bedrock reactivity [33].

Although the exact form of dissolved silica is not known, it is always measured as SiO_2 [35]. It has been calculated that the average concentration of silica in natural water ranges between 1.0 and 30.0 mL/L [36]. It has been also shown that water ascending from deep reservoirs contains higher silica values than those of shallow origin.

The silica content in the studied hot springs has the average concentration of 52.57 mg/L. This concentration is higher than the average calculated by [36]. This indicates that the source of the water reservoir is deep; however, it falls in the acceptable concentration of the World Health Organization standards for swimming water according to Table 4 above.

Salinity is a measure of the amount of salts in the water and conductivity is the ability of water to conduct an electrical current with dissolved ions as conductors [37]. The increase in conductivity infers to the increase in the salinity of the water [38]. The higher conductivity of hot spring water results from salinity or simply the concentrations of dissolved ions. The concentration of salinity is higher at location 1 compared to the sources of the hot springs; this led to the increase in conductivity at location 1. Three factors affecting the conductivity include: the concentrations of dissolved ions (salinity), the type of ions, and temperature. The temperature facilitates the mobility of ions. Hot springs' water is hot and this allows the dissolution of rocks in which they percolate hence increasing the concentrations of ions in the water and their conductivity. The results obtained from the

hot spring show that the conductivity (2440 $\mu\text{s}/\text{cm}$) and the salinity (1.25 ppt) fall within the acceptable range of World Health Organization standards for swimming water.

Iron occurs naturally in water; however, increased concentrations mostly lead to corrosion and affect the overall quality of the water. The study of [39] has shown that when iron concentrations exceed 0.2 parts per million (ppm), staining and clouding occur.

Phosphates are organic materials which come from various sources including dead plants and chemical fertilizers. If the phosphorus concentration is high in the water, it feeds the algae and allows them to grow and turns the water green and cloudy. Increased concentrations have possible health effects on swimmers.

Sulfates occur naturally and are abundant in the environment originating mainly from mineral deposits, soils, rocks, and the combustion of sulfur-related fuels according to [40]. Exposure to high sulfate concentrations has many health effects including reduced lung and heart function. Sulfates also lead to degrading visibility by scattering the light before it reaches the person.

The concentrations of iron, phosphate, and sulfate in the samples collected in the Nyamyumba hot springs are 0.2 mg/L, 0.31 mg/L, and 75.5 mg/L, respectively, and they all comply with the acceptable range of World Health Organization standards for swimming water.

People usually use hot springs' water in therapeutic activities. The study conducted by [41] has shown that bathing in hot springs increases the body temperature, which in turn increases the blood flow in the human body and eases the absorption capability of the intestines. When swimming in hot springs' water, people's skin is extensively exposed to chemicals and the dissolved minerals originating from the subsurface of the Earth and each mineral contains unique properties that may benefit or harm the human body.

Two pathways have been suggested for the transport of chemicals across the stratum corneum (outermost layer of skin): one for lipophilic chemicals and the other for hydrophilic chemicals [42]. The extent of uptake through the skin will depend on a range of factors, including the period of contact with the water, the temperature of the water, and the concentration of the chemical. For these reasons, the chemical composition of hot springs must be analyzed to understand the safety-related issues. All the parameters measured in this research are lower than the maximum values suggested by the WHO for recreational water (Figure 4).

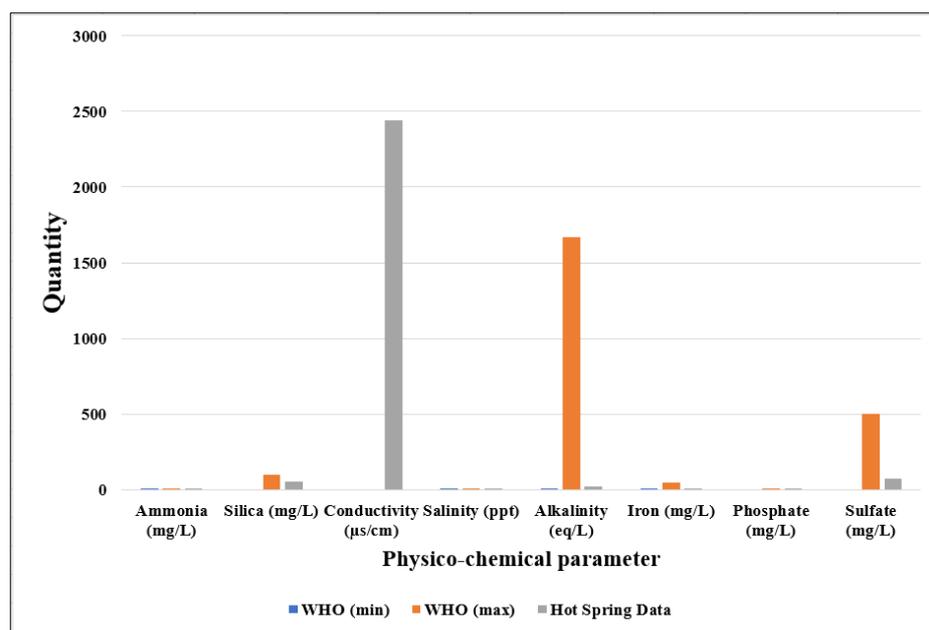


Figure 4. The comparison of the concentrations of chemicals in the hot springs and the WHO standards for swimming water.

Therefore, based on the measured parameters, swimming in hot springs can be trusted. However, this study focused on using only Nyamyumba hot springs' water for swimming and recreational purposes; detailed analysis for drinking purposes and continuous monitoring are recommended for people's safety. Furthermore, the relationship between Nyamyumba Hot Springs, active faults and volcanic activity around the Hot Springs is another issue we will discuss in next research.

6. Conclusions

This research aimed to study the concentrations of chemical compositions of Nyamyumba hot springs' water compared to those of Lake Kivu water. The location of Nyamyumba hot springs in the East African Rift System plays a great role in controlling the rising of the hot springs' water temperature. The heating of the water may be caused by a magmatic chamber in the underground area. Most of the chemicals in hot springs' water resulted from rock–water interactions. This has been influenced by the temperature of the hot springs. It has been identified that chemicals have higher concentrations at the source than in the lake water due to the dissolution of rocks by circulating hot water. The results also showed that the use of Nyamyumba hot springs for swimming is safe when compared to the WHO standard concentrations of chemicals in recreational waters. However, a detailed study is advised to consider other chemicals especially carcinogenic chemical elements and other infectious biological organisms.

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