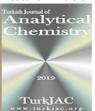
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Seasonal variations in tap water quality parameters in Çanakkale, Türkiye

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Abstract

Water pollutants are a major problem for the world. Especially, heavy metals are significant environmental pollutants due to their tendency to accumulate in organisms causing toxic effects on humans, animals, and aquatic organisms. Therefore, qualitative and quantitative analyses of pollutants are important. Especially the analysis of contaminants in tap water is important. In this study, unlike previous studies, physicochemical water quality parameters were also studied for the first time along with heavy metal analysis. Water-quality parameters (pH, temperature, conductivity, and dissolved oxygen) were determined in tap water by YSI 556 MPS, and other parameters such as biochemical oxygen demand (BOD) and chemical oxygen demand (COD) were determined by electrometric and titrimetric methods. Various anions were analyzed with the UV-VIS spectroscopy technique. Cd, Cr, Cu, Fe, Mn, Ni, Pb, and Zn were analyzed as pollutants by ICP-OES. Amounts of Cr, Cd, Ni, and Pb were found within detectable limits, but Cu, Fe, Mn, and Zn were found at 0.003 ppm, 0.173 ppm, 0.009 ppm, and 2.343 ppm, respectively. The pH of the tap water was in the interval between 6.64 and 7.12 (mean: 6.85 ± 0.12). Nitrite varied between 0.20 and 0.60 mg/L (average: 0.36 ± 0.001 mg/L) revealing that the tap waters exceeded the TSI standards for first quality drinking waters (0.20 mg/L). The data were evaluated using the relevant statistical analyses.

Keywords: Tap water, water quality, heavy metal, physicochemical, biochemical

1. Introduction

The development of industry and technology has provided better living conditions for humans, but previously unknown problems have emerged with the arrival of industry. Sometimes, wastewaters may contain high concentrations of heavy metals. After discharging these wastewaters, organisms in the receiving environment may be harmed. Also, wastewaters that have high concentrations of heavy metals may contaminate drinking waters, lake waters, or groundwater. It is estimated that approximately 80.0% of all diseases are caused by poor hygiene conditions and a lack of safe water. Millions of people die due to water pollution [1–4].

Metals that are denser than 5.00 g/mL are called heavy metals. More than 60 metals are included in this group, including Zn, Pb, Cd, Fe, Cu, Ni, and Co [5–7]. Heavy metals which are consumed by organisms via the food chain may cause bioaccumulation, and in some situations, there are toxic impacts when they exceed certain amounts [8–10]. Toxic heavy metals such as Ag,

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As, Ba, Cd, Cr, Pb, Mn, Hg, Ni, Se, V, and Zn are harmful to life [11]. Heavy metals with toxic properties enter the environment from various sources and constitute one of the most important causes of environmental pollution today [12]. Heavy metal accumulates in the body [8,9,13]. As, Cd, Cr, Ni, Pb, and Se are highly toxic, and have carcinogenic, mutagenic, and teratogenic effects [14]. Heavy metals create important problems in aquatic environments and for creatures living in these environments via the usage of natural resources and human activities [6–9,13,15].

The conductivity of substances that ionize in water depends on the temperature, salinity, and other water properties. Temperature influences the displacement speed of ions [16].

Determining the amount of dissolved oxygen in water is important both for living organisms and for determining whether industrial wastewaters have mixed with drinking waters or not. Dissolved oxygen (DO) is also a parameter used in the determination of the

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Fax +90 (286) 218 39 17 Received: April 30, 2022 Accepted: May 21, 2022 assimilation capacity of the receiving environment and pollution potential [17]. Chemical oxygen demand (COD) is one of the most used parameters in environmental pollution. The chemical oxygen demand of organic matter in water is mainly an indicator of the total amount of all organic materials [18]. Biochemical oxygen demand (BOD) in water is defined as the amount of oxygen required for the decomposition of organic matter used by microorganisms under aerobic conditions.

Sulfate is another anion widely known anion in water coming after bicarbonate and chloride. Sulfate in the soil passes into water systems over time. Among the water-insoluble sulfate salts, barium sulfate and calcium sulfate are the most resistant sulfate salts, and they are in the mid and low soluble groups. Sodium sulfate and magnesium sulfate have laxative effects in humans at 250 mg/L. Whereas animals are affected above the limit of 1000 mg/L. Sulfates also give water a bitter taste and form calcium sulfate and magnesium sulfate that precipitate in boiling water thereby their dissolved concentrations are very low in this type of water [19].

Nitrate and nitrite are commonly available in nature. Domestic and industrial wastes, especially nitrogen fertilizers commonly used in agriculture, contaminate soil and water systems in terms of nitrogen and so indirectly increase algal bloom levels in lakes, dams, and other freshwater resources which provide drinking water for humans' populations. In Türkiye, although there are many freshwater resources, most of these freshwater resources have been polluted with nitrogen from domestic, agricultural, and industrial wastewaters [20]. Freshwater resources such as the Atikhisar Dam in Çanakkale city are also reported to be exposed to pollution, especially in the mining industry.

Cyanide is an extremely volatile and toxic substance known as hydrocyanic acid. Hydrogen cyanide in the gas phase is colorless and has a bitter almond smell. Cyanide in the liquid phase is colorless, as in the gas phase. Cyanide is a toxic substance in the environment and used in rubber and fertilizer industries, the manufacture of plastics, gold, silver, certain chemicals, and pesticides. Apart from the non-tobacco smoke resulting from the combustion of - polymeric materials and structures containing nitrogen, cyanide is also found in some plants [21]. The upper limit value for cyanide is 0.200 mg/L in drinking water [22].

Heavy metals and bacteriological parameters were studied in tap water from Çanakkale (Türkiye) city [23]. Çanakkale tap water samples from 40 different points were collected between 20 April and 5 May 2007, and the water samples were analyzed for As, Pb, Cu, Zn Cd, and some microbiological parameters [23]. According to the study, Cd, Cu, Pb, and Zn were found to be under the

limit values (Cd: 0.95–11.9, Cu 0.004–0.580, Pb <0.08, and Zn 0.00–5.30 mg/L) [23]. In addition, according to microbiological analyses of the water samples which were completed using the membrane filter method, bacteria were found in 28 of 40 water samples, 18 of these contained coliform bacteria and one of them was *Escherichia coli* TS 266. Microbiological values for the water samples were higher than the Limit Values according to WHO, EPA, and EC [23]. In addition, there are a few studies about heavy metals combined with physicochemical parameters in the Strait of Çanakkale [24] and Biga Kocabaş River [25,26].

Although there are many studies on the analysis of heavy metals to determine the quality of drinking water in the Marmara region [27–29], there are a few studies on other quality parameters such as pH, dissolved oxygen (DO), biochemical oxygen demand (BOD) and chemical oxygen demand (COD), combined with heavy metals analysis in wastewater of Kocabaş Stream of Marmara basin [25,26].

Therefore, in this study, unlike previous studies, physicochemical (pH, temperature, conductivity, and DO, BOD, and COD water quality parameters in tap waters of the Çanakkale center were also studied for the first time along with heavy metal (Pb, Cd, Fe, Zn, Cu, Mn, Cr) analysis. Besides, nitrite (NO₂) and nitrate (NO₃) levels were studied in this study due to the toxic effect of nitrite.

2. Materials and Methods

2.1. Study area and period

The study area, Çanakkale, is located in the northwest of the Republic of Türkiye at 25°40'–27°30' east longitude and 39°27'–40°45' north latitude. In this study, a total of 80 tap water samples were analyzed. The tap water samples were collected seasonally (Spring: May 2012; Summer: July 2012; Autumn: October 2012; Winter: February 2013) from five different neighborhoods known as Esenler (St.ER), Cevatpaşa (St.CR), Ismetpaşa (St.IR) Kemalpaşa (St.KR) and Barbaros (St.BR) in Çanakkale province (Fig. 1).

2.2. Sampling and preservation

The tap water samples were taken 3 minutes after the tap was turned on. The water samples collected from taps were placed in clean polythene sample bottles (1.00 L) and stored in a deep freezer at -21.0 $^{\circ}$ C for measurements of sulfate, nitrite, and nitrate. For the analyses of cyanide anions, the water samples were stored in 250 mL PE flasks and preserved with sodium hydroxide solution (pH = 12.0) for ten days. The water samples for heavy metal measurements were preserved with nitric acid (HNO₃) and stored in clean polythene sample bottles

(250 mL) at 4.00 °C. In this study, a total of 80 samples collected from five sampling points were analyzed for each parameter. For samples at each station, three separate measurements were performed and the obtained values were averaged. Relative Standard Deviations (% RSD) of measurements were under 5.00%.



Figure 1. Sampling locations for drinking water quality parameters in Çanakkale, Türkiye

2.3. Reagents and apparatus

Analytical reagents used in all analyses were analytical grade (Merck, Germany). Double distilled deionized water (<0.100 µS/cm conductivity) was used throughout the experiments. In order to minimize contamination and interference, all the materials and glassware were carefully cleaned. The stock standard solution was prepared daily (AccuTrace, the reference Standard Merck).

2.4. Metal analysis using the ICP-OES method

Concentrations of heavy metals in tap waters were measured by a Perkin Elmer Optima 8000 Model (ICP-OES). Analyses of all samples were repeated three times. The ICP-OES device was calibrated before each Merck multi-element standard measurement via solution.

2.5. Temperature, conductivity, and pH

Water quality parameters such as temperature, pH, conductivity, and dissolved oxygen were measured with a water probe "YSI 556 MPS" and Hach Lange apparatus HQ40D in situ.

2.6. Sulfate, nitrite, and nitrate

Sulfate, nitrite, and nitrate anions were analysed according to the colorimetric method. A Hach-Lange DR2800 VIS spectrophotometer was used for the analyses.

2.7. Cyanide

Cyanide analyzed the was according spectrophotometric (colorimetric) method. For this, a Hach-Lange DR2800 VIS spectrophotometer was used. In the method, cyanide was converted to cyanogen chloride (CNCl) by the reaction of cyanide with chloramine-T at a pH of less than 8. After the reaction was completed color formation by the addition of pyridine-barbituric acid reagent was determined by the absorbance formed at 578 nm for the complex formed with pyridine-barbituric acid reagent and CNCl. To obtain colors of comparable intensity, it is essential to have the same salt content in both the sample and the standard [30].

2.8. Biochemical oxygen demand (BOD5)

For the BOD5 measurement, 250 mL tap water samples were placed in Winkler glass bottles (Wheaton-USA) and incubated in a dark location for 5 days.

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| Parameters | Device Used | Method | Reference* |
| Sulfate (mg/L) | Hach-Lange DR2800 Spectrophotometer | Turbidimetric Method at 420 nm wavelength | SM 4500-SO ₄ E. |
| Cyanide (mg/L) | Hach-Lange DR2800 Spectrophotometer | Colorimetric Method at 578 nm wavelength | SM 4500-CN- E. |
| Nitrite (mg/L) | Hach-Lange DR2800 Spectrophotometer | Colorimetric Method at 543 nm wavelength | SM 4500-NO ₂ - B. |
| Nitrate (mg/L) | Hach-Lange DR2800 Spectrophotometer | UV Spectrophotometric Method at 220–275 nm wavelength | SM 4500-NO ₃ - B. |
| BOD ₅ (mg/L) | Hach-Lange Limunescence Dissolved Oxygen Electrode | 5-Day BOD Test | SM 5210 B. |
| COD (mg/L) | Merck Pharo 100 Spectrophotometer | Closed Reflux, Colorimetric Method at 420 and/or 600 nm wavelength | SM 5520 D. |
| Temperature (°C) | TFA 30.1040 Thermemeter | Laboratory and Field Methods | SM 2550 B |
| рН | Hach-Lange pH-Meter | Electrometric Method | SM 4500 H+ B. |
| Conductivity (µS/cm) | Hach-Lange Conductivity Meter | Electrical Conductivity Method | SM 2510 B. |
| Dissolved Oxygen (mg/L) | Hach-Lange Limunescence Dissolved Oxygen Electrode | Limunescence Method | ASTM D888-12 |
| Heavy metals (mg/L) | ICP-OES Perkin Elmer Optima 8000" | ICP-OES Method | EPA 200.7 |

^{*}SM: Standard Methods for the Examination of Water and Wastewater. APHA, AWWA, WEF. 22nd Edition; ASTM: American Society for Testing and Materials

Table 2. Physicochemical parameters in the tap waters from Çanakkale, Türkiye

| Seasons | Sampling station | Temperature (°C) | pН | Conductivity (µS/cm) | Dissolved oxygen (mg/L) |
|---------|------------------|------------------|-----------------|----------------------|-------------------------|
| | Esenler Region | 10.1 | 6.98 | 360.0 | 7.60 |
| | Cevatpaşa Region | 9.4 | 7.01 | 327.0 | 7.30 |
| A7: 1 | Ismetpaşa Region | 7.3 | 6.95 | 362.0 | 8.40 |
| Winter | Kemalpaşa Region | 6.6 | 6.84 | 364.0 | 7.20 |
| | Barbaros Region | 7.9 | 6.72 | 364.0 | 6.90 |
| | Winter average | 8.3 ± 0.1 | 6.90 ± 0.12 | 355.0 ± 19.3 | 7.48 ± 0.17 |
| | Esenler Region | 12.7 | 6.76 | 412.0 | 7.10 |
| Spring | Cevatpaşa Region | 11.8 | 6.87 | 414.0 | 6.80 |
| | İsmetpaşa Region | 10.9 | 6.84 | 421.0 | 7.40 |
| | Kemalpaşa Region | 13.6 | 6.65 | 417.0 | 6.90 |
| | Barbaros Region | 11.8 | 6.89 | 431.0 | 6.80 |
| | Spring average | 12.2 ± 0.1 | 6.80 ± 0.12 | 419.0 ± 22.8 | 7.00 ± 0.16 |
| | Esenler Region | 20.1 | 6.91 | 530.0 | 6.90 |
| | Cevatpaşa Region | 18.7 | 6.99 | 526.0 | 6.40 |
| ummer | İsmetpaşa Region | 18.0 | 7.01 | 532.0 | 6.80 |
| ummer | Kemalpaşa Region | 21.2 | 7.12 | 483.0 | 6.10 |
| | Barbaros Region | 18.9 | 6.96 | 564.0 | 6.10 |
| | Summer average | 19.4 ± 0.2 | 7.00 ± 0.12 | 527.0 ± 28.2 | 6.46 ± 0.14 |
| | Esenler Region | 16.3 | 6.64 | 471.0 | 7.30 |
| | Cevatpaşa Region | 14.3 | 6.71 | 481.0 | 7.10 |
| | İsmetpaşa Region | 16.9 | 6.65 | 478.0 | 8.00 |
| Autumn | Kemalpaşa Region | 15.1 | 6.73 | 399.0 | 7.40 |
| | Barbaros Region | 14.5 | 6.79 | 452.0 | 7.10 |
| | Autumn average | 15.4 ± 0.2 | 6.70 ± 0.12 | 456.0 ± 24.8 | 7.38 ± 0.17 |
| | Annual average | 13.8 ± 0.2 | 6.85 ± 0.12 | 439.0 ± 23.8 | 7.08 ± 0.16 |

The BOD levels were analyzed by comparing the DO levels of a water sample taken immediately with the DO levels of tap water samples incubated in a dark location for 5 days. The difference between the two DO levels represents the amount of oxygen required for the decomposition of any organic material in the tap water samples. DO concentrations were measured using a dissolved oxygen test kit.

2.9. Chemical oxygen demand (COD)

For the chemical oxygen demand (COD), Winkler glass bottles (250 mL) acidified with hydrochloric acid were used. The tap water samples were preserved with sulphuric acid to a pH < 2 and maintained at 4 $^{\circ}\text{C}$ until analysis. Analyses of the samples began within 24 hours. In this application, quantitative analyses of COD were performed using the Hach-Lange DR2800 VIS

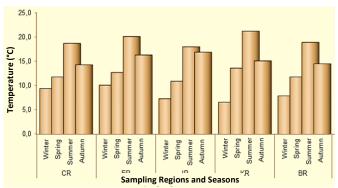


Figure 2. Seasonal and regional variations in temperatures in the tap waters of φ anakkale

spectrophotometer reading absorbance values between 200–400 nm and Merck Spectroquant COD mercury-free cell test.

2.10. Statistical analysis

Two-way analysis of variance (ANOVA) was used for statistical analysis.

2.11. Operation parameters for the methods

Devices and operation parameters for all methods are reported in Table 1.

3. Results and Discussion

3.1. Physicochemical parameters in the tap-waters

Results of the physicochemical parameter results such as temperature, conductivity, pH, and dissolved oxygen (DO) of the tap water samples which were collected from 5 different locations in Çanakkale are presented in Table 2. Moreover, drinking water quality limit values according to Turkish standard TS 266 and other international standards (limits) are presented in Table 3.

3.2. Temperature variations

The temperature variations in the tap water, which were taken from 5 various locations in Çanakkale, were given in Table 2 and Fig. 2. As expected, the temperature of the tap water samples from all sampling sources reached the highest levels in the summer.

Table 3. Turkish standard TS 266 and other international drinking water standards and limit values

| Parameters | Turkish Standards Istitute (TS 266; 2005) | Word Health Organization (WHO, 2011) | USA Environmental Protection Agency (EPA, 2008) | European Union (EC, 1998) |
|---------------------------------------|---|---|---|------------------------------|
| Temperature (°C) | 25.00 | - | - | - |
| pH | 6.50-9.50 | 6.50-8.00 | 6.50-8.50 | 6.50-9.50 |
| Dissolved Oxygen (DO) (mg/L) | 8.000 | - | - | - |
| Sulfate (mg/L) | 250.0 | 500.0 | 250.0 | 250.0 |
| Cyanide (mg/L) | 0.050 | 0.070 | 0.200 | 0.050 |
| Nitrite-N (mg/L) | 0.200 | - | - | - |
| Nitrate-N (mg/L) | 5.000 | - | - | - |
| Chemical Oxygen Demand (COD) (mg/L) | 25.00 | - | - | - |
| Biological Oxygen Demand (BOD) (mg/L) | 4.000 | - | - | - |
| Ni (mg/L) | 0.020 | 0.020 | - | - |
| Cd (mg/L) | 0.005 | 0.003 | 0.005 | 0.005 |
| Cr (mg/L) | 0.050 | 0.050 | 0.100 | 0.050 |
| Pb (mg/L) | 0.010 | 0.010 | 0.015 | 0.010 |
| NO ₃ - (mg/L) | 50.00 | 50.00 | 45.00 | 50.00 |
| Cu (mg/L) | 2.000 | 2.000 | 1.000 | 2.000 |
| Mn (mg/L) | 0.050 | 0.100 | 0.050 | 0.050 |
| Fe (mg/L) | 0.200 | 0.300 | 0.300 | 0.200 |
| TSS (mg/L) | - | 1000 | 500 | - |
| Zn (mg/L) | 2.000 | 3.000 | 5.000 | - |
| $NH_{4^+}(mg/L)$ | 0.500 | 1.500 | - | 0.500 |
| Hardness (as CaCO ₃) | - | 500.0 | - | - |
| Colour (PC Unit) | 20.00 | 15.00 | 15.00 | - |
| Turbidity (NTU) | 1.000 | 5.000 | 1.000 | 1.000 |

Both the highest (in summer: 21.20 °C) and the lowest temperature value (in winter 6.60 °C) were observed in the KR. However, while the seasonal temperature variations were high, the regional temperature variations were low (Table 2 and Fig. 2). When regional variations of average temperature in the tap waters during the year are examined, there were no important variations between sampling regions (Table 1). During the year, the annual average temperature in the tap waters was 13.80 ± 0.2 °C (Table 2). Both the average value and other values for temperature were acceptable according to Turkish standard TS 266 limit values (Table 3).

Although the drinking water temperature was not at a level that directly causes health problems, it is well known that as water temperature increases, the corrosive effect level increases on equipment within the water network system, and the solubility level of heavy metals increases. Therefore, temperature increases in drinking waters indirectly create a negative effect on human health, so monitoring of temperature levels of drinking waters is important for public health [31].

3.3. pH variations

pH variation in the tap water which was taken from 5 various locations in Çanakkale is presented in Table 2 and Fig. 3.

Seasonal variations of pH values of the Tap-Water in all regions showed that there was no linear correlation between samples. The highest pH value was detected in KR in winter (7.12) and the lowest value was detected in ER in autumn (6.64). The pH values measured in the tap water samples in Çanakkale were in the interval between minimum and maximum values of both national and international standards (Table 3 and Fig. 3). When both regional and seasonal variations in average pH in the tap waters during the year are examined, the values were under neutral pH value (pH: 7.00), except for summer values in the KR and IR (Table 3). During the year, all pH levels were under the neutral value for pH (7.00) (Fig. 3), and thereby the average pH value in samples was 6.85 ± 0.12 (Table 2). So, it can be concluded that the tap-waters of Çanakkale city were partly unhealthy in terms of pH or acidity.

Although the direct effect of low and high pH levels in drinking water on human health is generally unknown lower pH values are thought to treat many diseases, even cancer while acidic waters can cause cancer and various diseases directly.

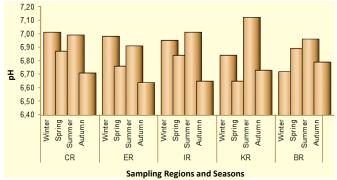


Figure 3. Seasonal changes in pH value in the tap waters of Çanakkale

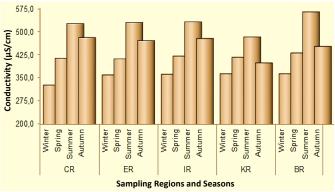


Figure 4. Seasonal changes of conductivity in the tap waters of Canakkale

However, there are some claims that while alkaline waters treat many diseases and even cancer, acidic waters cause various diseases, even cancer. But it is well known that acidity increases in drinking water both cause corrosion of equipment used in the water distribution system and increase the solubility of heavy metals conductivity and temperature. Considering this situation, although not directly, high acidity indirectly creates a negative effect on human health. Therefore, monitoring pH levels in drinking waters is important for public health [31].

3.4. Conductivity variations

The conductivity variations in the tap-water samples, which were collected from 5 different locations of Çanakkale, are presented in Table 2 and Fig. 4.

Conductivity levels reached their highest values in summer. The highest conductivity value was detected in BR in summer (564.0 μ S/cm) and the lowest value was detected in CR in winter (327.0 μ S/cm). The conductivity values measured in the tap water samples from Çanakkale, were in the interval between minimum and maximum values of both national and international standards (Table 2 and Fig. 4). When the regional variations of average conductivity in the tap waters during the year are examined, the values varied between 416.0 ± 22.1 in the KR and 453.0 ± 24.6 in the BR (Table 2). During the year, the annual average conductivity value in the tap waters was 439.0 ± 23.8 μ S/cm (Table 2).

An increase in conductivity causes corrosion and oxidation of equipment in the water distribution systems. Considering that the major source of heavy metals in drinking water is the corrosion and oxidation of water distribution equipment such as household plumbing, taps, and pipes, conductivity increases in drinking waters indirectly create a negative effect on human health. Therefore, monitoring conductivity values, in addition to temperature and pH measurements, in drinking waters is important for public health [31].

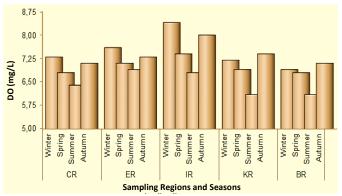


Figure 5. Seasonal changes in DO values in the tap waters of Canakkale

3.5. DO variations

DO variation in the tap-water samples, which were seasonally taken from 5 various points in Çanakkale, are presented in Table 2 and Fig. 5.

Seasonal variations in the DO values showed that there was an inverse (negative) correlation between both temperature and conductivity with DO values. When the samples had the lowest temperature and conductivity, DO concentrations were measured at the highest levels. The highest DO value was measured in the IR in winter (8.40 mg/L) and the lowest DO value was determined in BR and KR in summer (6.10 mg/L). The DO values measured in the tap water samples in Çanakkale, were in the interval between minimum and maximum values of both national and international standards (Table 2 and Fig. 5). When regional variations of average DO in the tap waters during the year are examined, the values were between 6.70 ± 0.15 in the BR and 7.65 ± 0.17 IR (Table 2). During the year, the annual average DO value in the tap waters was 7.08 ± 0.16 (Table 2).

High DO value has vital importance for life in the aquatic environment, and moreover, it has vital importance for public health in drinking water. High DO levels in drinking water are a good situation since it makes drinking water taste better. However, high DO speeds up corrosion of the water network systems, with high temperature, conductivity, and acidity levels. The level of oxygen in water is in close relation to temperature, pressure, and salinity. Many sources of drinking water such as lakes and ponds have anoxic conditions in the summer period, due to oxygen deficiency linked to water pollution and biological processes. Therefore, considering that there are positive and negative effects of low and high DO levels, it is desirable for public health that DO levels of drinking water are neither too low nor too high [32].

3.6. Seasonal changes of biochemical parameters in the tap waters

The biochemical parameter results such as sulfate, cyanide, nitrite, nitrate, BOD, and COD in the tap water

Table 4. Seasonal variations of sulfate, cyanide, nitrite, nitrate, BOD, and COD in tap waters from Çanakkale, (nd: not detected)

| Seasons | Sampling | Sulfate (mg/L) | Cyanide (mg/L) | Nitrite (mg/L) | Nitrate (mg/L) | BOD5 (mg/L) | COD (mg/L) |
|---------|------------------|-----------------|----------------|-----------------|-----------------|-----------------|-----------------|
| | Esenler Region | 73.0 | nd | 0.30 | 2.90 | 1.22 | 4.05 |
| | Cevatpaşa Region | 87.0 | nd | 0.40 | 2.10 | 3.08 | 12.80 |
| TA7* 1 | İsmetpaşa Region | 76.0 | nd | 0.30 | 3.00 | 2.41 | 8.26 |
| Winter | Kemalpaşa Region | 77.0 | nd | 0.40 | 2.60 | 2.64 | 8.50 |
| | Barbaros Region | 75.0 | nd | 0.30 | 2.40 | 3.02 | 9.28 |
| | Winter average | 77.6 ± 1.5 | nd | 0.34 ± 0.01 | 2.60 ± 0.05 | 2.47 ± 0.37 | 8.58 ± 0.81 |
| | Esenler Region | 96.0 | nd | 0.40 | 3.40 | 1.61 | 6.12 |
| | Cevatpaşa Region | 91.0 | nd | 0.30 | 3.40 | 2.84 | 10.1 |
| , . | İsmetpaşa Region | 82.0 | nd | 0.40 | 3.10 | 1.76 | 5.12 |
| Spring | Kemalpaşa Region | 92.0 | nd | 0.20 | 2.40 | 1.43 | 5.54 |
| | Barbaros Region | 90.0 | nd | 0.20 | 2.60 | 3.33 | 10.1 |
| | Spring average | 90.2 ± 1.7 | nd | 0.30 ± 0.01 | 2.98 ± 0.05 | 2.19 ± 0.32 | 7.40 ± 0.69 |
| | Esenler Region | 116.0 | nd | 0.30 | 4.10 | 1.88 | 5.12 |
| | Cevatpaşa Region | 103.0 | nd | 0.30 | 3.70 | 2.71 | 11.2 |
| | İsmetpaşa Region | 96.0 | nd | 0.30 | 3.80 | 1.93 | 7.12 |
| ummer | Kemalpaşa Region | 108.0 | nd | 0.20 | 3.60 | 3.01 | 6.24 |
| | Barbaros Region | 103.0 | nd | 0.40 | 3.70 | 2.36 | 8.16 |
| | Summer average | 105.2 ± 2.0 | nd | 0.30 ± 0.01 | 3.78 ± 0.07 | 2.38 ± 0.35 | 7.57 ± 0.71 |
| | Esenler Region | 101.0 | nd | 0.50 | 3.20 | 0.32 | 2.06 |
| | Cevatpaşa Region | 96.0 | nd | 0.60 | 3.20 | 1.74 | 8.04 |
| | İsmetpaşa Region | 86.0 | nd | 0.40 | 2.90 | 2.69 | 9.14 |
| Autumn | Kemalpaşa Region | 101.0 | nd | 0.50 | 3.40 | 2.06 | 4.68 |
| | Barbaros Region | 84.0 | nd | 0.40 | 3.10 | 1.99 | 7.13 |
| | Autumn average | 93.6 ± 1.8 | nd | 0.48 ± 0.02 | 3.16 ± 0.06 | 1.76 ± 0.26 | 6.21 ± 0.64 |
| | Annual average | 91.70 ± 1.74 | nd | 0.36 ± 0.01 | 3.13 ± 0.06 | 2.20 ± 0.33 | 7.44 ± 0.71 |

Table 5. Average values for sulfate, cyanide, nitrite, nitrate, BOD and COD in tap waters from Çanakkale, during the year (nd: not detected)

| Stations | Sulfate (mg/L) | Cyanide (mg/L) | Nitrite (mg/L) | Nitrate (mg/L) | BOD5 (mg/L) | COD (mg/L) |
|-------------------|----------------|----------------|-------------------|-----------------|-----------------|-----------------|
| ER | 96.5 ± 1.8 | nd | 0.40 ± 0.01 | 3.40 ± 0.06 | 1.26 ± 0.19 | 4.34 ± 0.41 |
| CR | 94.3 ± 1.8 | nd | 0.40 ± 0.01 | 3.10 ± 0.06 | 2.59 ± 0.39 | 10.5 ± 0.99 |
| IR | 85.0 ± 1.6 | nd | 0.30 ± 0.01 | 3.20 ± 0.06 | 2.20 ± 0.33 | 7.41 ± 0.70 |
| KR | 94.5 ± 1.8 | nd | 0.30 ± 0.01 | 3.00 ± 0.06 | 1.27 ± 0.34 | 6.22 ± 0.59 |
| BR | 88.0 ± 1.7 | nd | 0.30 ± 0.01 | 3.00 ± 0.06 | 2.68 ± 0.40 | 8.67 ± 0.88 |
| Annual Average | 91.7 ± 1.7 | nd | 0.360 ± 0.001 | 3.13 ± 0.06 | 2.20 ± 0.33 | 7.44 ± 0.71 |

samples, which were collected from 5 different locations in Çanakkale, are presented in Table 4 and Table 5.

3.7. Sulfate variations

The sulfate variations in the tap water, which were seasonally measured from 5 various locations in, were given in Table 4 and Fig. 6.

Sulfate is one of the most abundant anions in water. Sodium sulfate and magnesium sulfate have laxative effects in humans. Therefore, the sulfate concentration has an upper limit of 250 mg/L [19]. The highest sulfate concentration was detected in ER in the summer (116.0 mg/L) and the lowest sulfate concentration was determined in ER in winter (73.0 mg/L) among the Çanakkale city tap waters. Compared to TS 266 and other international drinking water standards, the sulfate

concentration findings were below the upper limit values (Table 3).

3.8. Nitrite and nitrate variations

The nitrite and nitrate variations in the tap-water samples, which were collected seasonally from 5 different locations in Çanakkale, are presented in Table 4, Fig. 7, and Fig. 8.

While the lowest nitrite concentration (0.20 mg/L) in this study was measured in KP in spring and summer and in BR in spring, concentration (0.60 mg/L) was found in CR in autumn. As is known, according to the drinking water quality classification of the TSI, nitrite values above 0.50 mg/L and over indicate 4th quality drinking waters.

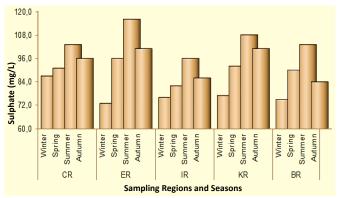


Figure 6. Seasonal changes of sulfate concentrations in the tap waters of Canakkale

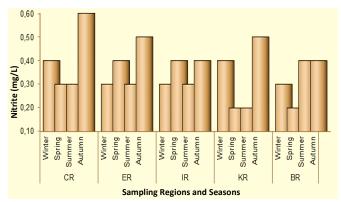


Figure 7. Seasonal changes in nitrite concentrations in the tap waters of Çanakkale

Average nitrite values in Table 4 ($0.360 \pm 0.001 \text{ mg/L}$) revealed that the tap waters from Çanakkale exceeded the TSI standards for first quality drinking waters (nitrite upper limit value: 0.200 mg/L) (Table 3). In other words, the nitrite findings were high above the upper limit values according to TS 266 drinking water standards (Table 3). It is known that nitrite interacts with hemoglobin and the formation of methemoglobin occurs in this way. As a result, the oxygen-carrying capacity of the blood decreases with the increase of nitrite in the blood [33]. It is known that nitrite has more toxicity than nitrate. For instance, it was reported that LD50 values of sodium nitrite are between 85 and 220 mg per kilogram of body weight in mice and rats [34,35].

The highest nitrate concentration was determined in ER in summer (4.10 mg/L) and the lowest nitrate concentration was determined in CR in winter (2.10 mg/L). The average nitrate (NO₃-) value in Table 4 (3.13 \pm 0.06 mg/L) revealed that the tap waters were lower than the upper Limit Values according to TS 266 Standard (Table 3).

As it is known, nitrate and nitrite are part of the nitrogen cycle and nitrate is the final product of the biochemical oxidation of nitrogen. Nitrate (NO₃-) is found in stable forms of combined nitrogen for systems rich in oxygen. Therefore, NO₃- does not have a direct toxic effect. NO₃- can be reduced by microbial action.

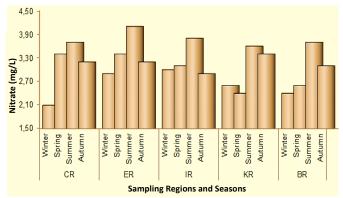


Figure 8. Seasonal changes in nitrate concentrations in the tap waters of Canakkale

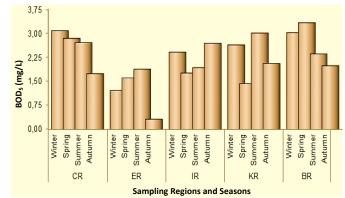


Figure 9. Seasonal changes in BOD in the tap waters of Canakkale

For instance, NO₃- is transformed into harmful nitrite ions by the activity of bacterial nitrate reductase [33]. Sometimes, the biochemical reaction can reduce nitrite ions (NO₂-) to different compounds or oxidize them to NO₃-[36].

3.9. BOD variations

The biological (biochemical) BOD and COD variations of the tap-waters, which were seasonally taken from 5 various locations of Çanakkale, are presented in Table 4, Fig. 9, and Fig. 10.

BOD is also called biological oxygen demand. BOD refers to the amount of organic matter which can be decomposed biologically under aerobic conditions at a certain temperature over a specific period [37,38]. The BOD value is most stated in terms of mg of oxygen consumed per liter of water sample during 5 days of incubation in the dark at 20.0 °C and is often used as a parameter for the degree of organic pollution in the water [37]. In this study, the highest BOD value was determined in BR in spring (3.33 mg/L) and the lowest value of BOD was determined in ER in autumn (0.32 mg/L) (Table 4 and Fig. 9).

The values for BOD vary proportionally with the values for COD. BOD is similar in function to COD, since both measures the level of organic compounds in water.

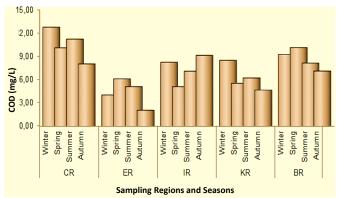


Figure 10. Seasonal changes in COD in the tap waters of Canakkale

However, because it measures everything that can be chemically oxidized, COD is less specific than BOD which refers only to levels of biodegradable organic matter. The highest value of COD was determined in CR in winter (12.80 mg/L) and the lowest value was determined in ER in autumn (2.06 mg/L) (Table 4 and Fig. 10). Most pristine rivers which have drinking water quality generally have BOD under 1.00 mg/L. Moderately polluted rivers may have BOD values in the range of 2-8 mg/L. Rivers are generally considered severely polluted when BOD values exceed 8.00 mg/L [39]. Both in this framework and according to national and international water standards (Table 3), the tap (drinking) waters of Çanakkale are moderately polluted in view of the average values for BOD $(2.20 \pm 0.33 \text{ mg/L})$ and COD $(7.44 \pm 0.76 \text{ mg/L})$ (Table 4, Fig. 9, and Fig. 10).

3.10. Seasonal variations of heavy metals in the tap waters

Seasonal and regional variations of heavy metals (Pb, Cd, Cr, Ni, Fe, Mn, Cu, Zn) in the tap water samples, which were collected from 5 different locations in Çanakkale, are presented in Table 6 and Table 7. Their seasonal variations are also illustrated in Fig. 11–Fig. 14.

3.11. Iron (Fe) variations

Fe variations in the tap waters which were seasonally taken from 5 different locations of Çanakkale, were given in Table 6 and Fig. 11.

During the study, minimum and maximum Fe concentration values were found between nd and 0.173 mg/L (Table 6). The highest Fe concentration value was found in IR and BR compared to other sampling regions (Fig. 11). The highest Fe value (0.173 mg/L) detected in the BR in spring was below the limit value in the TS 266 and other international drinking water standards such as WHO, EPA, and European Union (EC) (Table 3). In conclusion, according to the average Fe value in the drinking waters of Çanakkale, Türkiye (Fe: 0.040 \pm 0.055), there was no risk in terms of human health in the study area.

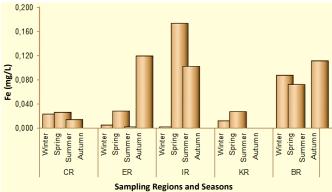


Figure 11. Seasonal changes in Fe concentrations in the tap waters of Canakkale

The average Fe amount in rivers was reported to be 0.70 mg/L. The concentrations in anaerobic groundwater are usually between 0.50 and 10.0 mg/L, but these concentrations can sometimes reach over 50.0 mg/L [39]. In the study area, even the highest Fe concentration (0.170 mg/L) did not reach this upper limit value (0.30 mg/L) for drinking waters. Fe is known to be an essential element for the human body. The minimum daily requirement for the element varies between 10 and 50 mg/day, depending on age, sex, physiological activity, and bioavailability [40,41].

3.12. Manganese (Mn) variations

Mn variations in the waters, which were collected from various locations in Çanakkale, were given in Table 6 and Fig. 12.

Mn is abundant in the earth's crust [42]. Mn is an essential element for both humans and animals because it is required for the functioning of many cellular enzymes (e.g., manganese superoxide dismutase, pyruvate carboxylase) and can activate many others such as kinases, decarboxylases, hydrolases, and transferases [43]. Manganese ion species are Mn²⁺, Mn⁴⁺ or Mn⁷⁺ [44].

During the study, minimum and maximum Mn concentrations varied from nd to 0.009 mg/L (Table 6). Mn concentrations were higher in IR and BR than in any other sampling region (Fig. 12). The highest Mn value (0.009 mg/L) detected in the IR region in spring (0.009 mg/L) was under the limit value of the WHO, USA, EPA, and EC (Table 3) and is suitable for human health.

It is known that Mn occurs naturally in many surface and ground water sources and soils which may enter these waters. Ambient manganese concentrations in fresh waters typically vary between 0.01 and 2.00 mg/L [45].

3.13. Copper (Cu) variations

Cu variations in water, which were seasonally taken from 5 various places in, were given in Table 6 and Fig. 13.

Table 6. Variations of metal concentrations in the tap-waters of Çanakkale, (nd: not detected)

| Seasons | Sampling | Pb | Cd | Cr | Ni | Fe | Mn | Cu | Zn |
|-----------|-----------|----|----|----|----|-------------------|-------------------|-------------------|-------------------|
| | Esenler | nd | nd | nd | nd | 0.005 | 0.003 | 0.002 | 0.262 |
| | Cevatpaşa | nd | nd | nd | nd | 0.023 | 0.003 | 0.001 | 0.087 |
| Winter | İsmetpaşa | nd | nd | nd | nd | 0.002 | 0.003 | 0.001 | nd |
| winter | Kemalpaşa | nd | nd | nd | nd | 0.012 | 0.003 | 0.003 | nd |
| | Barbaros | nd | nd | nd | nd | 0.087 | 0.006 | 0.001 | 0.229 |
| | Winter | nd | nd | nd | nd | 0.026 ± 0.035 | 0.004 ± 0.001 | 0.002 ± 0.001 | 0.116 ± 0.124 |
| | Esenler | nd | nd | nd | nd | 0.028 | 0.003 | nd | 0.489 |
| | Cevatpaşa | nd | nd | nd | nd | 0.026 | 0.002 | nd | 0.058 |
| Coning | İsmetpaşa | nd | nd | nd | nd | 0.173 | 0.009 | nd | 1.599 |
| Spring | Kemalpaşa | nd | nd | nd | nd | 0.027 | 0.002 | nd | nd |
| | Barbaros | nd | nd | nd | nd | 0.072 | 0.004 | nd | 0.052 |
| | Spring | nd | nd | nd | nd | 0.065 ± 0.063 | 0.004 ± 0.003 | nd | 0.440 ± 0.677 |
| | Esenler | nd | nd | nd | nd | 0.002 | 0.001 | nd | 0.084 |
| | Cevatpaşa | nd | nd | nd | nd | 0.014 | 0.001 | nd | 0.076 |
| Summer | İsmetpaşa | nd | nd | nd | nd | 0.102 | 0.008 | nd | 2.343 |
| Julilliei | Kemalpaşa | nd | nd | nd | nd | nd | nd | nd | 0.016 |
| | Barbaros | nd | nd | nd | nd | nd | nd | nd | 0.167 |
| | Summer | nd | nd | nd | nd | 0.024 ± 0.044 | 0.002 ± 0.003 | nd | 0.537 ± 1.011 |
| | Esenler | nd | nd | nd | nd | 0.119 | 0.002 | nd | 1.06 |
| | Cevatpaşa | nd | nd | nd | nd | nd | nd | nd | 0.084 |
| Autumn | İsmetpaşa | nd | nd | nd | nd | nd | nd | nd | 1.246 |
| | Kemalpaşa | nd | nd | nd | nd | nd | nd | nd | 0.031 |
| | Barbaros | nd | nd | nd | nd | 0.111 | 0.001 | nd | 0.401 |
| | Autumn | nd | nd | nd | nd | 0.046 ± 0.063 | 0.001 ± 0.001 | nd | 0.564 ± 0.560 |
| | Averages | nd | nd | nd | nd | 0.040 ± 0.052 | 0.003 ± 0.003 | 0.001 ± 0.001 | 0.414 ± 0.644 |

Table 7. Average values for metal amounts in the tap-waters of Çanakkale, Türkiye during the year (nd: not detected)

| Stations | Pb (mg/L) | Cd (mg/L) | Cr (mg/L) | Ni (mg/L) | Fe (mg/L) | Mn (mg/L) | Cu (mg/L) | Zn (mg/L) |
|----------------|-----------|-----------|-----------|--------------|-------------------|-------------------|--------------------|--------------------|
| ER | nd | nd | nd | nd | 0.039 | 0.002 | 0.001 | 0.474 |
| CR | nd | nd | nd | nd | 0.016 | 0.002 | nd | 0.076 |
| IR | nd | nd | nd | nd | 0.069 | 0.005 | nd | 1.297 |
| KR | nd | nd | nd | nd | 0.010 | 0.001 | 0.001 | 0.012 |
| BR | nd | nd | nd | nd | 0.068 | 0.003 | 0.001 | 0.212 |
| Annual Average | nd | nd | nd | nd | 0.040 ± 0.055 | 0.003 ± 0.001 | 0.0004 ± 0.001 | 0.4142 ± 0.524 |

In this study, Cu concentrations of the tap water in Çanakkale were found between nd and 0.003~mg/L (average: $0.001~\pm~0.001~\text{mg/L}$). The highest Cu concentration was determined in KR in winter with a level of 0.003~mg/L (Table 6 and Fig. 13). Fig. 13 revealed that Cu concentrations in KR (0.003~mg/L) and partly in ER (0.002~mg/L) were much higher than the values in any other region. Even these highest Cu values during the study were much lower than the limit value (2.00~mg/L) in the TS 266, WHO, and EPA.

Cu is present in surface water and groundwater in fixed concentrations [46]. Cu concentration in surface waters is 0.0005 to 1.000 mg/L in many studies in the USA [46]. In UK, the average Cu concentration in the river-stour was between 0.003 and 0.019 mg/L (average: 0.006 mg/L). Background concentration based on an upper control station was 0.001 mg/L. This six-fold increase in copper levels was due to the effect of a sewage treatment plant found downstream [47]. Cu levels varied between 0.0008 and 0.010 mg/L in an unpolluted location on the River Periyar in India [47]. Finally, Cu concentrations in drinking water vary widely

due to different water physicochemical characteristics [47–50].

On the one hand, the concentration of Cu is to be low [46, 51,52]. On the other hand, the Cu amount in drinking water is increasing [19]. Although Cu concentrations in our tap waters are much lower and suitable in terms of human health, the tap water in our region is slightly acidic (pH: 6.85 ± 0.12).

3.14. Zinc (Zn) variations

Zn variations in the tap-waters, which were seasonally taken from 5 various locations in Çanakkale were given in Table 6 and Fig. 14.

Zn is found in small amounts in almost all volcanic rocks. The fundamental zinc minerals are in the form of sulfides such as sphalerite and wurtzite [53]. The natural zinc level of the soil is generally between 1 and 300 mg/kg [54]. The concentration of Zn is usually under 10.0 $\mu g/L$ in surface waters and between 10–40 $\mu g/L$ in groundwaters [53].

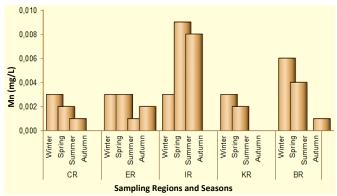


Figure 12. Seasonal changes in Mn concentrations in the tap waters of Canakkale

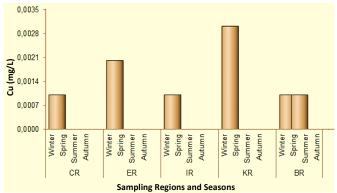


Figure 13. Seasonal changes of copper concentrations in the tap waters of Çanakkale

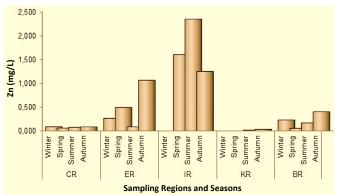


Figure 14. Seasonal changes in zinc concentrations in the tap waters of Çanakkale

In tap water, the Zn concentration can be much higher than usual as a result of the leaching of Zn from piping and relevant equipment [54]. The most corrosive waters in terms of Zn are ones that have low pH, high CO₂ content, and low mineral salts. In a study including 67% of public water supplies in Finland, the average Zn concentration in water samples taken from upstream and downstream parts of the water networks was below 20.0 μ g/L. There were much higher concentrations reaching 1.10 mg/L in tap water [55]. Even Zn levels in groundwater were much higher than in surface waters. For instance, there were higher concentrations of Zn reaching 24 mg/L in waters pumped from about 6000 wells in Finland [56].

In our study, the highest Zn concentration was determined in IR in summer with a level of 2.343 mg/L (Table 6 and Fig. 14). Fig. 14 revealed that Zn concentrations in IR and partly in ER were much higher than the values in any other region. The highest Zn value during the study was above the limit value (2.00 mg/L) for TS 266. However, the average value of Zn (0.414 \pm 0.644 mg/L) was below the limit values for one national (TS 266) and two international standards of the WHO and EPA. Therefore, Zn levels in the tap water in Çanakkale, Türkiye were within the range of acceptable values. Zn concentrations in the tap water samples in Çanakkale (0.414 \pm 0.644 mg/L) were much lower than the concentrations in tap waters from Finland (1.10 mg/L) [55].

3.15. Other toxic heavy metals

During the study, concentrations of Pb, Cd, Cr, and Ni, which are toxic heavy metals for human health, were not found at measurable values (not detection: nd). Considering that toxic heavy metal concentrations were found not detected (nd), there are no health risks for tap waters in Çanakkale for some metal amounts.

4. Conclusion

In this study, unlike previous studies, physicochemical water quality parameters were also studied for the first time along with heavy metal analysis. Results of this study about tap water quality parameters in Çanakkale, Türkiye, unfavorable situations were not detected in terms of physicochemical parameters, except for pH and nitrite. Although pH values measured in the tap water samples from Çanakkale were in the interval between minimum and maximum values of both national and international standards, the average pH value of the drinking waters (6.85 \pm 0.12) was under the neutral pH value (pH: 7.00) except for summer values in the KR and IR (Table 3 and Fig. 3). According to the drinking water quality classification of TS 266, nitrite values higher than 0.50 mg/L represent 4th quality drinking waters. The average nitrite value in this study (0.36 \pm 0.01 mg/L) revealed that the tap waters in Çanakkale exceeded the TSI standards for first quality drinking waters (Table 3). Therefore, the study showed that Çanakkale tap waters were unhealthy in terms of acidity and nitrite ions. As it is known, the presence of even a low nitrite level in drinking water reduces the quality of drinking water

Considering toxic heavy metals such as Pb, Cd, Cr, and Ni and non-toxic metals such as Fe, Mn, and Zn, it is clear that tap water in Çanakkale does not pose any health risks to human health. To summarize, heavy metal contamination, exceeding national or international

standards, was not found in the main water sources in Çanakkale province. However, the findings revealed that although heavy metal concentrations in our tap waters were very low and there was no risk in view of human health, it is necessary to consider that there may be an increase in heavy metal concentrations in the water network system, due to the fact that the tap-waters in our region are slightly acidic (pH: 6.85 ± 0.12). An increase in acidity and temperature is known to increase heavy metal solubility. As it is known that the major source of heavy metals such as lead, iron, copper, and zinc in drinking water is corrosion of household plumbing, taps, and water distribution pipe equipment. Water absorbs a lot of heavy metals such as copper and iron by leaching from plumbing materials such as pipes, fittings, and brass faucets. The number of heavy metals in the water depends on the types and amounts of minerals in the water, how long water stays in the pipes, and the water temperature, and acidity.

Finally, we suggest the public use cold water coming from the taps after running for one minute for drinking and cooking in order to decrease the level of some heavy metals that accumulate when household plumbing is not in use as heavy metals from plumbing corrosion can accumulate overnight. Moreover, because the dissolution of heavy metals is much easier in hot water than in cold water, hot water consumption should be limited, and it should not be used to prepare food for babies.

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Author Contributions

Hasan Kaçar: Collected the tap water samples and performed the analysis.

Selehattin Yılmaz: Conceived the analysis and wrote the paper.

Muhammet Türkoğlu: Performed statistical analysis and wrote the paper.

Murat Sadıkoğlu: Provided scientific contribution.

Conflicts of Interest

The authors declare no conflict of interest.

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