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ARAŞTIRMA MAKALESİ **RESEARCH PAPER**

A Comparative Study for Some Chemical Properties of Packaged Natural Mineral Water Sold in Sivas (Türkiye)

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Abstract: In the study, some chemical properties of packaged natural mineral water sold in Sivas were researched and compared with sticker values. The paired t-test statistic was used for comparisons between the sticker and laboratory findings. There were statistically significant differences for fluoride (p=0.042*), magnesium (p=0.009**) and sulfate (p=0.000**) between sticker and laboratory findings. The maximum values were observed to be as pH (8.30), calcium (85.60 ppm), chloride (6.60 ppm), fluoride (0.12 ppm), iron (0.03 ppm), magnesium (21.10 ppm), potassium (1.50 ppm), sulphate (8.00 ppm) in sticker findings and pH (8.14), calcium (39.98 ppm), chloride (4.00 ppm), fluoride (0.01 ppm), iron (0.30 ppm), magnesium (23.97 ppm), potassium (0.00 ppm), sulphate (0.00 ppm) in laboratory findings for water samples. These results obtained from all packaged natural mineral water samples were below the proposed standards of national and international for drinking water. At the same time, the fluoride content of packaged natural mineral waters in Sivas was determined to be fairly small which may be detrimental in the health of local inhabitants for dental carries. Hereby, the people of Sivas should be selective when deciding which packaged natural mineral waters to drink.

Keywords: Health, mineral, natural, packaged, water, quality.

Sivas (Türkiye)'da Satılan Ambalajlanmış Doğal Mineralli Suların Bazı Kimyasal Özellikleri için Karşılaştırmalı Bir Çalışma

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Öz: Bu çalışmada, Sivas'da satılan ambalajlanmış doğal mineralli suların bazı kimyasal özellikleri araştırılmış ve etiket değerleri ile karşılaştırılmıştır. Etiket ve laboratuvar bulguları arasındaki karşılaştırmalar için eşleştirilmiş t-testi kullanılmıştır. Etiket ve laboratuvar bulguları arasında florür (p=0.042*), magnezyum (p=0.009**) ve sülfat (p=0.000**) için istatistiksel olarak önemli farklar bulunmuştur. Maksimum değerler, etiket bulguları için pH (8.30), kalsiyum (85.60 ppm), klorür (6.60 ppm), florür (0.12 ppm), demir (0.03 ppm), magnezyum (21.10 ppm), potasyum (1.50 ppm), sülfat (8.00 ppm); laboratuvar bulguları için pH (8.14), kalsiyum (39.98 ppm), klorür (4.00 ppm), florür (0.01 ppm), demir (0.30 ppm), magnezyum (23.97 ppm), potasyum (0.00 ppm), sülfat (0.00 ppm) olarak belirlenmiştir. Tüm ambalajlanmış doğal mineralli su örneklerinden elde edilen bu sonuçlar, içme suyu için önerilen ulusal ve uluslararası standartların altındadır. Aynı zamanda, Sivas'da ambalajlanmış doğal mineralli suların florür içeriğinin oldukça düşük olduğu belirlenmiştir ve bu da yerel halk sağlığına diş çürümeleri şeklinde olumsuz etkiler yapabilir. Bu vesile ile Sivas halkı, hangi ambalajlı doğal mineralli suları içeceğine karar verirken seçici davranmalıdır.

Anahtar kelimeler: Ambalajlanmış, doğal, kalite, mineral, sağlık, su.

INTRODUCTION

One of the most important reasons that distinguishes the Earth from other planets and why it is a living environment for living things is the presence of water. Water is one of the indispensable needs of human life on earth. There are no calories, sugar, oil, cholesterol and caffeine in water. Water is of increasing strategic importance. Water, which is of vital importance for people and is also of great importance for health, must first of all be reliable. In this regard, individuals generally turn to consuming packaged water (Karakuş et al. 2016). Water consumption per capita in the world is around 800 m^3 per year on average. Approximately 1.4 billion people, corresponding to approximately 20 percent of the world's population, lack adequate drinking water, and 2.3 billion

people long for healthy water. Some estimates show that more than 3 billion people will face water scarcity by 2025. According to The Food and Agriculture Organization, in 2025 the ratio of the population experiencing water scarcity and water stress to the world population is estimated to increase by 34 percent and 15 percent, respectively (Tümer et al. 2016). Packaged waters are a source of drinking water that is increasingly used in both developing and developed countries. Due to the excessive variability in the mineral content of packaged waters, the public should have access to information about the minerals, since the total dissolved solids range from almost zero to several thousand milligrams per liter (WHO, 2009). Despite its economic costs and environmental damage, the use of packaged water is increasing worldwide. This is linked to the accessibility of packaged water, its convenience, taste and the belief that it is safer than tap water. Packaged water production in Türkiye in 2020 is 4.6 billion liters and this production amount is expected to increase year by year (Ulvi et al. 2022). Unfortunately, Türkiye is a country on the verge of water scarcity. Türkiye's rainfall rate is below the world average, and rapid increase in migration and climate changes cause the picture to gradually deteriorate. For this reason, water management must be in a way that eliminates the problem within the country (Ögenler and Okuyaz, 2017).

The quality of consumed water directly affects health (Şavik et al. 2012). Packaged drinking waters in Türkiye are defined in four different classes. These are natural spring water, drinking water, processed drinking water and natural mineral water. The natural mineral water is usually seen as a product that has medical value because of the high amounts of dissolved mineral (Güler, 2007). Natural mineral water is the groundwater, which is naturally occurring under suitable geological conditions at various depths of the earth's crust, is extracted by one or more sources from the earth spontaneously or by technical methods, is defined by mineral content, residue elements and other components and protected against all kinds of pollution risks. The most important feature that separates natural mineral waters from other drinking waters is that they contain a specific amount of mineral at the time they are obtained from the source. Natural mineral waters have different chemical compositions and their mineralization can range from a few milligrams to several grams per one liter of water. Due to some substances and minerals they carry, mineral waters have additional positive effects on health as compared to spring waters (Karagülle, 2004). To date, no studies have been conducted regarding the quality of packaged natural mineral waters sold in the province of Sivas. In the study, some chemical properties of six different brands of packaged natural mineral water widely consumed by the people of Sivas were researched and compared with sticker values.

MATERIAL AND METHOD

***Description of Study Area*:** The study area is Sivas province located in the central Anatolian part of Türkiye. According to the 2018-year address based population registration system, the population of the province center is 470589. The province covers an area of 27202 square kilometers. The altitude of the province is about 1285 meter. It is bordered by Erzincan to the east, Yozgat to the west, Ordu and Tokat to the north, Kahramanmaraş and Malatya to the south, Giresun to the northeast and Kayseri to the southwest. Türkiye's road and railway network linking the four side passes through Sivas. In Sivas, agriculture, husbandry, weaving, leatherwork, mining and small handcrafts are the most important sources of income (Doğan, 2007). Sivas is one of the coldest provinces in Türkiye. The winter months are freezing cold and the average winter temperature is around 0 °C in Sivas. It is observed that the temperature decreased to -36.4 °C. In summer the temperature is usually above 19 °C. However, it is seen that the temperature in summer is over 38 °C. As can be understood from this, the annual temperature difference shows a big difference of 74 °C. The average number of days the temperature drops below 0 °C is 132 days in Sivas (Pürlü, 2013).

Sampling Design and Procedure: Brands of packaged natural mineral waters were bought through randomly sampling from local markets and shops in Sivas province of Türkiye. All water samples were collected during May 2018 - April 2019. Container sizes of the collected samples are 0.5 L. The packaged mineral water samples of six different brands were collected in Sivas. Water samples are given a code number between 1 and 6 to keep their brand names secret. Permite numbers of packaged natural mineral water samples were recorded as MS.66.01 for brand number of 1, MS.54.08 for brand number of 2, MS.01.01 for brand number of 3, MS.80.01 for brand number of 4, MS.54.01 for brand number of 5 and MS.54.05 for brand number of 6. Similarly, permission dates of packaged natural mineral water samples were determined as 08 September 2009 for brand number of 1, 03 February 2012 for brand number of 2, 09 January 2010 for brand number of 3, 26 October 2016 for brand number of 4, 06 October 2009 for brand number of 5 and 20 July 2010 for brand number of 6. The analysis results on the stickers of the packaged mineral water samples were used as comparison data. Packaged water sticker values are the license values obtained from Provincial Health Directorates of the Türkiye Ministry of Health. Provincial Public Health Directorates make taken water samples analyzes. The consequences of the analysis are based on the mineral water samples taken from the final filling point, which is the last point to be used for water consumption in the labels of the natural mineral waters.

Concentrations of parameters are in ppm, except pH. In this study the amount of pH, calcium, chloride, fluoride, iron, magnesium, potassium, sulphate of six different brands were analyzed and compared with labeled values. The pH of the packaged mineral water samples was measured by a digital HI 2211 Hanna pH meter. Fluoride and iron were measured by colorimetric method using Chembio test kits CB5100 and CB5090 respectively. Potassium and sulphate of the packaged water samples were analyzed by titrimetric method using Chembio test kits CB5390 and CB5450, respectively. Chloride and total hardness were measured by titrimetric method using Aquamerck test kits 1.11106.0001 and 1.08039.0001 respectively. Calcium and magnesium of the packaged water samples values were reckoned according to the formula of total hardness change (Höll, 1979; Kasımlıoğlu & Yılmaz, 2014). Also, laboratory and sticker values of six packaged natural mineral water samples widely consumed in Sivas were compared and discussed with the international drinking water regulations (ECDWD, 1998; TWRHC, 2005; EPA, 2014; CDWQ, 2017; WHO, 2017).

Statistical Analysis: The dependent sample paired t-test was performed to assessment the differences of packaged natural mineral waters in Sivas between the sticker and laboratory findings. The Kolmogorov Smirnov test was used to evaluate the normal distribution of observations. Statistical analyzes were performed with IBM SPSS 22.0 package program. There was a statistically significant difference between the laboratory and sticker values in cases $p<0.05$ (at 95 percent confidence level) and $p<0.01$ (at 99 percent confidence level).

RESULTS AND DISCUSSION

Nowadays, people drink bottled waters by buying. Water is very important for human health. But it is even more important to know what is the property of drinking water is. However, it is very difficult to know the nature and property of the water flowing through the creek, fountain or any resource. The characteristics of packaged water are written on the stickers. Drinkers of packaged water should read the stickers well and know what they drink. The natural mineral water is rich in mineral content. Minerals in these waters help to meet the body's mineral needs. This type of water in the stickers "Natural Mineral Water" is written in capital letters. In addition, what are the minerals in these waters and their quantities are written on the stickers. When the natural mineral water is consumed, the body's water needs are mixed. In addition, the essential minerals in the body help us to meet our daily mineral needs by getting into the blood through rapid absorption in the intestines. It is not possible to supply water with other liquids and foods. Nothing replaces the water. The most important supporting evidence for this is that all minerals in natural mineral waters are present in certain proportions. The packaged natural mineral water is widely used for drinking due to its potential benefits. All of the samples researched packaged natural mineral water is produced in different cities of Türkiye. There is no packaged natural mineral water company in Sivas. Sold in Sivas, the mentioned packaged water resources are mostly located in Sakarya province of Türkiye followed by Adana, Osmaniye and Yozgat provinces. Samples of six brands of packaged natural mineral water of 0.5 litres capacity were collected randomly from Sivas. Descriptive statistics and comparisons of sticker and laboratory findings of some chemical properties in packaged natural mineral water samples are summarized in Table 1. The highest pH (8.30), calcium (85.60 ppm), chloride (6.60 ppm), fluoride (0.12 ppm), potassium (1.50 ppm) and sulphate (8.00 ppm) levels were determined in sticker findings of packaged natural mineral water, whereas the highest iron (0.30 ppm) and magnesium (23.97 ppm) levels were measured in the laboratory findings of packaged natural mineral water in Sivas (Table 1). It was found that calcium was the highest parameter in the sticker and laboratory findings. Also, sticker mean findings of pH, calcium, chloride, fluoride, potassium and sulphate were found to be higher than laboratory mean findings. However, sticker mean values of iron and magnesium were determined to be lower than in laboratory mean findings.

Table 1. Descriptive statistics and chemical of sticker and laboratory findings of packaged natural mineral waters.

Parameters	Sticker Findings				Laboratory Findings				Paired t-test	
	Min	Max	Mean	±SD	Min	Max.	Mean	±SD	t-value	p-value
pH	7.55	8.30	8.05	0.27	7.53	8.14	7.89	0.21	1.217	0.278
Calcium	19.37	85.60	39.36	23.66	18.56	39.98	29.75	8.11	0.955	0.384
Chloride	1.07	6.60	2.69	2.23	1.00	4.00	2.00	1.26	1.680	0.154
Fluoride	0.00	0.12	0.06	0.04	0.01	0.01	0.01	0.00	2.712	0.042*
Iron	0.00	0.03	0.01	0.01	0.00	0.30	0.10	0.12	-2.022	0.099
Magnesium	1.70	21.10	6.48	7.26	11.13	23.97	17.83	4.86	-4.098	0.009**
Potassium	0.00	1.50	0.36	0.56	0.00	0.00	0.00	0.00	1.581	0.175
Sulphate	2.74	8.00	5.83	1.77	0.00	0.00	0.00	0.00	8.052	0.000**

Min: Minimum, Max: Maximum, SD: Standard Deviation, Asterisks Notation (*: $p<0.05$; **: $p<0.01$) indicate the significance level of the comparisons in the Table 1.

According to the ECDWD (1998), TWRHC (2005) and EPA (2014) standards the tolerable range of pH in drinking water should be 6.50-9.50 unit. The pH ranged from 7.53 to 8.30 in sticker and laboratory findings in Sivas (Table 1) it also observed within the ECDWD (1998), TWRHC (2005) and EPA (2014) standards for drinking water. According to the results of the dependent sample t-test conducted to test whether the difference between the

mean values of the sticker and laboratory values of the chemical parameters were statistically significant (Table 1), there was no significant difference in pH between the sticker and the laboratory mean values (p=0.278). The mean pH (8.05±0.27) was determined in the stickers, whereas the mean pH (7.89±0.21) was measured in the laboratory (Table 1). Therefore, packaged natural mineral water samples are alkaline in Sivas. One of the most important factors determining the valuable of water for the body is pH. In recent years, various drops and carbonates have been recommended to make drinking water alkaline. On this subject, a comment can be made by reading the pH value of water in stickers. Alkaline water to pH 9.5 means that it is healthier for the body. In other words, the pH value of the packaged waters should be from 7.1 to 9.5 for public health. There is no need to use such drops and carbonates to make the water alkaline. When buying packaged natural mineral water, the pH value of 7.1 and above should be preferred for drinking. Therefore, it is crucial to check the pH value of the packaged natural mineral water in stickers.

In this study, calcium cation ranged from 18.56 to 85.60 ppm were determined in sticker and laboratory findings, whereas magnessium cation ranged from 1.70 to 23.97 ppm were determined in sticker and laboratory findings for packaged natural mineral water samples (Table 1). Calcium and magnesium are very important for human health. Calcium and magnesium help the vessels and muscles work regularly. Insufficient intake of the calcium and magnesium may adversely affect health. Food is the main source of calcium and magnesium. The available proof shows that, due to food habits, many people in most countries are unable to obtain the recommended intake of one or both of these nutrients from their diet. Calcium and magnesium concentrations in drinking water vary significantly from one source to another. In some populations, rich mineral drinking water can contribute significantly to the total intake of these nutrients (WHO, 2009). For calcium and magnesium have not suggested any guideline value by the WHO (2017). According to the results of the dependent sample t-test conducted to test whether the difference between the mean values of the sticker and laboratory values of the chemical parameters were statistically significant (Table 1), the mean difference between the sticker and the laboratory values of the magnesium value was found to be statistically important at the 99 percent confidence level (t=-4.098; p<0.01). For magnesium in Sivas, the sticker mean value ($\bar{x}$=6,48) is lower than the mean of the laboratory value ($\bar{x}$=17,83). However, there was no significant difference in calcium between the sticker and the laboratory mean values (p=0.384).

According to the by ECDWD (1998), TWRHC (2005), EPA (2014), CDWQ (2017) and WHO (2017) standards of chloride anion should not exceed 250 ppm for drinking water. High levels of chloride cause a salty taste in water and beverages. The taste levels for the chloride are due to the associated cation. Chloride values exceeding 250 ppm increase the likelihood of taste detection. A guideline value is not suggested for chloride (WHO, 2017). In this study, chloride anion varied from 1.00 to 6.60 ppm in sticker and laboratory findings for packaged natural mineral water samples (Table 1). The results show that the chloride anion in Sivas is below the standard limit. According to the results of the dependent sample t-test conducted to test whether the difference between the mean values of the sticker and laboratory values of the chemical parameters were statistically significant (Table 1), there was no significant difference in chloride between the sticker and the laboratory mean values (p=0.154).

According to the ECDWD (1998), TWRHC (2005), CDWQ (2017) and WHO (2017) standards the permissible guideline value of fluoride anion should be 1.50 ppm for drinking water. Fluoride is an element that has harmful effects on the body in its deficiency and excess. Low amounts of fluoride have negative effects on dental health, while high amounts of fluoride accumulate in teeth and bones and cause a disease known as fluorosis. The most important feature of fluoride is the protective effect against dental caries. Addition of fluoride to water sources at levels above 0.6 ppm indicates a decrease in tooth decay in children. Optimum beneficial effects of fluoride are realized at about 1.0 ppm. The parametric level for the fluoridated supplies is 1.0 ppm. However, the parametric level for the supplies with naturally occurring fluoride is 1.5 ppm (EPA, 2014). Fluoride is widely used in areas of high sugar intake to reduce tooth decay. These are tablets, mouthwashes, toothpaste, varnishes or gels. In addition, fluoride can be added to table salt or drinking water to provide protection against tooth decay. The amounts added to the drinking water are generally between 0.5 and 1 ppm. Above 1.5 ppm increase the risk of dental fluorosis. Higher levels cause skeletal fluorosis (WHO, 2017). According to Dissanayake (1991), if fluoride falls below 0.5 ppm in drinking water, it causes dental caries; If fluoride is between 0.5 and 1.5 ppm, it is optimal for dental health; if fluoride is between 1.5 and 4 ppm, it causes fluorosis in the teeth; If fluoride is between 4 and 10 ppm, it causes dental and skeletal fluorosis and if fluoride is above 10 ppm, it causes crippling fluorosis. In this study, fluoride anion varied from 0.00 to 0.12 ppm in sticker and laboratory findings for packaged natural mineral water samples (Table 1). The results show that the fluoride anion in Sivas is below the standard permissible limit by ECDWD (1998), TWRHC (2005) and WHO (2017). According to the results of the dependent sample t-test conducted to test whether the difference between the mean

values of the sticker and laboratory values of the chemical parameters were statistically significant (Table 1), the difference of fluoride value between the sticker value and laboratory value was found to be statistically significant at 95 percent confidence level (t=2.712; p<0.05). For fluoride in Sivas, the sticker mean value ($\bar{x}$=0,06) is higher than the mean of the laboratory value ($\bar{x}$=0,01). Such a low fluoride level may adversely affect public health because it is necessary for the body. The mean value of fluoride of packaged natural mineral waters samples in Sivas was found to be remarkably lower than the proposed level by Dissanayake (0.5-1.5 ppm) the best for dental health. Since the mean value of fluoride anion is low in Sivas, the use of these packaged natural mineral waters as a continuous drinking water can lead to an increase in frequency of tooth decay for the population. This should be seen as a warning on the stickers, or the addition of fluoride to the water, providing the best standard of 0.5-1.5 ppm necessary for the inhibition of tooth decay.

According to the ECDWD (1998), TWRHC (2005) and EPA (2014) allows maximum allowable level of iron cation is 0.20 ppm for drinking water. The iron is between 0.5 and 50 ppm in natural fresh waters. Although turbidity and color can be present, there is usually no noticeable taste at iron levels below 0.3 ppm. A guide value for iron was not suggested (WHO, 2017). In this study, iron cation observed from 0.00 to 0.30 ppm in sticker and laboratory findings for packaged natural mineral water samples (Table 1). The results show that the iron cation in Sivas is below the standard limit. Table 1, indicates there was no significant difference in iron between the sticker and the laboratory mean values (p=0.099).

In this study, the potassium anion observed from 0.00 to 1.50 ppm in sticker and laboratory findings for packaged natural mineral water samples (Table 1). Potassium is important for health in humans. Potassium in drinking water is rarely found in alarming levels in drinking water. It was not necessary to create for potassium a health based guideline value in drinking water (WHO, 2017). Table 1, indicates there was no significant difference in potassium between the sticker and the laboratory mean values (p=0.175) in Sivas.

According to the ECDWD (1998), TWRHC (2005) and WHO (2017) standards have based 250 ppm as the highest acceptable level of sulphate anion. Sulfate may cause a significant taste in drinking water. High levels of sulfate can have a laxative effect on consumers. The taste disorder varies according to the associated cation. The taste disorder is minimal below 250 ppm. No value was recommended for the sulphate (WHO, 2017). In this study, the sulphate anion observed from 0.00 to 8.00 ppm in sticker and laboratory findings for packaged natural mineral water samples (Table 1). The results show that the sulphate anion in Sivas is below the standard limit for ECDWD (1998), TWRHC (2005) and WHO (2017). According to the results of the dependent sample t-test conducted to test whether the difference between the mean values of the sticker and laboratory values of the chemical parameters were statistically significant (Table 1), the mean difference between the sticker and the laboratory values of the sulphate value was determined statistically significant at the 99 percent confidence level (t=8.052; p<0.01). For sulphate in Sivas, the sticker mean value ($\bar{x}$=5,83) is higher than the mean of the laboratory value ($\bar{x}$=0,00).

Also, a narrow variability of price was determined in the all samples of Sivas that the 0.5 L price of packaged natural mineral water ranged from a minimum of 0.50 Turkish Lira to a maximum of 1.00 Turkish Lira, while the average with standart deviation was 0.71±0.24 Turkish Lira. Even though it brings a financial burden, packaged natural mineral waters are one of the appropriate ways to meet the water needs of consumers in Sivas.

Most of the mean sticker values were higher in packaged natural mineral water samples than the mean laboratory values (Table 1). The comparison of the analyzed and recorded data using the dependent sample paired t-test indicated that the sticker findings were significantly higher than the laboratory findings for Fluoride (t=2.712; p=0.042*) and sulfate (t=8.052; p=0.000**). Conversely, according to the results of the dependent t-test comparison, laboratory findings were significantly lower than the label findings for magnesium (t=-4.098; p=0.009**). However, the pH (t=1.217; p=0.278), calcium (t=0.955; p=0.384), chloride (t=1.680; p=0.154), iron (t=-2.022; p=0.099) and potassium (t=1.581; p=0.175) in the sticker findings were not determined statistically different from laboratory findings. The water quality properties in the stickers are normally for water obtained at the resource, but the resource can change according to the quality over time. These differences between sticker and laboratory concentrations of packaged natural mineral water components presumably indicate that they are affected by post-packaging conditions. Especially, considering the low and high temperature in Sivas province may be as low -36.4 ºC and high 38 ºC, it may affect the concentrations of water components in packaged natural mineral water. Therefore, this study shows that packaged natural mineral water needs to be more strictly controlled in determining and monitoring its quality. Relevant authorities should strictly monitor the post-packaging conditions and quality of packaged natural mineral water provided to consumers to protect public health. In addition, further studies on microbiological tests and analysis of heavy metals are recommended for packaged natural mineral water.

CONCLUSION

Natural mineral waters contain many minerals such as calcium, magnesium, chloride, fluoride, iron. In this way, many minerals needed by the body are taken with drinking water. Because of the mineral content, natural mineral waters are a positive contribution to human health. This study highlighted that all packaged natural mineral water samples sold in Sivas were appropriate for drinking. Results like pH, calcium, chloride, fluoride, iron, magnesium, potassium and sulphate were in the range proposed by the ECDWD (1998), TWRHC (2005), EPA (2014), CDWQ (2017) and WHO (2017). On the other hand, the fluoride content of packaged natural mineral waters in Sivas was found to be significantly lower than the recommended level by Dissanayake (<0.5 ppm), and causing dental caries. For this reason, it is beneficial to pay attention to this situation when buying packaged natural mineral water in the province of Sivas.

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