



Effect on human health of the arsenic pollution and hydrogeochemistry of the Yazır Lake wetland (Çavdır-Burdur/Turkey)

Simge Varol¹ · İlknur Köse²

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Abstract

In this study, the physicochemical parameters, major ions and arsenic (As) contents of water resources in the Yazır lake wetland, were evaluated. In addition, water resources in this region were investigated from the point of water quality and health risk assessment. Thirty water samples were collected from the area in dry and wet seasons. Ca-Mg-HCO₃ and Ca-HCO₃ were the dominant water types. The Gibbs diagram suggests that most of the samples fall in rock-dominance zone, which indicates the groundwater interaction between rock chemistry. When compared to drinking water guidelines established by World Health Organization and Turkey, much greater attention should be paid to As, Fe, and Mn through varied chemicals above the critical values. According to the pH-ORP diagram, the predominant species is arsenate (H₂AsO₄⁻²). The high concentrations of As in the surface water and groundwater are related to oxidative and reductive dissolution reaction of Fe and Mn hydroxides within the Kızılcadağ ophiolite and melange. In addition, the seasonal changes in As concentrations depend on the increase in pH of water samples. The major toxic and carcinogenic chemical within water samples is As for groundwater and surface water. From the results of hazard index, it is verified that As which is taken by ingestion of water was the main contaminant, and toxic human risk in the study area. The obtained results will help define strategies for As problems in the water resources in future.

Keywords Hydrogeochemistry · Yazır Lake · Wetland · Arsenic · Health risk assessment

Introduction

The *water* is one of the main substances responsible for life on earth. There is now water scarcity in many parts of the world. The water constrained by increased agriculture, industry, and domestic use is an economically challenging factor for countries. In addition to these, desertification and erosion, caused by climate change in watersheds, increase the seriousness of these effects (Venkatramanan et al. 2015). Besides, with the increase in population who need water for drinking and

irrigation, water demand is increasing. Due to the inadequacy of existing surface waters, the importance of groundwater increases exponentially.

Knowing the chemical properties of groundwater is important for determining the suitability of water for use of drinking, domestic, irrigation, and industrial purposes. Interaction of the aquifer minerals with groundwater is important in the formation of groundwater chemistry. Also, the quality of groundwater is affected from the geological formations and anthropogenic activities (Kelepertsis 2000; Siegel 2002; Sullivan et al. 2005; Stamatis 2010; Gnanachandrasamy et al. 2015). In some cases, concentrations of toxic elements or bacteriological contaminants may increase in the water, which may have adverse effects on human health (Gnanachandrasamy et al. 2015). Depending on this, more than 50% of diseases and deaths in developing countries are associated with water pollution (UNESCO 2007; Nagarajan et al. 2010). One of the most important water pollution sources is arsenic (As). As species in surface water and groundwater have become worldwide health problems due to their carcinogenic and many other toxic features (Cho et al. 2011). As which is included

Responsible editor: Philippe Garrigues

✉ Simge Varol
simgevarol@sdu.edu.tr

İlknur Köse
ilknur_kafas@hotmail.com

¹ Water Institute, Suleyman Demirel University, Isparta, Turkey

² Department of Geology Engineering, Suleyman Demirel University, Isparta, Turkey

in the Group A classification by the United States Environmental Protection Agency (USEPA) is an important carcinogen.

As which threatens human health is increasing gradually in its water cycle today. This situation seriously increases the threats to human health. Researches on the effects of As on health have showed that As in drinking water mainly causes skin cancer, but it also causes wide variety of other cancer types like liver, bladder, and lung cancers. *Inorganic* As is more toxic than *organic* As (ATSDR 2000; Nguyen et al. 2009).

The water resources in the Yazır Lake wetland are mainly used as drinking water by the local people. In addition, the water resources in the area are also used in different aims for domestic and irrigation purposes. For this reason, this study is also very important in terms of public health. Depending on this, the hydrogeological and hydrogeochemical data of Yazır Lake wetland (Çavdır/Burdur) were investigated and used to determine the main factors and mechanisms controlling the chemistry of water in the area. We also aimed to determine the surface and groundwater quality and to determine the spatial interpolation of the major physical and chemical parameters of the water samples in the study area. In addition, we aimed to determine the extent of the As problem, qualitative assessment of health risk, and the concentrations of As and the provenance of As in groundwater in the study area.

Materials and methods

Study area

The Yazır Lake wetland (Çavdır/Burdur) is located in the southwest of Turkey (Fig. 1), and it has a 194.78-km² watershed area (Köse 2017). Generally, climate of the plain and its vicinity are affected from Mediterranean climate zone in the middle Anatolian climate zone. The mean rainfall is 406.08 mm and the mean evapotranspiration is 337.55 mm in the study area (Köse 2017). Kocadere stream is the most important surface flow in the study area. In addition, Yazır Lake is situated in the middle of the area and irrigation water is supplied from this lake. The drinking water of the settlement areas is supplied from the spring waters in the study area. Also, the irrigation waters are supplied from the well water and surface flows.

Geological and hydrogeological setting

The lithological formations and the duration of water contact with these formations determine the chemical composition of the surface water and groundwater. For this reason, firstly, lithological formations and their hydrogeological properties in the study area have been determined. The lithological units

in the study area were investigated in two groups. These are units settled as autochthonous and allochthonous (Köse 2017; Fig. 1). The allochthonous units are Marmaris peridotite (Çapan 1980); Kızılcadağ ophiolite and melange (Poisson 1977); Orhaniye formation which consists of dolomitic limestones and layered chert members (Meşhur et al. 1989); Dutdere limestone (Şenel et al. 1989; Bilgin et al. 1990); Karanasıflar formations which composed of limestone, claystone, and sandstone (Şenel et al. 1989); Yuvedağ formation which consists of neritic and dolomitic limestones; and Çatlıca formation which consists of limestone, dolomitic limestone, and cherty limestone. The autochthonous units in the study area are the Çameli formation which consists of claystone, sandstone, conglomerate, and limestones (Erakman et al. 1982) with also alluvium, slope debris, and accumulation. The formation of the geological structure of Yazır Lake wetland basin is related to tectonic activities (Koçyiğit et al. 2000).

The lithological units in the study area have different hydrogeological characteristics. These units are grouped according to the ability to keep water on area as impermeable (aquifuge), semipermeable (aquitard-1, aquitard-2, aquitard-3), permeable-1 (granular aquifer), and permeable-2 (karstic aquifer). Units with similar hydrogeological properties were collected in the same group. These units and their properties are given in Table 1. The alluvium, which is thought to be an important aquifer in the basin, has an area of about 34.84 km² and is named a granular aquifer. Karstic aquifer is the other important aquifer in the study area. The most of water resources in the study area are obtained from the olistoliths within Kızılcadağ ophiolite and melange (Fig. 1) (Köse 2017). The seasonal variation of groundwater level in the study area is related to two types of factors. These are natural factors such as precipitation, evaporation, stream, artificial factors such as withdrawal of groundwater from wells, and recharge by irrigation from Kozağacı dam. The main source of groundwater in the study area is precipitation waters. The depth to groundwater table varies between 3.70 and 75.00 m in November 2016 and 1.00 and 40.00 m in April 2017 (Köse 2017).

Sampling and analysis

In the research area, total 30 water samples from wells (5 samples), springs (8 samples), stream (1 sample), and lake water (1 sample) were analyzed in October 2016 (dry period) and April 2017 (wet period) for the determination of their major chemical characteristics. The geographic details of these sample locations and region are presented in Table 2. The water samples were collected in two plastic bottles, pre-washed with 0.5% nitric acid (HNO₃), and deionized water, from each sampling point. During sampling, two bottles were filled with water from each spot, filtered, and add few drops of HNO₃ in one water sample; pH of the samples achieved lower

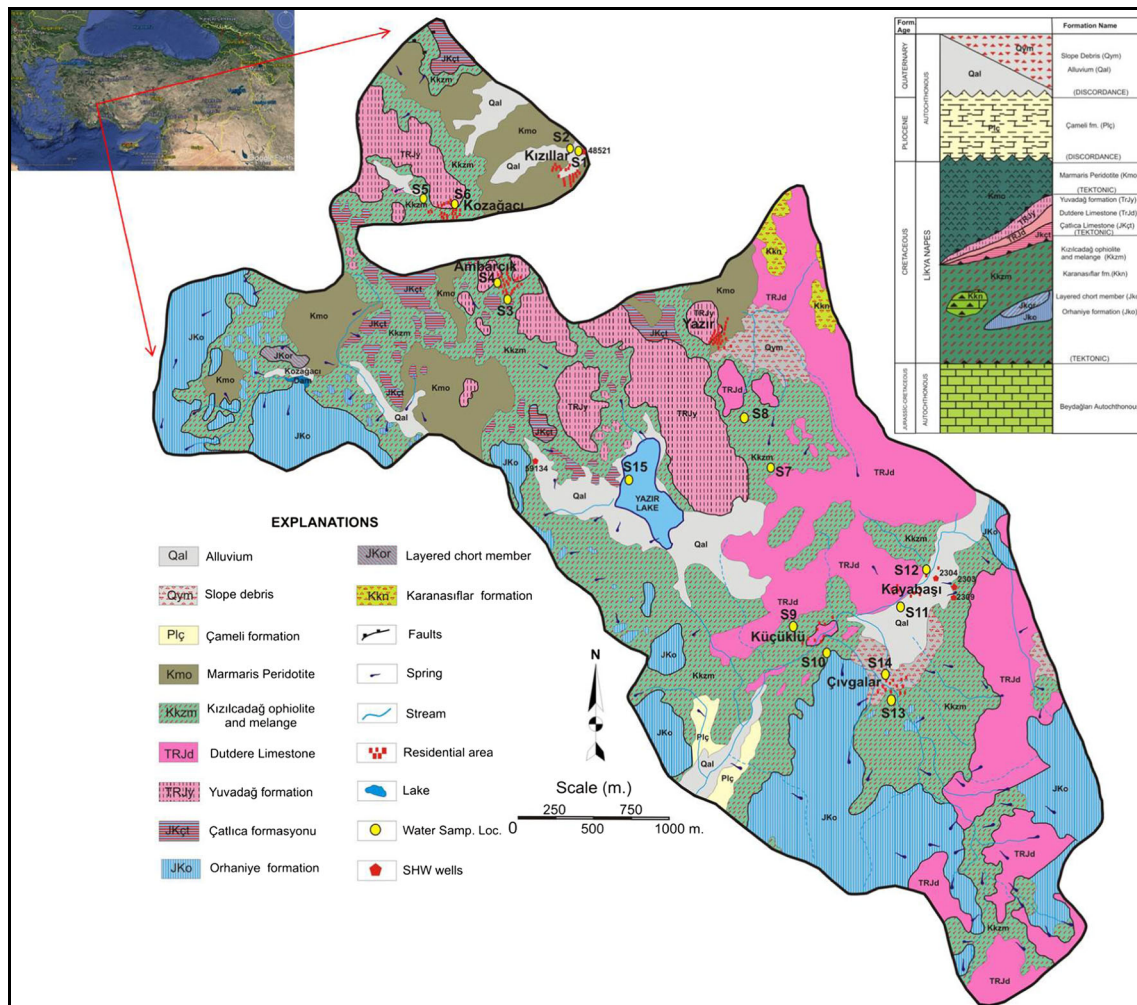


Fig. 1 Location and geological maps of the study area (Köse 2017)

than 2 that was used for major cations and As analysis, while second sample was not acidified and used for major anions. All the samples were transported and kept in the dark at 4 °C for analysis. The physical parameters like pH, EC, and discharge temperature (°C) were determined in situ by using YSI multi-parameter water quality sonde (YSI 6050).

The major chemical constituents like Ag, Al, As, Au, B, Be, Bi, Br, Ca, Cd, Ce, Cl, Co, Cr, Cs, Cu, Dy, Er, Eu, Fe, Ga, Gd, Ge, Hf, Hg, Ho, In, Ir, K, La, Li, Lu, Mg, Mn, Mo, Na, Nb, Nd, Ni, Os, P, Pb, Pd, Pr, Pt, Rb, Re, Rh, Ru, S, Sb, Sc, Se, Si, Sm, Sn, Sr, Ta, Tb, Te, Th, Ti, Tl, Tm, U, V, W, Y, Yb, Zn, and Zr were analyzed by inductively coupled plasma–mass spectrometer (ICP-MS) at the Bureau Veritas Commodities Canada Ltd. (ACME Laboratory Vancouver, Canada, an ISO 9002-accredited company). HCO₃⁻, CO₃⁻², and Cl⁻ concentrations were determined by titrimetric method; SO₄⁻², NO₃, NO₂, and NH₃ were determined by using ion chromatography in the Hacettepe University Water Chemistry Laboratory (Ankara, Turkey). The charge–balance error of the water samples was less than 5%, which is within the limits of acceptability.

Arsenic risk assessment

Exposure assessment

As enters into human body through several pathways including food chain, dermal contact and inhalation but in comparison with oral intake all others are negligible (ATSDR 2000). The average daily dose (ADD) through drinking water intake is calculated according to following formula (USEPA 1998; Eq. (1)):

$$ADD = C \times IR \times ED \times EF / BW \times AT \tag{1}$$

where C, IR, ED, EF, BW, and AT represent the As concentration (C) in water (µg L⁻¹), water ingestion rate (IR) 2 (L day⁻¹), exposure duration (ED) (assumed 15 years), exposure frequency (EF) (350 days year⁻¹), body weight (BW) (70 kg), and average lifetime (AT) (30/70 × 365 days), respectively.

Table 1 Lithostratigraphic relations of the geologic units and hydrogeological properties

Age	Formation	Lithology	Hydrogeological properties
Quaternary	Alluvium (Qal)	Gravel. Sand and mudstone	Permeable-1 (granular aquifer)
Quaternary	Slope alluvium (Qym)	Attached to the loose gravel. Sand and mudstone	Permeable-1 (granular aquifer)
Pliocene (Neogene)	Çameli formation (Plç)	Conglomerate. Sandstone. claystone. Clayey limestone. Marl. Conglomerate	Semipermeable (aquitard-1)
Aptian–Albian (Cretaceous)	Marmaris peridotite (Kmo)	Peridotite. Serpentinite and serpentinized peridotite	İmpermeable (aquifuge)
L. Cenoniyen (Cretaceous)	Kızılcadağ ophiolites and melange (Kkzm)	Chert, diabase, basalt, spilit, tuff, tuffit, gabro, diyabase, and serpentinite blocks within ophiolitic matrix. Limestone within the olistolites	Semipermeable (aquitard-3)
M.-L. Triassic	Dutdere limestone (TrJd)	Recrystallized limestone	Permeable-2 (karstic aquifer)
Jurassic-Cretaceous	Orhaniye formation (JKo)	Calciturbidite. Cherty limestone	Semipermeable (aquitard-2)
L. Eosen	Karanasıflar formation (Kkn)	Neritic limestone and cherty bresh	Permeable-2 (karstic aquifer)
Triassic-Cretaceous	Yuvadağ formation (TRJy)	Neritic limestone and dolomitic limestone	Permeable-2 (karstic aquifer)
	Çatlıca formation (JKçt)	Limestone, dolomitic limestone, and cherty limestone	Permeable-2 (karstic aquifer)

Human health risk assessment

In this study, both the chronic and carcinogenic risk levels were also assessed. Generally, the HQ can be calculated by the following equation (USEPA 1998; Eq. (2)):

$$HQ = ADD/RfD \quad (2)$$

where the As toxicity reference dose (RfD) is 0.0003 mg kg⁻¹ day⁻¹ (USEPA 2005). Non-cancer risk is represented in terms of hazard quotient (HQ) for a single substance for multiple substances and/or exposure pathways. If the exposure level of a substance exceeds the corresponding RfD, i.e., HQ exceeds 1, there may be concern for potential non-carcinogenic effects. The higher the value means the greater the likelihood of an adverse non-carcinogenic health effect (USEPA 1989; Khan et al. 2008; Muhammad et al. 2010; Qaiyum et al. 2011; Jamaludin et al. 2013; Varol and Davraz 2016).

The cancer risk (R_{cancer}) was calculated using the following formula (Eq. (3)):

$$R_{cancer} = ADD \times CSF \quad (3)$$

where R is the excess probability of developing cancer over a lifetime as a result of exposure to a contaminant (or carcinogenic risk), ADD is the chronic daily intake (mg/kg/day), and SF is the slope factor of the contaminant (mg/kg/day)⁻¹ (Kavcar et al. 2009; Eq. (3)). According to USEPA (2005) database, the SF that is the slope factor of the contaminant (SF) for As is 1.5 mg kg⁻¹ day⁻¹.

Statistical analysis

All mathematical calculations were calculated for physio-parameters using Excel 2007 (Microsoft Office). The statistical analysis that is the principle component analysis (PCA) was analyzed by using SPSS software version 15.

Result and discussion

Hydrogeochemistry

The groundwater quality depends on the properties of the aquifer rocks in the water (Subramani et al. 2005; Kumar et al. 2016). The chemical properties of the water determine the use status for domestic, industrial, or agricultural activities. In many developed and developing countries, water pollution is one of the most important factors of diseases and infant deaths (Kumar et al. 2016). In this study, the physicochemical, major ion, and quality evaluations of surface and groundwater were determined and detailed below under headings.

Seasonal evaluation of physical parameters

The pH of spring waters varied between 8.96 (S12) and 9.61 (S13) and 8.93 (S12) and 9.80 (S13), respectively, for dry and wet seasons. The pHs of the well waters were measured between 8.56 (S6) and 9.22 (S2) in dry season and 8.54 (S6) and 10.07 (S2) in wet season. Also, the pH of the surface waters varied sequentially, 9.27 in lake water (S15) and 9.91 in stream water (S14) for dry seasons and 9.53 in lake water (S15) and 9.62 in stream water (S14) for wet seasons. The pH values

Table 2 The geographic details of the sample locations and region

Sample no.	Sample type	Sample location	Sample location coordinates	Population ^a
S1	Spring water	Kızıllar village	35°S, 742,470°E/4,107,271°N	397
S2	Well		35°S, 741,037°E/4,107,274°N	
S3	Spring water	Ambarcık village	35°S, 739,532°E/4,103,409°N	452
S4	Well		35°S, 739,978°E/4,102,739°N	
S5	Spring water	Kozağacı village	35°S, 737,274°E/4,105,568°N	1203
S6	Well		35°S, 738,028°E/4,105,450°N	
S7	Spring water	Gölcük village	35°S, 746,531°E/4,094,239°N	310
S8	Well		35°S, 745,174°E/4,101,996°N	
S9	Spring water	Küçükklü village	35°S, 747,686°E/4,095,068°N	363
S10	Well		35°S, 747,633°E/4,094,643°N	
S11	Spring water	Kayabaş village	35°S, 749,784°E/4,095,876°N	407
S12	Spring water		35°S, 749,359°E/4,096,020°N	
S13	Spring water	Çıvgalar district	35°S, 749,596°E/4,093,460°N	103
S14	Stream water		35°S, 749,240°E/4,093,149°N	
S15	Lake water	Yazır Lake	35°S, 741,373°E/4,098,585°N	

^a TSI (Turkish Istatistical Institute) 2017 data

increased in wet season for water samples (Tables 3 and 4). This increase is due to the high interaction between rock and rain-water (Makwe and Chup 2013; Ngabirano et al. 2016).

The EC values of spring waters were measured between 183.4 (S13) and 434.6 $\mu\text{S}/\text{cm}$ (S3) in dry season and 139.3 (S13) and 394.00 $\mu\text{S}/\text{cm}$ (S12) in wet season. The EC values of well waters also varied in the range 119.2 (S6)–988.00 $\mu\text{S}/\text{cm}$ (S4) in dry season and 293.7 (S10)–968.00 $\mu\text{S}/\text{cm}$ (S6) in wet season. Also, the EC values of surface waters were measured in the range 214.00 in stream water (S14) and 241.8 $\mu\text{S}/\text{cm}$ in lake water (S15) in dry season and 253.7 in stream water (S14) and 297.8 $\mu\text{S}/\text{cm}$ in lake water (S15) in wet season (Tables 3 and 4). The high EC values for both dry and wet seasons indicated the spatial variability of leaching and dilution with recharging rainfall. Also, the higher EC values in wet season might be attributed to enhanced chemical weathering and longer residence time of groundwater in the aquifers (Oinam et al. 2011; Alam et al. 2016).

In addition, the T ($^{\circ}\text{C}$) values of spring waters were measured in the range 11.9 (S12)–17.3 $^{\circ}\text{C}$ (S3) in dry season and 9.2 (S13)–13.4 $^{\circ}\text{C}$ (S7) in wet season. The T ($^{\circ}\text{C}$) values of well waters also varied in the range 11.4 (S10)–17.5 $^{\circ}\text{C}$ (S6) in dry season and 11.0 (S8)–12.5 $^{\circ}\text{C}$ (S4) in wet season. Also, the T ($^{\circ}\text{C}$) values of surface waters were measured in the range 11.8 $^{\circ}\text{C}$ in lake water (S15) and 12.4 $^{\circ}\text{C}$ in stream water (S14) in dry season, and 9.4 in lake water (S15) and 11.2 $^{\circ}\text{C}$ in stream water (S14) in wet season (Tables 3 and 4). The temperature of the waters in the study area varied with the meteorological conditions in the dry and wet seasons.

Oxidation-reduction potential (ORP) is a measurement that indicates the degree to which a substance is capable of oxidizing or reducing another substance. It is also possible to determine the species of water chemistry and As by ORP values measured in water (APHA 1998). In the present study, ORP

values of spring waters were found ranging between 327.00 mV (S5) and 620.90 mV (S13) in dry season and 240.3 mV (S1) and 348.00 mV (S3) in wet season. The ORP values of well waters also varied in the range 320.2 mV (S2)–370.1 mV in dry season and 208 mV (S2)–325.6 mV (S6) in wet season. Also, ORP values of surface waters were measured in the range 338.9 mV (S15) in lake water and 410.9 mV (S14) in stream water in dry season, and 288.9 mV in lake water (S15) and 295.5 mV in stream water (S14) in wet season (Tables 3 and 4).

Seasonal evaluation of major ions

The most common ion in the groundwater is Ca^{2+} . Subsequently, Mg^{2+} , Na^{+} , and K^{+} ions are present in the composition in a lesser amount (Tables 3, 4, and 5). The sources of Ca^{2+} and Mg^{2+} in water are generally carbonate-rich rocks such as limestone and dolomitic limestone. The carbonates in these rocks are mixed in the groundwater with various ways such as irrigation and precipitation.

The increase in Mg^{2+} ion in dry and wet seasons was observed at spring waters (S1, S4, and S15) within the Kızılcadağ ophiolite and melange. The major source of Mg^{2+} in the groundwater is ion exchange of minerals in rocks with water. The increase in K^{+} was determined only for dry season at Ambarcık spring water (S3), which discharges at the contact of the layered chert member and Kızılcadağ ophiolite and melange. This increase is due to ion exchange due to water-rock interaction.

Bicarbonate (HCO_3^{-}) was the main anionic constituent of the groundwater samples ranging from 115.61 to 505.43 mg/L in dry season and 148.05 to 538.93 mg/L in wet season. HCO_3^{-} representing the major source of alkalinity generally prevails due to the dissolution of CO_2 and carbonates, reaction

Table 3 Physicochemical characteristics of groundwater and surface water in the study area (dry season)

Sample location	Site no.	T (°C)	pH	ORP (mV)	EC (µS/cm)	TDS (mg/L)	Ca (mg/L)	Mg (mg/L)	Na (mg/L)	K (mg/L)	HCO ₃ (mg/L)	CO ₃ (mg/L)	Cl (mg/L)	SO ₄ (mg/L)	NO ₃ (mg/L)	NO ₂ (mg/L)	NH ₄ (mg/L)	As (mg/L)	Mn (mg/L)	Fe (mg/L)
Kızillar (s.w)	S1	14.2	9.10	355.4	390.8	254.02	44.84	39.77	10.94	0.68	313.71	0.00	6.22	16.33	2.14	0.00	0.58	0.03	0.00	0
Kızillar (w)	S2	15.5	9.22	320.2	329.9	214.44	45.72	26.39	9.76	1.44	249.81	0.00	5.61	14.40	0.00	0.05	1.09	0.02	0.35	0.19
Ambarcık (s.w)	S3	17.3	9.15	338.7	434.6	282.49	51.04	42.41	5.07	0.38	325.33	0.00	4.56	5.99	6.32	0.04	0.59	0.06	0.00	0.01
Ambarcık (w)	S4	14.8	8.67	355.2	988	642.20	129.05	80.43	32.06	1.50	505.43	0.00	47.47	54.32	157.76	0.35	2.56	0.08	0.00	0
Kozağacı (s.w)	S5	17.3	9.12	327.0	270.1	175.57	65.75	3.23	2.91	0.41	203.33	0.00	2.04	3.17	2.33	0.00	0.16	0.08	0.00	0.11
Kozağacı (w)	S6	17.5	8.56	370.1	119.2	77.48	134.88	63.31	63.40	52.52	406.67	0.00	71.14	70.99	263.80	0.00	0.00	0.03	0.00	0
Gölcük (s.w)	S7	13.5	9.26	343.3	242.0	157.30	45.62	14.60	3.47	0.36	203.33	0.00	2.60	4.15	3.65	0.00	0.04	0.08	0.00	0
Gölcük (w)	S8	12.4	8.68	347.2	336.9	218.99	86.75	8.15	3.72	0.41	278.86	0.00	4.17	5.00	5.31	0.02	0.57	0.07	0.00	0.02
Küçükütlü (s.w)	S9	12.8	9.37	619.7	351.2	228.28	58.03	29.38	5.84	0.46	255.62	22.86	5.67	6.11	5.57	0.00	0.36	0.08	0.00	0
Küçükütlü (w)	S10	11.4	8.85	352.7	327.2	212.68	62.23	24.77	5.01	0.79	296.29	0.00	4.46	5.45	4.37	0.00	0.26	0.08	0.00	0
Kayabaş (s.w)	S11	13.4	9.41	513.1	200.0	130.00	50.87	3.46	3.29	0.46	139.43	17.14	2.21	3.11	2.80	0.00	0.25	0.07	0.00	0
Kayabaş (w)	S12	11.9	8.96	389.6	401.0	260.65	70.10	35.13	7.14	0.80	284.67	17.14	6.36	9.60	21.62	0.00	0.38	0.08	0.00	0
Çıvgalar (s.w)	S13	12.3	9.61	620.9	183.4	119.21	50.64	2.79	2.31	0.34	115.61	22.86	2.14	2.19	3.64	0.00	0.19	0.06	0.00	0
Çıvgalar (str.w.)	S14	12.4	9.91	410.9	214.0	139.10	38.56	30.61	5.53	0.33	156.86	22.86	2.14	5.79	0.56	0.00	0.20	0.06	0.11	5.52
Yazır Lake	S15	11.8	9.27	338.9	241.8	157.17	37.56	23.26	5.07	0.53	156.86	22.86	3.09	3.42	8.12	0.00	0.20	0.07	0.00	0
WHO (2011)															3	50	0.05–0.50	0.01	–	–
TSI 266 (2005)															0.05	50	0.05–0.50	0.01	0.05	0.2

s.w spring water, w well water, str:w stream water, WHO (2011) World Health Organization drinking water limit value, TSI 266 (2005) Turkish Standardization Institute drinking water limit value
 Permissible concentration values in water are presented in bold

Table 4 Physicochemical characteristics of groundwater and surface water in the study area (wet season)

Sample location	Site no.	T (°C)	pH	ORP (mV)	EC (µS/cm)	TDS (mg/L)	Ca (mg/L)	Mg (mg/L)	Na (mg/L)	K (mg/L)	HCO ₃ (mg/L)	CO ₃ (mg/L)	Cl (mg/L)	SO ₄ (mg/L)	NO ₃ (mg/L)	NO ₂ (mg/L)	NH ₄ (mg/L)	As (mg/L)	Mn (mg/L)	Fe (mg/L)
Kızılılar (s.w)	S1	11.6	9.67	240.3	363.9	236.54	47.34	41.82	10.02	0.80	313.88	11.65	6.77	21.17	3.68	0.00	0.00	0.04	0.01	0.01
Kızılılar (w)	S2	11.1	10.07	208	330.0	214.50	47.53	77.91	12.87	1.46	467.86	17.47	16.56	16.33	8.41	0.00	0.00	0.07	0.03	0.03
Ambarecik (s.w)	S3	10.7	9.29	348	354.7	230.56	62.47	30.69	6.46	0.36	307.96	17.47	5.78	7.28	6.86	0.00	0.10	0.08	0.04	0.01
Ambarecik (w)	S4	12.5	8.68	300	968.0	629.20	130.82	79.71	32.82	2.02	538.93	0.00	51.07	60.74	194.72	0.00	0.00	0.10	0.06	0.02
Kozağacı (s.w)	S5	10.3	9.54	285.6	209.5	136.18	58.98	2.18	1.57	0.25	213.20	0.00	1.50	2.96	1.55	0.00	0.03	0.08	0.04	0.01
Kozağacı (w)	S6	11.5	8.54	325.6	968.0	629.20	118.73	52.93	54.87	43.53	414.56	0.00	79.96	83.79	258.49	0.00	0.00	0.13	0.09	0.04
Gölcük (s.w)	S7	13.4	9.08	294.3	356.9	231.99	81.18	12.19	3.58	0.48	319.80	0.00	5.80	6.84	7.91	0.00	0.00	0.12	0.09	0.04
Gölcük (w)	S8	11.0	9.26	297.4	359.7	233.81	35.99	52.08	4.74	0.22	367.18	0.00	3.21	6.94	5.33	0.00	0.00	0.07	0.03	0.02
Küçükü (s.w)	S9	10.4	9.28	313.2	221.6	144.04	50.74	10.31	2.61	0.36	219.12	0.00	3.22	4.28	3.27	0.00	0.08	0.08	0.04	0.01
Kıçıklü (w)	S10	11.0	8.99	313.4	293.7	190.91	55.84	20.49	3.92	0.65	278.34	0.00	3.56	5.83	4.35	0.00	0.00	0.09	0.02	0.05
Kayabaş (s.w)	S11	12.6	9.21	322.8	224.0	145.60	46.32	2.98	2.70	0.46	189.51	0.00	1.99	3.56	2.53	0.00	0.00	0.10	0.09	0.02
Kayabaş (s.w)	S12	12.2	8.93	323.0	394.0	256.10	65.63	31.51	6.13	0.84	337.57	0.00	7.72	11.45	23.29	0.00	0.00	0.11	0.07	0.07
Çıvgalar (s.w)	S13	9.2	9.8	302.4	139.3	90.55	39.43	1.51	1.03	0.35	148.05	0.00	1.41	2.13	3.07	0.00	0.02	0.09	0.05	0.08
Çıvgalar (str.w.)	S14	11.2	9.62	295.5	253.7	164.91	45.58	12.92	4.02	0.29	183.59	23.30	2.07	5.21	0.77	0.00	0.03	0.11	0.07	0.30
Yazır Lake	S15	9.4	9.53	288.9	297.8	193.57	44.90	33.45	6.38	0.24	242.81	17.47	3.03	16.12	9.75	0.00	0.12	0.10	0.04	0.08
WHO (2011)												3			50	50	0.05-0.50	0.01	-	-
TSI 266 (2005)												0.05			50	50	0.05-0.50	0.01	0.05	0.2

s.w: spring water, w: well water, str:w: stream water, WHO (2011) World Health Organization drinking water limit value, TSI 266 (2005) Turkish Standardization Institute drinking water limit value
 Permissible concentration values in water are presented in bold

Table 5 The location and major ion sequences of water samples (Köse 2017)

Sample location	No.	Sample type	Dry season (October 2016)		Wet season (April 2017)	
			Cation sequence	Anion sequence	Cation sequence	Anion sequence
Kızıllar	S1	Spring	Mg > Ca > Na > K	HCO ₃ > Cl > SO ₄ > CO ₃	Mg > Ca > Na > K	HCO ₃ > SO ₄ > CO ₃ > Cl
Kızıllar	S2	Well water	Ca > Mg > Na > K	HCO ₃ > Cl > SO ₄ > CO ₃	Mg > Ca > Na > K	HCO ₃ > CO ₃ > Cl > SO ₄
Ambarcık	S3	Spring	Ca > Mg > Na > K	HCO ₃ > Cl > SO ₄ > CO ₃	Ca > Mg > Na > K	HCO ₃ > CO ₃ > Cl > SO ₄
Ambarcık	S4	Well water	Mg > Ca > Na > K	HCO ₃ > Cl > SO ₄ > CO ₃	Mg > Ca > Na > K	HCO ₃ > Cl > SO ₄ > CO ₃
Kozağacı	S5	Spring	Ca > Mg > Na > K	HCO ₃ > Cl > SO ₄ > CO ₃	Ca > Mg > Na > K	HCO ₃ > SO ₄ > Cl > CO ₃
Kozağacı	S6	Well water	Ca > Na > Mg > K	HCO ₃ > Cl > SO ₄ > CO ₃	Ca > Mg > Na > K	HCO ₃ > Cl > SO ₄ > CO ₃
Gölcük (Yazır)	S7	Spring	Ca > Mg > Na > K	HCO ₃ > Cl > SO ₄ > CO ₃	Ca > Mg > Na > K	HCO ₃ > Cl > SO ₄ > CO ₃
Gölcük (Yazır)	S8	Well water	Ca > Mg > Na > K	HCO ₃ > Cl > SO ₄ > CO ₃	Mg > Ca > Na > K	HCO ₃ > SO ₄ > Cl > CO ₃
Küçüküklü	S9	Spring	Ca > Mg > Na > K	HCO ₃ > CO ₃ > Cl > SO ₄	Ca > Mg > Na > K	HCO ₃ > Cl > SO ₄ > CO ₃
Küçüküklü	S10	Well water	Ca > Mg > Na > K	HCO ₃ > Cl > SO ₄ > CO ₃	Ca > Mg > Na > K	HCO ₃ > SO ₄ > Cl > CO ₃
Kayabaş	S11	Spring	Ca > Mg > Na > K	HCO ₃ > CO ₃ > Cl > SO ₄	Ca > Mg > Na > K	HCO ₃ > SO ₄ > Cl > CO ₃
Kayabaş	S12	Spring	Ca > Mg > Na > K	HCO ₃ > CO ₃ > Cl > SO ₄	Ca > Mg > Na > K	HCO ₃ > SO ₄ > Cl > CO ₃
Çıvgalar	S13	Spring	Ca > Mg > Na > K	HCO ₃ > CO ₃ > Cl > SO ₄	Ca > Mg > Na > K	HCO ₃ > SO ₄ > Cl > CO ₃
Çıvgalar	S14	Stream water	Ca > Mg > Na > K	HCO ₃ > CO ₃ > Cl > SO ₄	Ca > Mg > Na > K	HCO ₃ > CO ₃ > SO ₄ > Cl
Yazır Gölü	S15	Lake water	Mg > Ca > Na > K	HCO ₃ > CO ₃ > Cl > SO ₄	Mg > Ca > Na > K	HCO ₃ > CO ₃ > SO ₄ > Cl

It is meant to indicate the sample number and water type are presented in bold

of silicates with carbonic acid (Ranjan et al. 2013), and oxidation of organic matter (Jeong 2001). The Cl⁻ content increased at most of the samples in dry season in discharges at the contact of the Dutdere limestone and Kızılcadağ ophiolite and melange. This increase is probably related to water-rock interaction between Kızılcadağ ophiolite and melange with rainwater. Sulfate (SO₄²⁻) which originates from oxidation of sulfite (Ranjan et al. 2013) ranged between 3.11 and 70.99 mg/L in dry season and between 2.13 and 83.79 mg/L in wet season. NO₃⁻ ranged from 0 to 263.80 mg/L in dry season and 0.77 to 258.49 mg/L in wet season. Pollution originated from agricultural activities is the case for both water samples, where NO₃ concentrations are high.

Hydrogeochemical facies

Hydrogeochemical facies are a useful tool for determining the chemical history and origins of groundwater. It is used to show similarities and differences in the chemistry of groundwater samples based on dominant cations and anions (Piper 1944, 1953). Piper trilinear plots were made for the samples collected during dry and wet seasons. According to the Piper diagrams, Ca-Mg-HCO₃ and Ca-HCO₃ which are the dominant water types were observed in dry and wet seasons (Fig. 2). In the study area, only one sample (S6) is Ca-Na-HCO₃-Cl-SO₄ water type in dry season. This well is drilled within Yuvadağ formation which consists of dolomitic limestone and Kızılcadağ ophiolite and melange. The increase of Na and SO₄ ions in this well originated from water-rock interaction with the ophiolites.

Mechanism controlling the groundwater geochemistry

There are two types of scatter diagrams those have been proposed by Gibbs (1970). Gibbs's diagrams are widely employed to assess the functional sources of dissolved chemical constituents, such as precipitation dominance, rock dominance, and evaporation dominance (Gibbs 1970). The Gibbs ratios determined by Gibbs 1970 are calculated using the following formulas:

$$\text{Gibbs Ratio-I (for Anion)} = \text{Cl}^- / (\text{Cl}^- + \text{HCO}_3^-)$$

$$\text{Gibbs Ratio-II (for Cation)} = (\text{Na}^+ + \text{K}^+) / (\text{Na}^+ + \text{K}^+ + \text{Ca}^{2+})$$

Gibbs ratio-I values varied from 0.04 to 0.80 with an average value of 0.16, and Gibbs ratio-II values varied from 0.02 to 0.14 with an average value of 0.05 in dry season. Also, Gibbs ratio-I values varied from 0.02 to 0.37 with an average value of 0.11 and Gibbs ratio-II values varied from 0.01 to 0.25 with an average value of 0.05 in wet season. The chemical data of groundwater sample points of the area were plotted in Gibbs' diagrams (Fig. 3). Samples from both seasons fell in rock-dominance zone, suggesting precipitation-induced chemical weathering along with dissolution of rock-forming minerals.

Statistical analysis

Correlation analysis was applied to determine the relationship between physicochemical properties of water samples. It is possible to obtain information about the mineral and chemical

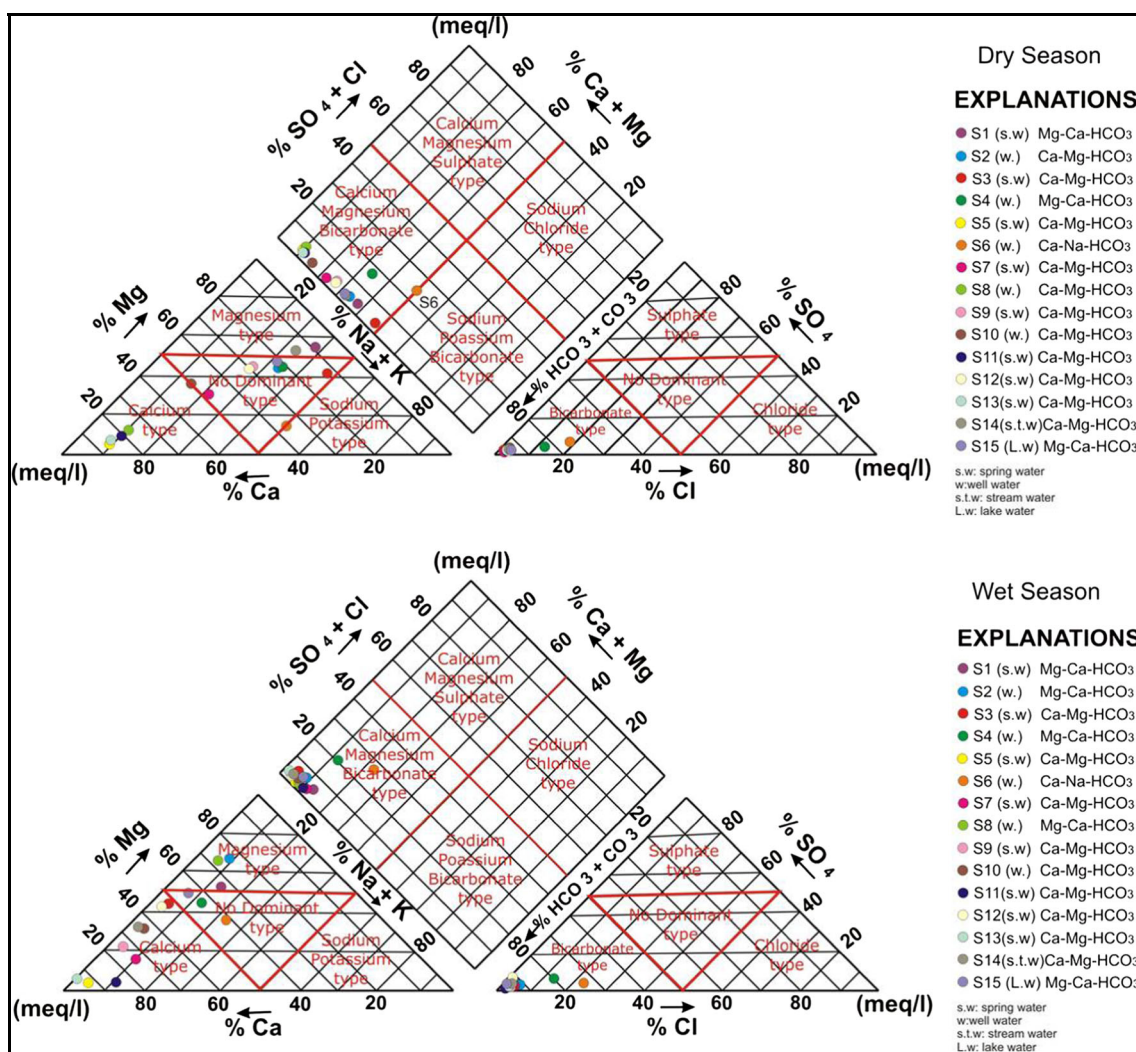


Fig. 2 Piper diagrams in dry and wet seasons (Piper 1944)

processes and chemical constituents of water by correlation analysis (Varol and Davraz 2015).

While the results of the correlation analysis are being evaluated, if the correlation coefficient is close to 1 or -1, it means a good positive relationship between the two variables. The near-zero values are significant if $p < 0.05$, but there is no relationship between them. Namely, while it is assumed that there is strong correlation between the parameters with r value > 0.7 , it is said that the r value is moderately correlated between 0.5 and 0.7 (Manish et al. 2006). To evaluate the potential relationship between various physiochemical parameters and trace elements, “Pearson correlation analysis (PCA)” was carried out. All the processes were performed using SPSS software version 15.0 for Windows. In addition, the Pearson correlation matrix was applied separately for the dry and wet seasons in order to study the changes in the relations between the parameters. The correlation matrix of the parameters is given in Table 6 as dry and wet seasons.

According to the physicochemical PCA results, T ($^{\circ}\text{C}$) showed positive correlation with $\text{EC-TDS-K}^+-\text{Cl}^-$ and showed negative correlation with NH_4 in wet season. In addition, this parameter showed negative correlation with CO_3^- in dry season. EC and TDS showed positive strong correlation with $\text{Mg}^{2+}-\text{Na}^+-\text{HCO}_3^- - \text{Cl}^- - \text{SO}_4^{2-} - \text{NO}_3^-$ in wet season and showed positive strong correlation with NH_4^+ in dry season. pH showed positive strong correlation with As in wet season. This parameter showed positive strong correlation with CO_3^- and As in dry season. No significant relationship between ORP and any parameters was observed during the wet and dry seasons (Table 6). The physicochemical PCA results represent water-rock interaction and the contribution of point and non-point source pollution from domestic and agricultural wastes.

According to the major cation PCA results, Mg showed positive strong correlation with $\text{Na}^+-\text{HCO}_3^- - \text{Cl}^- - \text{SO}_4^{2-} - \text{NO}_3^-$ in wet season. Mg showed positive strong correlation

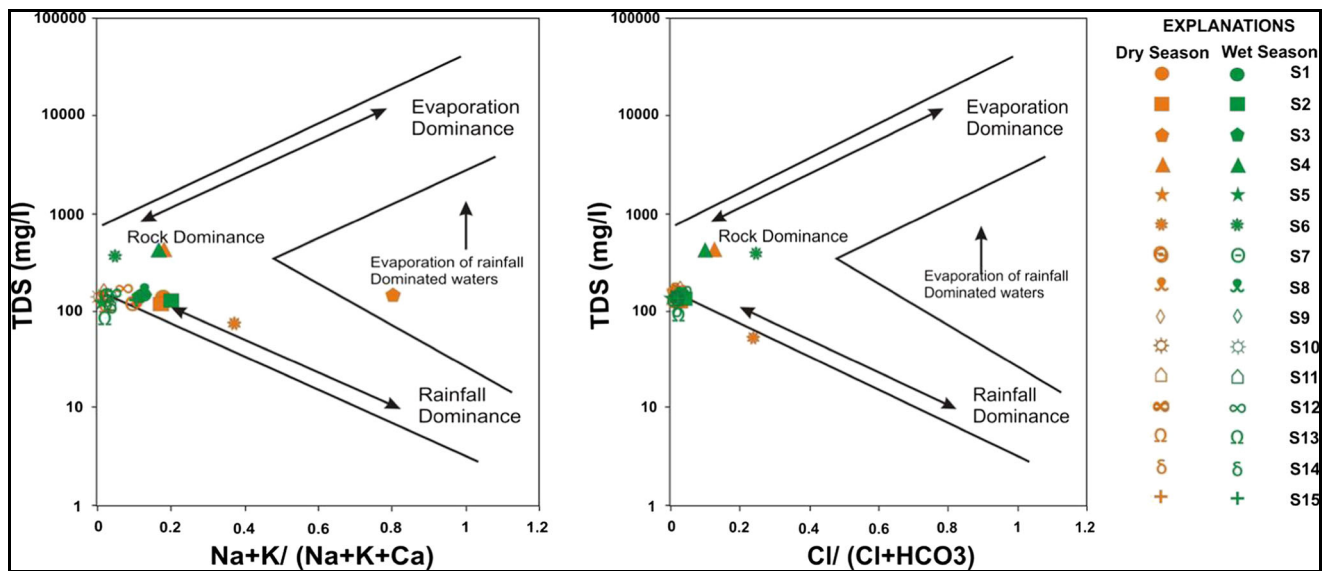


Fig. 3 Gibbs diagrams (Gibbs 1970), illustrating the mechanisms controlling the chemistry of groundwater samples

with $\text{Na}^+ - \text{K}^+ - \text{HCO}_3^- - \text{Cl}^- - \text{SO}_4^{2-}$ in dry season. Correlation of Mg ion with other ions indicated that they derived from dolomitic limestone and magnesium sulfate minerals within the Kızılcaadağ ophiolite and melange with water interaction and also agricultural processes in the study area. Na^+ showed positive strong correlation with $\text{HCO}_3^- - \text{Cl}^- - \text{SO}_4^{2-} - \text{NO}_3^-$ in wet season and showed positive strong correlation with $\text{K}^+ - \text{HCO}_3^- - \text{Cl}^- - \text{SO}_4^{2-}$ in dry season. The strong loading of Na^+ indicates natural weathering of rock minerals and various ion exchange processes in the groundwater system (Davraz and Özdemir 2014; Varol and Davraz 2016). In addition, correlation of Na ion with $\text{Cl}^- - \text{SO}_4^{2-} - \text{NO}_3^-$ ions indicated an anthropogenic pollution. K showed positive correlation with $\text{HCO}_3^- - \text{Cl}^- - \text{SO}_4^{2-} - \text{NO}_3^-$ and showed negative correlation with NH_4^+ in wet season. Also, this parameter showed positive correlation with $\text{HCO}_3^- - \text{Cl}^- - \text{SO}_4^{2-}$ in dry season (Table 6). The strong loading of Na^+ and K indicates natural weathering of rock minerals and various ion exchange processes in the groundwater system. In addition, correlation of Na and K ions with $\text{Cl}^- - \text{SO}_4^{2-} - \text{NO}_3^-$ ions indicated an anthropogenic pollution.

According to the major anion PCA results, HCO_3^- showed positive strong correlation with $\text{Cl}^- - \text{SO}_4^{2-} - \text{NO}_3^-$ in dry and wet seasons. Cl^- showed positive strong correlation with $\text{SO}_4^{2-} - \text{NO}_3^-$ in dry and wet seasons. SO_4^{2-} showed positive correlation with NO_3^- in wet season. No significant relationship between SO_4^{2-} and any parameters was observed during the dry season (Table 6). This situation can be explained by anthropogenic pollution in the study area (Varol and Davraz 2016).

According to the heavy metal and pollution parameters PCA results, As showed positive strong correlation with Mn and moderate correlation with Fe in wet season. This can be explained by natural hydrogeochemical evolution of water by

groundwater-rock interaction. The dissolution of rocks and minerals in Kızılcaadağ ophiolite and melange by chemical weathering can cause this. As may derive from reductive dissolution of iron and microbial oxidation of organic matter. No significant relationship between NO_3^- and any parameters was observed during the wet season. Likewise, no significant relationship between $\text{NO}_2^- - \text{Mn}$ and any parameters was observed during the wet season. But, NO_2^- shows positive strong correlation with NH_4^+ (Table 6). Correlation of NO_2^- ion with NH_4^+ indicated an anthropogenic pollution in the study area.

According to the result of statistical analysis, this situation is considered as a result of rock-water interaction depending on climatic conditions, ion exchange processes, and anthropogenic effects in the water resources.

Evaluation of the arsenic pollution in waters

As as a trace element is an important component in more than 200 minerals such as elemental As, sulfides, oxides, arsenates, and arsenites. Many of these minerals are ore minerals or alteration products. In the nature, these minerals are relatively less (Smedley and Kinniburgh 2013). As is a metalloid that has toxic effects and can be found both as a natural and anthropogenic origin in groundwater. Natural processes such as decomposition, erosion, biological activities, and anthropogenic activities such as petroleum refining, insect killers, semiconductors, dyes, metals, soaps, medicines, and herbicides pollute the groundwater considerably (Rasool et al. 2016).

In the study area, As concentrations in dry season were between 0.03 and 0.08 mg/L in spring waters, 0.02–0.08 mg/L in well waters, 0.06 mg/L in stream water, and 0.07 mg/L in lake water. In addition, As concentrations in wet season were

Table 6 Correlation matrix of dry and wet season parameters of waters in the study area

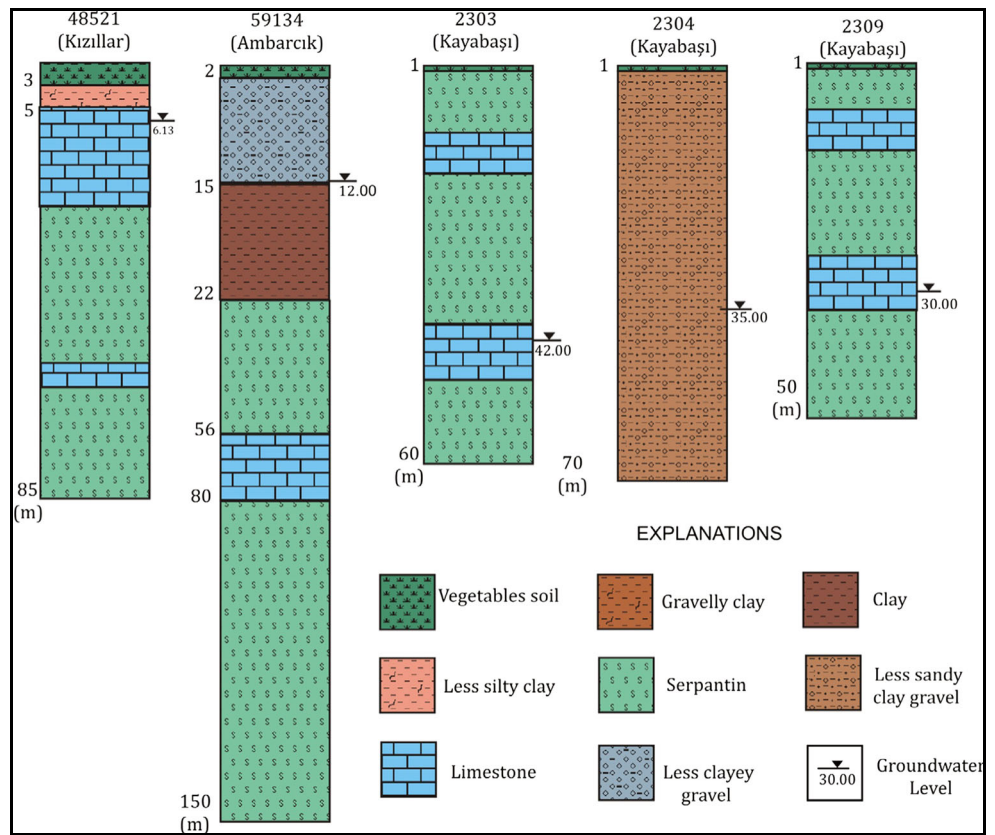
Dry	T	pH	ORP	EC	TDS	Ca	Mg	Na	K	HCO ₃	CO ₃	Cl	SO ₄	NO ₃	NO ₂	NH ₄	As	Mn	Fe
T	1.00	-0.28	-0.35	0.10	0.10	0.26	0.352	0.30	0.18	0.39	-0.58	0.24	0.41	-0.02	0.37	0.06	-0.33	0.08	0.27
pH	r	1.00	0.31	0.71	0.71	0.33	0.19	0.26	0.51	0.14	0.02	0.37	0.12	0.91	0.16	0.83	0.22	0.75	0.31
ORP	r		1.00	0.13	-0.40	-0.75	-0.47	-0.47	-0.067	-0.84	0.73	-0.66	-0.58	-0.53	-0.34	-0.26	0.75	0.34	0.14
EC	r			1.00	0.07	0.00	0.07	0.07	0.00	0.00	0.00	0.00	0.02	0.04	0.20	0.34	0.01	0.20	0.59
TDS	r				1.00	0.85	0.98	0.97	-0.12	-0.20	0.63	0.04	-0.04	0.09	-0.47	-0.20	0.03	-0.16	-0.45
Ca	r					0.25	0.50	0.38	0.66	0.46	0.01	0.87	0.87	0.74	0.07	0.47	0.89	0.54	0.08
Mg	r					0.25	0.50	0.15	0.25	0.62	-0.34	0.51	0.48	0.22	0.57	0.83	0.28	-0.12	0.03
Na	r					0.36	0.05	0.38	0.35	0.01	0.20	0.05	0.06	0.42	0.02	0.00	0.31	0.66	0.89
K	r					0.36	0.05	0.15	0.35	0.62	-0.34	0.51	0.48	0.22	0.57	0.83	0.28	-0.12	0.03
HCO ₃	r					1.00	0.36	0.46	0.46	0.59	-0.41	0.46	0.33	0.58	0.27	0.10	0.32	-0.39	-0.11
CO ₃	r						1.00	0.89	0.53	0.81	-0.25	0.08	0.21	0.02	0.32	0.70	0.23	0.14	0.67
Cl	r							1.00	0.72	0.70	-0.20	0.82	0.91	0.45	0.33	0.44	0.50	0.08	-0.10
SO ₄	r								1.00	0.61	-0.35	0.80	0.68	0.21	0.28	0.11	0.28	0.39	0.72
NO ₃	r									0.01	-0.64	0.83	0.00	0.11	0.40	0.22	0.88	-0.05	-0.34
NO ₂	r									1.00	0.00	0.00	0.00	0.03	0.10	0.07	0.96	0.39	0.58
NH ₄	r										1.00	0.23	-0.37	-0.02	-0.47	-0.25	0.05	0.10	-0.15
As	r											1.00	0.89	0.93	0.07	0.35	0.85	0.72	0.58
Mn	r												1.00	0.58	0.32	0.47	-0.09	-0.13	-0.37
Fe	r													0.02	0.23	0.07	0.72	0.62	0.16
Wet	T	pH	ORP	EC	TDS	Ca	Mg	Na	K	HCO ₃	CO ₃	Cl	SO ₄	NO ₃	NO ₂	NH ₄	As	Mn	Fe
T	1.00	-0.50	0.08	0.59	0.59	0.47	0.29	0.34	0.61	0.45	-0.17	0.53	0.37	0.32	0.37	0.46	0.72	0.48	-0.03
pH	r	1.00	0.75	0.02	0.02	0.07	0.28	0.21	0.01	0.09	0.52	0.03	0.17	0.02	0.23	0.00	0.12	0.07	0.90
ORP	r		1.00	-0.55	-0.55	-0.64	-0.25	-0.23	-0.43	-0.43	0.60	-0.46	-0.30	-0.54	0.42	0.88	0.88	-0.46	-0.02
EC	r			1.00	0.03	0.01	0.36	0.39	0.10	0.10	0.01	0.08	0.26	0.03	0.11	0.11	0.02	0.08	0.93
TDS	r				1.00	0.32	-0.11	0.01	0.21	-0.03	-0.35	0.11	-0.04	0.19	-0.00	-0.00	0.39	0.41	0.02
Ca	r					0.52	0.68	0.97	0.44	0.90	0.20	0.67	0.86	0.49	0.98	0.14	0.14	0.12	0.91
	r					0.52	0.82	0.83	0.60	0.86	-0.02	0.86	0.88	0.80	0.80	-0.54	0.21	0.09	-0.05
	r					0.04	0.00	0.00	0.01	0.92	0.00	0.00	0.00	0.80	0.03	0.03	0.44	0.72	0.83
	r					0.52	0.82	0.83	0.60	-0.02	-0.02	0.86	0.88	0.80	-0.54	-0.54	0.21	0.09	-0.05
	r					0.04	0.00	0.00	0.01	0.92	0.00	0.00	0.00	0.80	0.03	0.03	0.44	0.72	0.83
	r					1.00	0.25	0.36	0.69	-0.30	-0.30	0.68	0.41	0.54	-0.26	-0.26	0.41	0.36	-0.23

Table 6 (continued)

Dry	T	pH	ORP	EC	TDS	Ca	Mg	Na	K	HCO ₃	CO ₃	Cl	SO ₄	NO ₃	NO ₂	NH ₄	As	Mn	Fe
p							0.36	0.17	0.00	0.04	0.26	0.00	0.12	0.03	.	0.33	0.12	0.18	0.40
Mg	r				1.00		1.00	0.93	0.50	0.87	0.24	0.79	0.93	0.75	.	-0.39	-0.07	-0.22	-0.04
Na	r					1.00		0.00	0.05	0.00	0.37	0.00	0.00	0.00	.	0.14	0.78	0.41	0.88
K	r							1.00	0.59	0.78	0.37	0.82	0.96	0.75	.	-0.29	0.04	-0.08	-0.04
HCO ₃	r								0.02	0.00	0.17	0.00	0.00	0.00	.	0.28	0.87	0.75	0.87
CO ₃	r								1.00	0.62	-0.17	0.84	0.61	0.58	.	-0.64	0.25	0.19	-0.05
Cl	r									0.01	0.53	0.00	0.01	0.02	.	0.01	0.36	0.48	0.85
SO ₄	r									1.00	-0.09	0.88	0.83	0.83	.	-0.58	-0.01	-0.08	-0.18
NH ₄	r										0.74	0.00	0.00	0.00	.	0.02	0.96	0.75	0.51
As	r										1.00	0.00	0.25	-0.07	.	0.44	-0.19	-0.26	0.15
Mn	r											0.98	0.36	0.79	.	0.09	0.49	0.33	0.57
Fe	r											1.00	0.87	0.81	.	-0.52	0.14	0.02	-0.12
	p												0.00	0.00	.	0.04	0.59	0.91	0.65
													1.00	0.00	.	0.20	0.78	0.75	0.87
														0.00	.	-0.32	0.31	0.13	0.12
														1.00	.	0.24	0.26	0.62	0.65
															.	1.00	-0.09	-0.09	-0.00
																	0.73	0.74	0.98
																	1.00	0.86	0.61
																		0.00	0.01
																		1.00	0.33
																		1.00	0.21
																		1.00	1.00

The correlation matrix of the parameters and the parameters with a significant relationship between them are shown in bold

Fig. 4 Well logs opened by State Hydraulic Works (SHW) in the study area



between 0.04 and 0.12 mg/L in spring waters, 0.07 and 0.13 mg/L in well waters, 0.11 mg/L in stream water, and

0.10 mg/L in lake water (Tables 3 and 4). The permissible limit value of As for drinking water is 0.01 mg/L according to WHO

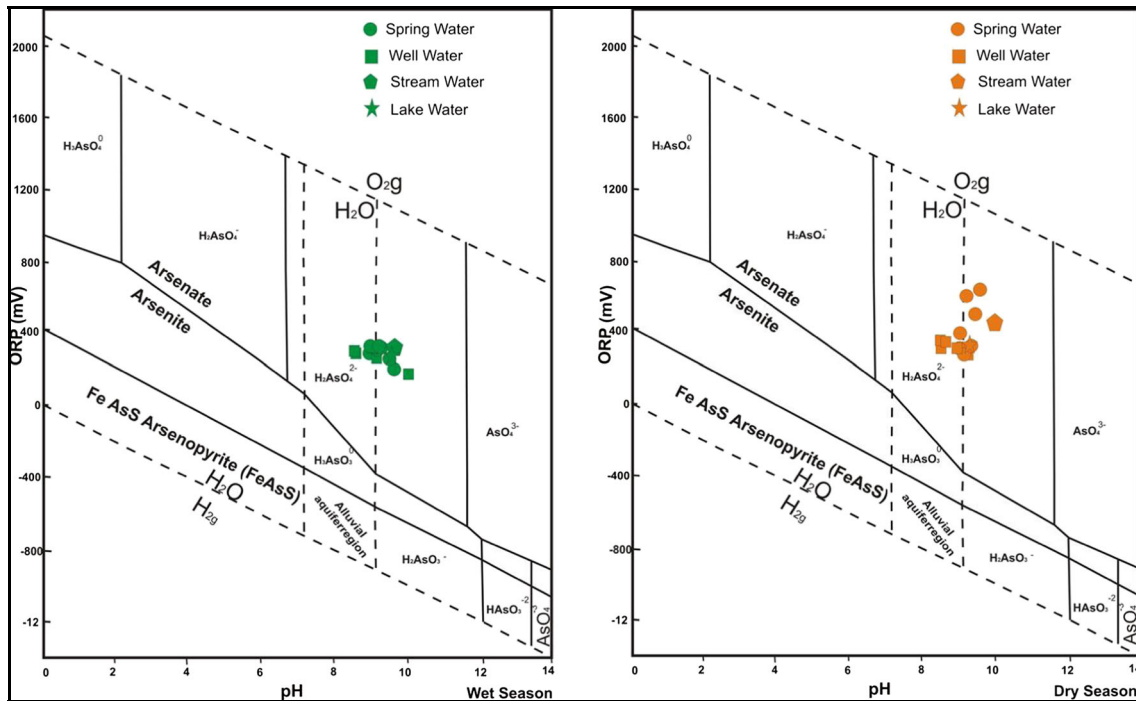


Fig. 5 ORP-pH diagram of aqueous arsenic species in water (Smedley and Kinniburgh 2002)

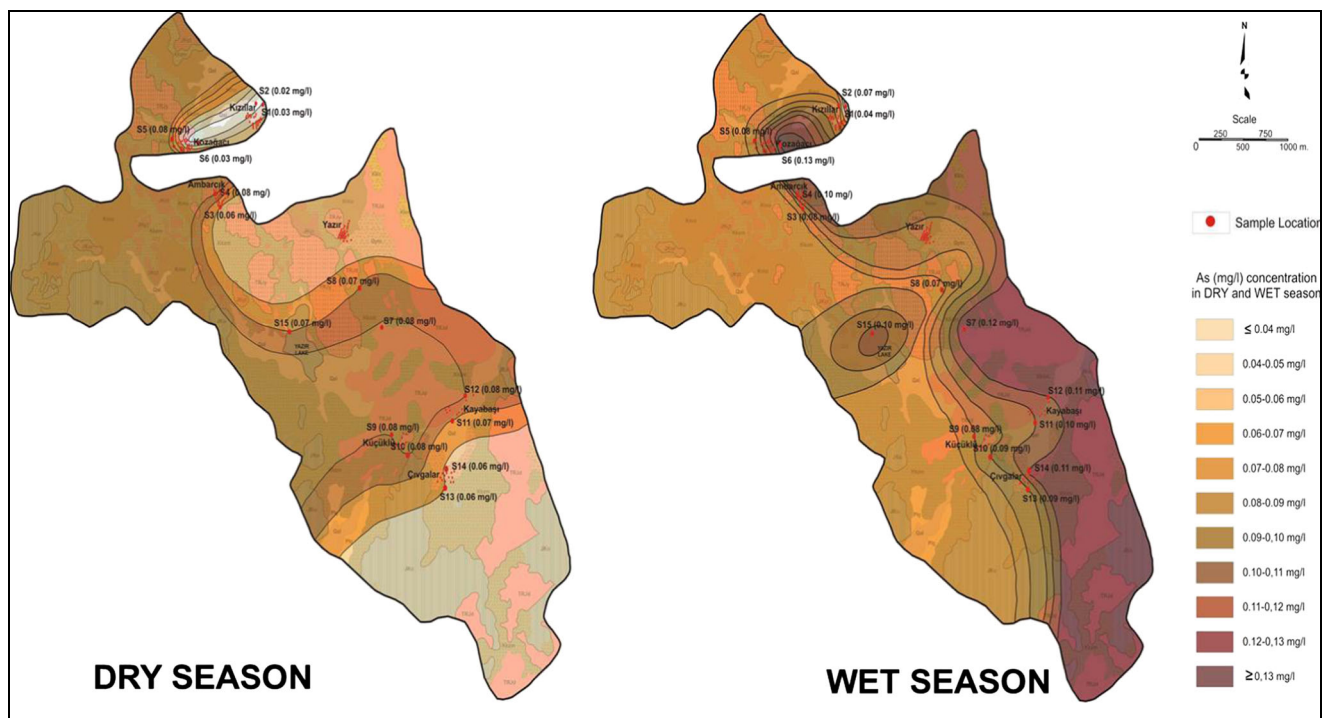


Fig. 6 Seasonal distribution map of the As concentration

(2008) and Turkish drinking water standards (TS 2005). According to results of this study, the As contents of all samples exceeded the permissible values.

The source of As and the mechanism of transport were investigated in this present study. Kızılcaadağ ophiolite and melange are sources of As in water in the study area, and As solubility is governed by hydrological and hydrochemical processes. The Kızılcaadağ ophiolite and melange show wide spread in the study area, and water is obtained from the olistolithic limestones in the ophiolitic mélangé (Fig. 4). This unit consists of heavily serpentinized ophiolitic exposures, and can be considered as a geogenic source of As in water resources. The As concentrations were high in all water samples especially S3 (spring water) and S4 (well water) samples (Ambarcık) in the basin. Furthermore, the positive

correlation between As, Fe, and Mn was determined in the wet season (Table 6). The manganese mineralizations have been detected in the Kızılcaadağ ophiolites around Ambarcık village in previous research (Teker 2010). Especially, Ambarcık manganese mineralization was determined to be hydrogenetic-hydrothermal in the ophiolitic radiolarites and limestone. In addition, according to the rock analyses carried out in the same study, the As concentration in Ambarcık manganese mineralization is between 17.7 and 43.5 ppm (Teker 2010). This research in the study area also supports our results.

There are four basic geochemical reactions for the presence of As in water. These reactions include oxidative and reductive dissolution (McArthur et al. 2001; Nickson et al. 1998), desorption, and concentration by evaporative enrichment (Welch et al. 2000). According to the above description, the

Fig. 7 Seasonal variation of arsenic concentrations in study area waters

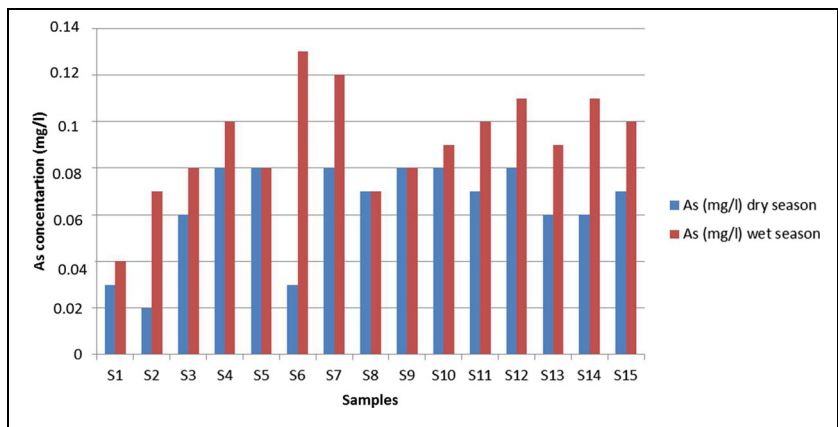
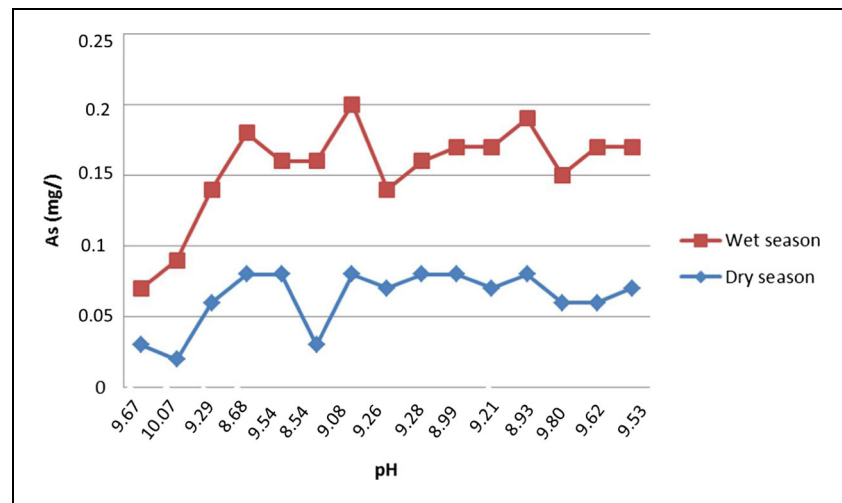


Fig. 8 Seasonal variation in arsenic concentrations due to pH



high concentration of As in the surface water and groundwater is a oxidative and reductive dissolution reaction of Fe and Mn hydroxides within the Kızılcaadağ ophiolitic melange.

Stream and lake waters in the study area may be polluted by geogenic pollution as well as anthropogenic pollution. Agricultural activities are widely used, and there are not enough sewerage infrastructures in the study area. It is known that significant increases in As concentrations in river and lake waters can also occur due to pollution from industrial or sewerage wastes.

As in nature exist in four oxidation states (+5, +3, 0, -3). The most common inorganic As species in water and aqueous media are arsenate (As^{5+}) and arsenite (As^{3+}) (Nguyen et al. 2009). Under oxidizing conditions, arsenate generally predominates, whereas arsenite predominates when conditions become sufficiently reducing. Under pH conditions of groundwater, arsenate exists as negatively charged oxyanions H_2AsO_4^- or HAsO_4^{2-} , and arsenite is present as the uncharged species H_3AsO_3 (Hem 1985). Because of the differences in their charge, the strength of adsorption and desorption reactions between these different As species and solid-phase surfaces in aquifers varies. Also, mobility of As could be affected by the redox reactions involving either aqueous or adsorbed As (Manning and Goldberg 1997). Both ORP and pH are important factors controlling on As speciation in the natural environment.

As species in water samples was determined by using the pH-ORP diagram (Fig. 5). According to this diagram, arsenate ($\text{H}_2\text{AsO}_4^{2-}$) is a predominant As species in the study area. The main forms of As in surface and subsurface waters are known arsenate (As^{5+}) and arsenite (As^{3+}). The increase of pH in water affects the solubility of As in water resources. As can be adsorbed to the colloidal iron oxides in an oxidizing environment with a pH value higher than 9.4 (Gasemi et al. 2014). As can cause pollution of water resources due to oxidation under alkaline conditions. The pH values of the water samples

in the study area ranged from 8.56 to 9.91, and all of the waters were alkaline in terms of pH. This situation creates a potential environment for As contamination in the study area.

Seasonal variation of arsenic concentrations in groundwater

The seasonal variations of As concentrations were investigated in groundwater. For this, As concentrations were compared in dry and wet seasons (Figs. 6 and 7). The slight increase in wet season was observed in As concentrations. It is known that, under acidic conditions, As is in a +5 oxidation state and is usually present in the form of arsenate anions. Arsenates are strong adsorbents and form strong complexes with the surface of ferric oxyhydroxides (Williams 2001; Casiot et al. 2003; Oli'as et al. 2004). This process depends on the pH. The degree of adsorption of arsenates is decreasing in more alkaline conditions (Smedley and Kinniburgh 2002). The slightly higher pH values during wet season can also influence the increase of arsenic in this period, favoring desorption of arsenate ions from the surface of the ferric oxyhydroxides. (Williams 2001; Oli'as et al. 2004). It has been observed that the pH values of the waters in the study area increased during the wet season. The increase in pH concentrations in the wet season are thought to cause an increase in As concentrations (Fig. 8). Furthermore, the concentration of As in water is remaining low in dry season due to strong surface adsorption on ferric oxyhydroxide (Oli'as et al. 2004).

Arsenic health risk assessment

Risk assessment is an effort to identify and measure the effects of various pollutants on human health. It also includes evaluating toxicity data for human exposure to chemicals and estimating the potential exposure levels. There are three main ways of exposure (ingestion, inhalation, and dermal absorption) (Varol and Davraz 2016). Only the ingestion path is

Table 7 Calculated carcinogenic and non-carcinogenic risks of drinking water (dry and wet seasons)

Substance	Water sample no.	C (mg/L)	IR (l/day)	ED (years)	EF days/years	BW (kg)	AT (days)	ADD (mg/kg)	RfD (mg/kg/day) ^a	SF (mg/kg/day) ^a	R _{cancer} ^{-1a}	HQ _{non-cancer}
As (dry season)	S1	0.03	2	15	350	70	30/70 × 365	0.03	3 × 10 ⁻⁴	1.5	0.04	95.89
	S2	0.02	2	15	350	70	30/70 × 365	0.02	3 × 10 ⁻⁴	1.5	0.03	63.93
	S3	0.06	2	15	350	70	30/70 × 365	0.06	3 × 10 ⁻⁴	1.5	0.09	191.78
	S4	0.08	2	15	350	70	30/70 × 365	0.08	3 × 10 ⁻⁴	1.5	0.12	255.71
	S5	0.08	2	15	350	70	30/70 × 365	0.08	3 × 10 ⁻⁴	1.5	0.12	255.71
	S6	0.03	2	15	350	70	30/70 × 365	0.03	3 × 10 ⁻⁴	1.5	0.04	95.89
	S7	0.08	2	15	350	70	30/70 × 365	0.08	3 × 10 ⁻⁴	1.5	0.12	255.71
	S8	0.07	2	15	350	70	30/70 × 365	0.07	3 × 10 ⁻⁴	1.5	0.10	223.74
	S9	0.08	2	15	350	70	30/70 × 365	0.08	3 × 10 ⁻⁴	1.5	0.12	255.71
	S10	0.08	2	15	350	70	30/70 × 365	0.08	3 × 10 ⁻⁴	1.5	0.12	255.71
As (wet season)	S11	0.07	2	15	350	70	30/70 × 365	0.07	3 × 10 ⁻⁴	1.5	0.10	223.74
	S12	0.08	2	15	350	70	30/70 × 365	0.08	3 × 10 ⁻⁴	1.5	0.12	255.71
	S13	0.06	2	15	350	70	30/70 × 365	0.06	3 × 10 ⁻⁴	1.5	0.09	191.78
	S14	0.06	2	15	350	70	30/70 × 365	0.06	3 × 10 ⁻⁴	1.5	0.09	191.78
	S15	0.07	2	15	350	70	30/70 × 365	0.07	3 × 10 ⁻⁴	1.5	0.10	223.74
	S1	0.04	2	15	350	70	30/70 × 365	0.04	3 × 10 ⁻⁴	1.5	0.06	127.85
	S2	0.07	2	15	350	70	30/70 × 365	0.07	3 × 10 ⁻⁴	1.5	0.10	223.74
	S3	0.08	2	15	350	70	30/70 × 365	0.08	3 × 10 ⁻⁴	1.5	0.12	255.71
	S4	0.10	2	15	350	70	30/70 × 365	0.10	3 × 10 ⁻⁴	1.5	0.14	319.63
	S5	0.08	2	15	350	70	30/70 × 365	0.08	3 × 10 ⁻⁴	1.5	0.12	255.71
	S6	0.13	2	15	350	70	30/70 × 365	0.12	3 × 10 ⁻⁴	1.5	0.19	415.53
	S7	0.12	2	15	350	70	30/70 × 365	0.12	3 × 10 ⁻⁴	1.5	0.17	383.56
	S8	0.07	2	15	350	70	30/70 × 365	0.07	3 × 10 ⁻⁴	1.5	0.10	223.74
	S9	0.08	2	15	350	70	30/70 × 365	0.08	3 × 10 ⁻⁴	1.5	0.12	255.71
	S10	0.09	2	15	350	70	30/70 × 365	0.09	3 × 10 ⁻⁴	1.5	0.13	287.67
S11	0.10	2	15	350	70	30/70 × 365	0.10	3 × 10 ⁻⁴	1.5	0.14	319.63	
S12	0.11	2	15	350	70	30/70 × 365	0.11	3 × 10 ⁻⁴	1.5	0.16	351.60	
S13	0.09	2	15	350	70	30/70 × 365	0.09	3 × 10 ⁻⁴	1.5	0.13	287.67	
S14	0.11	2	15	350	70	30/70 × 365	0.11	3 × 10 ⁻⁴	1.5	0.16	351.60	
S15	0.10	2	15	350	70	30/70 × 365	0.10	3 × 10 ⁻⁴	1.5	0.14	319.63	

^aData from USEPA (2005)

taken into account in this study. Ingestion was already reported to be the most important way for exposure to trace elements (Kavcar et al. 2009; Varol and Davraz 2016).

Generally, local people use both groundwater (springs) and surface water (streams) for drinking and domestic purposes in the study area. The As concentration has been used to calculate potential health risk assessment: chronic and carcinogenic effects like average daily dose (ADD), hazard quotient (HQ), and carcinogenic risk (CR). The calculated health risk values were presented in Table 7. The values of ADD were between 0.02 and 0.08 (mg/kg) in dry season and 0.04 and 0.12 (mg/kg) in wet season. The values of $HQ_{\text{non-cancer}}$ were between 63.93 and 255.71 in dry season and 127.85 and 415.53 in wet season. In addition, values of R_{cancer} were between 0.03 and 0.12 in dry season and 0.06 and 0.19 in wet season. Carcinogenic risk is the likelihood of developing any type of cancer against the risk of exposure to a substance that has a lifelong toxic effect on a person. The acceptable or tolerable risk for regulatory purposes is in the range of 10^{-6} to 10^{-4} (USEPA 2000; WHO 2004; Muhammad et al. 2010). Our calculated carcinogenic risk values in the basin exceeded acceptable or tolerable values of the carcinogenic risk of EPA. It indicates that the ingestion of water over a long lifetime could increase the probability of cancer.

Conclusion

Hydrochemical properties and As pollution in water with its effects on human health have been examined in the Yazır Lake wetland basin. In the study, total 30 water samples taken from wells, springs, stream, and lake water were analyzed in dry and wet seasons.

A Piper diagram was used to determine hydrogeochemical types of water samples. Accordingly, Ca-Mg- HCO_3 and Ca- HCO_3 were the dominant water types observed in dry and wet seasons. Generally, the water samples of the study area were dominated by calcium and bicarbonate ions. The chemical composition of water samples was strongly influenced by water-rock interaction. In the study area, the increasing of Na and SO_4 ions in only one sample were determined and this increasing was related to the ophiolites. In addition, the Gibbs diagram was used to determine the mechanism controlling the groundwater geochemistry. According to the diagrams, samples from both seasons fell in rock-dominance zone, suggesting precipitation-induced chemical weathering along with dissolution of rock-forming minerals.

A correlation analysis for T ($^{\circ}\text{C}$), pH, ORP, EC, TDS, NO_3^- , NO_2^{2-} , NH_4^+ , As, Mn, Fe, and major ions was performed with the bivariate method to describe the relation between hydrochemical parameters. In the study area, some groups of species showed moderate to strong correlation ($r < 0.7$). Weathering processes and anthropogenic inputs are

the two main contributors for changing the geochemical composition of the water in the study area. According to statistical analyses, the increases in EC and TDS values of water samples are related to nitrogen derivatives which are originated from agricultural activities. The positive correlation between Na, Fe, SO_4 , and Mg was determined. This suggests that the Mg increase is originated from rock-water interaction as relation to the Kızılcaadağ ophiolitic melange.

When compared with the drinking water guidelines established by WHO and TS, much greater attention should be paid to As ions that occur above the critical values. The positive correlation between As, Fe, and Mn was determined with correlation analysis. In the previous research, Mn mineralization was determined in Kızılcaadağ ophiolitic melange and the As increase was also established in this Mn mineralization with rock analysis. These explanations prove that As, Mn, and Fe increases observed in all water resources in the basin are related to the Kızılcaadağ ophiolitic melange due to water-rock interaction.

As in nature exists in four oxidation states. According to the ORP-pH graphs, As species in the water samples was arsenate. In addition, the results showed that the high concentration of As in groundwaters is a oxidative and reductive dissolution reaction of Fe and Mn hydroxides within the Kızılcaadağ ophiolitic melange. Stream and lake waters in the study area may be polluted by geogenic pollution as well as anthropogenic pollution. The seasonal variation of As concentrations in the study area is related to climatic conditions, rock-water interaction, and anthropogenic effects.

Risk assessment is an attempt to identify and quantify potential risks to human health resulting from exposure to various contaminants. In this study, only the ingestion route which is most important route was taken into consideration. As ion is the major toxic and carcinogenic chemical within drinking water in study area. This indicates that As poses serious health concerns to the local residents via oral intake.

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