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ASSESSMENT OF THE QUALITY OF BOTTLED WATER MARKETED IN ADDIS ABABA, ETHIOPIA

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ABSTRACT. The consumption of bottled drinking water has been increasing in the world. But, the quality of bottled water used for human consumption is not subjected to any stringent quality control measure in Ethiopia. The present study was carried out to determine the physicochemical quality parameters of twenty brands of bottled drinking water available in Addis Ababa and to compare with drinking water guidelines set by World Health Organization (WHO) and Compulsory Ethiopia Standards (CES). The samples were collected by random sampling technique. The physicochemical parameters such as pH; TDS and EC; total alkalinity, total hardness, HCO₃⁻, Cl⁻, SO₄⁻², NO₃⁻, F) and common cations (Ca²⁺, Mg²⁺, Na⁺, and K⁻) were determined by pH meter, conductivity meter, tirtation, UV-Visible spectrometry, flueride ion selective electrode and microwave plasmatomic emission spectrometry, respectively. The mean physicochemical concentration of TDS, total alkalinity, total hardness, HCO₃⁻, SO₄⁻², NO₃⁻, NO₂⁻, F⁻, Cl⁻, Ca²⁺, Mg²⁺, Na⁺, K⁺, pH and EC in bottled water samples were found in the range (4.67-139), (12-165), (4-97), (16.3-202), (0.05-3.84), (0.02-0.84), (0.08-0.28), (0.03-1.26), (4.28-20.8), (0.19-8.9), (0.02-2.5), (1.8-24.5), (0.14-8.08 mg/L), (6.43-7.69) and (9.8-289 µs/cm), respectively. The drinking water quality index (WQI) was calculated based on 15 important quality parameters and the results were found in the range 3.85-49.2. The mean concentrations of physicochemical parameters in almost all the bottled drinking water were below the permissible limit set by WHO and CES. Hence, all the tested bottled water samples.

KEY WORDS: Bottled water, Common cations, Common anions, Physicochemical parameters, Water quality index

INTRODUCTION

Water is one of the most important of all natural resources known on the earth. It is mainly used for drinking purposes which come from the surface and underground water sources [1]. 97% water exists in the oceans which are not suitable for drinking purpose and 3% is fresh water. Of the 3% fresh water 2.97% is comprised of glaciers and ice caps and remaining little portion of 0.3% are available as a surface and groundwater for human use [2]. A high quality water is required for drinking purposes for better health [3]. Freshwater is already a limiting resource in many parts of the world. In the next century, it will become even more limiting due to increased population, urbanization and climate changes. The safety of drinking water might be affected by various contaminants which includes chemical, physical and microbiological contaminants which can cause serious health problems to human [4]. Moreover, human beings depend on water for almost every developmental activity. Because of its importance, the pattern of human settlement throughout the history has often been determined by its availability [5].

Most of the diseases in developing countries are caused by the consumption of contaminated water. The people living in developing countries lack access to clean water due to environmental pollution [6]. The safety of drinking water is affected by various contamination of drinking water with chemical, microbiological and anthropogenic activities [7]. The chemical contaminants in potable water include: some heavy metals, non-metals, disinfection by-products, nitrate, nitrite, pesticides, and sulfate [8]. In most urban-rural communities in the developing countries especially the Sub-Sahara Africa, surface waters (rivers, streams, and lakes

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among others) have been the most available sources of water used for domestic purposes. The water from these sources is contaminated with domestic, agricultural, and industrial wastes and likely to cause water-related diseases. Impaired surface water quality always result in an unhealthy socio-economic environment [9]. The levels of chemicals in drinking water are sometimes high enough to cause acute health effects. Polluted drinking water causes many diseases as diarrhea, vomiting, gastroenteritis, dysentery, kidney problems [10].

Water quality parameters are the physical, chemical, and biological characteristics of water in association to the set of standards [1, 11]. Safe water is to say that drinking water need to be free from pathogenic organisms, toxic substances, an overdose of minerals and organic materials as well as it should be pleasant (free from color, turbidity, odor and taste) [1, 12]. Water quality parameters provide important information about the health of a water body. Besides, these parameters are used to find out the quality of water for drinking purpose [2]. The quality of water is the degree of its potability and is determined by the level of physicochemical, microbial and heavy metals (which include suspended and dissolved substances in the water, the degree of alkalinity (pH), temperature, appearance in term of color, taste, odor and the presence of non desirable microorganisms). Water for domestic purposes should therefore be free from these substances in order to prevent waterborne diseases [13]. World Health Organization (WHO) publishes guidelines for drinking water quality [14, 15] which many countries use as the basis to establish their own national standards. The guidelines represent a scientific assessment of the risks to health from biological and chemical constituents of drinking water.

The bottled water first began in 1970, and with the promotion of this product bottled water market has grown, and by late 1990 multiplied by three times the soft drinks market. Water consumed by a human comes in various forms and from the various sources, bottled water is one of that forms [16]. Recently it has been increasing dramatically due to the ever increasing contamination of water resources. Bottled water consumption has been growing steadily worldwide during the last three decades and is regarded as fastest growing and most dynamic sector of all the foods and beverage industries. Consumption of bottled water is a proper choice in the world when no safe water is available or water treatment is difficult, but the safety of bottled water is important and it is necessary to monitor toxic and trace metal contaminants [17, 18]. In order to protect health, people turn to consume bottled water and spend a lot of money to buy bottled water. The non-arrival of useful water for consumers, presence of odors and unpleasant taste in the water supplied by municipal, believe of consumers on the impact of medical and therapeutic bottled water gives a sense of height of the social level in addition to the promotion of media for the consumers [16].

Bottled water delivery service is an important part of the bottled water industry. For homes and offices, it is a convenient way to receive cost-effective; high quality drinking water on a regular basis. Studies have shown that use of purified water in the workplace increases productivity and improve the overall health of the workforce [19]. In the metropolitan cities, like Addis Ababa (Ethiopia), with large population size and the diplomatic headquarters of Africa enhances the commercialization and profitability of water as a product in the national and international market for Ethiopian bottled water manufacturers [18].

Several studies have been done on assessment of quality of bottled waters in the different cities in the world. These include determination of heavy metal profile in bottled water samples obtained from various markets in Lagos, Nigeria [8], classification and evaluation of commercial bottled drinking waters in Saudi Arabia [9], application of water quality index for assessment water quality in some bottled water in Erbil City, Kurdistan Region, Iraq [20], chemical assessment of bottled drinking waters by IC, GC, and ICP-MS [21], physical, chemical and microbial analysis of bottled drinking water [22], chemical water quality of bottled drinking water brands marketed in Mwanza City, Tanzania [23] and comparative study between bottled mineral and tap water in Italy [24].

Water quality index (WQI) is a very useful and efficient method for assessing the suitability of drinking water; it is also a very useful arithmetical tool used to transform large number of water quality data into a single cumulatively derived number and for communicating the information on the overall quality of water to the concerned citizens and policymakers [13, 25]. It, thus, becomes an important parameter for the assessment and management of water quality (both surface and groundwater). WQI reflects the composite influence of different water quality parameters and is calculated from the point of view of the suitability of both surface and groundwater [20].

Several studies have been reported on the determination of water quality index of different water bodies in different countries. Some of these studies are determination of water quality index and suitability of urban river for municipal water supply in Lagos, Nigeria [13], determinations of water quality index and suitability of an urbawater body in Shimago Town, Karnataka, India [26], water quality assessment in terms of water quality index [27, 28], mathematical computation of water quality index of Vea Dam in Upper East Region of Ghana [29], assessment of water quality index of Robertson Lake Jabalpur (India) and use of test result in remote sensing application [30], water quality index assessment of Koudiat Medouar Reservoir, Northeast Algeria using weighted arithmetic index method [31], and water quality index for assessment of water samples of different zones in Chandrapur City (India) [32].

In developing countries, including Ethiopia, the drinking water quality is continuously being contaminated and become hazardous for human use due to high growth of population, expansion of industries, disposal of wastewater and chemical effluents into canals and other water sources [2]. In addition to water contamination by pathogenic microbes, which is common in Ethiopia, chemical pollution is rising with industrialization and with the widespread use of agricultural chemicals [33].

Some studies have been reported on the mineral composition of lake water, river water, and irrigation waters in Ethiopia [11, 34, 35]. Physicochemical analysis of drinking water has also been studied to assess the quality of drinking water in different cities and regions of Ethiopia [7, 33, 36, 37]. A few studies have been conducted on the quality of bottled water in Ethiopia. These include the levels of common ions in bottled mineral waters consumed in Addis Ababa, Ethiopia [38] and the assessment of chemical quality of major brands of bottled water marketed in Gondar Town, Ethiopia [18]. However, water quality index has not been determined in any of the water bodies including river water, lake water, irrigation water, drinking water and bottled water in Ethiopia.

Recently consumption of bottled water has been increasing in every region of Ethiopia. But, the quality of bottled water used for human consumption is not subjected to any stringent quality control measures in Ethiopia. Therefore, the present study was carried out to assess the water quality index of bottled water marketed in Addis Ababa, Ethiopia, using measurements of major physiochemical parameters.

The main objective of this study was to determine water quality parameters and calculate the water quality index of bottled water marketed in Addis Ababa, Ethiopia and to compare with the values set by WHO. The specific objectives of the study were: (1) to determine (i) common cations (Na⁺, K⁺, Ca²⁺, Mg²⁺) and anions (F⁻, Cl⁻, SO₄²⁻, NO₃⁻, NO₂⁻, HCO₃) of bottled water and (ii) TDS, EC, hardness, pH, and alkalinity of bottled water; (2) to compare (i) the levels of common ions in bottled water marketed in Addis Ababa with WHO and CES guidelines for drinking water, (ii) the measured values of physiochemical parameters with the labeled values and (iii) the physicochemical parameters of different brands of bottled water and (3) to calculate the water quality index of bottled drinking water in Addis Ababa to assess the quality of water as excellent, good, poor, very poor or unsuitable for drinking.

EXPERIMENTAL

Chemicals

Analytical reagent grade sodium hydroxide, concentrated hydrochloric acid, concentrated sulfuric acid, anhydrous sodium sulfate, sulfanilamide, N-(1-naphthyl) ethylene diamine dihydro chloride, potassium chromate, sodium salicylate, nitric acid (69-72%), potassium sodium tartarate, diazotized sulfanilamide, silver nitrate, calcium carbonate, magnesium carbonate, sodium chloride, ethylene diamine tetra acetic acid (EDTA), glacial acetic acid, sodium fluoride, Eriochrome black T (EBT), bromocresol green (Sigma Aldrich, Germany), ethanol (99.99%, Fisher Scientific, UK), phenolphthalein (Scharlau, European Union), glycerol solution, ammonium chloride, magnesium sulfate, methyl orange (Scharlau, European Union) were used in this study. All the chemicals and reagents whose production companies are not listed were supplied by Sigma Aldrich, Germany.

Instruments

The instruments used in this study includes: UV-Visible Spectrophotometer (Spectra max plus 385, UK), Microwave Plasma-Atomic Emission Spectrophotometer (Agilent Technologies, 4200 USA); Hotplate; Kjeldahl apparatus (Gallenhamp, USA), drying oven (Binder, Germany), pH meter (Model CP-505, Zabrze ul, Poland), conductivity meter (Orion Model 145, USA), and fluoride ion selective electrode (Orion, model 940 expandable ion analyzer, USA).

Sampling area

The present study was carried out at Addis Ababa, which is a capital city of Ethiopia. The population of the city is currently estimated to be 4.6 million [39]. It is located in the geographical coordinate 3^{0} - 15^{0} N and 33^{0} - 48^{0} E with an estimated area of 1,104,300 sq km [40]. Besides, Addis Ababa is a city of political and economic significance being a base for African Union and many other international organizations. The inhabitants are receiving drinking water from Dire Dam situated at the air distance of 20 km, Legedadi Dam at the air distance of 19.50 km, Gefersa Dam situated at the air distance of 18.20 km apart from Addis Ababa Water Supply Administration (AAWSA) office and Akaki water supply system and spring waters and boreholes around the border of the city mainly Akaki sub-city [41]. In addition to tap water supply, the city has an easy access to different brands of bottled water, which are mainly used for drinking purpose.

Water samples collection

Twenty different brands of bottled waters (Bw₁, Bw₂, Bw₃, Bw₄, Bw₅, Bw₆, Bw₇ Bw₈, Bw₉, Bw₁₀, Bw₁₀, Bw₁₁, Bw₁₂, Bw₁₃, Bw₁₄, Bw₁₅, Bw₁₆ Bw₁₇, Bw₁₈, Bw₁₉, and Bw₂₀) were selected for the present study. Each of the drinking bottled water was randomly purchased from shops and supermarkets from different locations in Addis Ababa during January and February 2018. Once all the samples were brought to laboratory, it was kept at 4 °C in the refrigerator with their original sale containers until analysis was carried out. The holding capacities of bottled water containers were 0.5, 1 and 2 L. For each brand of bottled water, three identical samples of bottled water were mixed to make a bulk sample. Furthermore, the sampled water was classified as natural spring, natural mineral and purified water. The brands of bottled water and sources of bottled water used in this study are described in Table 1.

Analytical procedures for the determination of physicochemical analysis

Total dissolved solids and electrical conductivity. Total dissolved solids (TDS) and electrical conductivity (EC) measurements were made using Thermo Orion Model 145, USA. The

instrument was calibrated with distilled water. An aliquot of 30 mL water sample was transferred to a 50 mL plastic beaker and put on the plate having a magnetic stirrer, and the instrument sensor was inserted to the beaker to measure the parameters. During the measurement the solutions were stirred constantly in order to homogenize and the measurement was performed in triplicate.

Table 1. Types and sources of bottled mineral water used in the present study.

No.	Bottled water brand	Water type	Source of water
1	Bw ₁	Natural spring water	Entoto mountain, Sululta
2	Bw ₂	Springs water	Ayer Tena, outskirts of Addis Ababa
3	Bw ₃	Natural spring water	Burayu
4	Bw_4	Natural spring water	Debre Birhan
5	Bw ₅	Natural mineral water	Highlands of Sululta
6	Bw ₆	Natural spring water	Holrta Genet
7	Bw ₇	Natural purified water	Entoto mountain, Sululta
8	Bw_8	Purified spring water	Menagesha mountain
9	Bw9	Natural mineral water	Burayu, Oromia region
10	Bw ₁₀	Natural spring water	Gurage area
11	Bw11	Natural spring water	Gurage mountain
12	Bw ₁₂	Pure mineral water	Gefersamona, Tatek
13	Bw ₁₃	Purified natural water	Sululita, Oromia
14	Bw ₁₄	Purified natural water	Alemgena, Oromia
15	Bw ₁₅	Natural mineral water	Koce mountain, Sebeta
16	Bw ₁₆	Natural purified water	Sebeta, Oromia region
17	Bw ₁₇	Natural mineral water	Alemgena, Oromia
18	Bw ₁₈	Natural spring water	Mugo Highlands
19	Bw19	Natural purified water	Chained mountain of Gurage region
20	Bw_{20}	Natural purified water	Wechecha mountain

Note: Bw is bottled water; number 1-20 indicates sample number of bottled water.

pH. The pH values of the bottled water samples were measured using a pH meter (Model CP 505) in triplicate. The pH electrode was rinsed with deionized water and calibrated with pH 4.00, 7.00 and 10.00, standard buffer solutions. The pH electrode was rinsed with deionized water to avoid contamination of the buffers. The pH values of the bottled water samples were measured by inserting the electrode in 50 mL of the bottled water sample in a beaker using the pH meter in triplicate.

Total alkalinity. The alkalinity of the sample was determined by titrating with standard sulfuric acid (0.02 M) using phenolphthalein and bromocresol indicators. 25 mL of bottled sample was transferred into conical flask and phenolphthalein and bromocresol indicators were added to the flask. Finally, the solution was titrated with H_2SO_4 [42]. The concentration of total alkalinity was calculated by useing equation 1.

Total alkalinity as
$$CaCO_3 = \frac{Volume of sulfuric acid \times Molarity of sulfuric acid \times 50 \times 1000}{Volume of sample}$$
 (1)

Bicarbonate. The same procedure as alkalinity was used in measurement of bicarbonates as recommended by [42], except the one conversion factor change in the formula which is 61 instead of 50.

$$Bicarbonate = \frac{Volume of sulfuric acid \times Molarity of sulfuric acid \times 61 \times 1000}{Volume of sample}$$
(2)

Total hardness. The total hardness of the water was determined by complexometric titration using EBT indicator and ammonium-ammonium chloride buffer solution. The buffer solution was prepared by weight 1.17 g of EDTA, 16.9 g of ammonium chloride, 780 mg of magnesium sulfate and 143 mL of ammonium hydroxide solution and trnasfer to the beaker which contains 500 mL distilled water. The solution was stirred until all the solids were dissolved, and its pH was found to be 10. Before the titration, the burette was rinsed with EDTA solution and filled with 0.02 M EDTA standard solutions. An aliquot of 25 mL sample water was transferred to a conical flask and then 2 drops of EBT indicator and 1 mL buffer solution were added the beaker. Finally the solution was titrated against to EDTA solution which was filled in the burette. The same procedure was used for three blank samples. The total hardness of the water sample was calculated by using the equation 3 as calcium carbonate (mg/L) [43].

Total hardness as
$$CaCO_3 = \frac{Volume EDTA \times Molarity EDTA \times 50 \times 1000}{Volume of sample}$$
 (3)

Chloride. The concentration of chloride was determined by titration. 1 mL of potassium chromate indicator and 25 mL sample water was added in the beaker, and the solution was titrated with (0.1 M) silver nitrate. The solution was stirred by magnetic stirrer while titration was carried out. The performance of the method was checked by a standard solution of sodium chloride. The same analytical procedures were used for the determination of chloride in three blank samples. The chloride concentration was calculated by this formula [44].

$$Chloride = \frac{(Volume siliver nitrate - blank) \times Normality siliver nitrate \times MW of chloride \times 1000}{Volume of sample}$$
(4)

MW = molecular weight.

Fluoride. To determine the levels of fluoride in the sampled water, total ionic strength adjustment buffer (TISAB) was prepared by dissolving 58 g sodium chloride, 2 g EDTA, 7 g trisodium acetate and 57 mL glacial acetic acid in 500 mL deionized water. Finally, the pH was adjusted to 5.3 by using 5 M sodium hydroxide solution, and made up to one liter with deionized water. TISAB was used to obtain a pH of 5.2-5.4, which is the optimum pH range for fluoride determination. 10 mL of TISAB and 10 mL of calibration solution were mixed in a beaker for measurement of F in the solution. Similar amount of TISAB was used for instrument calibration. 0.5, 1, 5, 10 and 20 mg/L series standard solution of fluoride were prepared for the instrument calibration. The slope of the calibration curve found to be 58.6 mV which is within the acceptable range (54-60 mV). Similar procedure was used for the measurement of fluoride concentrations in sample water. Thus, equal volumes 10 mL of water samples and 10 mL of TISAB were mixed in a 50 mL plastic beaker and the mixture were stirred thoroughly using a magnetic stirrer while measuring. The concentration of fluoride was read directly from fluoride potentiometer and triplicate measurements were made.

Sulfate. The concentration of sulfate in the sample was determined by spectrophotometric method. A stock standard sulfate solution was prepared by dissolving 147.9 mg anhydrous sodium sulfate (Na₂SO₄) in 1000 mL of distilled water. Conditioning reagent was prepared by mixing of 50 mL glycerol solution, 30 mL concentrated hydrochloric acid (HCl), 100 mL 95% ethanol, 300 mL distilled water and 75 g sodium chloride (NaCl). 100 mL of the sample water and 5 mL of conditioning reagent were transferred to 250 mL flask, and then a spoonful (10 g) of barium chloride crystals was added, mixed and placed on a magnetic stirrer and stirred at a constant speed exactly for one minute. Finally, 1 mL amount of the solutions was poured on absorption cell of the photometer, and turbidity of the solution was measured at 30 second intervals for four minutes at 420 nm. The same analytical procedures were used for the determination of sulfate ions in three blank samples [42].

Nitrite. The concentration of nitrite was determined by a colorimetric method [42]. 25 mL bottled water sample, 1 mL of nitrite and 1 mL of sulfanilamide solution were added to a 250 mL conical flask and allowed for 5 min. 1 mL of N-(1-naphthyl)ethylenediamine solution was added and mixed well. After 10 min absorbance was measured using UV-Visible spectrophotometer at a wavelength of 543 nm. The same analytical procedure was used for the determination of nitrite in three blank samples. The concentration of NO₂⁻ was calculated from the standard calibration curve.

Nitrate. The concentration of nitrate was determined by sodium-salicylate method [42]. First, 10 mL of bottled water sample and 1 mL of sodium-salicylate were transferred into a 250 mL conical flask and kept it in an oven at a temperature of 95 °C. The residue was dissolved in 1 mL of H₂SO₄, swirling the conical flask carefully while it is still warm. Secondly, 50 mL of distilled water and 7 mL of NaOH tartrate solution were mixed and diluted it to 100 mL by distilled water. The analysis is based on the reaction of the nitrate with sodium salicylate in a sulfuric acid medium, which forms a yellow colored salt of nitro salicylic acid. The intensity of color is directly proportional to the nitrate concentration and the absorbance was measured using UV-Visible spectrophotometer at a wavelength of 410 nm. The same analytical procedures were employed for the determination of nitrate ion in three blank samples. Finally, the concentration of NO₃⁻ was calculated from the standard calibration curve.

Analytical procedures for the determination of common cations

For the determination of metal ions in the water sample, 50 mL of each water sample was acidified with 5 mL of nitric acid (69-72%), and the mixture was digested in a fume cupboard for one hour at 100 °C until a clear solution was seen. After cooling, the mixture was filtered and the filtrate was transferred to a 50 mL volumetric flask and diluted with deionized water until the mark. This digestion was carried out in triplicate manner. Finally, calcium, magnesium, potassium and sodium were determined using microwave plasma-atomic emission spectrophotometer (MP-AES). 0, 5, 10, 15, 20 mg/L standard solution of Mg²⁺, Ca²⁺, Na⁺ were prepared from the stock solution, similarly 0, 5, 10, 20, 40 mg/L of K⁺ standard solution was prepared from potassium stock solution (calibration standard, Buck Scientific, USA), by using 2% HNO₃. Using these standard solutions, the calibration curves for each metal were obtained. The correlation coefficients of the graphs were found to be higher than 0.999 for all the analytes. Immediately after calibration, the sample solutions were aspirated into the MP-AES instrument and the metal concentrations were recorded. Three replicate determinations were carried out for each sample. The same analytical procedures were employed for the determination of the metals in twelve blank samples.

Assessment of water quality index (WQI) of bottled water samples

The water quality index was calculated by using the standards procedure of drinking water quality guidelines set by the WHO. The weight arithmetic method was used for calculation of water quality index of the water system [45]. The WQI was calculated for the 15 physicochemical parameters (EC, TDS, pH, total alkalinity, total hardness, fluoride, chloride, nitrate, nitrite, sulfate, Ca^{+2} , Mg^{+2} , K^+ , and Na^+). The computed WQI values are classified into five categories as: excellent, good, poor, very poor and unstable water quality [26–31] (Table 2). Thus, water quality parameters and quality rating (qi) corresponding to n^{th} term parameter is a number reflecting the relative value of this parameter in the polluted water with respect to its standard permissible limit value [13, 20, 29, 31, 32].

$$qi = \frac{100 (Vn-Vi)}{(Vs-Vi)}$$
 (5)

where, Vs = standard value; Vi = ideal value. In most cases Vi = 0 except in certain parameters like pH, the calculation of quality rating for pH (Vi was not zero). q pH = 100 (V pH - 7.0) / (8.5-7.0).

Calculation of unit weight. The unit weight (Wi) of various water quality parameters is inversely proportional to the recommended standards for the corresponding parameters.

$$Wi = \frac{1}{si}$$
(6)

where, Wi = unit weight for the nth parameter and Si = standards permissible value of nth parameter. Water quality index (WQI) is calculated using equation 7:

$$WQI = \frac{\Sigma qi Wi}{\Sigma Wi}$$
(7)

Table 2. Water quality index levels, quality status and grading.

Water quality index level	Water quality status	Grading quality
0-25	Excellent water quality	A
26-50	Good water quality	В
51-75	Poor water quality	С
76-100	Very poor water quality	D
>100	Unsuitable for drinking	E

Statistical analysis

The relationships (correlation) among physicochemical parameters were assessed by Pearson correlation methods using statistical software (SPSS Version 21). Besides, the graphical expression was done using Microsoft excel 7 in addition to arrangements of different data sets.

RESULTS AND DISCUSSION

Accuracy and precision of results

Accuracy and precision are probably the most often quoted terms to express the extent of errors in a given analytical results. Analytical results must be evaluated to decide on the best values to report and attempt to establish the probable limits of errors of the values. The analyst is thus concerned with the question of precision (repeatability of results), that is the agreement between the set of results. It can be determined by standard deviation, variance, and coefficient of variance, and mean of measurements. In this study, the standard deviation and mean of the results of triplicate measurements for all the 15 parameters from each sample (n = 20) were used to know the precision of the method.

Method validation for metal analysis

The efficiency of the optimized procedure is checked by various methods. These are certified standard reference solution analyzing and spiking sample with a known concentration of the analyte. In this work, method validation was established by spiking experiments (recovery test). The spiked samples were prepared by adding, 0.68, 0.43, 0.22 and 0.79 mg/L standard metal solution to 50 mL of bottled water. The spiked and unspiked water samples were digested and analyzed in similar manner. The percentage recovery of the analyte was calculated by using equation 8.

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Recovery
$$\% = \frac{\text{Conc. of metal in the spiked sample - Conc. of metal unspiked samples}}{\text{Conc. of metal added for spiking}} x 100$$
 (8)

The results of percentage recoveries for the studied metals in bottled water samples were within the acceptable range 94.1-103%. Therefore, this verifies that the optimized digestion procedure and the measurement procedure were valid for the analysis of bottled water.

Levels of physicochemical parameters

Total dissolved solid (TDS). The concentration of total dissolved solid in the studied bottled water samples ranged 4.67-139 mg/L. The WHO [15] recommended TDS value below 500 mg/L as acceptable for drinking water, while the Ethiopian Standard extends the limit to 1000 mg/L [46]. When compared with other bottled waters investigated, the concentration of total dissolved solid was higher in Bw₁₅, Bw₄, Bw₁₉, Bw₉, and Bw₁₇ bottled waters than the other brand types (Table 3). Moreover, the concentration of total dissolved solid reported on the label of each of the bottled water particularly in Bw₁₈, Bw₁₄, Bw₇, Bw₅, Bw₁, Bw₁₂, Bw₁₇ and Bw₄ was much higher than the finding of the present study. The present study was in contrast with the finding of Jafari *et al.* [47] who reported the highest value of TDS (326-461 mg/L) while it remains in a range between 4.67-139 mg/L in the present study. In addition, Momani [21] also reported a higher value of TDS in a bottled water with a minimum value of 112 mg/L, whereas the lowest concentration in the present study was 4.67 mg/L. All the bottled water samples investigated in this study are safe to drink from the total dissolved solid point of view.

Table 3. The value of physicochemical parameters in the investigated bottled water (mean \pm SD).

Sample	TDS (mg/L)	pН	EC (µs/cm)	TH (mg/L)	TA (mg/L)
Bw_1	87.3±0.58	7.69±0.03	186±0.90	46.0±2.00	93.3±2.31
Bw_2	19.0±0.10	6.97±0.02	41.1±0.85	14.7±2.31	25.3±2.31
Bw ₃	66.3±1.15	7.03±0.25	139±1.27	20.0±2.00	78.7±2.31
Bw_4	123±0.58	7.11±0.01	256±2.65	97.3±2.31	82.7±4.62
Bw ₅	76.3±0.58	7.22±0.11	161±0.47	54.7±2.31	84.0±4.00
Bw_6	28.7±0.58	7.37±0.06	60.6±0.70	9.33±2.31	36.0±1.00
Bw ₇	66.0±0.60	7.35±0.05	139±0.23	60.0±2.00	85.3±2.31
Bw_8	7.00±0.10	7.20±0.09	15.1±0.17	8.00±0.20	12.0±0.30
Bw ₉	107±1.50	7.48±0.08	225±1.00	96.0±1.00	127±2.31
Bw_{10}	34.7±0.58	7.30±0.27	72.1±0.99	24.0±0.40	45.3±2.31
Bw11	31.0±0.70	6.94±0.04	65.9±0.53	26.7±1.15	33.3±2.31
Bw ₁₂	38.0±0.40	7.04±0.07	80.0±0.45	32.0±0.60	44.0±0.70
Bw13	46.0±0.80	7.60±0.05	96.3±0.06	16.0±0.20	57.3±2.31
Bw ₁₄	80.7±0.58	7.36±0.47	170±0.86	42.7±2.31	96.0±4.00
Bw15	139±1.53	7.29±0.02	289±2.51	44.0±4.00	165±2.31
Bw_{16}	18.0±0.60	6.43±0.15	38.9±0.32	12.0±0.10	27.3±4.16
Bw ₁₇	102±1.00	7.56±0.32	215±2.08	74.7±2.31	123±2.31
Bw ₁₈	37.3±0.58	7.03±0.12	78.7±0.78	17.3±5.03	41.3±2.31
Bw_{19}	127±1.53	7.55±0.05	266±3.00	46.7±2.31	143±2.31
Bw ₂₀	4.67±0.58	6.52±0.21	9.87±0.90	4.00±0.05	13.3±2.31

Electrical conductivity (EC). The highest and the lowest value of electrical conductivity recorded in the present study were 9.8 and 289 μ s/cm in Bw₂₀ and Bw₁₅ bottled water, respectively (Table 3). According to WHO [15] the maximum permissible limit of electrical conductivity in drinking water is 1000 μ s/cm. The electrical conductivity found in the present study is much lower as compared with the permissible limit set by WHO [15]. Jafari *et al.* [47] have reported electrical conductivity values ranged from 706 to 976 μ s/cm in drinking water which are higher than the finding of the present study. However, the finding of the present study

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is a similar with the finding of Reda [36] who reported the electrical conductivity value of drinking water ranged between 210 to 231 μ s/cm in Arba Minch. The investigated parameters of all the bottled mineral waters lie in an acceptable limit for drinking purpose as recommended by WHO [14].

pH. The pH of the 20 brands of bottled water ranged 6.43-7.69 where the lowest and the highest value was recorded for Bw_{16} and Bw_1 bottled mineral water, respectively (Table 3). According to the recommendation of WHO [15] and CES [46], the permissible range of pH in a drinking water should be in the range 6.5-8.5. In this regard, the pH value of Bw_{16} bottled mineral water is lower than from the minimum range drinking water set by WHO [15] and CES [46]. All the remaining 19 bottled mineral water have a pH value lies within the range recommended by WHO [15]. The finding of the present study is in agreement with the finding by Momani [21]. Similarly in Italy, Sasikaran *et al.* [22] have reported the pH value of bottled water above the middle of the permissible limit. At pH values less than 6.5, water is corrosive and dissolves plumbing components. This is especially a concern when water contacts brass and copper piping where copper, zinc, and lead dissolve into the drinking water. Whereas pH values ≥ 8.5 of drinking water can promote hardness scale, precipitation and make chlorine disinfectants more effective.

Total hardness. The total hardness value in bottled water samples were ranged from 4 mg/L to 97 mg/L as calcium carbonate in Bw_{20} and Bw_{4} , respectively (Table 3). The values of total hardness in all of the bottled water lie in permissible limits for drinking water (<500 mg/L) set by WHO [14]. Although for all the bottled water samples the total hardness is below the recommendations of WHO [14], there is no labeled values on all the bottled water measured in this study (except Bw_7 bottled water was 13.3 mg/L). In this study, the concentration of total hardness in Bw_7 bottled water was 60 mg/L as calcium carbonate which is higher than from the labeled values on it.

Total alkalinity. The mean concentration of total alkalinity ranged from 12 to 165 mg/L in Bw₈ and Bw₁₅, respectively (Table 3). The maximum concentration limit of total alkalinity described by WHO [15] and CES [46] is 200 mg/L. In this study the value of total alkalinity labeled at the bottled drinking water was found below the permissible limit. The total alkalinity labeled at the bottled drinking water were 0.69, 7, 8.33, 85, 104 mg/L in Bw₁₀, Bw₁₆, Bw₈, Bw₁₉, Bw₁₇, respectively, but other bottled water were not labeled the concentration of total alkalinity. The minimum value of total alkalinity in this study was recorded in the Bw₈ bottled water which is higher than the label at bottled water. On the other hand, the maximum value of total alkalinity of this study was 165 mg/L in Bw₁₅ bottled drinking water which is higher than the label at the Bw₁₅ bottled water. The concentration of alkalinity reported by Reimann *et al.* [37] range from 15 to 1070 mg/L in drinking water in Ethiopian Rift Valley where the maximum of the range was higher than WHO permissible limit [15]. The reported result by Reda [36] range from 88.6-181 mg/L thus the maximum and minimum range is above the values found in this study.

Levels of common cations and anions in the water samples

Calcium. The mean calcium concentration was found in the range 0.19-8.9 mg/L, and the results are summarized in Table 4. The highest concentration of calcium was found in Bw₉ bottled water samples while the lowest values were found in Bw₈. The mean calcium value found in Bw₁, Bw₃ and Bw₄ are lower than to the values reported by Seda *et al.* [38]. All the bottled water samples are below WHO permissible limit (200 mg/L) that confirms all bottled water are safe to drink. According to study conducted by Mihayo and Mkoma [23], maximum and the minimum levels of calcium were 14 mg/L and 3.4 mg/L, respectively, in which the concentration of calcium was higher than in the present study. Similarly, Mekonnen *et al.* [18]

have reported the mean concentrations of calcium range from 3 to 28.3 mg/L, this concentration of calcium is relatively higher than that found in the present study.

Table 4. The concentration of common cations (mg/L) in the bottled mineral waters investigated (mean \pm SD).

Sample	Calcium	Magnesium	Potassium	Sodium
Bw_1	4.30±0.41	1.11±0.07	1.93±0.29	20.2±3.65
Bw ₂	1.18±0.08	0.42±0.04	0.59±0.03	1.80±0.10
Bw ₃	1.54±0.37	0.45±0.03	2.14±0.23	19.7±2.10
Bw_4	7.93±0.84	2.30±0.19	2.05±0.19	7.39±0.53
Bw ₅	4.65±0.63	0.67±0.57	0.46±0.20	9.55±1.86
Bw ₆	0.34±0.01	0.55±0.41	0.38±0.07	9.51±0.54
Bw ₇	3.40±0.13	2.15±0.02	1.11±0.01	3.91±0.12
Bw ₈	0.19±0.03	0.02±0.01	0.14±0.03	4.20±0.91
Bw9	8.09±0.37	2.51±0.09	1.28±0.16	9.16±0.45
Bw ₁₀	1.72±0.16	0.56±0.05	1.43±0.13	4.68±0.48
Bw11	1.85±0.13	0.59±0.04	1.99±0.10	3.60±0.27
Bw ₁₂	2.41±0.02	0.78±0.06	1.95±0.16	3.74±0.37
Bw13	0.76±0.06	0.35±0.02	0.29±0.03	11.8±0.18
Bw ₁₄	3.34±0.42	1.01±0.10	3.90±0.25	13.2±1.03
Bw ₁₅	2.49±0.51	1.13±0.23	1.51±0.28	24.5±0.33
Bw ₁₆	0.44±0.08	0.10±0.03	0.77±0.08	7.28±0.57
Bw17	6.14±0.27	2.10±0.08	2.10±0.04	14.5±0.24
Bw18	1.26±0.01	0.56±0.02	3.15±0.06	6.87±0.23
Bw ₁₉	2.03±1.97	1.48±0.04	8.08±0.23	24.1±1.47
Bw ₂₀	0.26±0.01	0.02±0.01	0.21±0.04	2.45±0.01

Magnesium. The mean magnesium concentration in the bottled water sample ranged from 0.02 to 2.5 mg/L (Table 4). The lowest concentration of magnesium was found in Bw_{20} and Bw_{8} while the highest concentration of magnesium found in Bw_{9} . According to investigation by Mekonnen *et al.* [18] in different bottled waters in Gonder city, the mean concentration of magnesium ranged from 0.32 mg/L to 6.5 mg/L. The mean concentration of magnesium is relatively higher than this study. The mean concentration of magnesium ranged from 0.08-12 mg/L in Bw_{15} and Bw_{4} , respectively and also Bw_{13} , Bw_{16} , Bw_{18} almost similar concentration of magnesium in this study but the other bottled water samples were different compared to labeled bottled water. As it was reported by Seda *et al.* [38] the mean concentration of magnesium in Bw_{3} was 0.45 mg/L. Therefore, the concentration of Aquaddis bottled water is higher than this study.

Sodium. In this study, sodium concentration in the bottled water samples ranged from 1.8 to 24.5 mg/L in Bw_2 and Bw_{15} , respectively (Table 4). The concentration of sodium in Bw_1 , Bw_{15} , and Bw_{19} were above the WHO [15] permissible limit. The concentrations of Bw_1 , Bw_{15} and Bw_{19} were 20.2 mg/L, 24.5 mg/L, and 24.1 mg/L, respectively, whereas the recommendation of WHO is below 20 mg/L in drinking water so that Bw_1 , Bw_{15} and Bw_{19} are unfit for drinking purpose with respect to the concentration of sodium. To compare the labels of sodium in the bottled water samples maximum concentration was 24.5 mg/L in Bw_{15} and the minimum concentration were 0.05 mg/L in Bw_8 . The levels of sodium labeled on the bottled water Bw_{14} and Bw_{18} are almost similar with that found in this study but the levels of sodium labeled on the other bottled water are relatively highly different. On the other hand, Seda *et al.* [38] reported the mean concentration of sodium in Abyssinia, Aquaddis, and Aquasafe as 18.3 mg/L, 40.3 mg/L, and 23.2 mg/L, respectively. These reported concentrations of sodium in Aquaddis and

Aquasafe were higher than the concentration of sodium found in this study and are also above the WHO recommendation while in this study, the sodium levels in the Bw_3 and Bw_4 bottled drinking water are below the recommendation of WHO [15]. The results are summarized in Table 4.

Potassium. The mean concentration of potassium ranged 0.14 to 8.08 mg/L in Bw_8 and Bw_{18} , respectively (Table 4). The permissible limit of potassium is 10 mg/L and 1.5 mg/L, described by WHO [48] and CES [46], respectively. The concentrations of potassium in this study are below WHO permissible limit in some of the bottled water; however, it is above the limit set by CES. The concentrations of potassium at the labeled bottled water ranged 0.7 to 6.11 mg/L in Bw_1 and Bw_2 . In this study, Bw_1 and Bw_2 values are 1.93 mg/L and 0.59 mg/L, which are lower than the labeled at bottled water, respectively. Mekonnen *et al.* [18] have reported the concentration range of potassium from 1.2 to 4.1 mg/L. The minimum value of the bottled water is higher than the walue of this study.

Chloride. Chloride content of 20 brands of bottles water ranged from 4.28 to 20.8 mg/L. The lowest mean concentration of chloride was found in Bw_2 while the highest mean concentration of chloride was found in Bw_4 bottled water (Table 5), whereas the WHO [15] and CES [46] prescribed limit is 250 mg/L in drinking water. Thus all the bottled water samples are safe for drinking from the chloride point of view. To compare the concentration of chloride labeled in bottled waters relatively Bw_2 and Bw_4 have high chloride content than that of other brands, respectively. The chloride concentrations of bottled waters compared to the labeled of the company at the bottled water were ranged from 0.26-13 mg/L in Bw_8 and Bw_4 , respectively. The chloride concentration reported by Reda [36] ranged from 135 to 554 mg/L in three different samples, whereas the concentration of chloride in this study is lower than the reported value Reda [36].

Table 5. The concentration of common anions (mg/L) in the bottled water samples investigated (mean \pm SD).

Sample	Bicarbonate	Chloride	Fluoride	Nitrate	Nitrite	Sulfate
Bw_1	114±2.81	11.8±1.64	0.62±0.06	0.37±0.01	0.18±0.01	0.66±0.02
Bw ₂	30.9±2.82	4.73±0.82	0.09±0.01	0.06±0.02	0.16±0.03	0.05±0.01
Bw ₃	96.0±2.82	8.99±2.17	1.26±0.07	0.11±0.16	0.18±0.01	0.50±0.03
Bw ₄	101±5.63	20.8±0.82	0.27±0.02	0.84±0.52	0.16±0.01	1.05±0.08
Bw ₅	103±4.88	6.63±0.81	0.16±0.01	0.59±0.03	0.15±0.01	0.07±0.02
Bw ₆	43.9±1.00	8.05±2.96	0.09±0.01	0.04±0.01	0.17±0.02	0.41±0.05
Bw ₇	104±2.82	4.73±2.16	0.18±0.01	0.10±0.01	0.16±0.01	0.59±0.05
Bw ₈	14.6±0.80	5.21±1.64	0.03±0.01	0.30±0.03	0.16±0.14	1.73±0.64
Bw ₉	155±2.82	8.05±0.82	0.69±0.02	0.02±0.01	0.14±0.01	0.05±0.01
Bw ₁₀	55.3±2.81	5.68±2.45	0.24±0.01	0.04±0.01	0.15±0.01	0.14±0.05
Bw11	40.7±2.82	9.47±2.17	0.14±0.00	0.07 ± 0.08	0.08 ± 0.04	1.17±0.07
Bw ₁₂	53.7±0.70	6.63±0.82	0.44 ± 0.41	0.12±0.02	0.14 ± 0.01	3.84±0.02
Bw ₁₃	69.9±2.82	6.15±2.96	0.33±0.03	0.20±0.09	0.18±0.01	0.76±0.07
Bw ₁₄	117±4.88	7.57±0.82	0.18±0.01	0.10±0.04	0.16±0.01	0.50±0.05
Bw15	202±2.82	7.57±3.57	0.59±0.02	0.49±0.03	0.17±0.01	0.36±0.08
Bw ₁₆	33.3±5.08	4.26±2.46	0.05±0.01	0.13±0.04	0.18±0.01	0.44±0.03
Bw ₁₇	149±2.82	9.94±2.46	0.31±0.01	0.44±0.03	0.15±0.04	0.12±0.03
Bw ₁₈	50.4±2.82	5.68±2.46	0.31±0.02	0.28±0.07	0.28±0.01	1.76±0.26
Bw ₁₉	174±2.82	13.3±0.82	0.55±0.03	0.03±0.01	0.28 ± 0.07	0.39±0.03
Bw ₂₀	16.3±2.82	4.26±0.20	0.05±0.01	0.05±0.01	0.18±0.01	0.05±0.01

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Fluoride. The fluoride concentration of the 20 brands bottled water ranged 0.03-1.26 mg/L in Bw8 and Bw3, respectively (Table 5). The maximum concentration of fluoride recommended by WHO [15] and CES [46] is 1.5 mg/L. Therefore fluoride levels in all the bottled water samples are below the permissible limit. Fluoride concentration below 1 mg/L diminishes caries reduction whereas the concentration of fluoride in all the bottled water samples were below 1 mg/L except Bw₃ bottled water with 1.26 mg/L. The labeled bottled water fluoride content were 0.04, 0.1, 0.13, 0.27, 0.3, 0.91 mg/L in Bw₈, Bw₂, Bw₁₀, Bw₁, Bw₁₈, and Bw₁₉, respectively, while other bottled water did not mentioned the concentration of fluoride. The concentration of fluoride in Bw₁₉ packaged drinking water is higher than that of this study and concentration of fluoride in Bw1 brand is lower than that of this study whereas remaining bottled waters have almost similar fluoride concentration. Seda et al. [38] have reported the concentration of fluoride in bottled water in the range of 0.70 to 2.25 mg/L, in that study the concentration of fluoride was above the permissible limit in Aquaddis brand water. Mekonnen et al. [18] have reported the concentration of fluoride in six bottled water which ranged 0.03-0.15 mg/L. Therefore the minimum concentrations of both studies are equal but the maximum concentration found in the present study is higher than the maximum concentration of that study.

Nitrate. The mean concentration of nitrate ranged 0.02 to 0.84 mg/L in Bw_4 and Bw_9 , respectively (Table 5). Nitrate in all the bottled water samples were lower than the WHO recommended value of 50 mg/L [15]. The concentrations of nitrate labeled on the bottled water were (nil, 0.02, 0.03, 0.11, 0.14, 0.22, 0.45 and 3.1, mg/L) in Bw_{17} , Bw_2 , Bw_8 , Bw_{13} , Bw_{19} , Bw_{16} , Bw_{10} , and Bw_1 , respectively, while the other bottled water were not labeled. In this study, the level of nitrate found in the above 8 bottled water were (0.03, 0.04 0.06, 0.13, 0.2, 0.3, 0.37 and 0.44 mg/L) in Bw_{19} , Bw_{10} , Bw_2 , B_{16} , Bw_{13} , Bw_8 , Bw_1 , Bw_{17} , respectively (Table 5). The concentration of nitrate found in this study and nitrate labeled on bottled water are almost similar except Bw_1 and B_{17} of bottled water. The labeled value of Bw_1 bottled water was higher than the present study while the Bw_{17} bottled drinking water was nil but this study found 0.44 mg/L. According to Mekonnen *et al.* [18], analysis of six brands of bottled water, the nitrate levels of the three brands were 0.251 mg/L, 0.389 mg/L, 0.33 mg/L in brand 1, brand 2, brand 3, respectively, while in the other three brands nitrate was not detected but in this study nitrate was found in all the bottled waters. Nitrate content in Bw_1 and brand 2 mineral water are almost similar.

Nitrite. The mean concentration of nitrite was in the range 0.08-0.28 mg/L (Table 5). The highest mean concentration of nitrite was found in Bw₁₈ and Bw₁₉ whereas the lowest mean concentration of nitrite was found in Bw₁₁. The nitrite concentrations were not labeled on bottled water but this study observed the concentration of nitrite in bottled water. The recommendations of WHO [15] and CES [46] are below 3 mg/L nitrite. Thus the concentration of nitrite in bottled water is safe for drinking purpose. Simeonova *et al.* [49] have reported the concentration of nitrite in drinking water in the range 0.01-1.56 mg/L. The maximum concentration of nitrite in drinking water is higher than the present study of bottled drinking water. However, the minimum concentrations of those studies were 0.01 mg/L which is smaller than the value of this study.

Bicarbonate. The mean concentration of bicarbonate ranged 16.3 to 202 mg/L in Bw_{20} and Bw_{15} , respectively (Table 5). The maximum concentration limit of bicarbonate set by WHO is 500 mg/L [15]. The concentration of bicarbonate found in this study is acceptable for drinking purpose. Rakotondrabe *et al.* [25] have reported the concentration of bicarbonate ranged from 1.77 to 59.3 mg/L which are much lower than the concentration of bicarbonate found in drinking water of this study. This study has found a higher concentration of bicarbonate in bottled drinking water compared with the labeled on the bottled water.

Sulfate. The mean concentrations of sulfate ranged 0.05-3.84 mg/L in Bw_{20} , Bw_9 and Bw_2 (Table 5). The highest mean concentration of sulfate was found in Bw_2 while the lowest mean concentration was found in Bw_{20} and Bw_9 (Table 5). The WHO [15] and CES [46] have recommended sulfate concentration below 250 mg/L in drinking water. Thus the sulfate levels in all the bottled drinking water is below the permissible limit and the bottled water samples are safe for drinking purpose with respect to sulfate. Mihayo and Mkoma [23] have reported a maximum concentration of sulfate, 48.2 mg/L and the minimum concentration of sulfate, 2.5 mg/L; these concentrations of sulfate are higher than this study.

Correlation

Pearson correlation [50] for various water quality parameters in the bottled water brands studied in this work was evaluated which revealed strong correlations for most of the constituents suggesting that they may originate predominantly from the same source. Higher correlations (r^2 > 0.80) were observed in TDS with alkalinity, bicarbonate and hardness. Whereas EC and TDS had very high correlation ($r^2 = 1$). In addition there was also a high correlation of EC with total alkalinity, total hardness, bicarbonate and total hardness ($r^2 > 90$). EC has also correlation with calcium, magnesium, sodium and chloride. Total hardness had higher correlation with magnesium and calcium and also correlated with total alkalinity and bicarbonate. Total alkalinity was highly correlated with bicarbonate $(r^2 = 1)$ and also correlated with sodium, magnesium and calcium. pH correlated with electrical conductivity, total alkalinity, bicarbonate and also bicarbonate correlated with magnesium and calcium. Fluoride correlated with sodium on the other hand calcium highly correlated with magnesium and bicarbonate with total alkalinity were highly correlated indicates the concentration of total alkalinity described by bicarbonate. TDS have showed high positive correlation to all the individual parameters and its correlation is significant at 99% confidence level, except nitrite. pH with fluoride, chloride, and potassium pairs positively correlated significantly at 95% confidence level. CEC except nitrite and sulfate was highly positively correlated with most parameter significantly at 99% confidence level. Generally almost all parameters pairs were significantly correlated at 99% and 95% confidence levels.

The comparison of chemical and physical parameters of bottled water samples among each other

Among the 20 brands of bottled water samples, highest pH value was found in Bw_1 and the lowest pH value was found in Bw_{16} . Among the 20 brands of bottled water samples highest electrical conductivity and the highest total dissolved solid were found in Bw_{15} bottled water sample. Whereas electrical conductivity and total dissolved solid were lowest in Bw_8 and Bw_{20} bottled water samples, respectively. In addition, Bw_3 and Bw_{16} have almost similar electrical conductivity and total dissolved solid water samples was higher than the other bottled water samples and also the concentration of bicarbonate in Bw_{15} was higher than all the other bottled water samples. The total hardness in Bw_4 was higher than the other bottled water samples. The total hardness, alkalinity and bicarbonate concentration in Bw_8 and Bw_{20} bottled water samples were lower than the other bottled water samples.

The concentration of nitrate in Bw_4 and Bw_5 of bottled water was higher values compared to other bottled water samples. The highest concentration of sulfate was found in Bw_{12} bottled drinking water whereas the highest concentration of nitrite was found in Bw_{18} and Bw_{19} compared to other bottled water. The lowest concentration of sulfate was found in Bw_2 , Bw_5 , Bw_9 and Bw_{20} bottled drinking water. Concentration of fluoride in Bw_3 bottled water was higher and the lowest concentration of fluoride was found in Bw_{20} bottled water samples. Concentration of chloride in Bw_4 bottled water had higher value and the lowest

concentration of chloride was found in Bw_2 , Bw_8 , Bw_{16} and Bw_{20} bottled water samples. The concentrations of sodium in Bw_{15} and Bw_{19} bottled water were higher than the other bottled drinking water. The lowest concentration of sodium was found in Bw_2 and Bw_{20} bottled water. The concentration of magnesium was higher in Bw_4 and Bw_9 and the lowest concentration of magnesium was in Bw_8 , Bw_{16} and Bw_{20} when compared to all the bottled water samples. The concentration of potassium was higher in Bw_{19} while the lowest concentration was found in Bw_8 , Bw_{16} and Bw_{20} of bottled water.

Calculation of water quality index of bottled drinking water

The bottled drinking water can be classified into five quality levels (excellent, good, poor, very poor and unstable) according to the water quality index [26–31]. The water quality index (WQI) was calculated by using the standards of the drinking water quality recommended by the WHO. The water quality index was calculated based weighted arithmetic method using various physicochemical parameters in different bottled water [45]. The results of water quality index in this study were found in the range of 3.85-49.2. Thus, all the bottled water samples analyzed in this study were grouped in either excellent or good water quality level. The details of calculation of water quality index are given in Table 6. Figure 1 shows the water quality index of the studied bottles water. Among the 20 brands of bottled waters studied, 75% of bottled water samples fall in excellent water quality while 25% of bottled water lies within good water quality.

Table 6. Calculation of the water quality index of bottled water samples.

Samples	Bw_1	Bw ₂	Bw ₃	Bw ₄	Bw ₅	Bw ₆	Bw ₇	Bw ₈	Bw9	Bw ₁₀
ΣWi	1.33	1.33	1.33	1.33	1.33	1.33	1.33	1.33	1.33	1.33
ΣqiWi	43.1	7.19	65.6	19.1	12.0	9.64	12.2	4.39	36.4	15.1
$WQI = \Sigma qiWi / \Sigma Wi$	32.3	5.39	49.2	14.3	9.00	7.22	9.18	3.29	27.3	11.3
Grade	В	Α	В	Α	Α	Α	Α	Α	В	Α
Samples	Bw11	Bw ₁₂	Bw ₁₃	Bw_{14}	Bw ₁₅	Bw ₁₆	Bw17	Bw18	Bw19	Bw ₂₀
ΣWi	1.33	1.33	1.33	1.33	1.33	1.33	1.33	1.33	1.33	1.33
ΣqiWi	10.2	24.2	20.2	17.4	36.4	6.97	21.7	21.9	42.3	5.14
$WQI = \Sigma qiWi / \Sigma Wi$	7.61	18.1	15.1	13.0	27.3	5.2	16.3	16.4	31.7	3.85



Figure 1. The water quality index of bottled water samples.

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Comparison of the results of physicochemical parameters with the labeled values on the bottled water samples

Table 7. Comparison of physicochemical parameters (mg/L) found in this study with labeled values of the bottled water samples (EC in µs/cm).

	T	DS		pН		EC	T	Ή	T.	A	HC	CO3 ⁻	C	1-	F	-
Sample	М	L	N	1 I		М	М	L	M	L	М	L	М	L	Μ	L
Bw_1	87.3	149	7.6	59 7.	4	186	46.0	-	93.3	-	114	134	11.8	7.50	0.62	0.27
Bw_2	19.0	21.4	6.9	97 7.	00 4	41.1	14.7	-	25.3	-	30.9	6.00	4.73	1.99	0.09	0.10
Bw ₃	66.3	82.0	7.0	03 7.	00	139	20.0	-	78.7	-	96.0	73.2	8.99	7.60	1.26	-
Bw_4	123	170	7.1	11 7.1	20 2	256	97.3	-	82.7	-	101	84.0	20.8	13.0	0.27	-
Bw ₅	76.3	144	7.2	22 7.1	24	161	54.7	-	84.0	-	103	64.8	6.63	6.38	0.16	-
Bw_6	28.7	20.3	7.3	37 7.	00 6	60.6	9.33	-	360	-	43.9	3.01	8.05	6.07	0.09	-
Bw_7	66.0	135	7.3	35 7.	50	139	60.0	13.8	85.3	-	104		4.73	5.60	0.18	-
Bw ₈	7.00	8.43	7.2	20 7.	00 1	15.1	8.00	-	12.0	8.33	14.6	8.41	5.21	0.26	0.03	0.04
Bw ₉	107	85.0	7.4	48 7.	32 2	225	96.0	-	127	-	155	146	8.05	6.03	0.69	-
Bw_{10}	34.7	54.5	7.3	30 7.	92 7	72.1	24.0	-	45.0	0.69	55.0	-	5.68	1.7.0	0.24	0.13
Bw_{11}	31.0	70.0	6.9	94 7.	10 6	65.9	26.7	-	33.3	30	40.7	60.5	9.47	4.00	0.14	0.50
Bw_{12}	38.0	155	7.0	04 7.	8 00	30.0	32.0	-	44.0	-	53.7	-	6.63	-	0.44	-
Bw ₁₃	46.0	50.0	7.6	50 7.4	40 9	96.3	16.0	-	57.0	-	69.9	-	6.15	4.82	0.33	-
Bw ₁₄	80.7	134	7.3	36 7.	18	170	42.7	-	96.0	-	117	-	7.57	5.67	0.18	-
Bw ₁₅	139	33.0	7.2	29 7.	10 2	289	44.0	-	165	-	202	25.9	7.57	2.6	0.59	-
Bw ₁₆	18.0	30.0	6.4	43 7.	00 3	38.9	12.0	-	27.3	7	33.3	11.2	4.26	5.53	0.05	-
Bw17	102	155	7.5	56 7.	00 2	215	74.7	-	123	104	149	126	9.94	6.27	0.31	0.30
Bw_{18}	37.3	130	7.0	03 7.	00 7	78.7	17.3	-	41.3	-	50.4	12.1	5.68	10.64	0.31	0
Bw ₁₉	127	65.0	7.5	55 7.)5 2	266	46.7	-	143	85	174		13.3	4.50	0.55	0.91
Bw ₂₀	4.67	10.0	6.5	52 7.	00 9	9.87	4.00	-	13.3	-	16.3	1.00	4.26	1.4	0.05	-
		NO ₃ -		NO ₂ ⁻		SO_4	2-	Ca	a ²⁺	Μ	(g ²⁺		K^+		Na^+	
Sample	e M	L		М	Ν	1	L	М	L	М	L	М	L	Μ		L
Bw_1	0.3	7 3.1	0	0.18	0.6	66	-	4.30	12.0	1.11	3.40	1.93	0.70	20.2	: 5	.40
Bw_2	0.0	6 0.0	2	0.16	0.0	05	4.40	1.18	3.21	0.42	1.95	0.59	6.11	1.80) 3	.30
Bw ₃	0.1	1 -		0.18	0.5	50	-	1.54	3.20	0.45	1.95	2.14	1.65	19.7	1	.08
Bw_4	0.8	4 -		0.16	1.0	05	4.00	7.93	21.0	2.30	12.0	2.05	3.60	7.39	1	6.0
Bw ₅	0.5	9 -		0.15	0.0	07	-	4.65	15.4	0.67	1.92	0.46	0.68	9.55	8	.47
Bw ₆	0.0	4 -		0.17	0.4	41	-	0.34	0.20	0.55	0.10	0.38	1.05	9.51	0	.15
Bw_7	0.1	- 0		0.16	0.5	59	-	3.40	5.10	2.15	0.94	1.11	2.42	3.91	1	2.7
Bw_8	0.3	0 0.0	3	0.16	1.7	73	0.30	0.19	1.14	0.02	0.32	0.14	1.94	4.20) 0	.05
Bwo	0.0	2 -		0.14	0 (05	-	8 0 9	5 76	2.51	2.68	1.28	0.84	9 16	3	80

Bw_4	0.84	-	0.16	1.05	4.00	7.93	21.0	2.30	12.0	2.05	3.60	7.39	16.0
Bw ₅	0.59	-	0.15	0.07	-	4.65	15.4	0.67	1.92	0.46	0.68	9.55	8.47
Bw ₆	0.04	-	0.17	0.41	-	0.34	0.20	0.55	0.10	0.38	1.05	9.51	0.15
Bw ₇	0.10	-	0.16	0.59	-	3.40	5.10	2.15	0.94	1.11	2.42	3.91	12.7
Bw ₈	0.30	0.03	0.16	1.73	0.30	0.19	1.14	0.02	0.32	0.14	1.94	4.20	0.05
Bw9	0.02	-	0.14	0.05	-	8.09	5.76	2.51	2.68	1.28	0.84	9.16	3.80
Bw_{10}	0.04	0.45	0.15	0.14	0.10	1.72	5.74	0.56	1.39	1.43	0.39	4.68	2.60
Bw ₁₁	0.07	1.00	0.08	1.17	3.00	1.85	10.5	0.59	5.00	1.99	1.20	3.60	4.80
Bw ₁₂	0.12	-	0.14	3.84	-	2.41	17.5	0.78	4.60	1.95	3.60	3.74	7.20
Bw ₁₃	0.20	0.11	0.18	0.76	0	0.76	1.92	0.35	0.67	0.29	0.37	11.8	3.00
Bw ₁₄	0.10	-	0.16	0.50	-	3.34	1.50	1.01	0.40	3.90	3.85	13.2	12.6
Bw ₁₅	0.49	-	0.17	0.36	-	2.49	0.11	1.13	0.08	1.51	0.20	24.5	9.40
Bw ₁₆	0.13	0.22	0.18	0.44	-	0.44	0.21	0.10	0.14	0.77	0.28	7.28	2.70
Bw ₁₇	0.44	Nil	0.15	0.12	1.50	6.14	26.8	2.10	7.20	2.10	3.30	14.5	7.70
Bw ₁₈	0.28	-	0.28	1.76	-	1.26	3.15	0.56	0.80	3.15	2.00	6.87	3.65
Bw ₁₉	0.03	0.14	0.28	0.39	-	2.03	18.1	1.48	10.0	8.08	4.00	24.1	24.0
Bw ₂₀	0.05	-	0.18	0.05	1.20	0.26	1.20	0.02	0.80	0.21	0.70	2.45	1.20
				-						-			

M and L indicate measured in this study and labeled at bottled water, respectively.

The results of physicochemical parameters of the bottled water samples were compared with the values labeled in the bottles (Table 7). Some of the parameters such as electrical conductivity, total hardness, total alkanity and fluoride are either not labeled at all or labeled only in few

brands of bottled water. There is a wide variation in the values of physicochemical parameters of the bottled water samples between the measured values and labeled values. These variations clearly emphasize that there is need of regulation about the monitoring of labeled values.

Comparison of the physicochemical parameters of the bottled water samples of this study with literature values

The results of physicochemical parameters of the bottled water samples were compared with the permissible levels recommended by the guidelines for drinking water quality (Table 8). The comparison of this study with WHO [15] and Ethiopian standards [46] showed that the concentrations of physicochemical parameters in almost all the bottled drinking water are below the permissible limit except sodium, potassium, and pH.

Table 8. Comparison of physicochemical parameters (mg/L) of the present study with WHO and CES recommended values (EC in µs/cm).

Sample	TDS	pН	EC	TH	TA	HCO ₃ -	Cl-	F-	NO ₃ ⁻	NO ₂ ⁻	SO_4^{2-}	Ca ²⁺	Mg^{2+}	K^+	Na^+
Bw_1	87.3	7.69	186	46.0	93.3	114.	11.8	0.62	0.37	0.18	0.66	4.30	1.11	1.93	20.2
Bw_2	19.0	6.97	41.1	14.7	25.3	30.9	4.73	0.09	0.06	0.16	0.05	1.18	0.42	0.59	1.80
Bw ₃	66.3	7.03	139	20.0	78.7	96.0	8.99	1.26	0.11	0.18	0.50	1.54	0.45	2.14	19.7
Bw_4	123	7.11	256	97.3	82.7	101	20.8	0.27	0.84	0.16	1.05	7.93	2.30	2.05	7.39
Bw ₅	76.3	7.22	161	54.7	84.0	103	6.63	0.16	0.59	0.15	0.07	4.65	0.67	0.46	9.55
Bw_6	28.7	7.37	60.6	9.33	360	43.9	8.05	0.09	.04	0.17	0.41	0.34	0.55	0.38	9.51
Bw ₇	66.0	7.35	139	60.0	85.3	104	4.73	0.18	0.10	0.16	0.59	3.40	2.15	1.11	3.91
Bw_8	7.00	7.20	15.1	8.00	12.0	14.6	5.21	0.03	0.30	0.16	1.73	0.19	0.02	0.14	4.20
Bw ₉	107	7.48	225	96.0	127	155	8.05	0.69	0.02	0.14	0.05	8.09	2.51	1.28	9.16
Bw_{10}	34.7	7.30	72.1	24.0	45.0	55.0	5.68	0.24	0.04	0.15	0.14	1.72	0.56	1.43	4.68
Bw_{11}	31.0	6.94	65.9	26.7	33.3	40.7	9.47	0.14	0.07	0.08	1.17	1.85	0.59	1.99	3.60
Bw ₁₂	38.0	7.04	80.0	32.0	44.0	53.7	6.63	0.44	0.12	0.14	3.84	2.41	0.78	1.95	3.74
Bw ₁₃	46.0	7.60	96.3	16.0	57.0	69.9	6.15	0.33	0.20	0.18	0.76	0.76	0.35	0.29	11.8
Bw ₁₄	80.7	7.36	170	42.7	96.0	117	7.57	0.18	0.10	0.16	0.50	3.34	1.01	3.90	13.2
Bw ₁₅	139	7.29	289	44.0	165	202	7.57	0.59	0.49	0.17	0.36	2.49	1.13	1.51	24.5
Bw ₁₆	18.0	6.43	38.9	12.0	27.3	33.3	4.26	0.05	0.13	0.18	0.44	0.44	0.10	0.77	7.28
Bw_{17}	102	7.56	215	74.7	123	149	9.94	0.31	0.44	0.15	0.12	6.14	2.10	2.10	14.5
Bw ₁₈	37.3	7.03	78.7	17.3	41.3	50.4	5.68	0.31	0.28	0.28	1.76	1.26	0.56	3.15	6.87
Bw ₁₉	127	7.55	266	46.7	143	174.	13.3	0.55	0.03	0.28	0.39	2.03	1.48	8.08	24.1
Bw ₂₀	4.67	6.52	9.87	4.00	13.3	16.3	4.26	0.05	0.05	0.18	0.05	0.26	0.02	0.21	2.45
Mean	61.9	7.20	130	37.3	70.6	86.2	7.98	0.33	0.22	0.18	0.73	2.71	0.94	1.77	10.1
Max	139	7.69	289	97.0	116	202	20.8	1.26	0.84	0.28	3.84	8.09	2.51	8.08	24.5
Min	4.67	6.43	9.87	4.00	12.0	16.3	4.26	0.03	0.02	0.08	0.05	0.19	0.02	0.14	1.80
WHO	500	6.5-8.5	1000	500	200	500	250	1.50	50.0	3.00	250	200	150	10.0	20.0
CES	1000	6.5-8.5	-	300	200	-	250	1.50	50.0	3.00	250	75	50	1.50	200

Comparison of the results of a present study with the results reported in the literature is summarized in Table 9. There is a wide variation in the values of physicochemical parameters of the bottled water samples of the present study with that from different countries and with that of other studies in Ethiopia. This is due to the fact that guidelines vary from country to country but also due to the fact that the original source of water is different due to the variation in geographical and climatic conditions. These variations are despite of the fact that every country follows the general guidelines of WHO [15].

	TDS	pН	EC	TH	TA	HCO ₃ ⁻	Cl	F-	Reference
Country	4.67-139	6.43-7.69	9.87-289	4.00-97.0	12.0-116	16.3-202	4.26-20.8	0.031.26.	This study
Ethiopia	-	6.50-8.34	120-1788	-	-	-	3.57-48.1	0.00-0.92	[38]
Ethiopia	-	-	-	-	-	-	0.4-6.3	0.03-0.15	[18]
Tanzania	7.80-126	7.3-7.6	17.4-280	20-66.5	16.0-79.0	-	2.5-48.2	-	[23]
Mutah,	210-978	6.70-8.20	177-1494	3.90-136.9	12.9-289	-	4.6-150	0.01-1.60	[21]
Jordan									
Kirkuk –	90.0-260	7.30-8.00	140-464	45.0-190	44-190	-	5.97-28.8	-	[16]
Iraq									
Saudi	100-253	7.00-8.00	-	15.0-110	-	1.30-120	1.00-68.0	0.00-1.20	[9]
Arabia									
Italy	-	4.10-8.80	18.0 -4090	0.30-105	-	3.5-1875	0.20-323	0.01-7.93	[24]

Table 9. Comparison of the physicochemical parameters (mg/L) of the bottled water samples of this study with literature values (EC in µs/cm).

Country	NO ₃	NO ₂	SO4 ²⁻	Ca ²⁺	Mg ²⁺	K ⁺	Na ⁺	Reference
	0.02-0.84	0.08-028	0.05-3.84	0.19-8.09	0.02-2.51	0.14-8.08	1.80-24.5	This study
Ethiopia	0.00-13.9	-	0.00-90.6	1.51-51.4	1.30-21.9	1.19-36.3	2.77-195	[38]
Ethiopia	0.25-0.39	-	0.06-4.02	3.00-28.3	0.32-6.5	1.2-4.1	ND	[18]
Tanzania	0.10-3.50	-	5.90-87.7	3.40-14.0	0.1-8.9	-	-	[23]
Mutah, Jordan	0.01-129		0.54-157	0.29-26.8	0.02-17.5	0.25-44.3	3.8-106.1	[21]
Kirkuk– Iraq	-	-	9.88-65.5	16.0-59.3	0.92-20.4	0.10-0.60	1.0 - 13	[16]
Saudi Arabia	0.00-250		0.00-880	0.30-40	0.01-25	0-13.40	1.40-35	[9]
Italy	0.01-35.1	0.00-0.6	1.40-1278	1.26-474.	0.26-75.7	0.1-85.2	0.4 -774	[24]

CONCLUSION

This study assessed the physicochemical quality parameters of 20 brands of bottled water presently sold in Addis Ababa retail shops and supermarkets. The results showed that the bottled water samples analyzed are safe for human consumption except few brands which contained slightly higher levels of few indivividual parameters than the permissible level of WHO and CES. The physicochemical variables of the bottled drinking water studied showed large variation in different brands of bottled water which depends on the compositions of bottled water such as natural environment, the source of water composition and type of treatment/purification techniques during the production of bottled drinking water.

The number and type of parameters reported on the labels of bottled water showed a lack of uniformity. The comparison of parameters showed major cations and anions were highly variable with each brand. The concentrations of some of physicochemical parameters measured in this study were comparable to the values labeled on bottled water, whereas some other parameters were lower than and higher than values reported on the labels. The water quality index based on 15 important parameters has indicated that the bottled water samples has the lowest value of water quality index of 3.85 and the highest value of 49.2 which categorized them as excellent water quality and good water quality, respectively. In general this study recommends the regular need for a nationwide survey about the quality of waters including tap and river waters as well as groundwater and all marketed bottled waters be monitored for quality and identity by concerned authorities to safeguard consumers' health.

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