

LEVELS OF COMMON IONS IN BOTTLED MINERAL WATERS CONSUMED IN ADDIS ABABA, ETHIOPIA

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ABSTRACT: The inorganic compositions of nine brands of bottled mineral water (Ambo, Aquaddis, Highland, Abyssinia, Aqua Safe, Crystal, Kool, Oasis and Real Springs) and municipal water samples from different sources that are consumed in Addis Ababa (Ethiopia) were determined using standard methods. Flame atomic absorption spectrometry was used for the determination of heavy metals (Fe, Zn, Cu, Cr, Cd and Pb); Cu, Cr, Cd and Pb were not detected in all the samples. Ion chromatography was used for the determination of common cations (Na^+ , K^+ , Ca^{2+} , and Mg^{2+}) and common anions (F^- , Cl^- , NO_3^- , SO_4^{2-} and PO_4^{3-}) in the mineral water and municipal water samples. Common cations were found to be higher in Ambo mineral water than other mineral water samples, some of the anions were not detected in some of the mineral water samples; phosphate was not detected in all the samples. Fe and Zn were in higher levels in municipal water as compared to mineral waters and the reverse was true in the case of common cations and anions. Some brands of bottled mineral water were found not to comply with international guidelines for drinking water with respect to trace metals (*e.g.*, Zn, Cu, Cr, Cd and Pb) and some anions (*e.g.*, F^- , NO_3^- , PO_4^{3-}) and may not be suitable for babies and people suffering from heart or kidney diseases. Some physical parameters, pH and electrical conductivity of each water sample were also determined; pH in some samples was found to be below the lower limit of WHO's guideline.

Key words/phrases: Addis Ababa (Ethiopia), bottled mineral water, common cations and anions, heavy metals, physical parameters

INTRODUCTION

Next to oxygen, water is the most important substance for human existence. Water is essential for life on Earth. Because of its importance, the pattern of human settlement throughout history has often been determined by its availability (Saleh *et al.*, 2001) and it is an essential nutrient, which also sustains agriculture, allows aquatic life, supports industry, produces hydroelectricity, permits aquatic transport, insures personal hygiene and maintains clean environment (Falkenmark, 1982; Ministry of Water Resources, 1997). Certainly, humans get the benefits listed above from the entire water resources of the world which is estimated to be $1.4 \times 10^9 \text{ km}^3$ (Gross, 1987). However, for the most part, human existence mainly depends on fresh water supply which is less than 1% of the water available on Earth (Reagen and Bookins-Fisher, 1997). The fresh water of the world is obtained from the annual precipitation of about 10^5 km^3 (Gebre-Emanuel

Teka, 1977) out of which Ethiopia's yearly share is estimated to be 110 km^3 (Ministry of Water Resources, 1997). However, 75% of this water is lost through the borders toward neighbouring countries leaving behind 27.5 km^3 . On the other hand, since this water is not evenly distributed, arid and semiarid regions of the country are threatened by desertification (Environmental Protection Authority, 1998). In addition to the process of desertification, pollution is also reducing the volume of safe drinking water. For instance cancer mortality due to exposure of ground water to hazardous chemicals is increasing (Griffith *et al.*, 1989; Lipp and Rose, 1997). If heavy metals are present in drinking water, they may lead to severe effects that include reduced growth and development, cancer, organ damage, nervous system damage, and in extreme cases, death.

Water is an essential component for life and its analysis for chemical properties including heavy metal contents are very important for public health studies (Soylak *et al.*, 2002). Heavy metals occur in

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nature and most of them are advantageous to humans because of their vast usages in different industries, agriculture, and medicine. However, they may also pose health hazards to the public because of their presence in air, water and food (Shahtaheri *et al.*, 2006). Furthermore water pollution by heavy metals is mainly caused by point source emissions from mining activities and a wide variety of industries (Nazif *et al.*, 2006; Shahtaheri *et al.*, 2006). Heavy metals include essential elements such as Co, Cr, Cu, Fe, Mn, Mo, Se and Zn as well as toxic metals like Ag, Al, As, Cd, Pb and Ni. Certain essential trace elements can be toxic when concentrations are raised above specific cut-off levels (Fiket *et al.*, 2007) and the elements which are toxic when present in higher levels could be very useful in small amounts (Daskalova, 2007).

A number of chemical contaminants have been shown to cause adverse health effects in humans as a consequence of prolonged exposure through drinking water. Exposure to high levels of fluoride, which occurs naturally, can lead to mottling of teeth and, in severe cases, crippling skeletal fluorosis. Nitrate may arise from the excessive application of fertilizers or from leaching of wastewater or other organic wastes into surface water and groundwater. Particularly in areas with aggressive or acidic waters, the use of Pb pipes and fittings or solder can result in elevated Pb levels in drinking water, which cause adverse neurological effects. There are few chemicals for which the contribution from drinking water to overall intake is an important factor in preventing disease. One example is the effect of fluoride in drinking water in increasing prevention against dental caries (World Health Organization, 2004).

Drinking water may be contaminated by a range of chemical, microbial and physical hazards that could pose risks to health if they are present at high levels. Because of the large number of possible hazards in drinking water, the development of standards for drinking water requires significant resources and expertise, which many countries are unable to afford. Fortunately, guidance is available at the international level. International trade in bottled water has increased in recent years, both in quantity and diversity. Aside from water shortages, real and perceived needs to improve health have also contributed to a growing trade in bottled water. Increasingly it has been recognized that traditional suppliers of

drinking water such as public and private waterworks may not always be able to guarantee the microbiological, chemical and physical safety of their product to the extent previously thought possible (Codex Alimentarius, 2001).

The World Health Organization (WHO) publishes “*Guidelines for Drinking-water Quality*” 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 and of the effectiveness of associated control measures. WHO recommends that social, economic and environmental factors be taken into account through a risk-benefit approach when adapting the *Guideline* values to national standards. As the WHO *Guidelines* are meant to be the scientific point of departure for standards development, including bottled water; actual standards will sometimes vary from the *Guidelines* (Codex Alimentarius, 1985).

The chemical contaminants for which epidemiologic studies have suggested a risk associated with their presence in potable water include: Al, As, disinfection by-products, F⁻, Pb, NO₃⁻, pesticides, Cd, Hg and SO₄²⁻ (Calderon, 2000). These contaminants are of both inorganic and organic origin. Naturally occurring contaminants are generally the result of leaching from geologic formations and are found primarily in groundwater.

Toxic doses of chemical contaminants cause either acute or chronic health effects. An acute effect usually follows a large dose of a chemical and occurs almost immediately. The levels of chemicals in drinking water, however, are seldom high enough to cause acute health effects. They are most likely to cause chronic health effects that occur after long exposure to small amounts of a chemical. Good-quality drinking water may be consumed in any desired amount without adverse effect on health. Such water is called ‘potable’. It is free from harmful levels of impurities such as bacteria, viruses, minerals, and organic substances. It is also aesthetically acceptable and is free of unpleasant impurities, such as objectionable taste, colour, turbidity, and odour.

Bottled water usually tastes better than the municipal water (Versai *et al.*, 2002). However, the taste does not always indicate safeness. At the levels present in drinking water, most harmful substances (including some disease-causing

microorganisms, nitrates, trace amounts of Pb and Hg, and some pesticides and organic materials) have no taste. Differences in the taste among bottled waters are generally due to differing amounts of CO₂, Ca, Fe compounds, Na, and other minerals and mineral salts.

In Addis Ababa, different brands of indigenously produced bottled mineral waters are available in local markets and restaurants. These known mineral waters are Ambo, Highland, Aquaddis, Abyssinia, Aqua Safe, Kool, Crystal, Oasis, Real, *etc.*

It is important to determine and compare the content of mineral waters consumed in Addis Ababa with some international guidelines in general and with that of municipal water in particular. However, no systematic investigation has been carried out on the composition of bottled mineral waters in Ethiopia. Hence in this research, the levels of common cations and anions and some heavy metals in the nine brands of the most common commercially available bottled mineral water listed above were determined by standard methods. For comparison purpose samples of municipal water was collected from three locations in the College of Natural Sciences Campus of Addis Ababa University and analyzed. This research was carried out to clarify some of the concerns about the quality and safety of bottled drinking water which practically costs much higher than public drinking water.

MATERIALS AND METHODS

Cleaning apparatus

Apparatus such as volumetric flasks (both glass and plastic), plastic measuring cylinders, plastic bottles, and plastic beakers were washed with detergents and tap water, rinsed with distilled-deionised water, soaked in concentrated nitric acid

for 24 hours, then rinsed with distilled-deionised water, dried in oven and kept in clean place until needed for use.

Sampling

Nine brands of bottled mineral water samples: Ambo, Highland, Aquaddis, Abyssinia, Aqua Safe, Crystal, Kool, Oasis and Real Springs mineral water that are consumed in Addis Ababa were purchased from local supermarkets. Five bottles of 500 mL size were purchased for each brand. Seven brands of mineral waters are sold in sealed plastic bottles while two brands (Ambo and Crystal) of mineral waters are sold in sealed glass bottle. All the sampled bottles were kept sealed and refrigerated at 4°C until the time of analysis. Municipal water samples were randomly collected from three different locations in Arat Kilo (College of Natural Sciences, Addis Ababa University) in 1 L volumetric flask for the comparison purpose. Each water sample was collected from the tap used for drinking in the campus that was left running for more than 5 min before collecting the sample. The collected tap water samples were kept in the sealed flasks and refrigerated with those of bottled mineral waters at the same temperature until the time of analysis. General information of the bottled mineral water samples is given in Table 1.

Instrumentation

Flame atomic absorption spectroscopy (FAAS)

Flame Atomic Absorption Spectroscopy (FAAS), Buck Scientific Model 210VGP (East Norwalk, USA) equipped with deuterium arc background correctors, hollow cathode lamps for each respective metal, and air-acetylene flame were used for the determination of heavy metals. 69–72% HNO₃ (Spectrosol, BDH, England) was used for acidifying the water samples. Deionised water was used for dilution of standard solutions and rinsing the apparatus.

Table 1. Brand, description, package and manufacturer of bottled mineral water samples.

Brand	Description	Manufacturer
Ambo	Sparkling mineral water	At source
Highland	Natural mineral water	Apex Bottling Company
Aquaddis	Natural spring water	Burayu Spring Water PLC
Abyssinia Springs	Natural spring water	Great Abyssinia PLC
Aqua Safe	Pure natural spring water	Debre Birhan Natural Spring Water PLC
Crystal	Fortified mineral water	Coca-Cola Company
Kool	Natural mineral water	MOHA Drinks Industry SC
Oasis Spring	Pure natural water	Pacific Industries
Real Springs	Purified natural spring water	TGMD Trade Work PLC

Ion chromatography (IC)

For the analysis of common cations (Na^+ , K^+ , Mg^{2+} , Ca^{2+}) and common anions (SO_4^{2-} , PO_4^{3-} , NO_3^- , F^- , Cl^-); ion chromatography was carried out using a Dionex gradient HPLC system DX-600 Dionex USA, equipped with an ED50 Electrochemical detector, Dionex LC25 chromatography oven, Autosampler (Dionex AS50), Dionex GS50 Gradient pump and EG40 eluent generator and PeakNet6 software for data acquisition and instrument monitoring. Isocratic separation of both cations and anions were performed on IonpacCS12A cation exchange and IonpacAS17 anion exchange columns. Dedicated IonpacCG12A guard column for cationic and IonpacAG17 guard column for anionic were also used in connection with the analytical columns. H_2SO_4 (20 mM) was used as mobile phase for eluting cations while KOH (20 mM) was used as mobile phase for eluting the anions. Composite primary standard solutions of cations that contains (200 mg/L Na, 500 mg/L K, 250 mg/L Mg and 500 mg/L Ca) was used for the analysis of cations which was prepared from chloride salts of the cations by using ultra pure deionised water (99.9% water) and similar composite standard solution of anions that contains (20 mg/L F^- , 100 mg/L Cl^- , 100 mg/L NO_3^- , 100 mg/L SO_4^{2-} and 200 mg/L PO_4^{2-}) was used for the analysis of anions which were prepared by the Dionex company from sodium salts of the anions in ultra pure deionised water (99.9% water). These composite primary standards were used for preparing working standard solutions by diluting the primary standard solution in deionised water. The working standard solution of cations contain 50 mg/L Ca, 25 mg/L Mg, 50 mg/L K and 20 mg/L Na and similarly that of anions contain 2 mg/L F^- , 10 mg/L Cl^- , 10 mg/L NO_3^- , 10 mg/L SO_4^{2-} and 20 mg/L PO_4^{2-} were used for single point calibration standards.

pH and conductivity

The pH of the samples was measured using Denver Instrument model 250, pH-ISE conductivity meter and calibrated with pH 10.01 standard buffer solutions. The conductance of each water sample was also measured using ion conductivity electrode with standard buffer solutions. The

concentration of fluoride was determined by Orion Model, EA 940 Expandable Ion Analyze (USA) using Orion F^- ion selective electrode.

Determination of heavy metals and common ions

The levels of six heavy metals (Fe, Zn, Cu, Cr, Cd and Pb) in the nine bottled mineral water samples and municipal tap water samples were determined using FAAS. Stock solutions of the metals (Fe, Zn, Cu, Cr, Cd and Pb) 1000 mg/L (calibration standard Buck Scientific, USA, prepared as nitrates for each metal in 2% HNO_3) were used for preparing intermediate standard solutions (10 mg/L) in 100 mL volumetric flask and working standards using deionised water. Working standards of metal solutions were prepared in 50 mL volumetric flask by diluting with deionised water. Four points calibration curves were established by running the prepared standard solutions and the linear correlation coefficients greater than 0.999 were obtained for all the analytes. Immediately after calibration, the sample solutions were aspirated into the FAAS instrument and direct readings of the metal levels were recorded. Three replicate determinations were carried out on each sample.

Bottled mineral water and municipal tap water samples were analyzed for the common ions using ion chromatography. The column of the IC was washed by flushing with deionised water until the base line of the instrument is correctly determined or zero background noise is obtained. The eluents of both cations and anions were degassed in sonication bath for 50 min at 30°C and purged with argon gas for at least 10 min to remove dissolved gases. Composite standard solutions that were prepared from the primary standard solutions of cations and anions were injected into the column. The components were identified by comparison of retention times with documented standards in the software. After the instrument calibration was done with single point calibration standard, the samples were run for 10 min in the determination of anions and 15 min for that of cations. As soon as the run was over, the chromatogram was displayed on software and the peak area of the chromatogram was integrated and converted into concentration automatically by the instrument software and the results were recorded.

RESULTS AND DISCUSSION

Levels of heavy metals

The water samples were analyzed for six heavy metals (Fe, Zn, Cu, Cr, Cd and Pb) using flame atomic absorption spectrometry (FAAS). The results are presented as average of the determination of triplicate recording of the three sample solutions for each water sample ($n = 9$). The results are given in Table 2. Cu, Cr, Cd and Pb were below the detection limits in all the samples. Fe and Zn were detected at mg/L levels and are reported.

Table 2. Average levels of Fe and Zn in the bottled and tap water samples ($n = 9$).

Sample	Level (mean $\pm$ SD) of metal (mg/L)	
	Fe	Zn
Ambo	0.140 $\pm$ 0.012	0.200 $\pm$ 0.020
Highland	0.079 $\pm$ 0.005	0.170 $\pm$ 0.005
Aquaddis	0.093 $\pm$ 0.005	0.080 $\pm$ 0.006
Abyssinia	0.055 $\pm$ 0.001	0.124 $\pm$ 0.002
Aqua Safe	0.051 $\pm$ 0.003	0.088 $\pm$ 0.002
Crystal	0.071 $\pm$ 0.002	0.293 $\pm$ 0.004
Kool	0.049 $\pm$ 0.001	0.163 $\pm$ 0.006
Oasis	0.035 $\pm$ 0.002	0.137 $\pm$ 0.006
Real Springs	0.038 $\pm$ 0.003	0.096 $\pm$ 0.008
Tap water	0.160 $\pm$ 0.005	0.334 $\pm$ 0.010

Fe was detected appreciably in all the water samples. Fe level ranged from 0.035 to 0.16 mg/L. Fe was relatively higher in municipal tap water than in all of the bottled mineral water samples. Ambo mineral water had the highest Fe level among the bottled mineral waters with the concentration level of 0.14 mg/L, while Oasis had the lowest level of Fe (0.035 mg/L). But the amount found in Ambo mineral water sample is less than that in the municipal tap water sample. In mineralized spring water with a total dissolved solid content of 500 mg/L, the taste threshold value is 0.12 mg/L. The level of 2 mg/L of Fe recommended by WHO does not pose a hazard to

health. But the taste and appearance of drinking water will usually be affected below this level (Saleh *et al.*, 2001). Therefore, the bottled mineral water samples as well as the municipal tap water samples have no health effect on humans upon drinking these waters as far as Fe is concerned.

The levels of Zn in all the water samples were appreciable than all the other heavy metals determined. The levels ranged between 0.09–0.33 mg/L. Zn level was higher in the municipal tap water than the bottled mineral waters but the level range in all samples is within the levels recommended for good health, since the levels of Zn in all the water samples were much lower than the accepted value of WHO guideline, 5 mg/L for drinking water. It is interesting that the filtration of water during water treatment at the source actually increase the level of Zn in the tap water possibly due to leaching from materials used in manufacturing the filters and also from the leaching of galvanized pipes. According to the report from Egypt (Dabeka *et al.*, 2002), Zn imparts an undesirable astringent taste to water. Water containing Zn at levels in the range 3–5 mg/L also tends to appear opalescent and develops a greasy film when boiled. Therefore none of the analyzed water samples can show any of the problems mentioned above as the level of Zn in all the samples is very low.

Levels of common ions and some physical parameters

Determination of levels of common cations (Na^+ , K^+ , Mg^{2+} , Ca^{2+}) and anions (Cl^- , NO_3^- , SO_4^{2-} , and PO_4^{3-}) were carried out using ion chromatography. The level of fluoride (F^-) was determined using F-selective electrode. The results of these analyses are summarized in Tables 3 and 4. The results are reported as the averages of the triplicate results ($n = 3$) of each analyte in all the samples.

Table 3. Average levels of common metals (mg/L) and physical parameters of bottled and tap water samples.

Sample	Level (mean $\pm$ SD) common metal (mg/L) and pH and EC					
	Na	K	Mg	Ca	pH	EC ($\mu\text{S}/\text{cm}$)
Ambo	195 $\pm$ 1.5	21.9 $\pm$ 0.9	36.3 $\pm$ 3.4	51.4 $\pm$ 3.3	6.50 $\pm$ 0.010	1788 $\pm$ 6
Highland	18.3 $\pm$ 1.5	4.81 $\pm$ 0.19	8.21 $\pm$ 0.59	35.4 $\pm$ 0.36	7.94 $\pm$ 0.015	393 $\pm$ 2
Aquaddis	40.3 $\pm$ 2.6	2.82 $\pm$ 0.23	1.87 $\pm$ 0.100	5.41 $\pm$ 0.45	8.23 $\pm$ 0.01	296 $\pm$ 2
Abyssinia	18.3 $\pm$ 0.22	2.22 $\pm$ 0.064	2.47 $\pm$ 0.139	6.80 $\pm$ 0.22	7.96 $\pm$ 0.057	230 $\pm$ 2
Aqua Safe	23.2 $\pm$ 0.39	4.40 $\pm$ 0.044	1.91 $\pm$ 0.020	11.6 $\pm$ 0.25	8.34 $\pm$ 0.040	262 $\pm$ 5
Crystal	48.1 $\pm$ 0.45	1.23 $\pm$ 0.146	2.13 $\pm$ 0.091	1.51 $\pm$ 0.17	7.49 $\pm$ 0.006	579 $\pm$ 12
Kool	20.3 $\pm$ 0.54	1.19 $\pm$ 0.037	2.84 $\pm$ 0.015	5.10 $\pm$ 0.025	7.35 $\pm$ 0.040	124 $\pm$ 3
Oasis	9.63 $\pm$ 0.15	1.89 $\pm$ 0.060	3.42 $\pm$ 0.025	2.01 $\pm$ 0.044	7.53 $\pm$ 0.021	161 $\pm$ 1
Real Springs	7.69 $\pm$ 0.21	6.05 $\pm$ 0.113	1.30 $\pm$ 0.009	5.67 $\pm$ 0.22	8.05 $\pm$ 0.042	188 $\pm$ 2
Tap water	2.77 $\pm$ 0.10	1.25 $\pm$ 0.034	3.37 $\pm$ 0.034	3.70 $\pm$ 0.051	7.83 $\pm$ 0.017	120 $\pm$ 1

Table 4. Average levels of common anions (mg/L) in bottled and tap water samples.

Sample	Level (mean $\pm$ SD) of anion (mg/L)				
	F ⁻	Cl ⁻	NO ₃ ⁻	SO ₄ ²⁻	PO ₄ ³⁻
Ambo	0.70 $\pm$ 0.004	48.1 $\pm$ 1.9	13.9 $\pm$ 0.48	ND	ND
Highland	ND	3.57 $\pm$ 0.34	2.83 $\pm$ 0.12	2.63 $\pm$ 0.23	ND
Aquaddis	0.60 $\pm$ 0.005	6.67 $\pm$ 0.58	2.48 $\pm$ 0.19	6.45 $\pm$ 0.64	ND
Abyssinia	0.92 $\pm$ 0.036	8.31 $\pm$ 0.34	ND	9.06 $\pm$ 0.23	ND
Aqua Safe	0.59 $\pm$ 0.025	12.48 $\pm$ 0.24	0.31 $\pm$ 0.015	10.26 $\pm$ 0.17	ND
Crystal	ND	6.15 $\pm$ 0.17	8.38 $\pm$ 0.27	ND	ND
Kool	ND	13.24 $\pm$ 0.29	2.80 $\pm$ 0.13	5.29 $\pm$ 0.18	ND
Oasis	0.25 $\pm$ 0.012	4.84 $\pm$ 0.10	6.02 $\pm$ 0.19	1.62 $\pm$ 0.071	ND
Real Springs	0.80 $\pm$ 0.034	10.79 $\pm$ 0.13	0.72 $\pm$ 0.034	0.99 $\pm$ 0.048	ND
Tap water	ND	5.70 $\pm$ 0.20	1.67 $\pm$ 0.045	2.10 $\pm$ 0.083	ND

ND = not detected

Ion chromatography provides a straight forward method for the simultaneous determination of alkali and alkaline earth metal cations in drinking water as described in Clesceri *et al.* (1998). In this study two alkali metal cations (Na⁺ and K⁺) and two alkaline earth metal cations (Mg²⁺ and Ca²⁺) were determined in the bottled drinking water and the municipal tap water samples and the results are discussed below separately for each cation.

This study confirmed that the levels of common metal ions (Na⁺, Mg²⁺, K⁺ and Ca²⁺) in the bottled mineral waters vary with the sample analyzed. This may be due to the fact that the origins of these water samples are different, as they are from different sources. The highest levels of these metals were determined in the Ambo mineral water. When compared to the content of the metal ions in the tap water sample, the levels of these cations in most of the mineral waters are higher than in the tap water.

Na is very important for human body and regulates the water balance and the acid-base balance in the blood and tissue. Na in the drinking water is not a health concern for most people because in healthy people, excess Na is eliminated through the kidneys and the correct balance of Na and water is maintained. But for people with heart disease, hypertension, kidney disease and circulatory illness, it may be an issue of health concern because of the inability of such person to maintain the required body balance of Na (Lau and Luk, 2002; Mahajan *et al.*, 2006). The WHO and United States Environmental Protection Agency (USEPA) have restricted people with hypertension or those on Na-restricted diet to drink water with Na content not more than 20 mg/L and those on moderate restricted diet should not drink water containing more than 270 mg/L of Na (Lau and

Luk, 2002). The Food and Nutrition Board of the National Research Council of America recommends that Na intake be limited to no more than 2400 mg per day (Mahajan *et al.*, 2006).

In this study, the average level of Na is compared among the water samples. The maximum level was determined in Ambo mineral water and the lowest was in tap water sample. The level ranges from 2.02 in tap water to 195 mg/L in Ambo bottled mineral water. Among the drinking water samples, Highland mineral water and the municipal tap water samples are suitable for both groups of people with the stated health problems. Ambo, Aquaddis and Crystal mineral waters are not suitable for people with hypertension or Na restricted diet but are suitable for moderately restricted Na diet. According to WHO guidelines for maximum level of Na in the drinking water, only Ambo bottled water exceeded that recommendation. Na may affect the taste of drinking water at level above 200 mg/L (Saleh *et al.*, 2001). Therefore according to this study people with hypertension health problem are advised not to drink Ambo, Aquaddis and Crystal mineral waters. All the samples analyzed contain much lower level of Na than the maximum permissible limit of 200 mg/L suggested by WHO. Na⁺ is ubiquitous in water. Most water supplies contain less than 20 mg/L of Na, but in some countries levels can exceed 250 mg/L (Codex Alimentarius, 2001).

K is also very important for human body and like Na regulates the water balance and the acid-base balance in the blood and tissue. There is no fixed health guideline for the amount of K present in water that would be considered safe by the WHO. Drinking water is not the major dietary source of K, and the level in drinking water seldom reaches 10 mg/L. However, USEPA has set a

maximum level of 100 mg/L. In people on low K diets, stroke, high blood pressure, and diabetes occur more frequently than in those who consume sufficient or high K diets (Saleh *et al.*, 2001). The K content of drinking water varies greatly depending on its source. The content tends to be larger in mineral waters than in ordinary tap water. The Committee on Dietary Allowances recommends 1875–5625 mg per day of K in order to maintain adequate and safe levels of K balance (Mahajan *et al.*, 2006).

Levels of K in analyzed samples range from 1.19–21.9 mg/L. Bottled waters (except Kool) contain slightly higher levels of K than tap water. No health-based guideline values are proposed for K. But it has been recommended that water with K exceeding 12 mg/L is not suitable for regular drinking because it may cause kidney stress and possible kidney failure (Codex Alimentarius, 1985). Municipally treated drinking water may contain small levels of K. The use of water softeners containing KCl can significantly increase the levels of K in drinking water, even at water hardness levels considered to be acceptable (Federal-Provincial-Territorial Committee on Drinking Water, 2007).

The average level of K found to be higher in mineral water samples than in the tap water. The highest value was found in Ambo mineral water and the lowest in the tap water, ranging from 21.9 to 1.03 mg/L. Since getting adequate K in the diet is hampered by relatively few good sources, K in the water represents a potentially significant benefit to the majority of people. Considering this view, brands containing very low K are also not beneficial for human health. So people suffering from high blood pressure and relying upon only these bottled mineral waters are liable to aggravate their symptoms as all of the bottled water samples and the tap water are low in K.

WHO limits hardness for drinking water between 100–500 mg/L. Hardness of water which is due to the presence of Ca and Mg salts in water, does contribute towards total Ca and Mg human dietary needs, which has a beneficial effect on bone structure. Studies on water hardness and cardiovascular disease mortality have suggested a lower incidence of heart disease in communities drinking hard water. Extremely hard water (hardness > 500 mg/L) is also unfit for consumption because the constituent minerals such as Ca can deposit inside the body if present in high amounts leading to kidney or gall bladder stones. Consumption of

very soft water (hardness < 50 mg/L) lacking in essential minerals like Ca, Mg and other trace minerals is also harmful for the body because water low in mineral content would rob off the body's minerals. People drinking such treated water excrete huge amounts of Ca, Mg and other trace minerals in urine. The more the mineral loss, the greater the risk for osteoporosis, osteoarthritis, hypothyroidism, coronary artery disease, high blood pressure and a long list of degenerative disease generally associated with premature aging (Mahajan *et al.*, 2006).

There is a significant difference in Mg levels between mineral water analyzed in this study and the tap water sample. Ambo, Highland and Oasis have higher levels of Mg as compared to tap water. All the brands except Ambo had Mg level less than the lower limit of 30 mg/L as prescribed by the WHO. Thus these bottled water samples violate the lower limit of the WHO for Mg; hence drinking them alone may lead to the health problem due to Mg deficiency. But Ambo mineral water is suitable for people in need of Mg. The level of Mg ranges from 1.30 in Real Springs to 36.3 mg/L in Ambo bottled mineral water.

All the water samples analyzed had Ca less than the lower limit of 75 mg/L as prescribed by the WHO. The relative levels of Ca are higher in Ambo, Highland and Aqua Safe bottled waters than in the other samples analyzed. Aquaddis, Abyssinia, Crystal, Kool, Oasis and Real Springs showed very low levels of Ca as compared to the other three and the levels of Ca range from 1.51 in Crystal to 51.4 mg/L in Ambo water. Drinking only these bottled mineral waters may lead to some of the above mentioned health problems. Natural water sources typically contain levels up to 10 mg/L Ca. Hardness levels above 500 mg/L are generally considered to be aesthetically unacceptable. Ca is one of the major elements responsible for water hardness. Water containing less than 60 mg/L of Ca is considered as soft water (Saleh *et al.*, 2001; Mahajan *et al.*, 2006). Therefore none of the nine brands of mineral water samples are considered as hard water.

pH and electrical conductivity

pH has no direct adverse effect on human health, however, according to WHO guidelines, the maximum desirable limit of pH is 7.0–8.5, and USEPA established pH limits from 6.5–8.5 (Mahajan *et al.*, 2006). Waters with pH lower than 4 have a sour taste and above 8.5 an alkaline bitter taste.

High pH induces the formation of trihalomethanes, which are toxic. pH below 6.5 starts corrosion in pipes, thereby releasing toxic metals such as Zn, Pb, Cd, Cu, etc. The pH value of the bottled mineral waters was between 7.0 and 8.5. Ambo bottled mineral water had pH value below 7, which was just slightly acidic with pH value 6.5 and it was out of the range of pH given by WHO guideline, but all the other samples analyzed had pH within the prescribed limits recommended by USEPA guideline. The range of pH values in the bottled water samples was between 6.5 in Ambo and 8.34 in Aqua Safe. Therefore according to the USEPA guidelines all the bottled as well as the tap water samples are suitable for drinking. The results of pH value of each sample are given in Table 3.

Electrical conductivity (EC) is a measure of the ability of aqueous solution to carry an electric current that depends on the presence and total levels of ions, their mobility and valence (Clesceri *et al.*, 1998). The EC is a valuable measure of the amount of cations and anions in water. In this study, its value ranges from 120 $\mu\text{S}/\text{cm}$ in tap water to 1788 $\mu\text{S}/\text{cm}$ in Ambo bottled mineral water. All the bottled mineral waters have higher conductivity than the tap water. From conductivity values of each water sample, Ambo bottled mineral water contains higher minerals than the rest of bottled mineral waters as well as tap water samples. The maximum permissible value is 2500 $\mu\text{S}/\text{cm}$ (Drinking Water Inspectorate, 2006). Thus all the bottled mineral waters are safe for drinking.

Levels of common inorganic anions

The U.S. National Primary Drinking Water Standards specify a maximum contaminant level (MCL) for a number of inorganic anions including fluoride, nitrate and nitrite. The MCLs are specified to minimize potential health effects arising from the ingestion of these anions in drinking water. Consequently, the analyses of these anions in drinking water samples are mandatory. Other common anions such as chloride and sulphate are considered secondary contaminants. The secondary standards are guidelines with regards to taste, colour, odour and certain aesthetic effects.

Ion chromatography has been recommended for compliance monitoring of these common inorganic anions in drinking water. In this study the common anions including fluoride, chloride, bromide, nitrate, sulphate and phosphate were determined in the bottled mineral water and the

municipal tap water samples. The results are summarized in Table 4. All the samples analyzed showed no phosphate at all.

Fluoride is recognized as having a beneficial effect on the development of children's teeth with 1.0 mg/L being the optimum level. Fluoride supplements are recommended for children between 3 and 13 years age if the level of fluoride in drinking water is below 0.3 mg/L (Mahajan *et al.*, 2006). However, level over 1.5 mg/L may damage children's teeth causing staining, mottling or cavities, the condition known as dental fluorosis. Fluoride levels were found to be in the range of ND–0.92 mg/L. The levels of fluoride in all the bottled mineral waters were below permissible level of WHO (1.5 mg/L). Fluoride was not detected in Highland, Crystal, Kool and tap water. Thus, those people, especially children consuming only these waters for drinking purposes may need to supplement their fluoride intake, such as the use of fluorinated toothpastes. In areas with relatively high fluoride levels in groundwater, drinking water becomes increasingly important as a source of fluoride (National Health and Medical Research Council, 2004).

Chloride level of the bottled mineral waters ranged from 3.57 in Highland to 48.1 mg/L in Ambo mineral water. None of the drinking water samples analyzed for chloride exceeded the limit permitted by WHO (250 mg/L). Thus all the bottled water samples are safe for drinking from the chloride point of view. All bottled mineral waters contain higher chloride concentrations than tap water, except Oasis and Highland bottled mineral waters. Chloride in water may be considerably increased by treatment processes in which chlorine or chloride is used. If a daily water consumption of 2 L and an average chloride level in drinking water of 10 mg/L are assumed, the average daily intake of chloride from drinking water would be approximately 20 mg per person, but a figure of approximately 100 mg/day has also been suggested (Protecting Our Environment, 2006).

Nitrate in the investigated water samples was found in the range of ND–13.9 mg/L. The highest value was obtained for Ambo whereas it was not detected in Abyssinia. In all the samples, nitrate was found below the proposed WHO safe drinking water standards of 50 mg/L and hence does not pose much health concern. When compared to municipal tap water, the nitrate contents of most of the bottled mineral water samples were higher. The nitrate concentration in surface water is

normally low (ND–18 mg/L) but can reach high levels as a result of agricultural runoff, refuse dump runoff, or contamination with human or animal wastes. The concentration often fluctuates with the season and may increase when the river is fed by nitrate-rich aquifers (Kovács *et al.*, 2004).

Sulphate was not detected in Ambo and Crystal mineral water. Relatively significant sulphate level was detected in Aqua Safe and Abyssinia mineral water. The level of sulphate in tap water was lower in some while higher in other mineral waters. Sulphate is one of the least toxic anions. The lethal dose for human as K or Zn sulphate is 45 g. The major physiological effects resulting from the ingestion of large quantities of sulphate are catharsis, dehydration, and gastrointestinal irritation. No health-based guideline value for sulphate in drinking water is proposed by WHO. However, because of the gastrointestinal effects resulting from the ingestion of drinking water containing high sulphate levels, it is recommended that health authorities be notified the sources of drinking water that contain sulphate level in excess of 500 mg/L (Saleh *et al.*, 2001). Sulphate concentration in the drinking water samples were in range of ND–10.26 mg/L. Sulphate was not detected in Ambo and Crystal mineral waters. All the analysed samples contain very low levels of sulphate than the maximum permissible limit of 250 mg/L by WHO. The average daily intake of sulphate from drinking water, air and food is approximately 500 mg, food being the major source. However, in areas with drinking water supplies containing high levels of sulphate, drinking water may constitute the principal source of intake (Bağ *et al.*, 2006).

Phosphate was not detected in all the bottled mineral water and the municipal tap water samples. No health-based guideline values are proposed for the phosphate.

Analysis of variation in composition of bottled mineral water samples

To know whether the composition of the samples of mineral water are significantly different or not it is important to use the application of analysis of variance (ANOVA). ANOVA is an extremely powerful statistical technique which can be used to separate and estimate the different causes of variation (Rayner *et al.*, 2007). The

calculation of one-way ANOVA for practical purposes can be done on computer using Minitab, Excel and SPSS 13.0 software (Miller and Miller, 2000). In this study SPSS 13.0 software was used to calculate the ANOVA for testing the significant differences in the composition of bottled mineral water samples. There were significant differences observed between the means of the determinations for all the analytes except Zn in Ambo and Highland, Fe in Ambo and Aquaddis and Fe in Highland and Aquaddis, Fe in Aqua Safe and Kool, Zn in Aqua Safe and Real Springs, K in Crystal and Kool, and Ca in Crystal and Oasis.

Comparison of present study with results from other countries

There are several reports from different countries on the analysis of mineral waters for the content of inorganic ions and physical parameters. It is important to compare the results obtained from the analysis of mineral water in Ethiopia with those of other countries to know the difference in the composition, suitability for drinking and their deviation from international guidelines outlined for drinking water. The results of present study have been compared with the composition of mineral waters from twenty countries (Lau and Luk, 2002). From each country the number of brands reported were as follows: Egypt, five brands; Australia, six brands; Canada, two brands; China, eight brands; France, eight brands; Germany, one brand; Hong Kong, five brands; Iceland, one brand; Indonesia, six brands; Italy, three brands; Japan, three brands; Malaysia, three brands; Portugal, two brands; Scotland, two brands; Sweden, one brand; Thailand, two brands; Turkey, one brand; UK, four brands and USA, two brands. The reports show that, all mineral water samples were analyzed for common ions (F^- , Cl^- , NO_3^- , SO_4^{2-} , Na^+ , K^+ , Mg^{2+} , Ca^{2+}). The comparison of present study was done with these reported results as ranges of the results of all brands for each ion as given in Table 5. The levels of common ions in the Ethiopian mineral waters are in the middle of the ranges reported for the mineral waters of other countries. Except few results, the composition of the bottled mineral water samples from different countries show more or less similar compositions. These results are shown in bold prints in Table 5.

Table 5. Comparison of the results obtained in the present study with results from other countries (Lau and Luk, 2002).

Country	Analytes (mg/L)							
	F ⁻	Cl ⁻	NO ₃ ⁻	SO ₄ ²⁻	Na ⁺	K ⁺	Ca ²⁺	Mg ²⁺
Ethiopia (present study)	0.0–0.92	3.57–48.1	0.0–13.9	0.0–9.06	2.77– 195.4	1.19– 36.3	1.51–51.4	1.30–21.9
Egypt	0.12–0.48	11.1– 221.1	0.1–18.7	10.4–68.12	4.49– 169	0.11–18.5	1.39–44.8	1.54–23.3
Australia	0.10–0.22	5.9–47.4	0.6–24.8	0.7–6.8	2.4–34.5	0.7–20.0	0.5–4.6	5.7–38.6
Belgium	0.03–0.19	5.7–11.4	0.2–1.3	4.0–4.7	7.2–7.9	1.2–1.3	6.0	1.2
Canada	0.2–0.36	0.0	0.2–1.0	1.7–6.7	0.0–1.5	0.2–6.0	3.0–7.9	0.0–0.7
China	0.07–0.79	0.0–67.0	0.9– 35.9	0.5– 177.0	8.1–31.4	0.4–24.1	0.7– 171.4	1.0–12.5
France	0.0–0.49	4.3– 125.3	0.0–18.3	7.2– 1039.0	7.5–49.0	5.0– 58.9	6.5– 468.6	2.2–21.0
Germany	0.0	45.8	1.1	47.0	227.0	170.5	113.0	50.5
Hong Kong	0.0–0.44	7.9–80.9	0.0–4.2	0.0–98.7	0.0–44.0	0.0–6.4	2.4–22.0	0.2– 47.3
Iceland	0.04	15.9	0.7	2.6	14.7	1.1	4.7	0.0
Indonesia	0.0–0.31	0.0–26.4	0.7– 38.1	1.3–27.2	9.1–40.0	4.3–70.7	2.8–21.4	2.8–10.3
Italy	0.0–1.2	0.0–19.4	5.1–9.1	4.8–41.5	3.3–30.9	0.5– 26.5	6.3–40.0	0.8– 48.0
Japan	0.0–0.02	3.0–7.8	0.5–1.5	0.4–2.3	7.9–8.4	0.9–2.8	0.4–2.1	0.4–2.1
Malaysia	0.05– 2.62	2.9–22.9	0.0–13.5	0.9–3.2	5.8–30.8	0.0–3.4	4.9–18.3	4.0–5.8
Portugal	0.0–0.05	8.6–15.8	1.3–1.9	0.7–1.3	7.6–11.8	1.5–13.6	0.0–22.1	3.0–4.3
Scotland	0.05–0.1	8.6– 138.6	1.1–4.8	0.8–8.0	8.1–58.0	13.3–17.9	47.7– 110.0	2.9–3.1
Sweden	2.4	26.5	0.7	9.3	225.0	0.6	5.5	1.9
Thailand	0.03–1.81	30.7– 133.2	0.2–0.4	1.7–33.9	68.0–69.2	0.2–14.9	31.6–38.0	15.1–25.0
Turkey	0.0	0.0	1.7	3.8	0.4	4.6	31.0	0.1
UK	0.0–0.1	15.1–33.5	0.3–15.0	3.6–70.0	10.3–30.0	1.4– 26.0	54.6– 140.0	0.8–5.0
USA	0.0–0.25	7.2– 214.1	0.4–0.6	6.0–106.1	0.0–11.1	0.2–3.7	9.1–79.7	3.8–4.3

Bold font indicates values exceeding limit of standard.

Comparison of present results with some national and international standards

Drinking water may be contaminated by a range of chemical, microbial and physical hazards that could pose risks to health if they are present at high levels. Examples of chemical hazards include Pb, As, Cr, Cd, F⁻, *etc.* and microbial hazards, include bacteria, viruses and parasites, such as *Vibrio cholerae*, hepatitis A virus, and *Cryptosporidium parvum*, while physical hazards include glass chips and metal fragments. Because of the large number of possible hazards in drinking water, the development of standards for drinking water requires significant resources and expertise, which many countries are unable to afford. Fortunately, guidance is available at the international level. The World Health Organization (WHO) publishes guidelines for drinking water quality 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 and of the effectiveness of associated control measures. WHO recommends that social, economic and environmental factors be taken into account through a risk-benefit approach when adapting the

guideline values to national standards. As the WHO guidelines for drinking water quality are meant to be the scientific point of departure for standards development, including bottled water; actual standards will sometimes vary from the guidelines (Lau and Luk, 2002).

Ten national and international guidelines were considered for comparing the results obtained in this study to check whether the results are within the limits of guidelines of drinking water or not (for knowing the suitability of these water sample for drinking). The results are summarized in Tables 6 and 7. These standard guidelines are WHO, UK, EU, USEPA, Pakistan, Canada, Australia, New Zealand, China and Ethiopia. The Ethiopian standard was developed in March 2002 for drinking water based on the WHO guidance and considering the geographical, economical and cultural values of the country (Ministry of Water Resources, 2002). According to these standard guidelines, no analytes are above the given standards except Na and K in Ambo which exceeded the UK maximum contaminant level. Therefore all the water samples can be used for drinking as it is within the limits of all the guideline.

Table 6. Comparison of results from the present study (heavy metals) with some national and international standards.

Standards	Analyte (mg/L)					
	Cd	Cr	Cu	Fe	Pb	Zn
WHO (World Health Organization, 2004)	0.003	0.05	2	0.2	0.01	5
Australia (National Health and Medical Research Council and Natural Resource Ministerial Council of Australia and New Zealand, 1996)	0.002	0.05	2	0.3	0.01	3
Canada (Abulude <i>et al.</i> , 2007)	0.005	0.05	≤1	≤0.3	0.01	≤ 5
USEPA (1998) ^a	0.005	0.05	1	–	0.005	5
New Zealand (Ministry of Health, 2005)	0.004	0.05	2	–	0.01	–
Pakistan (Government of Pakistan Ministry of Health, 2005)	0.003–0.01	0.05–0.1	1–2	–	–	–
Codex (Codex Alimentarius, 2001)	0.003	0.05	1	–	0.01	–
UK (MCL) (Fawell, 2007)	–	–	–	–	–	–
EU/EC (1998) ^b	–	–	–	0.2	0.1	–
China (1985) ^c	–	–	–	–	–	–
Ethiopia (Ministry of Water Resources, 2002)	0.003	0.1	2	0.4	0.01	6
This study						
Ambo	–	–	–	0.11	–	0.20
Highland	–	–	–	0.079	–	0.17
Aquaddis	–	–	–	0.093	–	0.08
Abyssinia	–	–	–	0.055	–	0.124
Aqua Safe	–	–	–	0.051	–	0.088
Crystal	–	–	–	0.071	–	0.293
Kool	–	–	–	0.049	–	0.163
Oasis	–	–	–	0.035	–	0.137
Real Springs	–	–	–	0.038	–	0.096
Tap water	–	–	–	0.086	–	0.334

Note: – The limit is not given; ^aUSEPA = United States Environmental Protection Agency Maximum Contaminant Level (MCL) (1998); ^bEU/EC = European Community Council Directive 98/83/EC, Parametric Values (1998), ^cChina for the standard of People Republic of China GB 5749-85 (1985).

Table 7. Comparison of results of the present study (common ions) with some national and international guidelines.

Standards	Analyte (mg/L)								
	Na ⁺	K ⁺	Mg ²⁺	Ca ²⁺	F ⁻	Cl ⁻	NO ₃ ⁻	SO ₄ ²⁻	PO ₄ ³⁻
WHO (World Health Organization, 2004)	200	–	–	–	1.5	250	50	250	–
Australia (National Health and Medical Research Council and Natural Resource Ministerial Council of Australia and New Zealand, 1996)	180	–	–	–	1.5	250	50	500	–
Canada (Abulude <i>et al.</i> , 2007)	≤ 200	–	–	–	1.5	≤ 250	45	≤ 500	–
USEPA (1998) ^a	–	–	–	–	1.5	–	10	250	–
New Zealand (Ministry of Health, 2005)	–	–	–	–	1.5	–	–	–	–
Pakistan (Government of Pakistan Ministry of Health, 2005)	–	–	–	–	1.5–4.0	250–400	–	–	–
Codex (Codex Alimentarius, 2001)	–	–	–	–	2	–	50	–	–
UK (MCL) (Fawell, 2007)	150	12	50	250	1.5	400	50	250	–
EU/EC (1998) ^b	200	–	–	–	1.5	250	50	250	–
China (1985) ^c	–	–	–	–	1	250	88	250	–
Ethiopia (Ministry of Water Resources, 2002)	350	–	–	–	3	533	50	450	–
This study									
Ambo	195.00	21.90	36.30	51.40	0.70	48.10	13.90	ND	ND
Highland	18.30	4.81	8.21	35.4	ND	3.57	2.83	2.63	ND
Aquaddis	40.30	2.82	1.87	5.41	0.6	6.67	2.48	6.45	ND
Abyssinia	18.31	2.22	2.47	6.80	0.92	8.31	–	9.06	ND
Aqua Safe	23.00	4.40	1.19	11.59	0.59	12.38	0.31	10.26	ND
Crystal	48.03	1.23	2.13	1.51	ND	6.15	8.38	ND	ND
Kool	20.32	1.19	2.84	5.10	ND	16.24	2.80	5.29	ND
Oasis	9.63	1.89	3.42	2.01	0.25	4.84	6.02	1.62	ND
Real Springs	7.69	6.05	1.30	5.67	0.80	10.79	0.72	0.99	ND
Tap water	2.77	1.25	3.37	3.70	ND	5.70	1.67	2.10	ND

Note: ND = Not detected; other notes as in Table 6.

CONCLUSIONS

The inorganic composition of a number of mineral water samples from different sources in Addis Ababa were determined and compared with that of municipal tap water sample. The levels of common cations (Na^+ , K^+ , Ca^{2+} and Mg^{2+}) were higher in mineral waters than in tap water. Among the heavy metals only Fe and Zn were detected in all the water samples. Tap water contains higher levels of these metals than bottled mineral water samples. Levels of Cr, Cd, Cu and Pb were below the detection limit in all the samples. The levels of anions varied among the samples, however the levels of these anions were higher in mineral water samples as compared to tap water. Phosphate was not detected in all the samples. Generally, mineral water samples contain higher minerals especially common ions than tap water. This was confirmed by conductivity measurement. It is advisable for people who have a problem of blood pressure, kidney diseases, heart disease, and circulatory illness to drink tap water in preference to mineral water. There is a large variation in the bottled mineral water composition among the brands compared, thus the consumers can choose the mineral water according to their preference.

All the parameters determined were below the guideline limits, except K level which exceeds the MCL laid by United Kingdom (UK). The importance of the quality of water for human consumption with regard to the health makes it necessary to establish norms to regulate it, including limits for all the parameters that directly affect human health and deteriorate water quality. The results of this study were also compared with the results of other selected countries' mineral waters. Except a few, the composition of mineral waters in Ethiopia is more or less similar to those of the other countries.

This study has shown that all the nine brands of mineral waters and the tap (municipal) water meet the national and international limits for the trace metals and common ions. This means that the bottled waters are safe to drink. However, regular monitoring and testing for chemical compositions should be ensured by the authorities concerned.

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